Proceedings on the Conference on THE PHYSICS of BREEDING October 19-21, 1959 ANL-6122

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PROCEEDINGS OF

THE CONFERENCE ON THE PHYSICS OF BREEDING

October 19-21, 1959

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Argonne National Laboratory Lemont, Illinois













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PREFACE

The Conference on The Physics of Breeding was sponsored by Argonne National Laboratory in an effort to effect a sophisticated technical exchange of information on this important topic in an atmosphere conducive to free and complete discussion. The program was divided approximately between fast and thermal reactors. For each type an examination of the microscopic data basic to the specification of reactor behavior was scheduled first to be followed by a description of the performance anticipated for the various systems which have received considerable study and are thought to have some potential for breeding. Efforts to obtain international participation resulted in an attendance list sprinkled with foreign guests, but almost all of the formal presentations were made by representatives of American installations.

The proceedings have been edited by the session secretaries namely, Messrs. G. Fischer, C. Kelber, D. Meneghetti, P. Persiani, D. Shaftman and A. B. Smith. A very considerable acknowledgement of their services is hereby made. It is hoped that no major misstatements have occurred during the process of editing the phonic transcription. A major acknowledgement is also made to Mr. A. B. Krisciunas of the ANL Laboratory Director's Office for an excellent job in the administrative arrangements for the conference.

The papers are published in the chronological order of presentation at the conference. Discussion of each paper is printed immediately after the conclusion of its formal presentation.

> David Okrent Program Chairman

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ARGONNE NATIONAL LABORATORY CONFERENCE ON THE PHYSICS OF BREEDING

Introduction by Norman Hilberry, Director

There is so much paper in this day and age that nobody can wade through all of it. You never know whether you have seen the right piece of paper or not.

In the business of creating new ideas, there is simply no substitute for conversation - for questions and answers and for getting together. Sometimes I think that the laboratories have been a bit remiss in not promoting more of the kind of symposia that this meeting represents. I believe that the development of the research and development program, not only in the Commission but on a broad national basis, depends precisely on setting up symposia of this type, where one can have the ease of interchange of ideas upon which new concepts are going to develop. I am sure no more appropriate subject could have been chosen for discussion today than the Physics of Breeding.

I think as time goes on it is becoming more and more evident that we can allow ourselves the luxury of burner-uppers only for those peculiar situations in which the situation itself requires this type of approach. Whether or not the atomic energy business makes any sense as far as central station power is concerned, in my estimation, depends entirely on our success in conquering the breeding program. As far as Argonne is concerned, you all know this stems way back. The discussions on which our program is based began in 1944, if not earlier, with Fermi and Zinn. We have moved ahead, perhaps not as rapidly as we should have upon occasion, but at least we have moved ahead continuously with a certain amount of grim determination to try to prove out this breeding principle as far as the fast breeders are concerned. I don't believe that this has been enough. The uranium-plutonium cycle is part of the picture; but the uranium-233thorium cycle is just as much a part of the picture. If one is going to take the long look, then it is clearly necessary that we establish both of these as effective instruments of energy production.

At this Laboratory, we have taken one further step because we have the peculiar idea that breeding only makes sense if it is a complete breeding cycle from reactor back into the reactor. We feel that the chemical processing and the metallurgical fabrication steps are just as important as the actual breeding within the reactor itself. Achieving a large breeding factor in a reactor and a short doubling time is a big help. However, I think we have to keep an eye on the entire cycle as well as to make maximum use of the possibilities, potentialities which the nuclei themselves offer. So, I am hoping that this meeting will turn out to be as useful and inspiring as it is possible for us to make it.

BURNING THE ROCKS

Alvin M. Weinberg

Introductory Remarks and Paper Read by H. G. MacPherson Oak Ridge National Laboratory Oak Ridge, Tennessee

Dr. Weinberg would have dearly loved to be here. As many of you know, he is greatly interested in breeder reactors. I would like to repeat some of his ideas with a small amount of editing to suit the present audience. Alvin Weinberg in turn would not claim that the thoughts presented here are original. Generally, all have been presented in different places. The assemblance of these into one talk seems to point up the longrange importance of breeding in a peculiarly potent way.

My purpose in these remarks is to speculate on the role of energy in the "Asymptotic State of Humanity" - that is, the state toward which we are moving, inexorably, because man's urge to multiply is limitless, whereas his resources are finite. In my talk I draw very heavily from many authors - in particular, Palmer Putnam,⁽¹⁾ Hans Thirring⁽²⁾ and, above all, Harrison Brown,⁽³⁾ who has given much ingenious thought to the matters which I discuss. I choose to dwell on energy, because the character of the asymptotic state of mankind - whether it will be a bare existence or a passably abundant life - will depend centrally on our capturing an inexhaustible supply of energy, either by learning how to burn the seas (fusion) or to burn the rocks (fission) or to trap the energy of the sun in a practical way.

That the asymptotic state of humanity depends, for its shape, on energy has been stated perhaps most strikingly by Sir Charles Darwin,⁽⁴⁾ who points out that, if the human doubling time of about 100 years persists, then in the year 2959 there will be about 2.7 x 10^{12} persons on earth; in 3959, 2.7 x 10^{15} . In fact, at this rate the mass of humanity would equal the mass of the earth by about 6500 AD, which is, of course, absurd. Evidently, one way or another, the population of the earth will stabilize. For my purpose I shall assume a stabilized human population of $7 \ge 10^9$ - the figure suggested by Brown, Bonner, and Weir in their much less ambitious, but more factual, The Next Hundred Years.⁽⁵⁾ But no matter what asymptotic population one chooses, the demand for energy will continually increase. For, as our natural resources dwindle, as we are forced to extract metals from ores of lower grade, or water from the sea, or liquid fuel from carbonates and water, we shall have to pay more and more in energy simply to do what we have been doing, let alone to improve our lot. Eventually, as Harrison Brown has stressed, mankind will have to make do with only four basic raw materials: the sea, the rocks (of average composition since

true ores will have been exhausted), the air, and the sun. (If we equate the sun to fire, these are essentially Aristotle's four elements!) The question really is not whether we shall reach this state - it is merely when we shall reach it.

Professor Brown and his associates have drawn up an energy balance sheet for such an asymptotic society of seven billion people who must eventually subsist on the sea, the rocks, the air, and the sun (Table I). The total projected yearly energy consumption is 1.9×10^{18} Btu or 6.3×10^7 Mw years heat, i.e., the equivalent of 70×10^9 tons of coal per year, or 10 tons per person per year. This is about 18 times the present equivalent energy input of 0.35×10^7 Mwy heat, i.e., the equivalent of four billion tons of coal. At this ultimate rate, the present fossil fuel reserves, of perhaps 2400 $\times 10^9$ tons, would hardly last 35 years. The nuclear component of the yearly energy input, according to Brown's estimate, amounts to about 45 $\times 10^9$ tons of coal equivalent or 4×10^7 Mwy heat.

TABLE I

Source	Equivalent Metric Tons	Equivalent Heat Energy	
· ·	of Coal (Billions)	10 ¹⁸ Btu	Heat (Mwy)
Solar energy (for 2/3 of space heating)	15.6	0.42	140 x 10 ⁵
Hydroelectricity	4.2	0.10	38×10^5
Wood for lumber and paper	2.7	0.07	24×10^{5}
Wood for conversion to liquid fuels and chemicals	2.3	0.06	21 x 10 ⁵
Liquid fuels and "petro" chemicals produced via nuclear energy	10.0	0.27	90×10^5
Nuclear electricity	35.2	0.96	320×10^{5}
	70.0	1.88	633×10^{5}

Asymptotic Yearly Energy Input Pattern* (World Population of 7 x 10⁹ Persons)

*From Brown, Bonner, and Weir, Ref. 5, p. 113

In this asymptotic state one can visualize the energy economy being divided into three major sectors: sunlight, primary nuclear sources, and energy converters. It is certain that sunlight will be used to produce food and, according to Brown, for much of our space heating. I shall consider later whether it will also become a primary source. The primary nuclear sources - fission and fusion - probably will be centered in great power plants, possibly, on the average, 20 times larger than the largest present-day coal-fired steam plants, since nuclear plants are so much less expensive in large size than in small. These plants would supply energy for direct use; they would also be used to supply energy for conversion to more convenient form, or for chemical reduction. For example, the reduction of iron oxide to metallic iron, which now uses about one-fourth of our coal, (6) can also be done either directly by electrolysis or a little less directly by electrolysis of water and reduction of FeO with the hydrogen which is produced. If the energy cost is 1/2 cent/electric kilowatt hour (kwhe), the additional energy charge would amount to only 1/2 cent/pound of iron. Similar considerations apply to all other metals: they appear in nature in oxidized form, and electricity can be used to reduce the ores to metals.

The primary energy source can be used to provide small-scale mobile energy - in principle, either by electrical storage systems or by chemical storage systems. An example of a simple chemical storage system would be electrolytically produced hydrogen; this could be used in the production of liquid fuel hydrocarbons from carbonate rocks even after our coal <u>per se</u> is gone. The energy cost is rather high, but not outside the realm of ultimate feasibility.

We thus see that an asymptotic state of civilization, stabilized at, say, a population of 7×10^9 , can be based upon the rocks, the seas, the air, and the sun - provided only that we have available a primary energy source and that we have worked out good methods for converting energy into convenient packages. This search for new primary energy sources - and for new energy converters - has become an enormous scientific frontier.

There is, of course, the possibility that the sun's energy can be used as the primary source. In the projected energy economy it represents 22 per cent of the total input, in addition to its use for production of food and wood. But the diluteness of the sun's energy and its unpredictability militate against its use as a primary source in large power stations. The solar energy striking the earth is 1.7×10^{14} kw and to produce all of the energy required in our energy balance would require collectors occupying about 35,000 square miles, assuming an efficiency of collection of 100 per cent and of conversion to electricity of 25 per cent. Actually the efficiency of collection and conversion to electricity, according to Palmer Putnam,(7)is only about 7 per cent, so that the total required area may be as high as 10^5 square miles.⁽⁸⁾ This is perhaps not entirely out of the question, though it does seem extremely unwieldy. Thus to quote Putnam, "The direct collection of solar energy on a vast scale by myriads of tracking mirrors, thermocouples, or other devices, its overnight storage, its conversion to transportable electricity, and its delivery at low cost from

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Arizona to Pittsburgh or from the Sahara to the Midlands appear remote in the light of what we know today." At the presently estimated capital cost of \$1000/kw,⁽⁹⁾ the ultimate electrical energy input would cost about four trillion dollars - a large sum, but in the ultimate span of human history not impossible (World War II was estimated to have cost $l\frac{1}{2}$ trillion dollars). A more concentrated, long-term energy source based on either the rocks or the seas thus seems to be extremely worthwhile, if not absolutely essential.

Sea-burning and Rock-burning

Where, then, do we stand in our efforts to burn the sea and to burn the rocks? First, I consider the availability of the raw materials. In the case of fusion based on a D-D reaction, the raw material is found almost entirely in the sea. Assuming that if the D-D reaction goes, then so will the D-T and D-He³, we have the following overall energy and material balance:

$$3D \rightarrow He^4 + p + n + 21.6$$
 Mev; 4 Megawatt days of heat/g of D. (1)

If the D-D and D-T reactions can be made to go, but, because of the higher Coulomb barrier, the temperatures required for the $D-He^3$ reaction are not achieved, the balance is

$$2.5D \rightarrow \frac{1}{2}He^3 + \frac{1}{2}He^4 + n + \frac{1}{2}p + 12.4$$
 Mev; 2.8 Mwd heat/g of D. (2)

If only the D-T process can be made to go, Li^6 is also a basic raw material, and the overall balance is

$$D + Li^6 - 2He^4 + 22.3 \text{ Mev}; 4 \text{ Mwd heat/g of } Li^6.$$
 (3)

In these reactions, the neutrons, He^3 , and tritium produced in the intervening reactions are used up again - they act as catalysts much as the carbon acts as a catalyst in the carbon cycle.

By comparison, the fission reaction is

$$U + n \rightarrow fission products + 200 Mev; 0.9 Mwd heat/g of U.$$
 (4)

The surprising result is that per gram of raw material, fission gives as much as 1/3 to 1/5 the energy of the deuterium-tritium-He³ cycle (Equations 1 and 2).

The amounts of deuterium, Li^6 , and U + Th contained in the seas and in the earth's crust, together with their energy content, and the length of time they will last at the asymptotic rate of 4×10^7 Mw heat, are shown in Table II. In making this table, I assumed reaction (1) for the deuterium, and reaction (3) for the lithium.

TABLE II

Source	ppm	Total Mass (Tons x 10 ⁻¹³)	Energy Content (Megawatt Days of Heat x 10 ⁻²⁰)	Will Last at 4×10^7 Mw (Heat) Rate (yr x 10^{-10})
Sea	33	6	2.4	1.7
Crust	5	21	8	5.4
Crust	4	16	1.5	1.0
Crust	12	50	4.5	3
Crust	16	66	6	4
	Source Sea Crust Crust Crust Crust	Source ppm Sea 33 Crust 5 Crust 4 Crust 12 Crust 16	SourceppmTotal Mass (Tons x 10^{-13})Sea336Crust521Crust416Crust1250Crust1666	SourceppmTotal Mass (Tons x 10^{-13})Energy Content (Megawatt Days of Heat x 10^{-20})Sea336 2.4 Crust5218Crust4161.5Crust12504.5Crust16666

Amounts of Deuterium, Li⁶, U, and Th on Earth*

*The basic data in this table are from B. Mason, <u>Principles of</u> Geochemistry, John Wiley, New York (1952) p. 41.

Before assuming, from Table II, that either sea-burning or rockburning would forever fulfill our energy requirements (the solar system is hardly expected to last longer than 10^{10} years), we must ascertain that less energy is required to extract the raw materials D, Li⁶, and U + Th from their asymptotic natural environments than is returned by burning these fuels. In the case of deuterium, the balance is clearly very favorable. Perhaps it is easiest to see in terms of monetary cost of extracting deuterium from water. The present cost of deuterium is about \$28/1b of D₂O or 30 cents/gram of deuterium. If one gram of deuterium is burned, by reaction (2), say, 3 Mwd of heat or about 22,500 kwh of electricity, worth about \$100, are produced. The fuel cost of the deuterium on this basis is less than 0.013 mill/kwhe, which is almost, but not quite, negligible.

With respect to U + Th, the situation is also favorable, provided we burn all the U and Th, not just U^{235} . About 1/5 of the U + Th, or 3 grams per ton, is contained in rather easily leachable portions of the granites, according to Brown and Silver.⁽¹⁰⁾ The energy content of this "easily" recoverable U + Th is equivalent to about 10 tons of coal or 3 Mwd heat/ton of granite. The energy required to recover this 3 grams/ton of U + Th is estimated by Brown and Silver to be equivalent to that from 25-50 lb of coal, as seen in Table III.

Brown estimates the asymptotic cost of treating one ton of granite to be from 1.00-2.25 - this amounts to about 30-80 cents/gram of U + Th or 0.05-0.12 mill/kwhe fuel burnup cost, assuming that all of the extractable U + Th can be burned by the process of breeding. The burnup cost is relatively small even if, as suggested by Keith Brown, ⁽¹¹⁾ the asymptotic cost per gram of U + Th is as high as 1.00-3.00/gram - in that case (assuming the extreme of 3.00/gm) the fuel burnup cost would be 0.5 mill/kwhe, which is still very low. The situation is unfavorable if only the U²³⁵ is burned; in that case the energy recovery is only 1/300th as much and this would hardly pay for extracting the U + Th.

TABLE III

Quarrying, crushing, hauling	7-9 kwh/ton
Acids	10-25 kwh/ton
Other direct charges (water pumping, etc.)	l kwh/ton
Hidden costs	0.7-2.3 kwh/ton
	19-37 kwh/ton 25-48 lb coal

Energy Required to Process Granite*

*Brown and Silver, Ref. 10.

The total amount of energy "practically" available from U + Th in the rocks is of course a good deal less than that given in Table II. We ought not to count the parts of the crust under the seas, nor the material more than, say, 3 km below the surface, nor the granites which carry a very great overburden of sediment. On the other hand, since the energy balance is so favorable, one could mine rocks with even 0.3 gram/ton U + Th and still get some 20 times more energy than is required to extract the fissionable material. One therefore cannot escape the impression that the extractable reserve of fissionable material is large enough to sustain humanity for the indefinite future.

Just how large a mining operation would be required to maintain an energy output of 4×10^7 Mw heat? Since one gram of fissionable material burned each day would maintain a heat rate of 1 megawatt, the total U + Th burned per day would be about 40 tons. To obtain this amount of fissionable material would require the mining of about 10^7 tons of rock per day. This may be compared with the world's daily production of coal and lignite, which in 1953 was 0.6×10^7 tons.⁽¹²⁾ Thus the whole mining operation required to sustain the asymptotic energy economy would be on about the same scale as the mining operation which now sustains our much smaller fossil fuel-based energy economy.

I know of no studies relating to recovery of low-grade Li ores. I should suppose that, if anything, Li would be easier to extract than U or Th, and that the asymptotic fuel cost of Li, just as the asymptotic fuel cost of U + Th, will always be negligible. However, the total amount of Li recoverable would probably be of the same order as the total amount of U + Th which is recoverable; if we must rely on the $D + Li^6$ cycle, then the amount of Li would probably limit the total energy recoverable from deuterium. In this case the extraction of lithium from the rocks would involve a mining operation of the same order as the extraction of the residual fissionable materials.

Problems of Sea-burning

The essential point of the foregoing remarks is that either seaburning or rock-burning could in principle be made the primary asymptotic energy source for the rest of mankind's history, and the cost of the energy produced could be at least within striking distance of the range of today's energy costs for a time which is very long compared to the present span of human history. The advantage of sea-burning, therefore, is not, as is often assumed, that deuterium is the only essentially inexhaustible fuel. The advantages are seen to be rather less fundamental - perhaps most important is that fusion is a relatively cleaner process than fission. The radioactive wastes associated with deuterium reactors ought to be much less troublesome than those associated with uranium reactors. Whether this is a crucial advantage is certainly very difficult to say at present.

Granted that achievement of sea-burning would represent a major advance - comparable to the discovery of fission - there remains the question of where we now stand in this quest for a successful deuterium reactor.

The extraordinary difficulty of confining the plasma may be judged by considering the pressure in the plasma. At a density of 10^{15} nuclei/cm³ and a temperature of 40,000 ev (4 x 10⁸ °K), which are the ignition conditions for the D-D reaction, the plasma pressure (P_p = nkT) is 60 atmospheres; this is a pressure which is usually held by stout steel walls - what must be done in Sherwood is to hold this pressure by magnetic lines of force!

It would be terribly premature to say that Sherwood - i.e., seaburning - is impossible, especially since the mirror geometry (on which ORNL's and USSR's OGRA is based) has thus far shown no instability. On the other hand, it would be equally incorrect to assume that mankind's future energy supply is assured on the basis of what we now know about the problem of sea-burning. The fair-sized experimental program being pursued in the U.S. (amounting to about $$38 \times 10^6$ in the next fiscal year) and the apparently comparable program in the USSR are in my view well justified; yet it would be a gross error if our effort at sea-burning were to divert us from a full-fledged effort aimed at the much more imminent rock-burning.

Problems of Rock-burning

The problems of rock-burning are of an entirely different order than are the problems of sea-burning. We certainly have not shown that we can ever burn any fraction of the deuterium; burning U^{235} , on the other hand, is rather a routine process. But, in the asymptotic state, burning U^{235} is not sufficient; in order to make the extraction of U + Th from granites energetically feasible, we must burn considerably more than the U^{235} . Beyond this, in order to make the ultimate fuel burnup costs even reasonably low, say less than 1 mill/kwh, we must burn not less than about 1/10 of the U + Th which we assume to be available at the previously quoted asymptotic figure of 30-80 cents/gram, that is, we must burn about 60 times as much U^{238} and Th²³² as there is initial U^{235} .

In order to burn more than the initial reservoir of U^{235} it is necessary to breed.

From the very long-term point of view which we are adopting here, all that is necessary to burn all the uranium and thorium is to achieve a breeding ratio of unity. However, the breeding ratio must refer to all the fissionable material burned in the whole energy system. Since there will undoubtedly always be some nuclear plants which, for compactness, must forego any breeding, it will be necessary to make up for these plants with plants which produce more fissionable material than they burn. Thus it seems inescapable that the solution to the ultimate energy problem by way of rock-burning depends on reducing to practice the nuclear breeding process, i.e., making practical breeder reactors with reasonably short doubling times, of the order of 10 years.

Fast Breeding and Thermal Breeding

Breeding cycles can in principle be based on either uranium as the raw material or on thorium as the raw material. From the asymptotic standpoint, thorium is preferable since it is three times as abundant and therefore should be three times as cheap as uranium. On the other hand, because of its more favorable geochemistry, uranium in readily workable deposits seems to be three or four times as abundant as thorium and so, in the short run, uranium breeding may be preferable. Serious work on both breeding cycles is now being pursued.

Although the problems of breeding, of course, are the subject of details of this conference I would like to mention one aspect of breeding that applies to breeders in this asymptotic state of society. This concerns the matter of inventory of fertile material. At present the fast breeder EBR-II being built by Argonne National Laboratory is rated at about 300 kwe/ton of natural uranium, including both the U²³⁸ in the blanket and the natural uranium needed to supply an initial charge of U²³⁵ for the core.

If the asymptotic price is $1-\frac{3}{gm}$, this amounts to $3000-\frac{9000}{kwe}$ for the installed fuel. At this price fast neutron breeding would begin to be as expensive as solar energy, and evidently great improvements would be needed. At such high inventory costs, solar energy indeed becomes a very serious competitor.⁽¹³⁾

Two factors make the situation much more hopeful, however. First, the rating of 300 kwe/ton is surely much lower than will be ultimately achieved, especially since the fueling of the core will eventually be done with bred Pu^{239} , not with U^{235} extracted from natural uranium. Second, and possibly more important, there is probably enough low-cost uranium and thorium available to start the asymptotic energy system at reasonable cost; the very expensive material from the granite would be needed only as make-up for fissionable material which has been burned. As make-up, \$3.00/gram uranium would add only 0.5 mill/kwhe to the cost of electricity. Even at the very low rating of 300 kwe/ton, the amount of uranium and thorium required as initial inventory for Brown's asymptotic nuclear energy system (4 x 10^{10} kwh) is only about 30 x 10^{6} tons. This may be compared with recent estimated potential reserves of uranium and thorium available at $30-50/1b - 20 \times 10^6$ tons for uranium, and 5×10^6 tons for thorium. The inventory cost, even at $\frac{50}{lb}$ (10 cents per gram), thus amounts to about \$300/kwe, which is a serious cost but certainly not impossible from the very long-term standpoint. Rather, I consider it remarkable that we see at hand a way of starting our asymptotic energy system with materials that are even now practically available, and that we can keep the system supplied with fuel for essentially all time by means of a mining operation only somewhat larger than the present coal mining operation of the world!

In the thermal breeding system based on the thorium- U^{233} cycle, it seems likely that an output of about 15,000 kwe/ton of thorium can be achieved fairly readily. In this case, Brown's entire asymptotic nuclear system could be fueled with only about 7 million tons of thorium initially, an amount which is very likely available at this figure of 10 cents/gram of thorium. At this price, inventory charge is only about \$70 per kilowatt. Thus, if thermal breeding in the thorium cycle in systems of low holdup can be achieved, this system can serve as the asymptotic energy source for mankind.

Do We Have a Responsibility to Future Generations?

It is fair to ask why this generation should have any particular responsibility to generations many, many years hence - why should we bother to develop an asymptotic energy source? I think there are several reasons, some practical, others moral, why we should pursue aggressively the ultimate energy source. First the practical reason: it is merely that asymptotic breeder reactors could be as economical as any other reactor once they are fully developed. Thus the motivation for pursuing the breeder systems, as compared to other reactor systems, at present has an economic base - even in the current economic framework.

As for the moral reasons, I see at least two. The March 1959 issue of "Population Bulletin" puts it aptly: the next twenty-five years may see the world's population rise from 2.5×10^9 to 4×10^9 . "....we should do well to ponder the significance of this development in terms of the destiny of our species.

"These next twenty-five years form part of a process which began some 200,000 years ago and which is about to culminate in man's full possession of the earth.

"The growth of world population during the next twenty-five years, therefore, has an importance which transcends economic and social considerations. It is at the very heart of the problem of our existence." $^{(14)}$

It is the lot of our generation to see clearly that the threshold into the asymptotic population state will surely be crossed - in this sense it is our generation or the next which probably will first witness the culmination of man's history on earth. I suppose each person has his own personal reaction to this knowledge; mine is that I would somehow feel more comfortable if, as a member of the generation which first sees the asymptotic population state approached, I could also leave to future generations the means to live relatively abundantly in this asymptotic state.

Finally, there is the broad political implication of this vision of an asymptotic, energy-abundant world. Should we succeed in supplying energy really cheaply from the rocks or, with good luck, from the seas, on as vast a scale as I contemplate, then the problem of have-not nations ought to become much less acute than it is now. Much of what countries do internationally nowadays is intended to forestall future actions of neighbors beset with population and raw materials problems. But everyone has granite, and air, and sea, and sun. One would hope that solving the problem of living relatively abundantly with only these raw materials would help to dispel these historic causes for strife among men and that, in the wake of such development, mankind could turn its energies to those peaceful pursuits which are the true expression of the human spirit.

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- 8. Thirring, <u>op. cit.</u>, p. 275. This figure agrees with Thirring's estimate that two square miles of collector would be required to produce 10⁹ kwh/year of electricity from the sun.
- 9. F. Daniels and J. A. Duffie, <u>Solar Energy Research</u>, p. 9, University of Wisconsin Press, Madison (1955). <u>Ibid</u>, p. 89. H. C. Hottel estimates a cost of \$1130/kw or more but in making this estimate assumes that Carnot efficiency rather than Rankine efficiency is available. If the costs of transmission (say from the Sahara to London) and storage are included, this figure could be much higher perhaps \$2000/kw or more.
- H. Brown and L. T. Silver, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, VIII, 129, United Nations, New York (1956).
- Keith Brown, private communication. If these higher estimates prove correct, the burnup cost is still small; however, the inventory costs would become very troublesome unless the inventory is derived from relatively abundant sources of U + Th.
- Department of Economic and Social Affairs, United Nations, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, I, 3, United Nations, New York (1956).

- 13. Perhaps it should be pointed out that if only the D-T reaction can be made to go, so that Li⁶ must be used as a blanket material in fusion reactors, the same problem of high inventory costs will ultimately face fusion, as well as fission, reactors.
- 14. "Population Bulletin," XV, 21-22, Washington, D.C. (March 1959).

Discussion of Paper Presented by Mr. MacPherson

MR. OKRENT:

Thank you, Dr. MacPherson, for the very interesting talk. Are there any comments from the floor?

MR. CRANBERG:

I would just like to remark that there is a contingent possibility which represents a combination of the burning of the rocks and the oceans which has received practically no public attention. The basic idea can be described as a plasma-target-fusion machine with a uranium blanket. The basic idea is that when one accelerates deuterons into a hot tritium plasma, 14-Mev neutrons are produced. These then produce fast fission in the uranium blanket. The raw materials for such a cycle are deuterium, lithium-6, and uranium or thorium. One of the distinctive features of this system is that the temperature which has to be reached in the plasma to make this system energy reproductive is in the neighborhood of a few electron volts, in contrast to the 40 kv which is necessary for the self-perpetuating thermonuclear reaction.

MR. OKRENT:

Would you like to volunteer, Dr. Spinrad?

MR. SPINRAD:

I would like to ask a question about one thing that seems to be an apparent contradiction in this analysis. That is the assumption of an asymptotic state of energy production and the further assumption that at this asymptotic state one will require breeding. In fact, it seems to me that if you are talking about an asymptotic state, you are talking about a state in which one has a sufficient supply of energy sources, machines and reactors. In this situation one requires only one-to-one conversion, so that one can put in feed materials and extract fission products and that is that. The breeding is required during the preparatory period.

MR. MacPHERSON:

The answer, which is at least mentioned in this paper, is that in this asymptotic state there will be uses for fission materials which are outside of breeders. An example is that of mobile power. Obviously, this could come from a secondary chemical source. However, today we don't seem content with chemical sources for mobile power and one can anticipate that in the future man will find need for small devices using fissionable materials. The point is well taken, however, it is a matter of speculation as to how important the additional fissionable material will be at that time.

MR. GREEBLER:

I didn't understand the time scale here. Just how important is it that we go all out on a breeding program right now as compared with starting with fifty years from now? What would this mean to us in terms of cost of neutrons, for example ?

MR. MacPHERSON:

That's a specific question and I am not sure that I can give a very specific answer. The thing which is clearest is the population trend, and even this cannot be extrapolated indefinitely. At present the world's population is growing at the rate of about one per cent a year. If this were to persist for a hundred years we would have the seven billion people on earth.

It is really with great difficulty that the food supplies of the earth, without a great deal of intensive effort in developing such things as algae sources, would supply these seven billion people. As to the usage of energy, as far as I know the best review of this was in the publication that Zinn and his associates got out on reactors. I interpret from this that, once nuclear power becomes cheaper than power from fossil fuel, there will be a very rapid increase in the nuclear power industry and that two or three decades of this rapid expansion could see a nuclear power industry which was large enough so that the reactors then in existence in their normal life time would use up our readily available supplies of uranium and thorium. I would like to point out that something of the order of three or four decades from now we will need reactors which are able to operate on a much higher cost uranium supply.

ON THE DEFINITION OF BREEDING

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In discussions of breeding of fissile material, the question of defining the subject occupies an inordinate role. In the hope of eliminating misunderstanding, we have therefore inserted this discussion at an early point in the program of this Conference. The fanciful titles given to the various definitions are mnemonic devides, and are to be treated as such.

Fool's Breeding

I know that nobody here is so foolish, but the initial conversion ratio (ICR) has been often used to define breeding. Since I have some responsibility for the ICR as a parameter, I might say, right off, that it is intended to be used only as a scoping index for calculations of reactivity change. Nevertheless, whenever ICR is >1, it is sometimes confused with breeding; that is to say,

$$BR (Fools) = \left[\begin{array}{c} Atoms new and fissionable made \\ Atoms original and fissionable destroyed \end{array} \right] C, t = 0 ,$$
(1)

where the subscripts refer to criticality and a time close enough to zero so that one gets over xenon and samarium jitters.

To illustrate why I have called this a foolish definition, I can merely point to the imaginary experiment of constructing a large, high moderator temperature, heavy water reactor fueled with U^{235} and with very low parasitic absorption. In principle, given U^{238} fertile material, one can make use of the fast effect to insure ICR >1. This looks like breeding, but it isn't, and for two reasons. The first reason is that the fissionable material made, Pu^{239} , isn't the same as the one, U^{235} , which is destroyed. The second follows from the first and has to do with the fact that, even if one removes all fission product poisons and hoards any Pu^{239} which is not needed for criticality, ultimately the reactor will run down. The Pu^{239} and other isotopes of Pu will simply not sustain a chain reaction unless a small feed of fresh fissionable is constantly injected.

For a counter example, I can propose the same reactor with Th^{232} as fertile material, and an ICR of 0.95. In spite of the need of adding still more U^{235} in the early states of the life cycle of this reactor, it will ultimately become self-sufficient and produce an excess of (bred) uranium with a feed consisting solely of Th^{232} .

From these examples, we can adduce two axioms of breeding, and we need still one more definition.

<u>Axiom 1</u>. The term "breeding" has a meaning only when the fissionable material produced is the same as the fissionable material destroyed.

 $\underline{Axiom 2}$. Breeding is a process definable only for a fuel cycle at a steady state.

<u>Definition 1.</u> "Steady state" for the purpose of evaluating breeding is a long-time average. Steady-state feed injection rate is the limit for increasingly great time of the ratio of total feed injected to time. Steady-state fuel removal rate is the limit of (total fuel withdrawn/time). Thus

Feed rate $= \frac{\lim_{t \to \infty} \frac{\text{Total feed added}}{t} \quad (a)}{t}$ Fuel removal rate $= \frac{\lim_{t \to \infty} \frac{\text{Total fuel removed}}{t} \quad (b)}{t} \quad (2)$ Inventory $= \frac{\lim_{t \to \infty} \frac{\int_{0}^{t} (\text{fuel in system}) \, dt}{t} \quad (c)}{t}$

This definition of steady state allows us to consider reactors (such as most heterogeneous types) which require and deliver fuel in a pulsed fashion. The corollaries (2) of the definition are clear as to feed rate and fuel removal rate, and inventory must be similarly defined. For the moment, I will ignore difficulties caused by leaving the word "system" undefined in the description of inventory. I wish, however, to point out that the inventory of the reactor is not necessarily the same as investment. In the case of "inventory," I am defining a property of the steady-state system. I now define "investment," which is a property of the method selected to achieve the steady state.

Definition 2. The investment of a system is the amount of fissionable material fed into the system in order to build up inventory. The investment need not be the same material as the inventory. The investment may indeed be a mixture, but in describing it, we must remember that one orange and one apple do not make two orples. If the invested material is not the inventory material, the investment may be more or less than the inventory.

Freshman Chemist's Breeding

I have been tempted to give this concept the alternate title of "Price Schedule" breeding, because it is based on the premise, still to be found in some of the price schedules, that we cannot distinguish one isotope from another. This definition may be a useful concept, indeed, in simplifying chemical and metallurgical process thinking. I therefore include it, even though it may give us reactor physicists some pain. Briefly, the method consists of working on the assumption that all isotopes of a given element -- fissionable, fertile, or scrap -- are indistinguishable. Therefore, at steady state, we have (say) U as the reactor fuel and reactor product. We may characterize this fuel by a value of $\overline{\sigma}_a$, giving the total absorption cross section per mean atom; $\overline{\sigma}_d$ as the cross section for destruction (i.e., chemical change) per mean atom, ν_c as the neutron production per atom chemically changed, η_c as the neutron production per neutron destroyed, E_c as the energy production per atom destroyed; and θ as the atoms destroyed/neutron destroyed ($\theta = \overline{\sigma}_d/\overline{\sigma}_a$).

The accompanying chart (Table I) illustrates how a mixture of isotopes as may be found at steady state in a reactor can be summed up to yield the gross properties of the fissionable element. Please do not criticize input data or roundoff, as this is an invented case, and the choice of invention is privileged. You may note that we have two modes of chemical destruction fission of the appropriate isotopes, and neutron absorption by the last element in the chain. In this model, both types of destruction are called "fission" and the energy per "fission" and neutrons per "fission" are accordingly reduced. Note also that some of the radiative capture is treated as a spooky neutron loss, without equivalent destruction of the element

TABLE I

	U ²³³	U ^{2 34}	U ^{2 35}	U ²³⁶	Total
σ (b)*	600	100	700	30	
$N \frac{atoms 1sotope}{atom element}$	0 519	0 311	0 045	0 125	1 000
N ^o a (b)	311 4	31 1	31 5	375	378
α *	0 10	œ	020	~	
η^*	2.3	0	2 0 5	0	
ν^*	2 53	0	2 46	0	
σ _f (b)	545 45	0	583 33	0	
σ _c (b)	54 54	100	116 67	30	
σ _d (b)	545 45	0	583 33	30	
$\nu \sigma_{\rm f} = \eta \sigma_{\rm a}$	1380	0	1435	0	
Νσ _d (b)	283	0	2.63	3.75	289
No _f (b)	283	0	3	0	286
$\eta N\sigma_a = \nu N\sigma_f (b)$	716 2	0	632	0	779
$N\sigma_a/N\sigma_d$ = neutrons produced/atom destroyed					2.69
$N\sigma_a/N\sigma_d$ = neutrons absorbed/atom destroyed					1.31
Fissions/atom destroyed					0.99
Atoms destroyed/neutron destroyed					0.76
Neutrons produced/neutrons destroyed					2 05
Fission/neutron destroyed					0 75
$E_c @ E_f = 200 Mev$					198 Mev

Illustrative Example of Gross Properties of U in a Th Breeder

*Assumed Values

25

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Obviously, now

BR (Freshman Chemist) =

\begin{bmatrix} Atoms of fissile element produced \\ Atoms of fissile element destroyed \end{bmatrix} steady state, critical (3)
```

I have omitted from Table I any indication of maximum breeding ratio. One may subtract 1.31 from 2.69 to get a number, but this number may be incompatible with the operation of the reactor. This is because the isotopic ratios (I can now mention that word) are themselves determined by the process of breeding.

Steady State

Equilibrium is a situation of stasis: nothing happens unless a push is applied. Steady state is a situation of flow: something is always coming in, and, particularly, something is always coming out. In the case of a breeder reactor, fertile material comes in and fissile material and fission products come out. We throw away the fission products, permanently, and sell the fissile material. ("We hope" should be attached to both these clauses.)

To describe a steady-state situation, we must define the processes of change inside and outside the reactor. Let me again use the thorium cycle as an example. Th²³² is fed in. The rate at which it is fed is such that none is ever removed even though the contents of the reactor remain constant. The reason is, of course, that it is transformed at the same rate at which it is fed. I will use the symbol $\delta/\delta t$ to indicate insertion or extraction rate. Thus

$$\frac{\delta N_{02}}{\delta t} = + N_{02} \sigma_{02} \phi \qquad (4a)$$

Forgetting the difficulties of Pa holdup and Th²³³ decay, we get

$$N_{02} \sigma_{02}^{\text{eff}} = N_{23} \sigma_{23}^{\text{eff}} + \frac{\delta N_{23}}{\delta t} , \qquad (4b)$$

and so on:

$$N_{23}\sigma_{23}^{\text{eff}}\phi \frac{\alpha_{23}}{1+\alpha_{23}} = N_{24}\sigma_{24}^{\text{eff}}\phi + \frac{\delta N_{24}}{\delta t}$$
(4c)

$$N_{24}\sigma_{24}^{eff}\phi = N_{25}\sigma_{25}^{eff}\phi + \frac{\delta N_{25}}{\delta t}$$
(4d)

$$N_{25}\sigma_{25}^{eff} \frac{\alpha_{25}}{1+\alpha_{25}}\phi = N_{26}\sigma_{26}^{eff}\phi + \frac{\delta N_{26}}{\delta t}$$
(4e)

For criticality,

$$N_{23}\sigma_{23}^{\text{eff}}(\eta_{23}-1) + N_{25}\sigma_{25}^{\text{eff}}(\eta_{25}-1) - N_{02}\sigma_{02}^{\text{eff}} - N_{24}\sigma_{\text{eff}}^{24} - N_{26}\sigma_{\text{eff}}^{26} > 0 \quad , \quad (5)$$

the > sign meaning that there must be some allowance for leakage, parasitic capture, etc

Basing extraction rates on feed rates, we may cancel out δt from (4) and get

$$\frac{\delta N_{23}}{\delta N_{02}} = 1 - \frac{N_{23}\sigma_{23}^{\text{eff}}}{N_{02}\sigma_{02}^{\text{eff}}}$$
(6a)

$$\frac{\delta N_{24}}{\delta N_{02}} = \frac{1}{N_{02} \sigma_{02}^{\text{eff}}} \left[N_{23} \sigma_{23}^{\text{eff}} \frac{\alpha_{23}}{1 + \alpha_{23}} - N_{24} \sigma_{24}^{\text{eff}} \right]$$
(6b)

$$\frac{\delta N_{25}}{\delta N_{02}} = \frac{1}{N_{02} \sigma_{02}^{\text{eff}}} \left[N_{24} \sigma_{24}^{\text{eff}} - N_{25} \sigma_{25}^{\text{eff}} \right]$$
(5c)

$$\frac{\delta N_{26}}{\delta N_{02}} = \frac{1}{N_{02} \sigma_{02}^{\text{eff}}} \left[N_{25} \sigma_{25}^{\text{eff}} \frac{\alpha_{25}}{1 + \alpha_{25}} - N_{26} \sigma_{26}^{\text{eff}} \right]$$
(6d)

The left-hand side of each of equations (6) must be greater than zero (1 e, no net feed of any type of uranium) The system is of course also subject to the criticality condition

In order to sharpen up the criticality concept, we replace ">0" in (5) with "= $\beta N_{02} \sigma_{02}^{\text{eff}}$ = L " That is to say, that neutron losses (L) can be expressed as a ratio (β) of lost neutrons to neutrons productively absorbed With this replacement, we can now define things quantitatively

Doubling and Honest Measure

The concept of doubling is based on the idea that, if we have a breeder reactor, its operation for a sufficiently long enough time will provide the inventory of a new reactor built and operated just like the original I propose to be picayune and to take the statement of identity literally In this case, the output must be a sample of the inventory Since the concept of breeding and doubling are intrinsically related we may turn this into a definition <u>Definition 3</u>. The output of a breeder reactor must be a sample of its inventory. Otherwise stated, the buyer of bred material must get an honest measure.

With this definition, we can proceed further with our algebra. We now require that, for our example, in the chemist's system, where all U is fuel,

$$\frac{N_{02}}{N_{23}}\frac{\delta N_{23}}{\delta N_{02}} = \frac{N_{02}}{N_{24}}\frac{\delta N_{24}}{\delta N_{02}} = \frac{N_{02}}{N_{25}}\frac{\delta N_{25}}{\delta N_{02}} = \frac{N_{02}}{N_{26}}\frac{\delta N_{26}}{\delta N_{02}} = \frac{b}{\sigma \inf_{02}} (a \text{ constant}) \quad . \quad (7)$$

The constant b can be envisaged as an "extraction" cross section for the ith isotope. Notice that extraction in this sense enters the mathematics homogeneously. Without reproducing the algebra we quickly derive:

Let β be the fraction of fertile absorption represented by poison absorption and leakage (nonfissionable isotopes of the fissionable element not counting as poison), and b/ σ_i be the removal rate of ith isotope/absorption of a neutron by ith isotope.

For the Th-U breeders, then

$$1 + \beta = \frac{\sigma_{23}}{\sigma_{23} + b} \left\{ \eta_{23} - 1 - \frac{\sigma_c^{23}}{\sigma_{23}} \frac{\sigma_{24}}{\sigma_{24} + b} \left[1 - \frac{\delta_{25}}{\delta_{25} + b} \left(\eta_{25} - 1 - \frac{\sigma_{25}^c}{\sigma_{25}} \frac{\sigma_{26}}{\sigma_{26} + b} \right) \right] \right\} .$$
(8)

The situation is analogous for other fertile-fissile combinations. To get b from $\beta,$ the equation

$$\mathbf{b} = \frac{\sigma_{23}}{1+\beta} \left\{ \eta_{23} - 2 - \beta - \frac{\sigma_{23}^{23}}{\sigma_{23}} \frac{\sigma_{24}}{\sigma_{24} + \mathbf{b}} \left[1 - \frac{\sigma_{25}}{\sigma_{25} + \mathbf{b}} \left(\eta_{25} - 1 - \frac{\sigma_{25}^{C}}{\sigma_{25}} \frac{\sigma_{26}}{\sigma_{26} + \mathbf{b}} \right) \right] \right\}$$
(9)

illustrates an iterative rule which always converges rapidly.

Partial Breeding (Mass Spectroscopists' Breeding)

Up to now, most of the discussion has been an attempt to illustrate the fact that, when breeding occurs, a number of isotopes reach the steady state together. For each isotope, a partial breeding process exists.

We can isolate partial breeding ratios (PBR)

$$(PBR)_{23} = \frac{N_{02}\sigma_{02}^{eff}}{N_{23}\sigma_{23}^{eff}}$$
(10a)

$$(PBR)_{24} = [N_{28}\sigma_{23}^{eff} \alpha_{23}/(1+c_{23})]/N_{24}\sigma_{24}^{eff}$$
(10b)

$$(PBR)_{25} = N_{24}\sigma_{24}^{eff} / N_{25}\sigma_{25}^{eff}$$
(10c)

$$(PBR)_{26} = [N_{25}\sigma_{25}^{eff} c_{25}/(1+c_{25})]/N_{26}\sigma_{26}^{eff}$$
(10d)

These are the ratios of formation to destruction for each isotope.

Conditions on equations (6) are transformable into a statement that each PBR in (10) is greater than or equal to unity. This is never a restrictive condition on performance, that is to say, a given reactor may, for a given feed rate of thorium, have several possible modes of fuel management. So long as the PBR for each isotope of the bred material is greater than unity, the system is a breeder. The PBR's may be different.

Physicists' Breeding

We have now exhausted the possibilities in taking a chemical attitude towards reactor breeding. The key word in all our attitude is the word "fuel." In the foregoing, we have considered uranium to be fuel and used our knowledge of isotopes merely to define the specialized properties which our fissionable fuel will have for criticality with a given loss-to-fertile neutron ratio. However, we do not have to take a naive view of chemists' breeding to get the proper answer, and for certain types of fuel management other definitions may be appropriate. Let me use another example.

When the fertile material is kept separate from the fuel, it is possible to "milk" bred fissionable material and have that as the only product. This means that when the system settles down to a steady state, all terms of equations (6), except for (6a), are zero. In essence, the higher isotopes of the chain are built in the system and are not considered as fuel at all.

I think that this gives us enough of a handle to go back into Definition 3 and fix up its remaining loose end with a new definition.

<u>Definition 4.</u> The fuel of a breeder is its content of material whose chemical and isotopic constitution is identical with the material extracted. Any other material which is in the reactor as a result of the investment is a converted investment.

Since physicists consider it a trivial exercise to milk fresh fissionable out of the fertile material in a reactor, I have taken this definition of breeding as the physicists:

BR(Physicists) =

Here, using again the Th-U chain, "fissionable" is now only U^{233} . In consequence, of the steady state, higher U isotopes reach a constant concentration. After some algebraic manipulations, the analogue to equations (8) and (9) is obtained as

$$b = \sigma_{23} / \left(1 + \beta \left\{ \eta_{23} - 1 - \beta - (\sigma_{23}^{c} / \sigma_{23}) [2 - \eta_{25} + (\sigma_{25}^{c} / \sigma_{25})] \right\} \right)$$
(12)

There are, of course, blends between chemists' and physicists' breeding - imperfect milking, for example. In these cases, the consistent uses of all our definitions will allow specification of: fuel, inventory, investment, and steady-state processes. From these specifications, one can determine a breeding ratio which is the same for either chemists' or physicists' definitions.

Some Remarks on Mixtures, Management, and Economics

The foregoing remarks, except for the definitions, argue by implication and application from very simple examples. Two points of complexity require some statement: one is the question of mixtures of reactors or of breeding systems (including breeder-burner reactor complexes which produce a net output); the other is the influence of fuel and reactor management on breeding performance.

The first of these questions is a knotty one and can only be resolved by building the investment definition back into the breeding chain. Let me again use an illustrative example.

We couple a fast Pu breeder to a series of Pu-Th converters. The complex has a feed of U^{238} and Th and a net output of U^{233} - yet there is no U^{233} in the system!

To get around this, one must consider the closure of the chain, i.e., the reinvestment of the U^{233} formed into the complex. In other words, the system I have just described is not a breeding system. We must consider how the U^{233} produced is to be invested in new plant. When we do that, we discover that this investment is equivalent to internal feedback of the U^{233} within the base system. When the cycle is completely closed, we have a breeding system, in other words.

I have not had time to work through all the ramifications implied here. I will stop this line of thought, therefore, with the remark that the closure of the reinvestment chain always brings us back to a definable steady-state system whose breeding ratio is completely definite.

As to techniques of fuel management, I hope that it is understood that the complete arts of reactor physics must be brought to bear on the definitions (herein implied for a very simplified case) of flux, cross section, and criticality. There is no quick substitute for the detailed understanding of the life-cycle of the isotopes in this case, but only a realization that by appropriate definitions one can select these tricky properties so that, on the average, reaction rates, feed rates, and output rates are correctly given. It goes without saying, for example, that cross sections must be flux weighted to get proper reaction rates, and importance-weighted for criticality, so that losses may not necessarily be obtained by looking at cross section. Similarly, fluxes must be adjusted for out-of-pile holdup, and process losses counted in destruction cross sections.

As to the economics of breeding, I leave that to the economists. This much must be clear. There are two physical problems involved in breeding: first, the specification of the steady-state processes, and second, the specification of the method of producing the inventory from the investment. Similarly, the economic problem may be divided into analysis of the steadystate costs and returns, and of the economic investment which accompanies the physical investment.

Summary

Breeding is a closed-cycle, steady-state process. A system is a breeder so long as its net output is reinvestable in a specified way so as to reproduce the fuel content of the original system. For multistage systems, this is equivalent to specifying a definite internal feedback.

A steady-state breeder is specified by the absence of fuel input to the system and a net output of fuel from the system. The minimum specifications on performance are any pair of ratios of:

> feed rate fission or power output fuel extraction rate.

Fuel may be definied in various ways:

- 1. as a chemical element;
- 2. as fissionable materials arising from a single-step fertile atom capture; and
- 3. as a specified mixture of elements and/or 'isotopes.

If the fuel cycle is closed (i.e., fuel supply uniformly expanding) the third definition is preferred. The most useful definition of breeding ratio is then

BR = $\frac{\text{Atoms of fuel extracted}}{\text{Atoms of feed inserted}}$

Fission Ratio = Fissions Atoms of feed inserted

Using a bit of tautology, fuel may be defined as "extracted material," since closure of the breeding chain will determine what this material is.
The detailed balance of isotopic behavior in a system is a problem in reactor physics under the postulated fuel management. Similarly, the fissions achieved per atom of feed input is a variable under the same conditions. The values of η , α , etc., for the various fissionable isotopes are only qualitatively. useful in describing breeding capabilities.

THE STATUS OF MEASUREMENTS OF $\overline{\nu}$ AND $\sigma(n, \gamma)$ FOR FAST NEUTRONS

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The average number of neutrons per fission $(\overline{\nu})$ and capture cross sections are both of great importance in the design of reactors. It is only in the last few years that effective methods have been developed for accurate measurements of these quantities over a wide range of energies and, for capture, of elements. The amount of data now accumulating is almost embarrassingly large, and certainly too much for me to cover in my talk. I shall deal only with $\overline{\nu}$ and capture cross sections for fast neutrons and mostly with our own data and methods.

The liquid scintillator tank, which was developed originally to detect neutrinos, has become very useful in measuring $\overline{\nu}$ and capture cross sections. Figure 1 shows one of the tanks we have used, a cylinder of 30-in. diameter filled with exotic liquid and almost entirely surrounded by photo-tubes. On the right is a massive collimator for neutrons, surrounding the Van de Graaff target tube. This early model was not very well shielded, but we have found it to be advantageous to surround the scintillator tank with 30,000 pounds of lead and borated paraffin.



Fig. 1. Schematic Diagram of Liquid Scintillator Tanks

The neutrons, from some such source as the P-T reaction, are collimated so as to pass down a $2\frac{1}{2}$ -in. hole in the scintillator without striking it. If $\overline{\nu}$ is to be measured, a fission counter is introduced into this hole. When a fission occurs, a large pulse is produced in the fission counter. The fission neutrons pass into the liquid scintillator, are rapidly slowed down, and are eventually captured, one by one. Figure 2 shows scope traces of these delayed capture pulses. These captures would take place in hydrogen, producing 2.2-Mev gamma rays over a time range of several hundred microseconds, except that we introduce cadmium into this solution. This gives large, 9-Mev pulses which occur mostly in the first ten microseconds following the fission. The first trace was triggered by a fission and shows the prompt pulse produced by fission gammas and the slowing down of neutrons, followed by the delayed capture of three fission neutrons. The second trace was tripped at a random time and shows one small background pulse, too small to be counted.



Fig. 2. Oscilloscope Trace of Delayed Capture Pulse

The principal advantage of the liquid scintillator in these experiments is its high efficiency (approaching 100 per cent) for both neutrons and gamma rays. Of course, there are many other ways of measuring $\overline{\nu}$, such as by carbon piles, less efficient counters, or measurements on critical assemblies. Figure 3 shows values of $\overline{\nu}$ for U^{235} and Pu^{239} as determined at a number of laboratories. The points for U^{235} show an increase of about 1 neutron per 7-Mev change in excitation, which is roughly what one would expect and is apparently about right for U²³⁸, U²³³, and Th²³² fission also. The points for Pu²³⁹ seem to give a curve with less slope, about 1 neutron in an 8 or 9-Mev change. Although more data have become available since I prepared this figure two years ago, particularly at 14 Mev, the slopes still seem to be about the same all the way to 14 Mev, giving values of $\overline{\nu}$ of about 4.5. It is impossible to make precise predictions for these slopes, because of changes in fission product distributions as neutron energy changes, and because of the lack of any very detailed theory of the fission process. One could not, for instance, predict values of $\overline{\nu}$ for any unmeasured nuclide with much certainty, and all calculations concerning $\overline{\nu}$ so far published are based on normalization to measured values.



$\overline{\nu}$ for U ²³⁵	
BNL-325 (1958)	2.47 ± 0.03
Diven, Martin, Taschek and Terrell	2.426 ± 0.06*
Hansen and Keepin	2.37 ± 0.07*
Kenward, Richmond, and Sanders	2.420 ± 0.037
Moat, McTaggart, and Mather	2.331 ± 0.053**
Colvin and Sowerby	2.418 ± 0.039
Average	2.42 ± 0.02

(Thermal values, including 0.016 delayed neutron per fission) *Corrected to thermal assuming $d\overline{\nu}/dE = 0.15 \pm 0.01$.

**Note added in proof: It appears that the aldermaston $\overline{\nu}$ value in Table I (2.331 ± 0.053) will be revised upward by several per cent when corrected for neutron spectrum effects (A. Moat, Private Communication).

There is reason to think, however, that the normalization may be to slightly wrong values. As shown in Table I, critical-assembly measurements by Hansen at Los Alamos indicate a value of 2.37 for $\overline{\nu}$ of U²³⁵, allowing for a reasonable increase of $\overline{\nu}$ with energy. This is 3 or 4 per cent lower than the BNL-325 value. I understand that other critical mass measurements give similar results. When we (Diven, Martin, Taschek, and I) measured $\overline{\nu}$ on an absolute basis several years ago, using our liquid scintillator, we found an absolute value for U²³⁵ of 2.426, about 2 per cent lower than the accepted value of 2.46 or 2.47. This fact we reported in our published paper, but we normalized our values to the BNL-325 figure. Now we are more inclined to believe that the correct figure is lower, in the light of several recent experiments. Kenward, Richmond, and Sanders at Harwell have obtained an absolute value, using a paraffin cylinder filled with BF₃ counters, of 2.420. Colvin and Sowerby, also at Harwell, have obtained 2.418 with a large graphite pile. Moat, McTaggart, and Mather at Aldermaston, using a large liquid scintillator, have obtained the lowest value:2.331. All of these figures are corrected to include delayed neutrons and are extrapolated to thermal energy if necessary.

The weighted average is 2.42 ± 0.02 , using the uncertainties given in the table. Thus it is probable that the standard value 2.47 of $\overline{\nu}$ for U²³⁵ is 2 or 3 per cent too high. Moat <u>et al</u>. think it is 6 per cent high. Since most $\overline{\nu}$ values are normalized to the U²³⁵ value, it may be that most quoted values are a little high.

As to $\overline{\nu}$ for Pu²³⁹, there is a noticeable spread to the reported values. The ratio to U²³⁵ ranges from the value of 1.174 of BNL-325 to that of 1.23 ± .01 of DeSaussure and Silver, a 5 per cent range of $\overline{\nu}$. Taking the $\overline{\nu}$ value for U²³⁵ as 2.42, this puts $\overline{\nu}$ for Pu²³⁹ in the range from 2.84 to 2.98. It may well be that the value of 2.90 ± 0.04 in BNL-325 is about right. This is also in agreement with the figure obtained from criticality measurements by Hansen. However, it would be desirable to pin this number down a little more firmly.

Cross sections for the capture of fast neutrons may be measured with the same liquid scintillator equipment I have described. Of course, one can measure these same cross sections by transmission measurements, or by reactivity methods, or in some cases by activation. The liquid scintillator method may be used even when no radioactivity is produced, and does not require extensive corrections.

Figure 4 shows a 19-in. scintillator tank used for some measurements of capture cross sections, in particular to measure the capture cross section for U^{235} . As in the $\overline{\nu}$ measurements, a collimated flux of neutrons is passed through an aperture in the scintillator. In this flux is placed a capture sample. When fast capture takes place, most of the gamma-ray energy produced is absorbed in the tank, giving a large pulse. The ideal situation would be that in which all of the gamma rays would be absorbed in the tank, giving a large, perfectly uniform pulse corresponding to the binding energy of the neutron captured plus its kinetic energy.

This pulse for fast capture may be separated from other events by pulsing the neutron beam and observing the rate at which such pulses occur in coincidence with the beam pulse. Figure 5 shows schematically the time distribution of pulses in our tank. In measuring the capture cross section for U^{235} it is necessary to separate captures and fissions, since both events produce large prompt pulses. This may readily be done by detection of the delayed neutron captures following fission. Scattered neutrons are not a



Fig. 4. Schematic Diagram of Apparatus for Measurement of Capture Cross Sections

serious problem for us. They produce an immediate pulse, due to recoil protons in the solution, which we bias out of our data. A bias of about 1 Mev, for instance, would be sufficient to exclude all the prompt pulses from scattering of neutrons of 1-Mev energy or less, whether elastic or inelastic. The scattered neutrons do produce delayed capture pulses, which raise our background rate, but not seriously. Figure 6 shows our results for the capture cross section of U^{235} , expressed as α , the ratio to the fission cross section. There is a continuous decrease in the range from 175 kev to 1 Mev. Recent data taken at Oak Ridge give values for α , in the region of a few tens of kev, as high as 0.4, so that the straight line we have drawn through our data should by no means be extended into this region of lower energy.



Fig. 5. Time Distribution of Pulses

Fig. 6. Values of α for U²³⁵

Measurement of the capture cross section of a nonfissionable material is much easier for us, so long as it is more than a few millibarns and the material is not too radioactive. In Fig. 7 is shown the time distribution of capture pulses in our scintillator for tantalum. These data are obtained by means of a time-to-pulse height converter. The range of time covered in the figure is one microsecond. The neutron burst lasts perhaps 0.02μ sec, or longer if we desire. For tantalum, which has a 242-mb capture cross section at 400 kev, a very nicely defined peak in counts appears at the time of the neutron burst; there is also an earlier peak from target gamma rays. For polyethylene, which has almost no capture cross section, no peak is visible, but there is a small effect due to scattered neutrons.



Fig. 7 Time Distribution of Capture Pulses

The capture cross section is obtained by rapid interchanges of samples. The area under the peak is proportional to the capture cross section and the number of atoms per cm^2 in the capture sample. Thus ratios of capture cross sections are obtained, and we have taken the well-measured capture-plus-fission cross section of U^{235} as our standard.

There are several sources of error which must be considered in such experiments. First, the samples must be thin, both to keep the attenuation of the neutron beam low and to minimize absorption of capture gamma rays. To keep this absorption of gammas low, the samples should also be small in diameter. We have used sets of $1\frac{1}{2}$ -in. discs, usually of 5 or 10-mil thickness. The discs are spread out so as to minimize gamma absorption.

The pulse height distribution obtained from silver in our scintillator appears in Fig. 8. The loss of capture pulses below bias settings could be one of the most important sources of error in such an experiment. Hence, a considerable amount of time has been given to investigating the pulse height distributions from our capture samples. There is always a broad distribution of pulse heights extending up to 7 or 8 Mev, with, usually, a peak at the point corresponding to the total energy release, for the 40-in. scintillator. In the 19-in. scintillator the probability of gamma-ray escape is greater and the pulse height distribution much worse, for cross section purposes. However, the consistency of cross-section results between the two scintillators gives a useful indication of the uncertainty in the value of the cross section.



Fig. 8 Pulse Height Distribution for Silver

Figure 9 shows similar data for cadmium. In this case some isotopes have a high binding energy and produce a secondary peak at about 9 Mev. The procedure in measuring cross sections is to bias at about 3 Mev, so as to eliminate the 2.2-Mev background. This causes loss of perhaps 20 per cent of the pulses, but knowledge of the pulse height distribution allows correction for this loss.

Not all our pulse height distributions are as good as these. In some cases, including that of gold, there is a somewhat wider pulse height distribution. This is presumably due to high-energy gamma rays, which are more likely to escape. For very small cross sections, also, it is difficult to measure these distributions, and the correction for 3-Mev bias is less certain.



Fig. 9 Pulse Height Distribution for Cadmium

The results of our measurements (by Diven, Hemmendinger, and myself) are shown in Figs. 10, 11, and 12, where the capture cross sections are shown as functions of neutron energy in the range from 175 to 1000 kev. The neutron energy spreads are indicated below. Relative errors are shown on all the points, with the exception of those at 400 kev, to which the other points are normalized. In absolute terms the cross sections at 400 kev are known to about 2 mb for small cross sections, and 5 to 10 per cent for larger cross sections. There is a general downward trend to the capture cross sections as a function of energy, but this is obviously not always the case.

A considerable amount of capture cross section data has been taken at Oak Ridge National Laboratory. Figure 13 shows some of the data of Gibbons, Macklin, Miller, and Neiler for tantalum. They have taken data in the energy range from 10 to 100 kev, where we have not yet attempted to work. Their techniques and results are similar to ours. However, in this case and in apparently every case where the two sets of data overlap, there is about a 20 per cent discrepancy, the Oak Ridge data being lower. Figure 14 shows the data for platinum from both laboratories. We have had a number of discussions on this point, but have not yet agreed as to the reason for the difference.



Capture Cross Sections



Fig. 11 Capture Cross Sections



5 X 10³ Τа 6.43 X 10²¹ atoms/cm² 0 2 00 $\left< \frac{\Gamma_n^0}{D} \right> = 1.9 \times 10^{-4}$ 10³ $\Gamma_{\rm y} = 56 \times 10^{-3} \, {\rm ev}$ σ_c (mb) $D_{1} = 12.4 \text{ ev}$ 5 1 1 1 1 1 • ORNL Li (p, n) o ♦ ORNL T (ρ, n)
♦ ORNL THRESHOLD OR THIN TARGET Ť. 2 $\sigma_{I=0}$ DIVEN, et al = 9 ev D 10² 10² 10³ 5 10 2 5 2 5 En(kev)

Fig. 13 Capture Cross Section for Tantalum

Fig. 12 Capture Cross Sections



Fig. 14 Capture Cross Section for Platinum

Capture data are also being taken at Duke, Livermore, and Wisconsin. A great mass of it is rapidly accumulating, and theoretical fits are being made with reasonable success.

Figure 15 shows our 400-kev cross sections as functions of A, the average mass number. There is a general upward trend to the cross sections in the region below mass 100, and thereafter no obvious trend. The most spectacular deviations occur for magic numbers of neutrons or protons in the target nucleus, in which case the cross section is usually quite low. Also, odd-Z target nuclides tend to have cross sections that are perhaps higher by a factor of two than for even-Z cases. These facts are in agreement with the ideas of Bethe and Hurwitz, who have suggested that the level density in the compound nucleus does not depend on the true excitation energy, but on an effective excitation energy, measured from some reference mass surface which includes no effects due to magic numbers or even-odd effects. Hence, target nuclei which are odd-odd result in unusually high level densities and effective excitations; magic nuclides tend to give unusually low level densities.

Since the capture cross section, in the region of a few hundred kev, is more or less proportional to the product of level density and the radiation width, the Bethe-Hurwitz idea allows a rough prediction of the capture cross section. This would be of use where there is not enough data on strength functions and level densities to allow a more precise calculation. The usual level density expression, exponential in $(AE)^{\frac{1}{2}}$ gives the general trend of the cross-section data when allowance is made for decreasing binding energy with increasing mass. The magic and even-Z nuclides give low effective excitations and hence low cross sections.

Figure 16 shows a number of accurate measurements of radiation width as compiled by Stolovy and Harvey. The radiation width is more or less constant in the vicinity of 0.1 ev. A semiclassical calculation of the



Fig. 15. Capture Cross Section at 400 kev

radiation width, based on Weisskopf's $E^{3}R^{2}$ transition probability and the usual expression for level density, gives an approximate $A^{-\frac{5}{6}}$ dependence, which more or less fits the general trend of $\Gamma\gamma$.



Fig. 16. Radiation Widths

Figure 17 shows our 400-kev capture cross section data, plotted against $(AE^*)^{\frac{1}{2}}$. The value of E*, or the effective excitation energy, was calculated by the use of Cameron's mass surface for odd-odd nuclides, without pairing or shell corrections, and the actual mass of the target nuclide. The correlation produced is not bad. For most nuclides the points



Fig. 17. 400 kev Neutron Capture

fall within a factor of about two of the straight line. The principal exceptions are the heaviest nuclides; it is possible that the mass surface used was not suitable in this region. Some minor factors involving A and E^* have not been included in this correlation. Their inclusion gives a slope of almost the theoretical value, but does not improve the fit.

Presumably this rough calculation works fairly well at a few hundred kev because both S and p-wave contributions are apt to be nearly saturatedthat is, independent of the neutron strength function - and because inclusion of d-wave and inelastic scattering effects should not change the result by a factor larger than 2.

Figure 17 has been included both to indicate that the Bethe-Hurwitz idea works fairly well on a quantitative basis, and to allow a reasonable estimate of a totally unknown capture cross section. I do not intend to go into competition with the rather precise calculations which can now be made of capture cross sections when enough data are available.

Discussion of Paper Presented by Mr. Terrell

MR. KATO:

I just wondered, how do your 400-kev cross sections as a function of A compare with the fast-activation cross section that Hughes <u>et al</u>. did a few years ago? These were also plotted as a function of A and also showed the magic number effect.

MR. TERRELL:

You're quite right. Plotting capture cross sections as a function of A is not new. I think there have been three or four papers that have had similar curves. I have not tried to compare them point by point, but they certainly look very much the same. The magic target nuclide yields low capture cross sections.

MR. KATO:

How do you determine the counter efficiency by your capture gammaray work?

MR. TERRELL:

Fortunately our efficiency is rather high, perhaps 95 per cent. We extrapolate down to zero pulse height and make certain corrections having to do with the pulse height distribution, and a few other minor factors. But basically we simply use ratios relative to the capture-plus-fission cross section of U^{235} . Does that answer your question?

MR. BARTELS:

What is the relationship between your value of $\overline{\nu}$ and a value one might infer from de Saussure's work? I gathered that you referred to his work on the reactor neutrons detected by the manganese sulfate method.

MR. TERRELL:

I gather you are speaking of the $\overline{\nu}$ for Pu^{239} - I wasn't aware of any absolute values. In terms of the ratio of 49 and 25 we obtained practically the same number which was measured by the Oak Ridge people, de Saussure and Silver. I haven't heard about an absolute measurement at ORNL.

MR. BARTELS:

I really was talking about an inferred value, not a direct absolute measurement, of $\overline{\nu}$. I'm not quite sure how far along they are. I think de Saussure is here. Would he care to comment on his work?

MR. DE SAUSSURE:

Absolute measurements of η have been made, which, I think, could lead to a higher value of $\overline{\nu}$ if one believes the alpha measurement of, I think, 0.118 for alpha of U²³⁵. Does that answer your question?

MR. SANDERS:

Could you say some more about the measurement of $\overline{\nu}$ based on the critical experiment by Hansen?

MR. TERRELL:

Yes, what Hansen did was to determine an average value of $\overline{\nu}$ for a fairly fast spectrum, that is, of the order of $l\frac{1}{2}$ -Mev neutrons. He found a value which, I believe, was 2.58. In order to get a thermal value this has to be extrapolated at some reasonable slope back to thermal energies. If you use the slope which seems to hold between zero and 14 Mev for most cases, namely, about 0.14 or 0.15 neutron per Mev, a value of $\overline{\nu} = 2.37$ is obtained. It does not seem possible to arrive at 2.46 by use of a reasonable slope. Does that answer your question?

MR. SANDERS:

I want to really know how they got the original data.

MR. TERRELL:

I'm sorry.

MR. SANDERS:

This is the critical sphere experiment of something.

MR. TERRELL:

Oh, yes. This is based on the measurement of the fast critical mass. I really don't know much about the work, in personal terms, but it is based on the measurement of critical mass, and on a calculation of the critical mass. They have to throw in a value of alpha, of course, and certain errors exist on various other quantities used.

VOICE FROM THE FLOOR:

I would like to know if you determined $\overline{\nu^2}$. If so, will you comment on it? What is its value?

MR. TERRELL:

We did make many measurements of this sort of thing, and the results have been fully reported in the Physical Review. Generally speaking, $\overline{\nu}^2$ has a value which is less than one would expect for a Poisson distribution, and, as a matter of fact, $\overline{\nu}^2$ is such that $(\overline{\nu}^2 - \overline{\nu})/(\overline{\nu})^2$ is of the order of 0.8. This seems to hold roughly true for a good many cases. This figure would be 1.0 in the case of a Poisson distribution.

MR. OKRENT:

With regard to the $\overline{\nu}$ measurements, I wonder whether you would care to comment on the corrections made for leakage of neutrons from these measurements. It looks like a fairly complex problem.

MR. TERRELL:

You are quite right. I think this is true of all of these measurements of $\overline{\nu}$. Certainly it is true of the liquid scintillator measurements. It involves some thought being given to the fractions of neutrons which are not counted and whether this fraction might vary from isotope to isotope. I believe that with a reasonably large scintillator, 30 or 40 inches in diameter, one would expect no more than perhaps 5 per cent of a fission spectrum to escape. Because the fission spectrum energy does not vary greatly, it would seem that you have very little change in this number.

MR. OKRENT:

I'd like to ask another question, more specifically, if you please. Has there been any experimental evaluation in the particular geometry of neutrons? In other words, how well can you quote the absolute levels?

MR. TERRELL:

Oh, yes. We have, at least in our other work, measured the loss of neutrons from our tank as a function of energy of the neutrons. We can get an absolute value of the efficiency at various angles and at various energies. We have done this and we have found that the losses run just about what one would calculate. We have also made Monte Carlo calculations of this thing. They seem to check out. VOICE FROM THE FLOOR:

Have you measured the values of the gamma-gamma?

MR. TERRELL:

We have not measured the values of gamma-gamma; I merely showed that for your information. It's done by means of fast choppers and such apparatus.

VOICE FROM THE FLOOR:

In other words, that was just for quite low energies.

MR. TERRELL:

Yes. The values for gamma-gamma were for thermal or epithermal energies. One might assume that the value would go as E^3 .

MR. PATTENDEN:

I think you may have answered this question already. I was going to ask it with respect to your capture cross-section measurements of U^{235} ; in this case you used the neutrons - delayed neutrons - detected in your tank and you used some sort of anticoincidence method to distinguish between captures and fissions. Is that right?

MR. TERRELL:

Certainly, we do look at the delayed neutrons. What we actually do is to analyze the number of pulses following a coincidence pulse. We count the pulses in a certain gated interval following the fission or the capture or whatever the event is. We do this under various conditions: with no beam, with no sample, with various types of scattering samples. We can thus arrive at fairly complete information as to the distribution of pulse numbers from background events, from fission events, from scattering events, and from capture events. By putting all the data together in a fairly obvious way, we can allow for the fission events.

VOICE FROM THE FLOOR:

Did the statement which you made about the geometrical efficiency of neutron counting as far as $\overline{\nu}$ is concerned also apply to the neutrons counted in the capture work?

MR. TERRELL:

We have used three different tanks. The calculations I mentioned were primarily for the 30-in. tank. For the 40-in. tank we have not done Monte Carlo calculations. But since the Monte Carlo calculations on the 30-in. tank gave results which could have been calculated fairly well by hand, we have not repeated them. The losses are quite a bit smaller in the 40-in. tank. At the end for the 19-in. tank they are somewhat more serious. But we did not worry about this, because the only neutrons we were interested in were fission neutrons, and we simply put a fission counter in the tank and measured some of these quantities directly without having to depend on a calculation.

MR. LEONARD:

I would like to point out that your experiments, say on α and on $\overline{\nu}$, are systematically related by correcting those fission events for which you get no neutrons. This has to be correct in the terms of a Poisson distribution, doesn't it? Could you give us some idea of the size of this effect?

MR. TERRELL:

It is my recollection that in our 19-in. tank (this was the one which we used to measure α) we found that between either 18 or 19 per cent of the fission events were followed by no pulse, except perhaps background pulses. This checks with calculations obtained from our measurements of $\overline{\nu}$. We did not depend, in the alpha work, on our previous measurements in this respect. We measured the quantity directly. Does that answer your question?

MR. LEONARD:

My recollection is that there was a paper submitted to the Geneva Conference which showed the same variation of $\overline{\nu}$ with energy for various isotopes. It reported getting a reasonable fit with the same slope for the various isotopes. Is my recollection correct?

MR. TERRELL:

I think you must be speaking about Leachman's paper. Well, several years ago Bob calculated some slopes based on a temperature of 1.0 or 1.4 Mev and he got about $\frac{d\nu}{dE} = 0.125$. Now with a lower temperature, like 0.7 Mev, one would get a somewhat higher slope of, say, 0.14. What Bob did in his most recent paper, the one you are thinking of, was to show these calculated values of his without trying to fit the points to the various $\overline{\nu}$ values.

There is quite a wide spread in some of the reported values. I don't think he tried to fit the points; that is my recollection. However, they would fit fairly well, since slopes between 0.17 and 0.10 can pass through some of these points.

MR. OKRENT:

If he were to correct back with a slope of 0.11 instead of 0.15, his $\overline{\nu}$ value would have been somewhat higher.

MR. TERRELL:

That is true. Let me see. The corrections of Hansen's data were of the order of about 0.19, I think, and if a lower slope had been used the correction might have been 0.15 or something of the sort. It might have brought Hansen's figure up to 2.4, or even higher. However, I am sure that the $\overline{\nu}$ data for U²³⁵ indicate a slope between 0.14 and 0.15. One does not know some of the details. For instance, between 5 and 14 Mev there is essentially no information.

FAST-NEUTRON SCATTERING BY U²³⁵, Pu²³⁹, AND U²³⁸

Lawrence Cranberg Los Alamos Scientific Laboratory Los Alamos, New Mexico

Two years ago at the Columbia Conference on the Interaction of Neutrons and Nuclei, I gave a talk on the topic for today and presented a fair amount of what was then new material. This material was obtained with recently developed techniques of fast-neutron spectroscopy, utilizing the measurement of the time-of-flight of the scattered neutrons.

Those results differed rather markedly from many of the measurements which had been reported previously by other methods. The older work depended, for example, on measuring the transmission of monoenergetic neutrons through spheres, and on methods which employed high-biased detectors to observe the differential scattering cross section. The discrepancies were factors of 2 or more in some cases.

In contrast to the situation described by the previous speaker on neutron capture, there is by no means a profusion of data from active workers in this field and there are actually little new data to report. But the new data which are available are perhaps of interest because I think they shed some light on the discrepancies that were apparent between our results and the results reported earlier. I hope that as a result of this talk you will have a clearer notion as to which set of data is to be taken the more seriously. If you don't, then I think the indication is clear that other people should take an interest in this field, and at the next meeting the paper on this theme should be given by some other individual.

In connection with the subject of discrepancies, let me say at the outset that I have great respect for much of the work which was done by the older methods, the sphere transmission approach, for example. I think the main reason for the existence of discrepancies is that the fissionable nuclei happen to have strongly excited low-lying excited states - a circumstance which presents a particularly unfavorable experimental situation for the sphere technique.

I propose, first, to review briefly the information which I presented in preliminary form at the Columbia Conference, and which has not changed much in the interim. Secondly, I will discuss in detail some of the supplementary work which we have done by way of cross-checking our results. Thirdly, I will indicate what other information has become available on this general subject. Finally, I will briefly describe what we plan to do in the future in this field. The level schemes of these nuclei are illustrated in Fig. 1. This shows the level scheme, at least as far as the low-lying levels are concerned, for the three nuclides under discussion; U^{238} , U^{235} , and Pu^{239} . These data were obtained by different experimental techniques. In all these three nuclides there is a level within 50 kev of the ground state. I emphasize this point because one must prepare for trouble, it seems to me, when discussing inelastic neutron scattering in these nuclei, if one is incapable of making a sharp distinction between elastic scattering corresponding to neutrons which leave the target nucleus in the ground state and inelastic scattering which corresponds to leaving the nucleus in a low-lying excited state.

Figure 2 illustrates the kind of raw data on which our results are based. These raw data are time spectra which were obtained in the following way. Monoenergetic neutrons were produced in an accelerator in the form of very short bursts of a few m μ sec duration. These neutrons were scattered by the scatterer under investigation, and at a distance of one or two meters with a fast neutron detector the spectrum of arrival times of the neutrons in that distant detector were measured.



Fig. 1. Energy levels of U^{235} , U^{238} , and Pu^{239} .



What I show here are some time spectra which are favorable cases - that is, they represent situations in which we resolved fairly completely what is known about the level structure of the nuclei involved. In the case where 545-kev neutrons are incident on a scatterer of U²³⁸, the structure can be resolved into two groups. The main group corresponds to elastic scattering, which leaves the U^{238} nucleus in its ground state, and the second group corresponds to excitation of the 44-kev level. The special point of interest connected with this figure is the fact that it illustrates the strength of the excitation of the first excited state in this nucleus at this energy. This is an observation at about 90-degree scattering angle and it can be seen that the inelastic scattering is comparable in magnitude to the elastic scattering. We think excitation of the 44-kev level continues to be very important, even up to 1 Mev. Under the condition of shorter flight paths the ground state can no longer be resolved, but the neutron group which corresponds to excitation of second excited states at 147 kev in U²³⁸ can be seen. These are illustrative of the raw data upon which our results are based.

Our results are given in Fig. 3. The energy of the monoenergetic neutrons which are incident on the scatterer is given. We have covered the range from 550 kev to about 2 Mev. The positions of the levels as we have determined them from our own measurements of the spectra, and the differential scattering cross sections observed at 90 degrees for the excitation of each of these levels, are shown.

Fig. 3.

Inelastic N	Neutron	Scattering	from	U ²³⁸	at	90°
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(Barns per steradıan)

	I = 2	I = 4	I = 6			<u> </u>	•	
-Q(kev)								
E _n (kev)	44 ±4	146±6	300±30	730±30	980±20	1060±30	1260 ± 40	1400 - 1750
550±10	0 135±0 015	0 023±0 05	< 0 01					
1000±24	0 07 ±0 02	0 05 ±0 015	0 007±0 004	0 041±0 006				
1500±36				0 022±0 002	0 037±0 004	0 037±0 004		
1890±30				0 007±0 003	0 041±0 004	0 035±0 004	0 008±0 004	
2000±40						0 103±0 01		0 087± 0 008

Figure 4 presents a summary of all the partial cross sections - cross sections for all the various things that can happen when monoenergetic neutrons strike a sample of U^{238} . The incident neutron energies are 550 kev, 1 Mev, and 2 Mev. The capture cross sections are based on the work reported by Terrell. The fission cross sections are out of BNL-325. The inelastic cross sections and elastic cross sections are our work.

Fig. 4. Partial and Total Cross Sections for U^{238}

E _n (kev)	Elastic	Inelastic	Fission ^a	Capture ^a	Sum	σa t
550	6.27 ± 0.3	1.72 ± 0.2	<0.01	0.14	8.20 ± 0.4	8.2 ± 0.1
1000	4.55 ± 0.5^{b}	2.02 ± 0.4^{c}	0.015	0.10	6.69 ± 0.6	7.0 ± 0.1
2000	3.93 ± 0.1	2.38 ± 0.2 ^c	0.56	0.05	6.92 ± 0.3	7.1 ± 0.1

1	-				١.	
	- H =	2 7*	n	C		
ſ.	200	**	**	9		

^a Reference 14

^b Reference 13 after subtracting 0.9 barns as the estimate for the contribution due to the 44 kev level.

^C Does not include levels within 250 kev of the primary neutron energy.

Incidentally, Fig. 3 shows differential scattering cross sections measured at 90 degrees, and Fig. 4 shows total inelastic cross sections. We have made the assumption that inelastic scattering is very closely isotropic. Now when we compare the sum of these cross sections with the total cross section the agreement is reasonable. There is a difference which is in a satisfactory direction. By satisfactory I mean the following: our neutron detector is incapable of detecting neutrons which have an energy less than 250 kev. So we don't see all the inelastically scattered neutrons. We expect that there will be some discrepancy here in the direction indicated. When I come to the detailed discussion of discrepancies, we shall look in greater detail at the 1-Mev situation for U²³⁸.

Figure 5 gives a summary of the results on U^{235} and Pu^{239} for incident neutrons of energies of 550 kev, 1 Mev, and 2 Mev. Here we have made no attempt to resolve individual levels. I have grouped the inelastically scattered neutrons into energy groups. Under the circumstances of the experiment, we couldn't tell the difference between neutrons which had lost 60 kev of energy and those which had lost no energy.

The fission cross sections and the capture cross section are shown and we add up across to get sum cross sections. At the lowest neutron energies the agreement between the sum and the total is quite good; only at 2 Mev do we feel that there is a significant difference which indicates inelastic scattering of neutrons we can't see because they are too low in energy.

Fig. 5.

PRELIMINARY

Neutron Scattering from U^{235} and Pu^{239}

(Barns)

$E_n = 5$	550 ź	E 10) kev
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-Q(kev)	350-275	275-60	60-0		Fission ^a	Capture ^b	Sum	σta
Element				·	-199-24-4-18-1-		-	
U ²³⁵	<0.03	0.53	5.30		1.20	0.15	7.18	7.3 ± 0.1
Pu ²³⁹	0.14	0.35	6.18		1.70	0.15	8.52	8.4
			$E_n = 10$)00 ± 24	ł			
	750-500	500-150	150-0					
U ²³⁵	0.33	0.33	4.70		1.27	0.1	6.73	6.6
Pu ²³⁹	<0.06	0.16	5.06		1.85	0.1	7.17	7.2
			$E_n = 20$	000 ± 40)			
	1750-1500	1500-1000	1000-500	500-0				
U ²³⁵	0.42	0.67	0.27	3.87	1.32	0.05	6.60	7.0
Pu ²³⁹	0.24	0.24	0.19	4.00	2.00	0.05	6.72	7.2

^a Brookhaven National Laboratory Report BNL-325

^b Estimated

Now we come to discrepancies. Figure 6 shows the data for the case we shall discuss in detail: the inelastic cross section for U^{238} for l-Mev incident neutron energy. We get the largest collection of results if we define the inelastic cross section as not involving excitation of the first two levels. Most of the workers were incapable of resolving neutrons which correspond to excitation of the first two levels, from neutrons which correspond to elastic scattering.

You can see that the data from various sources give values for the inelastic scattering cross section which is in the neighborhood of about twice our result.

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Inelastic Cross Sections of U²³⁸ at 1 Mev Not Including the First Two Levels (Barns)

1.40 ± 0.20 1.2 1.1 ± 0.2 1.35 ± 0.20	Allen Walt and Barschall Batchelor Bouston Walt, and Salmi
1.35 ± 0.20	Beyster, Walt, and Salmi
0.60 ± 0.15	Cranberg and Levin
	1

Partial Cross Sections for U²³⁸ at 1 Mev (Barns)

Total "Elastic"	Inelastic	Capture	Fission	Σ	Total
6.48 ± 0.2	0.60 ± 0.15	0.14	0.015	7.23 ± 0.3	7.0 ± 0.1

The best way we could think of to check our result was to measure the differential elastic scattering cross sections using time-of-flight techniques. Our result for the total elastic cross section including the first two levels is 6.48 barns. The inelastic cross section was 0.6 barn. Adding capture cross section and fission cross section we get 7.2 ± 0.3 barns, and 7.0 ± 0.1 for the total out of BNL-325. This is the result of carefully repeated checks with several samples of U²³⁸. The details of the procedure and results are given in Los Alamos report LA-2177.

One might ask, are the discrepancies between our results and those reported by others in the right direction to be accountable in any plausible way? I think the answer to that question is "yes." I might say I have discussed these discrepancies with the workers involved, and I think that they are in substantial agreement with the following interpretation of the discrepancy with the results of sphere transmission measurements.

The analysis of the results of the sphere method is contingent on the assumption that the resolution and bias of the detector are such as to distinguish sharply between elastic and inelastic scattering. If this condition is not met, then the calculated nonelastic cross section tends to be high because multiple inelastic scattering can degrade neutrons below the detector bias. This effect is enhanced in measurements with thick shells, and in cases in which the level which is not resolved from the ground state is strongly excited, both of which circumstances apply to the measurements of U^{238} .

Now there are some recent data from other sources on this particular point of inelastic scattering in U^{238} . One source of the recent data is Mr. Batchelor, who is the source of some of the earlier results. He has recently used time-of-flight spectrometry to report results in quite good agreement with our own. Other data come from a recent study, as yet unpublished, by Day and Lind at Los Alamos, who have observed excitation of the gamma rays in U^{238} which accompany inelastic scattering in those nuclei.

Figure 7 shows the level scheme which they have been able to infer from this work for U^{238} . The dotted line means that those gamma rays were superimposed on others with essentially the same energy. Figure 8 shows the excitation function of each of those gamma rays, expressed in barns, as a function of incident neutron energy, over the range in which we have been interested. Now we can get a figure which corresponds to our 0.6 barn by simply looking at 1 Mev and adding up all these partial cross sections. The result is 0.7 barn, with about a 20 per cent error, and in satisfactory agreement with our result.



Fig. 7.

I cannot go into details on the subject of discrepancies in the nuclides U^{235} and Pu^{239} , but these are in the same direction. They are comparable to those I discussed for U²³⁸, and I am inclined to surmise that they have the same origin, but I don't say this with absolute conviction, because of the possibilities of systematic errors in our work in allowing for the effects of fission in the strongly fissioning nuclides. Detailed discussion of these matters is also given in LA-2177. I think it is quite clearly desirable that somebody else move into this field and do some work to obtain independent checks on these discrepancies.

There are no new data for U^{235} , but there are some calculations which have been reported in the October <u>Physical Review</u> of last year by Rae, Margolis, and Troubetzkoy, based on plausible guesses as to what the level structure is in U^{235} . Figure 9 shows the results. The experimental points are shown and the calculated cross sections for energy loss in excess of 60 kev are given. The calculated prediction is indicated on the basis of various assumptions as to the level structure of U^{235} .





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The most plausible surmise as to what the level structure of U^{235} actually is like corresponds to curve "c" and the results are in quite good agreement with our own.

It is clear that there are considerably more data to be obtained, particularly at higher incident neutron energies. We have been rather slow in going into this field because of the increasing difficulties attendant on separating fission neutrons from inelastically scattered neutrons at higher incident neutron energy. It is our intention, however, to proceed to higher energy and at least to get data for 4-Mev incident neutron energy.

At 4 Mev, on the basis of work we have done on other heavy nuclei, we expect to see something like a Maxwell distribution for the spectrum of inelastically scattered neutrons, and the results will presumably be expressed in terms of a temperature. As a matter of fact, we have already reported for U^{238} at 2.5 Mev a temperature of 0.3 Mev.

Figure 10 covers a somewhat more general area, namely, the question of inelastic neutron scattering in general, under conditions where the incident neutron energy and the mass number of the target nucleus are such that one would obtain essentially a continuous spectrum for the inelastically scattered neutrons.





Variation of a with Mass Number (Preliminary)

This nuclear temperature T, which is inferred from the shape of the spectrum for a given incident neutron energy and given scattering material, is related to a parameter "a" which is the parameter "a" in the expression for the level density as given by Blatt and Weisskopf. In treating the nucleus as a Fermi gas, "a" has been predicted to be equal to the mass number "A" over 10.5. That is what the line shown represents. These various points have been obtained from an analysis of the spectra from each nuclide by Mr. David Thomson. The results given here are primarily at 7.0 Mev. We also have a fair number of points at 4 Mev and a few at 6.3 Mev. In general, 7 Mev is as high as we can go and yet stay below the (n,2n) threshold in all nuclides studied so that we are observing pure inelastic scattering. The essential feature of the result is the following. As we go up in incident neutron energy, we always seem to be approaching the Fermi gas prediction, although in many instances we are still an appreciable way from it.

These data provide a basis for systematizing a picture of inelastic scattering and, hopefully, for making some extrapolations into the region of the fissionable nuclei.

Discussion of Paper

Presented by Mr. Cranberg

MR. OKRENT:

Are there any comments or questions?

MR. TOPPEL:

You mentioned comments about the discrepancies that might appear in the sphere method but in the case of the helium 3 spectrometer these comments perhaps don't apply. Can you say something about that?

MR. CRANBERG:

Yes. When Batchelor used the spectrometer, he used the geometry of the sphere experiment. A helium 3 spectrometer has a very low efficiency. Much less than 1 per cent. To get satisfactory yield, he surrounded the helium 3 spectrometer by a fairly thick shell of U^{238} . It seems not much attention was paid to the role of multiple scattering in that thick sample when the cross section was evaluated.

MR. OKRENT:

I believe you made a comment on the fact that isotropy should hold up to 7 Mev --

MR. CRANBERG:

To all practical purposes --

MR. OKRENT:

Could you define practical then?

MR. CRANBERG:

That would really be presumptuous. Well, 20 per cent is unlikely. The reason I am hesitating is that we just measured the excitation to first level of Pb^{206} at 3 Mev and got a 2 to 1 isotropy. I think that the 206 situation is exceptional, it is only on the first excited state. The indication is that for the second and third excited states of Pb^{206} there is no appreciable anti-isotropy.

MR. TERRELL:

That last slide that you showed, Larry, showing the values of "a" as a function of "A," that change of the "a" toward the Fermi gas model as you increase the excitation, isn't that just what you would expect on the Hurwitz idea?

MR. CRANBERG:

I think you would expect it on any idea.

MR. DE SAUSSURE:

Would you care to comment on the region applicable to the socalled Snell experiment?

MR. CRANBERG:

I heard about this connection very recently and the tie-in was made by someone here at Argonne. I think whoever that person is, is much better qualified to comment than I am.

MR. HUMMEL:

There does seem to be a painful discrepancy between calculated and measured values of the diffusion length in the Snell experiment when the Cranberg inelastic scattering data are used. It would be presumptuous on our part to say that it had anything to do with the inelastic scattering measurements, however, as there are uncertainties in the analysis.

MR. WIGNER:

The diffusion length in the Snell experiment is perhaps a little difficult to interpret. But the total amount of secondary fissions seems to be clear and there are also the calculations in general of the total amount of the so-called fast effect, and they don't involve a good agreement with inelastic cross sections as obtained before. Something should be done on this question.

MR. CRANBERG:

If I had shown a comparison of Beyster's results with ours for what he called the inelastic cross section which represents an extrapolation corresponding to the case where you really resolve all the inelastic scattering from the elastic scattering, his result at 1 Mev for U^{238} is 1.65 barns and ours is 2 barns, plus or minus a few tenths. You see the data that I showed is the case where you lumped the first two levels of the elastic; if we try to get the maximum information out of it, if we try to resolve the inelastic completely, we are, if anything, high. So it seems to me that the discrepancies hinge largely on the role of these states.

VOICE FROM FLOOR:

With regards to the relation of this low inelastic scattering to this Snell experiment, I might point out that for that type of experimental use of the data, probably the old Beyster data is good enough, because one is talking about events that take place in a relatively small distance of uranium even for the multiple scattering effects and what you are talking about is an effective removal cross section from a group, which is properly given by Beyster's data.

MR. CRANBERG:

It seems to me if what is involved is the calculation which is based on microscopic cross sections, then if our data don't give the right answer there is something wrong with our data or the analysis.

MR. SMITH:

We have measured U^{238} scattering of 550 kev neutrons. If anything, your answer from the two plus level is 8 to 10 per cent higher than ours. This holds true right straight across, right up to 1.4 Mev, so I have a reason to believe that, if anything, your measurements are a little higher than ours, rather than too low. Thorium, according to our work, is about 10 to 12 per cent lower than that of U^{238} , from 550 kev to 1.4 Mev.



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SESSION II

Monday Afternoon, October 19, 1959

Chairman: W. J. MacCarthy Secretary: D. Meneghetti

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ON THE ESTIMATION OF FAST NEUTRON CROSS SECTIONS P. Moldauer, ANL
A COMPUTATIONAL SURVEY OF IDEALIZED FAST BREEDER REACTORS W. H. Roach, LASL
THE SENSITIVITY OF BREEDING RATIO IN FAST REACTORS TO UNCERTAINTIES IN CROSS SECTIONS D. Okrent, ANL
BREEDING: INTERNAL OR EXTERNAL R. M. Kiehn, LASL
PARAMETRIC ANALYSIS OF A PuO ₂ -UO ₂ -FUELED FAST BREEDER P. Greebler and P. Aline, G. E., San Jose

After Dinner Speaker:

Dr. Harold B. Finger, Chief, Nuclear Engine Programs National Aeronautics and Space Administration. "The Applications of Nuclear Reactors in Space."*

^{*}Not reproduced in proceedings.



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Peter A. Moldauer Argonne National Laboratory Lemont, Illinois

I should like to discuss the problem of estimating inelastic fast neutron cross sections which may be needed in reactor calculations but which have not yet been measured and may be difficult to measure.

These considerations will be restricted to neutron energies between a few hundred electron volts and several Mev and to target nuclei which are not too light (i.e., A>50) and not too magic. Under these conditions it appears justified to assume non-elastic neutron-induced reactions to proceed largely through compound nucleus formation; the associated cross sections will then consist of contributions from local resonances. One is here, however, not interested in the details of the resonance structure but in the energy average - taken over many resonances - of the cross section. Performing such an average over Breit - Wigner resonances for the cross section describing a non-elastic process x initiated by a neutron, one obtains the well-known expression:(1)

$$\langle \sigma_{nx} \rangle = \frac{\lambda^2}{2} \sum_{I j J} g_J \langle \rho_J \rangle \langle \frac{\Gamma_n^{I j J} \Gamma_x^{J}}{\Gamma^J} \rangle , \qquad (1)$$

where λ is the incident neutron wave length, 1 refers to the orbital angular momentum of the neutron, j to the combined spin of neutron and target nucleus, and J to the total angular momentum of the system. The factor g_J depends, besides J, on the target spin I; further, ρ_J is the density of compound nucleus resonance levels with angular momentum J and the proper parity. The total width of a spin J resonance is denoted by Γ^J and the partial widths for the neutron emission and the reaction channels are denoted by Γ_n^{IjJ} and Γ_x^{J} , respectively. The reaction channels x of principal interest here are radiation (γ), inelastic neutron emission (n'), and fission (f). All brackets in eq. (1) indicate an average over many resonances. The last bracket in eq. (1) can be expressed as a certain function of the average widths $\langle \Gamma_n^{IjJ} \rangle \langle \Gamma_x^{J} \rangle$ and $\langle \Gamma^{J} \rangle$.⁽²⁾ The form of this function depends on the statistical fluctuations of all the widths within the averaging interval. A code has been prepared for the GEORGE computer⁽³⁾ to calculate such functions and the average cross sections (1).

To perform these calculations one must have information on the average level densities and the averages and fluctuations of the widths. The best source of these parameters is, of course, actual measurements.
Relevant types of experiments are neutron resonance scattering experiments using fast choppers and other velocity selectors, average total neutron cross-section measurements, and average neutron capture, inelastic, or fission cross-section measurements. In cases where adequate data are not available to define the required parameters for a nuclide and energy range of interest, it is often possible to use the parameter systematics revealed by measurements on similar nuclides combined with theoretical predictions to obtain estimates.

I should like to review briefly the amount, quality and sources of information on resonance parameters and what this information reveals regarding the systematic variations of these parameters. Almost all chopper and velocity selector results are summarized in BNL-325.⁽⁴⁾ There are listed some fifty nuclides for which four or more adjacent s-wave neutron resonances have been observed, from which one obtains quite accurate values of the s-wave neutron resonance level density at neutron binding. These results can easily be systematized in the form of a Fermi gas model with single nucleon level spacings of the size expected in a potential well of nuclear radius.⁽⁵⁾ Unfortunately, the nuclear level spacings are extremely sensitive to changes in the parameters of such a model, so that even the most refined estimates (6) cannot be expected to yield predictions to within better than a factor of two. As a result, no reliable estimates of average reaction cross sections can be made without a measured resonance level density, except perhaps in cases where one averages over cross sections for a large number of nuclides and the uncertainties in the individual level densities of the nuclide may be expected to average out. It is further necessary to know the dependence of the level density upon J and energy. Experimental information on these comes mainly from average reaction cross sections and particle spectra. Theoretical treatments and analyses of data have been given by a number of authors.⁽⁷⁾ As a result, it is to be expected that the level density behaves like (2J+1) exp $\left[-(J+\frac{1}{2})^2/2\tau^2\right]$, where τ is of the order of 4 or greater. The energy dependence of ρ is more complicated. One may expect roughly a ten per cent increase in the level density for every 100-kev-increase in excitation energy at neutron binding for the heavier nuclei and a smaller increase for the lighter nuclei.

The average s-wave neutron width has been obtained for some 60 nuclides by resonance scattering experiments, ⁽⁴⁾ as well as by analyses of average total neutron cross sections⁽⁸⁾ which yield the s-wave neutron strength function $\langle \rho \rangle \langle \Gamma_n^{oJ} / \sqrt{E_n} \rangle$, where E_n is the neutron energy. This strength function has been of interest in the construction of complex well models for neutron scattering.⁽⁹⁾ The fluctuations in neutron widths are well known and follow the Porter-Thomas distribution.⁽¹⁰⁾ So far little is known of the values of the neutron widths for higher orbital angular momenta. Results obtained from the analyses of capture cross sections will be described a little later. The widths for neutron emission to inelastic levels are almost totally unknown. Their determination will have to await a larger amount of data on inelastic neutron scattering.

A large number of total radiation widths have been measured.⁽⁴⁾ Attempts to systematize the measured values have been made.⁽¹¹⁾ However, a theoretical understanding of total radiation widths has lagged behind that of the other parameters. Fortunately, variations in the values of Γ_{γ} are considerably less than those of $\langle \rho \rangle$ and $\langle \Gamma_n \rangle$. Except for the lighter nuclei, almost all measured values of $\langle \Gamma_{\gamma} \rangle$ are within a factor of ten of each other. Methods of determining $\langle \rho \rangle \langle \Gamma_{\gamma} \rangle$ from the analysis of average reaction cross sections will be discussed later. It is thought - and this conforms with most data - that for a given nuclide Γ_{γ} does not fluctuate appreciably from resonance to resonance.⁽¹⁰⁾ A formula for the energy dependence of $\langle \Gamma_{\gamma} \rangle$ has been given.⁽¹²⁾

Measurements of resonance parameters of four fissionable isotopes are listed in BNL-325. Again, such parameters, and in particular $\langle \rho \rangle \langle \Gamma_f \rangle$, can be obtained from analysis of fast neutron reaction cross sections, such as has been done by Rae, Margolis and Troubetzkoy.⁽¹³⁾ Analysis of the fluctuations in fission widths indicate that these are distributed according to a chi-squared distribution law with between two and four degrees of freedom.^(10,14)

As an example of the use of fast neutron reaction cross-sectional data in obtaining resonance parameters, I should like to discuss briefly the analysis of capture cross sections. How this depends on the resonance parameters is best seen by forgetting for the present about the effects of width fluctuations, the energy dependence of ρ or Γ_{γ} , and the remission of inelastic neutrons or neutrons with changed orbital angular momentum. Equation (1) gives then for the average capture cross section of neutrons with orbital angular momentum 1 the following limiting expressions:

$$\overline{\sigma}_{I} \sim \frac{\lambda^{2}}{2} \sum_{jJ} g_{J} \langle \rho_{J} \rangle \langle \Gamma_{n}^{1 j J} \rangle \text{ for } \langle \Gamma_{n}^{1 j J} \rangle \langle \Gamma_{\gamma}^{J} \rangle ; \qquad (2a)$$

$$\overline{\sigma}_{1} \sim \frac{\lambda^{2}}{2} \sum_{jJ} g_{J} \langle \rho J \rangle \Gamma_{\gamma}^{J} \text{ for } \left\langle \Gamma_{n}^{1 j J} \right\rangle >> \Gamma_{\gamma}^{J} \quad .$$
(2b)

We assume Γ_{γ}^{J} to be energy independent and $\langle \rho_{J} \rangle \langle \Gamma_{n}^{1 j J} \rangle$ to vary with energy as the neutron penetrability P_{ℓ} , which increases with neutron energy roughly as $E_{n}^{1+\frac{1}{2}}$ below about 100 kev and more slowly at higher energies. Then we see that eq. (2a) applies to the low-energy limit where, accordingly, $\overline{\sigma}_{\ell}$ varies as $E_{n}^{1-\frac{1}{2}}$, and eq. (2b) applies to the high-energy limit where $\overline{\sigma}_{l}$ varies as E_{n}^{-1} . The transition, containing a maximum in $\overline{\sigma}_{l}$ (except for l = 0), occurs in the region where $\langle \Gamma_{n}^{1 j J} \rangle \sim \Gamma_{\gamma}^{J}$. The qualitative features of this behavior are illustrated in Fig. 1, where $\overline{\sigma}_0$ is seen to change from an $E_n^{-\frac{1}{2}}$ to an E_n^{-1} behavior, while $\overline{\sigma}_1$ changes from $E_n^{\frac{1}{2}}$ to E_n^{-1} , etc. The ratio of the saturated (E_n^{-1}) values of $\overline{\sigma}_0$ and $\overline{\sigma}_1$ in Fig. 1 varies from five for I = 0 to about two for high spin targets, assuming $\langle \rho_J \rangle \Gamma_\gamma^J$ proportional to 2J+1. On the abscissa are indicated typical locations of the 10 and 100-kev points with respect to the features of the graph. The energy dependence of the total capture cross section is seen to reflect the successive entrances of p and higher orbital waves. Since Γ_γ^J can usually be assumed to be known at least approximately, the energy at which $\overline{\sigma}_1$ enters depends principally on $\langle \Gamma_n^{1jJ} \rangle$ or, better, on the 1-wave neutron strength function $\langle \rho_J \rangle \langle \Gamma_n^{1jJ} \rangle / 2P_1$. This picture is modified increasingly with increasing energy by the effects that have been neglected above. Competition with inelastic and neglected



Fig. 1. Schematic Behavior of Energy Dependence of σ_1 .

elastic scattering will tend to make the cross section fall off more sharply while the increasing level density tends to increase the cross section. At neutron energies above a few hundred kilovolts it is often difficult to unsnarl the contributions of so many factors to the observed energy dependence of a capture cross section. But at lower neutron energies the shape of the cross section is quite sensitive to the p-wave and often d-wave strength functions as well as the level density law and the presence of the inelastic levels.

Some fifteen nuclides for which good data of capture cross sections over a wide range of neutron energies between one and several hundred kilovolts are available have been analysed using the abovementioned code. A typical example

is shown in Fig. 2, which is an Oak Ridge drawing⁽¹⁵⁾ representing a summary of the capture cross-section data for iodine compared to the expected s-wave capture cross section. The line through the points shows my fit of the data using reasonable parameters. The effects of p and of d-wave capture, as well as those of the inelastic levels at 57 and 203 kev, are apparent. A summary of the information on p-wave strength functions obtained so far is shown in Fig. 3, where it is compared with the predictions of the black nucleus model and a simple square well complex potential model. The results tend to be in qualitative agreement with the latter model.



Fig. 2 Theoretical Fit of the Fast Neutron Capture Cross Section in Iodine. Experimental Data by R. L. Macklin <u>et al.(15)</u>



Fig. 3 Neutron p-wave Strength Functions Reduced from Fast Neutron Capture Cross Sections

The apparent splitting of the peak at atomic number 100 has also been noted by Newson⁽¹⁶⁾ and attributed to an expected spin orbit force. Results on the d-wave strength function, though sparse, also tend to agree with a complex well model. This work is in mid-stream right now since large amounts of new capture cross-section data have recently become available.⁽¹⁷⁾

Similar discussions apply to the analysis of inelastic neutron cross sections and fission cross sections, and analyses of such data are now underway. The analysis of the U^{235} fission, inelastic and capture cross sections by Rae, Margolis and Troubetzkoy⁽¹³⁾ has already been mentioned.

The code for the evaluation of eq. (1) has been used, in combination with the above-mentioned information on resonance parameters, for the estimation of a number of fast neutron cross sections. I would like to restrict myself today to reporting an estimate of the fission product capture cross section. For this purpose 47 separate calculations were performed to estimate the average capture cross sections of the 84 nuclides having yields of one atom or more per thousand atoms of Pu²³⁹ fissioned after irradiation of plutonium for one year by 2-Mev neutrons.⁽¹⁸⁾ Of these 84 nuclides there are four, representing 3 per cent of the total yield, for which detailed fast neutron capture data were available and which were included in the previously discussed resonance parameter analysis. For another eleven nuclides, representing 16 per cent of the yield, information on s-wave resonance parameters, particularly level spacings, was available. Where measured parameters were not available, they were estimated in the following way. For the radiation widths the systematic trend described by Hughes and Zimmerman⁽¹¹⁾ was followed. For neutron widths the observed trend in the s-wave strength function (as presented in Ref. 11) and complex well models for the higher 1 waves, as modified by the abovementioned results, were used. Black nucleus widths were used for inelastic neutron emission. For the unmeasured level densities, a two Fermi gas model formula, similar to that of Ross,⁽⁵⁾ was employed, taking shell model effects into account. The value of the parameter τ in the expression for the J dependence of ρ was taken to be eight. Fast neutron capture cross-section measurements relevant to nuclides representing 36 per cent of the yield were available for comparison with the calculation. In most cases, however, they were in the form of elemental cross sections representing quite different isotopic compositions from those obtaining in the fission products.

The results are shown in Fig. 4, where the fission product capture cross sections, $\sum_{i} Y_{i} \sigma_{i} / \sum_{i} Y_{i}$ (the σ_{i} are the isotopic cross sections and the Y_{i} the isotopic yields) are plotted against neutron energy from 100 ev to several Mev. The calculations have been carried to only 1 Mev. The curve shown above 1 Mev represents an out-and-out guess based on the observed general trend of capture cross sections of some 25 nuclides in that region.⁽¹⁹⁾ The curve is shown dashed above 200 kev to indicate the decreased reliability of the calculations at higher energies because of the badly known energy

dependence of the level density. For comparison, older estimates of the fission product capture cross sections by Greebler, Hurwitz and Storm⁽²⁰⁾ and by Businaro, Gallone and Morgan⁽²¹⁾ are shown.



Fig. 4. Fission Product Capture Cross Section $\sigma = \frac{\Sigma Y_i \sigma_i}{\Sigma Y_i}$

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Discussion of Paper Presented by Mr. Moldauer

MR. BOLLINGER:

If I understood your treatment correctly, in the calculations of the capture cross section you did not take into account the effect of the widening of the total width by the inelastic processes. Now, we just had a talk which you could not hear, because you were sick, in which it was claimed that it was essential to take this into account. I wonder if you have a comment on this.

MR. MOLDAUER:

I think I mentioned when I drew the picture (Fig. 1) that this is modified by inelastic scattering. This effect must certainly be added.

MR. BOLLINGER:

You feel that it is an important effect.

MR. MOLDAUER:

Oh, extremely important. In the calculations the effect of the inelastic levels has been taken into account, also the effects of fluctuations in the widths and the energy dependence of the level density.

MR. WIGNER:

Did you try to calculate the p-wave strength function by the forward bias of the inelastic scattering?

MR. MOLDAUER:

No. Do you know of such an analysis?

MR. WIGNER:

We did calculate it by the forward bias of scattering, which is perhaps a little easier because, as you point out, all the parameters of the s-wave cross section vary. It is rather difficult to tell in which way they vary. They are not as smooth as one would like them to be. It is rather difficult to attribute part of the total cross section to p-waves and part of it to s-waves. It is difficult to get measurements that accurate. We thought that the angular distribution would be the direct way to obtain it and we did get maxima, but I must admit that our results for the p-wave strength function also showed considerable fluctuations.

MR. MOLDAUER:

Thank you, I am very interested in this. I might just mention that in the region of some tens of kilovolts the s-wave capture cross section is already essentially saturated. The neutron width is large compared to the gamma width and therefore the result is not very sensitive to the s-wave strength function. It is sensitive only to the product of radiation width and level density and the p-wave strength function. The s-wave strength function does not really affect it much. Of course the dependence on radiation width and level density is a drawback. That is true.

A COMPUTATIONAL SURVEY OF IDEALIZED FAST BREEDER REACTORS

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The paper to be presented is excerpted from a more comprehensive article to appear in Nuclear Science and Engineering and gives some results of multigroup calculations on a series of idealized fast breeder reactors. A few remarks on the multigroup specifications used in these calculations will be followed by some comparisons between experimental and computed systems. The main body of the paper gives the extent of the calculational survey and some of the results thereof.

All of the systems in the survey were set up as critical radii problems and were solved using the S_4 approximation in the SNG code for the IBM-704. The machine print-out is detailed enough so that one can obtain essentially all of the relevant parameters regarding the computed system, including the initial breeding ratio.

The energy spectrum was broken up into sixteen groups as shown in Table I. This choice of energies was made so that one set of cross sections could be used for both fast systems and those of intermediate energies. Results of the present calculations show that a quite small fraction of the total flux is below group 9, that is, in the region where resonance effects of the fuel and fertile materials are most strongly felt.

Group	Energy Range	∆u	v(cm/shake)	x Fission Spectrum	
1	3 - ∞ Mev		28.5	0.204	
2	1.4 - 3 Mev	0.762	19.9	0.344	
3	0.9 - 1.4 Mev	0.442	14.7	0.168	
4	0.4 - 0.9 Mev	0.811	11.0	0.180	
5	0.1 - 0.4 Mev	1.386	6.7	0.090	
6	17 - 100 kev	1.772	2.70	0.014	
7	3 - 17 kev	1.735	1.14	0	
8	0.55 - 3 kev	1.696	0.480	0	
9	100 - 550 ev	1.705	0.206	0	
10	30 - 100 ev	1.204	0.101	0	
11	10 - 30 ev	1.099	0.0566	0	
12	3 - 10 ev	1.204	0.0319	0	
13	1 - 3 ev	1.099	0.0179	0	
14	0.4 - 1 ev	0.916	0.0109	0	
15	0.1 - 0.4 ev	1.386	0.00606	0	
16	Thermal (0.025)		0.00218	0	

TABLE I

Nevertheless, in the calculations, resonance shielding in all fissile and fertile materials was taken into account. A more detailed explanation of resonance effects and generation of sixteen-group cross sections will be found in the previously mentioned article.

The uncertainties in existing data on cross sections and in reducing these data to a multigroup set of cross sections lead, typically, to approximately a 5% uncertainty in computed radii of fast neutron critical spheres. Small adjustments to the multigroup cross sections as dictated by comparison between computations and experiment leads to the set used in these calculations, which predict radii of fast metal systems to about 2% and yet do not violate extant data on cross sections. Table II shows some comparisons between experimental and computed radii for critical and near-critical assemblies. Those assemblies marked with a superscript have been used in the adjustment procedure just mentioned. Quite good agreement between experiment and calculation is obtained for assemblies using U^{235} and U^{238} ; since there is a wealth of experimental data on such systems and many calculations, one can place reasonable confidence in calculations on proposed systems using these materials. Less confidence is felt in calculations on systems using Pu^{239} , where the experimental data are less, and on systems using U^{233} and Th^{232} , where essentially no critical data are available. Given in the table is a comparison between measured and computed values of the spectral index, the ratio of the fission rate of U^{235} to U^{238} . Good agreement is noted for fast metal $U^{235} - U^{238}$ systems, with only fair agreement for diluted $U^{235} - U^{238}$ assemblies and for Pu^{239} assemblies. The subcritical assemblies were extrapolated to critical, with good agreement between extrapolated and calculated radii. Such experimental checks as these give primarily a check on the fission and capture cross sections of the fissile and fertile materials used, since these cross sections govern, to a large extent, the neutron economy in the S_n calculations.

In the survey, three types of reactors are considered: 1) U^{235} depleted uranium; 2) Pu²³⁹-depleted uranium; and 3) U²³³-thorium. All of these are taken as homogeneous spherical cores, composed of fissile, fertile, structural, and coolant materials, surrounded by a 50-cm thick reflector of fertile, structural, and coolant material. The core composition is varied from one calculation to the next, while the reflector always contains 40 v/o fertile material, 40 v/o coolant (represented by sodium), and 20 v/o structural material (represented by stainless steel). Sodium and stainless steel (ss) are present in the core in one of the fixed volume percentages as follows: 1) 0, 0, i.e., no sodium or ss; 2) $33\frac{1}{3}$ v/o Na, $16\frac{2}{3}$ v/o ss; or 3) 50 v/o Na, 25 v/o ss. The three reactor types given represent the bulk of the survey, but several other systems are investigated to some extent. Thorium replaces the depleted uranium in several of the Pu-U systems, and a few of the U²³³-thorium reactors are calculated using a secondary reflector of 60 cm of graphite. Table III gives the reactors computed and the factors affecting breeding ratios.

TABLE II

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Computed vs	Experimental	Parameters	of Various	Assemblies

	Radius of spherical	equivalent core – cm	$\overline{\sigma}_{f}(U^{235})/at ce$	σ _f (U ²³⁸) nter
	Exp	Calc	Exp	Calc
U ²³⁵ - U ²³⁸ undiluted				
Godiva (bare Oy-94%) ^a Topsy (Oy-94% in thick U) ^a Jemima Oy-53-1/2% (bare cyl) ^a Jemima Oy-37-1/2% (bare cyl) ^a Jemima Oy-29% (bare cyl) ^a Oy-16% in 3" U (cyl)	8.71 6.04 12.14 14.62 16.85 20.4	8.67 5.98 12.13 14.77 17.28 20.64	6.5 7.3 8.7 10.8 ~13. 18.2	6.4 7.0 8.6 10.5 12.4 18.3
U ²³⁵ - U ²³⁸ diluted				
ZPR-III, No. 6F (30 v/o Oy-46-1/2%) ^a ZPR-III, No. 9A (50 v/o Oy-23-1/2%) ZPR-III, No. 11 (81 v/o Oy-11-1/2%, cyl)	23.0 25.4 32.4	22.8 25.4 32.5	14.5 19.6 26.	12.0 17.1 24.3
Pu ²³⁹ - U ²³⁸ undiluted				
Jezebel (bare Pu) ^a Popsy (Pu in thick U) ^a	6.29 4.44	6.25 4.43	5.0 5.7	5.4 6.2
Pu-depleted U Pu ²³⁹ and U(depleted) plates in 7.5" U(depleted), (cyl)	29.85	31.06	-	12.6
Pu ²³⁹ - Th ²³² composite				
Pu ²³⁹ and Th ²³² plates in 4.5" U(depleted), (cyl) Pu ²³⁹ and Th ²³² plates in 7.5" U(depleted), (cyl)	21.85 29.08	22.02 29.50	-	9.2 9.3
U ²³³ and Th ²³² (subcritical)				
10 kg U^{233} in U 10 kg U^{233} in Oy-93% 7.5 kg U^{233} in U 7.5 kg U^{233} in Oy-93% Oy-94% in 1.8" Th	5.04 5.04 4.60 4.60 7.80	5.00 5.02 4.55 4.55 7.64	- - - -	5.3 5.2 5.5 5.2 6.5

^a Used for adjustments of S_n parameters within their ranges of experimental error.

Syste Reac-	System	/stem with 50 cm Std Refl ⁽¹⁾ Core (in v/o)			Fuel Absorption,	Diluent Absorption, Core and	Leakage, System	Fertile Capture, Core and Befl	Conversion Ratio,	$\alpha = \left[\frac{Fuel Captures}{Fuel Fissions}\right]$
	U ^{2 3 5}	U ²³⁸	38 Na Fe ⁽²⁾ Core Reflector		Bystem	(Conversion Ratio)	Refl			
703	8	92	0	0	1.000	0.017	0.066	1.594	1.660	0.166
704	16	84	0	0	1.000	0.035	0.208	1.417	1.625	0.155
705	30	70	0	0	1.000	0.042	0.296	1.296	1.592	0.144
706	60	40	0	0	1.000	0.046	0.355	1.195	1.550	0.130
707	100	0	0	0	1.000	0.048	0.380	1.134	1.514	0,120
708	4	46	33-1/3	16-2/3	1.000	0.051	0.030	1.449	1.479	0.185
709	8	42	33-1/3	16-2/3	1.000	0.055	0.151	1.349	1.500	0.169
710	15	35	33-1/3	16-2/3	1.000	0.054	0.241	1.263	1.504	0.155
711	30	20	33-1/3	16-2/3	1.000	0.053	0.313	1.181	1.494	0.139
712	50	0	33-1/3	16-2/3	1.000	0.051	0.351	1.128	1.479	0.128
713	20	13-1/3	50	16-2/3	1.000	0.056	0.283	1.176	1.459	0.146
714	3	22	50	25	1.000	0.093	0.055	1.228	1.283	0.209
715	4	21	50	25	1.000	0.086	0.090	1.229	1.319	0.192
716	7-1/2	17-1/2	50	25	1.000	0.072	0.180	1.194	1.374	0.171
717	15	10	50	25	1.000	0.063	0.258	1.152	1.410	0.152
718	25	0	50	25	1.000	0.058	0.301	1.120	1.421	0.139

TABLE III A

(1) Standard Reflector 40 v/o depleted U 40 v/o Na

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20 v/o Fe, density adjusted to simulate stainless steel

(2) Fe density adjusted to simulate stainless steel

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Reac-	System	System with 50 cm Std Refl ⁽¹⁾ Core (in v/o)			Fuel Absorption,	Diluent Absorption,	Leakage,	Fertile Capture, Core and	Breeding Ratio,	$\alpha = \begin{bmatrix} Fuel Captures \\ Fuel Fissions \end{bmatrix}$
tor	Pu ²³⁹	U ²³⁸	Na	Fe ⁽²⁾	Core	Reflector	System	(Breeding Ratio)	Refl	[fuel fissions]
719	8	92	0	0	1.000	0.039	0.207	2.108	2.315	0.097
720	16	84	0	0	1.000	0.057	0.378	1.911	2.289	0.087
721	30	70	0	0	1.000	0.062	0.484	1.787	2.271	0.076
722	60	40	0	0	1.000	0.064	0.562	1.687	2.249	0.065
723	100	0	0	0	1.000	0.064	0.603	1.633	2.236	0.057
724	4	46	33-1/3	16-2/3	1.000	0.071	0.129	1.980	2.109	0.117
725	8	42	33-1/3	16-2/3	1.000	0.074	0.294	1.855	2.149	0.100
726	15	35	33-1/3	16-2/3	1.000	0.073	0.411	1.762	2.173	0.086
727	30	20	33-1/3	16-2/3	1.000	0.071	0.504	1.683	2.187	0.073
728	50	0	33-1/3	16-2/3	1.000	0.069	0.556	1.637	2.193	0.064
729	20	13-1/3	50	16-2/3	1.000	0.075	0.464	1.680	2.144	0.079
730	2	23	50	25	1.000	0.127	0.049	1.735	1.784	0.158
731	4	21	50	25	1.000	0.104	0.187	1.742	1.929	0.125
732	7-1/2	17-1/2	50	25	1.000	0.090	0.315	1.705	2.020	0.103
733	15	10	50	25	1.000	0.080	0.427	1.662	2.089	0.084
734	25	0	50	25	1.000	0.076	0.493	1.632	2.125	0.073
730A	2	23(3)	50	25	1.000	0.129	0.014	1.772	1.786	0.158
758	2	23(4)	50	25	1.000	0.130	0.095	1.366	1.461	0.152
759	2	23(5)	50	25	1.000	0.146	0.020	1.425	-	0.152
					(Cab	sorption inclue	ded)			

TABLE III B

(1) Standard Reflector: 40 v/o depleted U

40 v/o Na

20 v/o Fe, density adjusted to simulate stainless steel

(2) Fe density adjusted to simulate stainless steel

(3) 71 cm standard reflector

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(4) U^{238} replaced by Th²³² in core; depleted U replaced by Th²³² in reflector

(5) U²³⁸ replaced by Th²³² in core; depleted U replaced by Th²³² in reflector; 60 cm C blanket enclosing system

TABL	٦E	11	I	С

Reac- tor	System with 50 cm Std Refl(1) Core (v/o)			Fuel Absorption,	Diluent Absorption,	Leakage,	Fertile Capture, Core and Patl	Breeding Ratio,	$a = \left[\frac{\text{Fuel Captures}}{\text{Fuel Fissions}} \right]$	
tor	U ^{2 3 3}	Th ^{2 3 2}	Na	Fe ⁽²⁾	Core	Reflector	System	(Breeding Ratio)	Refl	[ruel fissions]
752 735 736 737 738 739 753 740 741 742 743 744 745 754 746 747	$ \begin{array}{c} 8\\ 12\\ 24\\ 36\\ 60\\ 100\\ 4\\ 6\\ 12\\ 18\\ 30\\ 50\\ 20\\ 2\\ 3\\ 6\\ 9\end{array} $	92 88 76 64 40 0 46 44 38 32 20 0 13-1/3 23 22 19 16	$\begin{array}{c} 0\\ 0\\ 0\\ 0\\ 0\\ 33-1/3\\ 33-1/3\\ 33-1/3\\ 33-1/3\\ 33-1/3\\ 33-1/3\\ 50\\ 50\\ 50\\ 50\\ 50\\ 50\\ 50\\ 50\\ \end{array}$	0 0 0 0 0 0 16-2/3 16-2/3 16-2/3 16-2/3 16-2/3 16-2/3 16-2/3 16-2/3 25 25 25 25 25	1.000 1.000	$\begin{array}{c} 0.034\\ 0.042\\ 0.044\\ 0.048\\ 0.048\\ 0.047\\ 0.061\\ 0.059\\ 0.056\\ 0.056\\ 0.055\\ 0.055\\ 0.053\\ 0.058\\ 0.108\\ 0.095\\ 0.077\\ 0.069\end{array}$	$\begin{array}{c} 0.291 \\ 0.394 \\ 0.528 \\ 0.593 \\ 0.651 \\ 0.690 \\ 0.218 \\ 0.321 \\ 0.468 \\ 0.531 \\ 0.606 \\ 0.653 \\ 0.562 \\ 0.135 \\ 0.230 \\ 0.381 \\ 0.452 \end{array}$	1.177 1.076 0.958 0.904 0.860 0.837 1.176 1.091 0.984 0.932 0.877 0.849 0.899 1.147 1.089 0.996 0.955	1.468 1.470 1.485 1.498 1.511 1.526 1.394 1.412 1.451 1.462 1.482 1.502 1.461 1.282 1.319 1.377 1.407	$\begin{array}{c} 0.067\\ 0.064\\ 0.059\\ 0.056\\ 0.053\\ 0.050\\ 0.075\\ 0.071\\ 0.064\\ 0.061\\ 0.056\\ 0.053\\ 0.059\\ 0.090\\ 0.083\\ 0.072\\ 0.067\end{array}$
748 749 750 755 751 756 757	9 15 25 8 12 4 2	10 0 92 88 46 23	50 50 50 0 33-1/3 50	$25 \\ 25 \\ 0(3) \\ 0(3) \\ 16 - 2/3(3) \\ 25(3)$	$ \begin{array}{c} 1.000\\ 1.000\\ 1.000\\ 1.000\\ 1.000\\ 1.000\\ 1.000\\ 1.000 \end{array} $	0.089 0.064 0.060 0.081(4) 0.103(4) 0.097(4) 0.130(4)	0.452 0.529 0.594 0.068 0.090 0.049 0.026	0.955 0.909 0.873 1.353 1.340 1.309 1.241	1.407 1.439 1.467 - - -	0.087 0.062 0.057 0.067 0.064 0.075 0.090

(1) Standard Reflector: 40 v/o Th²³² 40 v/o Na

20 v/o Fe, density adjusted to simulate stainless steel

(2) Fe density adjusted to simulate stainless steel

(3) 60 cm C blanket enclosing system

(4) C Absorption included

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Figure 1 shows the critical mass of U^{235} in the $U^{235}-U^{238}$ reactors as a function of v/o fuel in the fissile plus fertile material in the core, for three different volume fractions of inert diluents. The upper curve is for the most dilute systems investigated; the point at the upper left corresponds to 3 v/o of fissile material in a total core volume of ~3000 liters. The legend gives the concentration of inert diluents for each curve. It should be noted that the critical mass increases rapidly with decreasing v/o fuel in the minimum fuel region of the curves. This region is of particular interest since it is here that one obtains the highest breeding ratios for systems with finite reflectors and, as will be seen further on, the breeding ratios increase more slowly, percentage-wise, than the critical masses for the same change in fuel loading. The experimental points in the figure are from ZPR-III and LASL data; all of the ZPR-III assemblies and some of the Los Alamos assem-



Fig. 1. Critical Mass as a Function of Volume Fractions

blies have been converted to the types of systems studied here, the conversion process making use of the Topsy assembly dilution exponents. The points have been included chiefly to show the applicability of such dilution exponents in giving the relative worths of various materials in experimental or computed systems.

Figures 2 and 3 show the computed critical masses of various Pudepleted U and U²³³-thorium reactors. The one point in Fig. 2 near the upper curve where thorium has been substituted for the uranium is of some interest. The critical mass of Pu in this Pu-Th system increases by about 30% from the comparable system with uranium; as will be noted in the discussion on breeding ratios, the decrease of breeding ratio is about 20%. In Fig. 3, the critical mass of U²³³ in these systems is apparently less than that of Pu in the Pu-U systems of Fig. 2 for the same v/o fuel in the fissile plus fertile core material. This is due, of course, to the reduced density of thorium compared to the other fissile and fertile materials and in taking the abcissa as v/o rather than a/o.



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Fig. 2. Critical Mass in Pu-depleted U Reactors

Fig. 3. Critical Mass in Th-depleted U Reactors

Figure 4 gives the initial conversion ratios for $U^{235}-U^{238}$ systems. Each of the differently lettered curves is for reactors wherein the volume fraction of inert diluents in the core is constant, as explained in the legend; the numbered subscripts refer to systems with either a 50-cm or an infinite reflector. Thus, for example, curve A1 gives conversion ratios for systems having no Na or ss in the core, with a 50-cm reflector composed of 40 v/odepleted uranium, 40 v/o Na, and 20 v/o ss; curve A_2 gives the conversion ratios for systems of the same composition but with an infinite reflector of the same composition. The intersection point of the curves for finite and infinite reflectors gives the approximate minimum volume fraction of fuel for criticality. Comparing these curves with the critical mass curves, one sees that, in this general region of minimum volume fraction of fuel, the critical dimensions of the core are very large and the neutron leakages into the reflectors correspondingly small. Thus, increasing the thickness of the reflector has little effect on the conversion ratio, i.e., breeding is done mainly in the core. The conversion ratios decrease as the core becomes more highly diluted with inert diluents. This is due to increased parasitic capture in the inert diluents and to spectral softening, which decreases the fast fission contribution to the conversion ratio from the fertile U²³⁸. The



Fig. 4. Conversion Ratio in U²³⁵-U²³⁸ Systems

importance of leakage is indicated by the difference in conversion ratios for like systems having finite or infinite reflectors. The infinite reflector curves were generated, in fact, by addition of the finite system leakage term to the fertile capture term. Increasing the core fissile material at the expense of fertile material tends to decrease the conversion ratio, due to a reduction in fertile capture in the core. For highly dilute cores, however, where the core fuel to fertile ratio is small, this reduction is more than compensated for by the increase in the reflector fertile capture, as indicated by the rise in conversion ratio in the C_1 curve. Such overcompensation can be expected in such highly dilute systems where U^{238} is the fertile material, whereas, for systems using thorium, one would not expect it. The points identified with a (1), originating near the C_1

curve and rising in increasing value with volume fraction of fuel, are conversion ratios for systems whose cores are identical in composition to those in the C systems but which have a 50-cm reflector of fertile material (U^{238}) only. For such dilute systems it seems advantageous to concentrate the fertile material in the reflector.

The breeding ratios for Pu-U reactors, as shown in Fig. 5, are considerably greater than for the $U^{235}-U^{238}$ systems, due to the greater number of neutrons emitted per fission in Pu than in U^{235} . Other than this one feature, the curves are very similar to the ones in Fig. 4, even to the slight rise in breeding ratio exhibited in the lower curve. Substitution of thorium for the uranium decreases the breeding ratio by about 20%, as mentioned previously, due to the poorer fast fissioning properties of thorium compared to U^{238} . The two points near the bottom of the figure give breeding ratios for such a replacement: the lower point for a type-C system, the upper point for the same system plus 60 cm of graphite.

Figure 6 shows the breeding ratios for U²³³-Th reactors, which in general are much smaller than for either of the systems just discussed and quite different in shape for highly dilute systems. Thorium is, of course, inferior



Fig. 5. Conversion Ratio in Pu-U Systems



to U^{238} as a fast fissioner; in addition, the smaller density of thorium compared to U^{238} makes it a poorer reflector. It is obvious that 50 cm of reflector is insufficient here to prevent a large fractional loss of neutrons by leakage. (Compare the curves for finite and infinite reflectors.) Improvement would be noted in the breeding ratios if the reflector were thickened or if a secondary reflector were used, as shown by the points (1), (2), and (3) where a 60-cm secondary graphite reflector has been added.

The uncertainties in the computed cross sections for the inert diluents leads to the greatest error in calculated breeding ratio for the most highly diluted systems. However, as seen in Table III, increasing the parasitic capture by 50% gives less than a 5% decrease in breeding ratio, assuming that all increased capture by inert diluents comes from fertile capture.

An estimate has been made of how Pa^{233} in U^{233} -Th systems affects the initial breeding ratio. Two expressions for breeding ratio follow, the first called (B.R.)₀, which is the breeding ratio for a system where the effect of Pa^{233} is not considered, i.e., "Zero Power" systems.



In the first equation, the first term in the numerator gives the fuel contribution, the second term the fast fission contribution from the fertile material, and the third term is the parasitic capture in the inert diluents. The second equation gives the average breeding gain due to the Pa^{233} , where τ_P is the mean lifetime of Pa, about 39 days. In both equations leakage is neglected.

If one assumes that Pa^{233} has a fission cross section like Np^{237} and a capture cross section like Th²³² except at thermal energy, then one can estimate the change in initial breeding ratio for any flux level. One of the U^{233} -Th reactors in the survey has been used in the study of Pa, a low v/o fuel system, standard reflector, surrounded by 60 cm of graphite. The thick graphite provides a substantial thermal flux to the outer region of the primary reflector; since the thermal capture cross section of Pa²³³ is about 70 barns, and since the large amount of diluent reduces the flux fraction above the assumed fission threshold, this system, of all those calculated, should give a maximum loss in initial breeding ratio. Evaluation of the second equation for the system, using flux print-outs from the calculated problem, gives, for an assumed flux of 10^{16} at the center, a value of (B.R.) about $\frac{1}{2}$ % less than (B.R.)₀. If the core flux is assumed to have an average value of 10¹⁶, then the change is about $-l\frac{1}{2}\%$. The decrease is about 5% for a central flux of 10¹⁷. A flux of this magnitude is somewhat higher than those presently envisioned for fast breeders. One concludes that the effect of Pa²³³ on breeding ratio in fast reactors is small.

Discussion of Paper Presented by Mr. Roach

MR. WIGNER:

I was very interested at the accuracy with which you can calculate the critical radius. You can calculate it apparently to one-half a millimeter. Is that correct?

MR. ROACH:

I should have said that calculations on fast metal systems, which were used for purposes of adjusting cross sections have given results which agree with experimental values within about two per cent.

MR. WIGNER:

A half a millimeter?

MR. ROACH:

Yes, the agreement in Table II is good to half a millimeter in most cases. Such results might be expected, however, since agreement between experiment and calculation is required in the procedure for adjusting the cross sections.

MR. WIGNER:

Could you say something more about this, how you obtained the cross sections ?

MR. ROACH:

It is quite a lengthy procedure. I would be very happy to show you what we have afterwards, if you care to see it.

MR. MacCARTHY:

I had a question of my own about the values of alpha for plutonium which you used. There is quite a selection available.

MR. ROACH:

I do not have the alpha values for plutonium listed. However, in comparing our dilute plutonium system calculations to those of Loewenstein and Okrent on similar systems, we noted best agreement between our breeding ratio values and theirs where their "low α " values for plutonium had been used. So I would say that our plutonium α values probably compare with the "low α " set given in Loewenstein and Okrent's Geneva paper.

VOICE FROM FLOOR:

Will they be in the paper?

MR. ROACH:

The alpha values for the computed systems will be given in the paper. Alpha for plutonium can be obtained from the Nuclear Science and Engineering article referred to previously.

MR. WIGNER:

Could you tell us why the breeding ratio increases with increasing U^{233} concentration? In Fig. 6 you had the breeding ratio plotted against U^{233} concentration in thorium. It increased with increasing concentration. What is the effect that is essential here?

MR. ROACH:

The curves you refer to are those for systems having an infinite reflector, where it has been assumed in the calculations that all neutrons formerly lost through leakage, for the finite reflector case, are now captured in the thorium. So what one is looking at here is the sum of the fertile capture rate, decreasing in the core and increasing in the reflector as fuel replaces fertile material in the core, and the leakage rate which increases with greater core fuel concentration.

MR. MacCARTHY:

Isn't the reactor getting small going up that hill? It is a spectrum effect probably. There is more thorium fission, probably, which in the particular definition of conversion ratio used by Mr. Roach makes the breeding ratio go higher.

MR. WIGNER:

You assume that the neutrons which leak out are completely utilized.

MR. ROACH:

Right. As I mentioned, this is not exactly true. For an infinite reflector of the same composition as those listed for the finite reflected cases, about 95% of the neutrons are captured in the thorium. For the graphite-reflected systems, about 80% of the neutrons returning from the graphite are captured in fertile material.

MR. OKRENT:

It seems to me that the rise is a spectrum effect. In general, the contribution of thorium fissions to breeding ratio (always a small number) is larger as the number of thorium atoms per fissionable atom goes up. Hence, it is not an increase in the number of fissions in thorium that is doing this. However, as you put more and more U^{233} into the core per everything else, the number of parasitic captures in sodium, in iron, and in U^{233} itself goes down. This will give you a higher breeding ratio. Also ν will go up as the fission energy goes up. The rising ν and decreasing alpha with fission energy in the U^{233} itself would have to be the reason for the rise in the first case, where only thorium and U^{233} were present.

MR. ROACH:

It is correct that ν for U^{233} increases, and α decreases, as the core fuel becomes more concentrated. For the C₂ curve in Fig. 6, ν rises from a value of about 2.55 for the most dilute system to about 2.63 for the most concentrated system, while α drops from 0.090 to 0.057. These two effects are possibly enough to account for the exhibited rise in breeding ratio.

MR. HAEFELE:

You calculated quite a lot of fast breeders. Did you employ for all of these calculations the same set of group constants or did you recalculate for each breeder according to the different spectra as a group constant?

MR. ROACH:

I mentioned this in the first part of the talk, concerning resonance effects. Is this what you are concerned about ?

MR. HAEFELE:

Well, not really resonance effect, but, according to these contributions, you can have different spectrums varying from 100 kev up to say 300 kev.

MR. ROACH:

Very true. We have used the same set of cross sections throughout.

MR. HAEFELE:

Well, of course, of the input cross section, but I mean with respect to the group constants here an average spectrum process is included. Of course, I suppose that you have always employed the same set of microscopic input data, but in addition to these input data, do you use a spectrum to average them in order to obtain the group constants? Have you employed always the same spectrum in averaging these input microscopic crosssection data? Did you employ for every breeder a different averaging spectrum?

MR. ROACH:

The only spectrum we used is the fission spectrum that I showed in Table I.

MR. HAEFELE:

Well, I think there must be a point of misunderstanding, because in averaging these microscopic input data, you have to employ the finer spectrum in the reactor.

MR. WIGNER:

May I try to explain Dr. Haefele's point? He says that there is a variation of the neutron flux within one group and this variation is different in different cases that you consider. The question is: did you take that into account or, did you just calculate within the group an average cross section, the same for every reactor?

MR. ROACH:

In the computation of the cross sections we did calculate an average value throughout each energy region. These were used as primary input data to the SNG calculations. Inasmuch as the cross-section sets had been adjusted to give proper results for a variety of critical assemblies whose spectra ranged from fast to epithermal, it was felt that further adjustment of the cross sections to take into account the variation in spectra exhibited by the breeders considered here was unnecessary.

MR. HAEFELE:

So if I understand you correctly, you employed for all these different breeder calculations the same set of group constants.

MR. ROACH:

Yes.

I want to bring up again the point that Dr. Wigner asked about this effect of rising breeding ratio with changing concentration of uranium. I think it is solely due, if you review your numbers again, to a leakage term. That is in reference to the assumption that all the neutrons that leak out are absorbed in fertile material and do not reflect back into the system. We met up with this difficulty in our calculations and I think it is solely a leakage effect that you are looking at.

THE SENSITIVITY OF BREEDING RATIO IN FAST REACTORS TO UNCERTAINTIES IN CROSS SECTIONS

David Okrent Argonne National Laboratory Lemont, Illinois

It seemed pertinent to have some idea as to just how sensitive the performances of various systems would be to the uncertainties in the cross sections which now face us in calculations involving fast reactors.

A series of rather simple calculations have been performed, using old-fashioned diffusion theory, few groups, and rather crude cross sections. In general, the things that were investigated were: the effects of various types of structural materials, the effects of different assumptions on inelastic scattering, the effects of different assumptions of alpha, and finally, the effects of fission products and of different assumptions on their capture properties.

I believe that, when we have examined some of the uncertainties assumed herein, it may justify the previous speaker's assumption in not recomputing microscopic cross sections for each reactor. We don't know things well enough yet, except in very specific applications, to warrant such a recalculation each time. In principal it can be done, but it takes effort which seems hardly worth while now.

All of the calculations were done on simple spherical systems. Always uranium and Pu^{239} plus a coolant (sodium) and a structural material, which will be steel, if not stated otherwise, were used. The blanket was the same on all these reactors, namely, 60 per cent uranium, 20 per cent steel, and 20 per cent sodium. This is considered to be reasonable, engineeringwise; it is similar to what the Fermi reactor and the EBR-II reactor have. It was made thick enough so that very few neutrons were leaking out. This was only about 50 or 55 centimeters. Actually, of course, various types of blanket optimizations can be made, but this was unimportant for what we were doing here. The core was what was variable.

In general, the size of the core was taken as 1500 liters, unless the effects of size itself were being studied. This is a fairly large core compared to those presently under construction, but in some sense it is representative of the thinking for future reactors.

Figure 1 shows the six different compositions used. The first system is a metal system in that the fuel alloy is U^{238} plus plutonium, in this case 30 per cent uranium by volume of fuel alloy, which is about what one finds in the EBR-II core. These are typical numbers for a reactor which will have a performance of about one megawatt per liter.



Reactor No.	Fuel Alloy	Structure	Na Coolant
1	30% U + Pu	20%	50%
2	45% U + Pu	20%	35%
3	30% UO ₂ + PuO ₂	20%	50%
4	55% UO ₂ + PuO ₂	15%	30%
5	30% UC _{1.75} + PuC _{1.75}	20%	50%
6	30% Pu - Fe	20%	50%

The second system is one where the amount of fuel alloy has been increased at the expense of the coolant. This would, therefore, be a system having a lower performance in the sense of megawatts per liter.

The third and fourth systems are two possible oxide systems. The oxide here is taken to have 0.7 of the theoretical density, as I recall. The fifth system is a carbide system; it is really not expected to be much different from an oxide system.

 $PuC_{1.75}$ is not supposed to be any magic carbide that we are working on furiously; in fact, the metallurgists say that UC is the most likely to succeed. This system represents one with more carbon, and therefore one with a softer spectrum than one may actually get. It thus will accentuate the spectrum shift due to the carbon.

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The last system is a poor representation of a system in which there is no internal breeding, where some kind of relatively innocuous diluent is added. In this case we call it iron, the iron having a very low capture cross section relatively, though nonzero. The structure is taken separately here so that one could have, for example, a tantalum structure along with the iron diluent in the plutonium.

In Fig. 2 the critical mass (M_c) , the breeding ratio (B.R.) and the average alpha of the plutonium for these various systems are given as a function of size. I will discuss primarily the breeding ratios. Fifteen hundred liters is the case we will look at most. There is really only slight variation from 800 to 2500 liters, as you can see.

In these relatively practical systems one expects breeding

Fig	. 2
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 $\mbox{M}_{\mbox{C}}, \mbox{ B.R. AND } \alpha$ for various core compositions and volumes

Boactor Tuno	80	0 Liter		15	00 Lite	r	2500 Liter		
Keacion Type	Mc	B.R.	α	Mc	B .R.	α	M _c	B.R.	α
30% U + Pu	508	1.64	.22	839	1.62	.23	1290	1.61	.24
45% U + Pu				998	1.73	.21			
30% Oxide	387	1.39	.28	587	1.32	.30	846	1.29	.32
55% Oxide				671	1.47	.28			
30% Carbide				668	1.35	.28			
30% Pu + Fe	303	1.08	.33	403	<i>.</i> 93	.37	514	.79	.40

ratios of 1.6 to 1.7 for the metal system and of 1.3 to 1.45 for the oxide system. This particular carbide system falls in between the two oxides. In system six we have completely lost any bonus from fast fission inside the core, although there was still some small bonus from the blanket. The spectrum is quite soft because we have a relatively low ratio of plutonium to other types of atoms. Thus, alpha in system six is the largest of all six systems. As a consequence of these two effects the breeding ratio has dropped below 1. Dr. Kiehn will show us later what is required to keep the breeding ratio for such a system high; hence I will not go into it further.

Fig 3

REACTIVITY CHANGE (8k/k) ON REMOVING 40% OF Na IN 1500 LITER CORE

Reactor Type	Steel Structure	Mo Structure	Ta Structure
30% U + Pu	+.0083	+.0111	
45% U + Pu	+.0089		
30% Oxide	0127	0048	
55% Oxide	0010		
30% Carbide	0030	+.0016	
30% Pu + Fe	1346		+.0057
L	L	<u> </u>	

The metal systems, which look the best here, have a problem of their own in this size, as is demonstrated in Fig. 3, where can be seen the results of a very simple calculation of what I would call the coolant removal effect. If one were to take some of the sodium out of the core, representing either a change in density of the coolant or in some accidental condition, one would like to lose reactivity. Within these crude calculations the two metal systems seem to have gained reactivity, whereas the others, in this case, have lost varying amounts. Some of the losses are close enough to zero to be

questionably negative. These particular metal systems might be alterable geometrically so as to maintain good breeding while exhibiting a negative sodium coefficient. A significant increase in core leakage coupled with an associated increase in the concentration of fissionable material might do this. This effect was, to my knowledge, first noted by the people at APDA and they have a paper at the November ANS meeting covering some work on this subject, so I will not go into it further.

Figure 4 lists some of the important cross sections used in the analysis. The capture in iron is very small. The tantalum and the molybdenum cross sections correspond to those reported in Diven's Geneva paper, where measurements were available. The cross section for tantalum at low energies is based on one rather questionable measurement in BNL-325, but I think more recent measurements evidence some trend for the cross section of tantalum to rise more rapidly than that of molybdenum. In

Fig. 4

CAPTURE AND INELASTIC CROSS SECTIONS IN BARNS

Group	Energy Range	$\sigma_{\rm c}^{\rm Fe}$	σc ^{Mo} c	$\sigma_{\rm c}^{\rm Ta}$	$\sigma_{ m in}^{ m U^{238}}$	$\sigma_{ ext{in}}^{ ext{Pu}}$
1	10 - 1.35 Mev	.005	.020	.090	2.3	0.7
2	1.35 - 0.5	.005	.040	.180	0.55	0.2
3	0.5-0.18	.006	.055	.290	0.5	0.2
4	0. 18 - 0.067	.008	.065	.500	0.2	0
5	0.067 - 0.009	.018	.090	.700	0	0
6	0.009 - 0	.014	.150	1.500	-	-

Groups 1, 2 and 3 the ratio of capture in these two materials is 4 or 5. At lower energies, the ratio seems to increase, however.

The inelastic cross sections generally conform with Dr. Cranberg's measurements, I am sure he will be happy to hear. The variations assumed

later on are in the direction of larger inelastic cross sections since this is the direction in which other measurements go. These are then relatively low inelastic cross sections. They represent group effects, of course. All the individual levels have been very crudely molded into the cross-section set, and they are not supposed to be the last word or hardly even the first word in accuracy.

Fig. 5

MC AND B.R. AS FUNCTION OF STRUCTURAL MATERIAL FOR 1500 LITER CORE

	F	e	Мо		Ta	
Reactor Type	Mc	B.R.	Mc	B.R.	м _с	B.R.
30% U + Pu 45% U + Pu 30% Oxide 55% Oxide 30% Carbide 30% Pu + Fe	839 998 587 671 668 403	1.62 1.73 1.32 1.47 1.35 .93	889 1042 672 721 746 554	1.48 1.61 1.16 1.35 1.20 .80	1359 1485 1220 1121 1295 1199	.92 1.07 .65 .86 .68 .50

Figure 5 shows what happens to breeding ratios for the 1500-liter reactor upon change of structural material, going from a weak absorber to a moderate absorber and on to a strong absorber. As may be seen in such a system the use of tantalum is rather fatal to the breeding ratio.

The use of molybdenum is painful but not impossible. With this structural material one can get rather decent breeding ratios with all of the systems except the last.

Figure 6 shows the effect of raising $\sigma_{\text{inelastic}}$ to values more in conformity with those of experimenters other than Cranberg. The actual change that has been made in getting the comparison between what is called regular inelastic and high inelastic cross section, is to double all the inelastic cross-section measurements as used in these group cross sections

for U²³⁸, except for the first group. We have not changed the amount of scattering from above the fission threshold to below, since this is less subject to uncertainty for a variety of reasons The plutonium inelastic cross sections were also doubled, but most of these systems have so little plutonium that this change was not a major effect. In a typical system there is a change of about 0.05 in breeding ratio. The 30 per centoxide system has less uranium hence the change in spectrum was not near-

Fig. 6

EFFECT OF HIGH INELASTIC ON MC, B.R. AND & FOR 1500 LITER CORE

Reactor Type	Regu	ılar İnela	stic	High Inelastic			
	Mc	B .R.	α	M _C	B.R.	α	
30% U + Pu	839	1.62	.23	877	1.57	.25	
45% U + Pu	998	1.73	.21	1067	1.66	.24	
30% Oxide	587	1.32	.30	595	1.31	.31	
55% Oxide	671	1.47	.28	685	1.44	.29	
30% Carbide	668	1.35	.31	679	1.33	.32	
30% Pu + Fe	403	.93	.37	409	.93	.37	

ly as great. There is not a very major shift due to inelastic scattering and one might, therefore, ask why we bothered even having Dr. Cranberg on the program.

You will recall, however, that I have just mentioned a serious problem in connection with the sodium reactivity effect. This is almost completely a spectral effect. In order to understand it, we will certainly have to know the spectrum. It will, indeed, be very difficult to calculate the sodium effect accurately without having good data on inelastic scattering, and the life of specific reactor designs may hang on the sign of this effect. Furthermore, I think you will see, in a minute, that the variations in alpha that I have assumed do not produce much greater changes in breeding ratio. The breeding ratio is most sensitive to composition, not to uncertainties in cross section.

Figure 7 shows the assumed variation in alpha of plutonium. I believe we see all the monoenergetic and reactor-averaged measurements of alpha except those in EBR-I in this figure. The Sampson-Molino curve is the one that was chosen to correspond to this data, while giving a rough agreement with the radial variation of alpha as measured in the core and blanket of EBR-I. The low alpha and the high alpha are intended to provide some basis for estimating the effects of these uncertainties on breeding ratio.



Fig. 7 Alpha Pu²³⁹ as a Function of Energy

Figure 8 gives the results. For the metal system, for example, we have results for a regular alpha, a high alpha, and a low alpha. We see here that a drop in breeding ratio of about 0.1 results on going to high alphas. If we incorporate into the calculations the high inelastic cross sections, in addition, the spectrum is degraded in accordance with the high inelastic cross sections and there will be a further drop down to 1.45 from an original

EFFECT OF VARIATION IN α ON M_C AND B.R. FOR 1500 LITER CORE (B.R. in parens corresponds to high inelastics)

Reactor Type	Re	Regular $lpha$			High $lpha$			Low $lpha$		
	Mc	B.R.	α	Mc	B.R.	α	Mc	B.R.	α	
30% U + Pu	839	1.62	.23	854	1.52 (1.45)	.28	816	1.78	.16	
45% U + Pu	998	1.73	.21	1010	1.64	.25	975	1.89	.14	
30% Oxide	587	1.32	.30	608	1.22 (1.20)	.36	557	1.50	.21	
55% Oxide	671	1.47	.28	689	1.36 (1.32)	.34	644	1.65	.19	
30% Carbide	668	1.35	.31	690	1.24	.38	637	1.53	.21	
30% Pu + Fe	403	.93	.37	432	.85	.44	363	1.07	.26	

value of 1.62. So, this is the combined effect of both high alpha and high inelastic cross sections. If we use the low alpha curve, the value of 1.62 rises to 1.78. Generally speaking, the results are similar for all the systems. The critical mass mostly goes along with the breeding ratio. It won't always vary, because sometimes there are compensating effects.

The last effect I would like to discuss is that of fission

products on breeding ratio. When we started worrying about the performance of fast power reactors, many years ago, we were not in the relatively happy state that Dr. Moldauer now finds us, where we have many nuclides measured, and he is about to be able to calculate the behavior of these. There was almost nothing known about fission product capture for high neutron energies. We thought it would be nice if we could make up some kind of a mixture that could be used for measuring the effects of fission products in our fast critical facility. We didn't want to use radioactive materials nor did we want to use separated isotopes in preparing this mixture, since this would raise the cost very much. A mixture of natural elements was sought which would represent the fission products reasonably well. The following course was pursued. Take the fission product spectrum which remains in a reactor after it has been running for 100 or 200 days so that you are not considering the short half-lives. Substitute natural elements for this group of fission products, using the following simple rules: we will conserve the number of magic isotopes present, we will assume that the capture cross section is increasing slowly (monotonically) with nuclear radius or atomic number, and we will assume that an even-even isotope is about the same as a neighboring even-even isotope, and that an even-odd isotope is about the same as a neighboring even-odd or odd-even isotope. One can then make up a mixture of natural elements which, within these assumptions, should have about the same average capture cross section. And one would think the inelastic scattering properties were similar, too, since about the same rules would apply. One can thereby find many "equivalent" mixtures. We chose to make up two, the compositions of which are shown in Fig. 9

We have called these Physicum, not to be confused with fissium, if you should hear this word tomorrow. We have added these various elements in their natural form, and as you see for Physicum I and II, there are radically different amounts. For example, molybdenum varies by a factor of 3 one way, and ruthenium by 2 the other way. These do look very different. The hope was that they would act pretty much the same, of course.

Fig. 9

Atom		Mass of Metal or Compound Added, g		Atom		Mass of Metal or Compound Added, g	
Interest	Added As	Physicum I	Physicum II	Interest	Auleu As	Physicum I	Physicum II
Se Rb Sr Zr Nb Mo Ru Rh	Se Metal Rb_2SO_4 $SrCO_3$ ZrO_2 Nb_2O_5 MoO_3 Ru Metal Rh Metal	0 67 	7.99 9.65 6.28 5.13 81 02 60 70 222.93 32 46	Te I Cs Ba La Ce Pr Nd	Te Metal Cs I Cs ₂ SO ₄ BaCO ₃ La ₂ O ₃ CeO ₂ Pr_6O_{11} Nd ₂ O ₃	81 96 - 42 98 199.11 10 92 57.54 22.82 22 56	105 35 71,30 94 11 132 72 71 61 34 97 96,99
Pd Ag Cd In Sn Sb	Pd Metal Ag Metal Cd Metal In Metal Sn Metal Sb Metal	23 17 46.22 2.00 6.43 1 51 79 85	23.22 21.55 2.01 6.44 1.51 2.28	Sm Gd Total Gra Average F Ave, Atom of Ph M	Sm ₂ O ₃ Gd ₂ O ₃ ms Formula ic Wgt letal	$ \begin{array}{r} 164 27 \\ 4 60 \\ 1203 89 \\ Ph_{8,91} O_{13} 53 \\ 110 7 \end{array} $	0 77 1157 43 Ph _{8.55} O _{9 8} 117.0

NOMINAL COMPOSITIONS OF PHYSICUM I AND II MIXTURES

Figure 10 shows their behaviors in the relatively few experiments that have been done with them so far. The first experiments were done in

Fig. 10

REACTIVITY WORTH RELATIVE TO VOID AT CORE CENTER, INHOURS/GRAM ATOM

	Mo	Nb	Ph I	Ph II	Ta	^x *PhI	^x ằh ∏
EBR-Ⅲ	-1.3	-2.0	-2.3	-2.7	-5.3	.27	.36
Fermi	-1.0	-	-2.8	-	-4.5	.53	-
9:1 U ²³⁸ /U ²³⁵ no coolant	-1.5	-1.9	-2.3	-3.0	-5.9	.20	.34

^{*}x. Ta + (1-x) Mo = Ph

an assembly which looked like the EBR-II. For orientation purposes, we also show the results obtained from molybdenum, niobium (which is a somewhat stronger capturing material), and tantalum. As you see, Physicum I and II had about the same reactivity effect in the EBR-II reactor. Somehow, in the Fermi reactor, only Physicum I was measured, and it disagrees somewhat with the other results, suggesting a possible error. The 9:1 assembly gave a slightly wider dispersion between the two mixtures than was obtained for the EBR-II, but the range of the results

is similar All, without exception I believe, fall between niobium and tantalum. One can express the reactivity effect of Physicum I or II as some fraction, x, times that of tantalum, plus (1-x) times that of molybdenum One thus learns what mixture of molybdenum and tantalum acts like Physicum, within whatever phenomena we have measured here. One finds that this value of x varies from about 0.2 to 0.5. These measurements were all made on systems which have a rather harder spectrum than the large reactors I have reported on earlier. I have already mentioned that at the very low energies the ratio of capture in tantalum relative to molybdenum seems to increase quite a bit. So one is being rather conservative if extrapolation of this sort of measurement down to lower energy is made in this fashion.

Finally, I might just note one thing as a matter of orientation. If we assume that the reactors in ZPR-III have an average spectrum energy of, say, 200 kev, the capture in molybdenum at this energy would be about 65 mb, while for tantalum it would be about 350 mb, giving the factor of about 5 previously mentioned. The Physicum would fall somewhere between 120 and 200 mb, let's say. The Hurwitz-Greebler estimate would fall below the molybdenum, at about 50 mb. So it would appear that Physicum possesses something like two and one half to three times the absorption predicted by the Hurwitz-Greebler calculation. This is about the ratio that Dr. Moldauer showed this afternoon, although the two results were arrived at independently, of course.

Fig. 11

EFFECT OF FISSION PRODUCTS EQUIVALENT TO BURNUP OF 10% OF TOTAL (U + Pu) ATOMS ON CRITICAL MASS AND BREEDING RATES FOR 1500 LITER CORE

Deactor Type	No F. P.		F. P. ≈10%	Ta + 90% Mo	F. P. ≈ 50% Ta + 50% Mo		
Reactor Type	Mc	B.R.	Mc	B.R.	Mc	B.R.	
30% U + Pu	839	1.62	839	1.50	896	1.38	
45% U + Pu	998	1.73	1001	1.58	1077	1.45	
30% Oxide	587	1.32	585	1.26	612	1.21	
55% Oxide	671	1.47	670	1.39	708	1.31	
30% Carbide	668	1.35	665	1.27	706	1.20	

Figure 11 shows what happens if you calculate the effect of fission products, in one case represented by a mixture half tantalum and half molvbdenum, and in the other case by a more weakly absorbing fission product, 10 per cent tantalum and 90 per cent molybdenum. We assume that 10 per cent of the total uranium plus plutonium in the core has been burned, making for a peak burnup of 200,000 Mwd/t, a number not quite achievable, metallurgically, at the moment. For the more weakly absorbing condition of x = 0.1, this much burn-

up reduces the breeding by 0.1 to 0.15 for the metal systems, less for the others. If we take the more pessimistic assumption about fission product capture, the breeding ratio drops by 0.2 to 0.25. Hence, fission products do not have a radical effect on the performance of such a system, for these are very large numbers of fission products indeed.

BREEDING: INTERNAL OR EXTERNAL?

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Introduction

The problems of internal or external breeding have to be evaluated for every breeder system. Material composition, geometrical shapes, spectrum changes, lattice arrangements and their effects on new fuel production and heat transfer have to be examined for each system configuration. For some classes of breeder systems, internal breeding is the most attractive method of obtaining high specific power and short doubling time; for other classes, external breeding is more attractive. The work presented here attempts to point out some of the nuclear properties and problems associated with the various schemes of internal and external breeder design. Some of the various topics of breeding physics to be discussed may be described in terms of a few simple relations involving spectrum-averaged nuclear parameters. These relations will first be developed and then applied to a specific class of breeder systems. For the class systems studied, no clear-cut nuclear advantage was obtained for either internal or external breeding. The methods that were utilized indicate that, for some other classes of systems, external breeding may be advantageous from a nuclear viewpoint.

If a critical configuration is assumed to consist of a fuel region and a fertile blanket region, and is assumed to be so large that leakage loss of neutrons is negligible, then a relatively simple analysis may be made based upon a detailed neutron balance. The assembly will determine some spaceenergy distribution of neutrons from which it is possible to derive spectrumaveraged parameters for use in such a detailed neutron balance. It is important to note that the blanket region may have a significant multiplication, in the sense that one neutron injected into the blanket will produce M_b total blanket events (absorptions). The external breeding ratio will turn out to be strongly dependent upon the blanket multiplication.

In the fuel, or core, region there will be produced η_c neutrons per core region absorption, leaving η_c - l neutrons to enter the blanket. If f is the fuel utilization factor (fraction of fuel region absorptions which occur in fuel) then $(\eta_c - 1)/f$ neutrons are injected into the blanket per fuel absorption in the fuel region.

For some given incident neutron spectrum, the blanket, of given size, will have a multiplication M_b . If the fissionable isotopes in the blanket have threshold characteristics, as is true of the two most important fertile isotopes, then the blanket multiplication will be sensitive to the incident neutron spectrum. If the blanket is extremely thick, the spectrum in the

blanket will be the blanket equilibrium spectrum; for smaller blankets the spectrum (asymptotic) is determined more by the core spectrum, and may be more energetic on the average than the equilibrium spectrum. Proper design (cells) will allow a preservation of the asymptotic rather than of the equilibrium spectrum. Furthermore, the addition of a small amount of fuel material to the blanket will harden the blanket spectrum significantly, and the blanket multiplication will no longer strongly depend on the threshold fissionable isotopes.

For a given blanket multiplication, M_b , there will be a total of $(\eta_c - 1)M_b/f$ blanket events per core region fuel absorption. If A is the fraction of total blanket events which are fertile captures, and B is the fraction of total blanket events which are absorptions in fissionable (fuel) material, and IPR is the ratio of fertile captures, if any, to fuel absorptions in the core, then the assembly potential production ratio, PR, defined as the total fertile captures per fuel absorptions, is

$$PR = \frac{\frac{(\eta_c - 1)M_bA}{f}}{1 + \frac{(\eta_c - 1)M_bB}{f}} + IPR$$

Noting that the blanket multiplication may be written as $1/(1 - \eta_b)$, the above formula may be written in terms of cross-section ratios, yielding a useful form for desk speculations. For example, for the Pu²³⁹-U²³⁸ cycle, and ignoring internal breeding and parasitic losses,

$$PR = \frac{(\eta_c - 1)/f}{1 + (\sigma_f^{28}/\sigma_c^{28})(1 - \nu) + (N^{49}/N^{28})_{bl} \{ [1 + (\eta_c - 1)/f](\sigma_a^{49}/\sigma_c^{28}) - \nu(\sigma_f^{49}/\sigma_c^{28}) \}}$$

Except for the ratio $\sigma_f^{28}/\sigma_c^{28}$, the other cross-section ratios are relatively slowly varying functions of average neutron energy.

The exploitation of the above concepts will be demonstrated by a number of examples based upon a series of multigroup S_N calculations, including a particular system of interest to LASL. This particular system consists of a molten Pu-Co-Ce alloy contained in tantalum and cooled by sodium. The core volume proportions, which were chosen from a heat transfer analysis, are nominally 20% Ta, 33% Na, and 47% fuel alloy. The core volume fractions were adjusted, of course, as various core compositions and configurations were studied. The blankets for all systems studied consist of a 60% U, 10% Fe, 30% Na structure, unless specified otherwise. The computations were done for spherical systems, unless otherwise noted.

Tantalum, although a relatively serious fast reactor poison, is one of the few possible container materials for molten plutonium. As the value of the tantalum capture cross section strongly influences the system production ratio, it is felt necessary to present the actual tantalum capture cross sections used in the calculations These group parameters are compared to the experimental data in Fig 1



Fig 1 Tantalum Group Capture Cross Sections The histogram represents the group cross-section parameters The data are are from BNL-325.

The other isotopic multigroup cross sections used in the following studies are improvements on the set developed in 1956 and published in Nuclear Science and Engineering (1) Specifically, more attention has been given to the recent measurements of capture cross section for U²³⁸, and the value of α^{49} has been increased slightly in the lower energy groups These values of α^{49} would be called "high" to "high-standard" in comparison to Okrent's values (2)

Effects of Internal Poisons

As tantalum is a relatively serious poison, the effects of increasing the ratio of core tantalum to plutonium were investigated for an external breeder As the tantalum/plutonium ratio is increased in the set of systems examined, a more favorable heat rating (specific power) is obtained, but because the value of $(\eta_c - 1)/f$ decreases, due to an increasing amount of parasitic absorption in tantalum, a compromise must be established between high production gain and high specific power The use of the simple formulas given above results in an effective, standardized
method of evaluating the nuclear characteristics of the various systems. Figure 2 portrays the appropriate nuclear parameters for the plutoniumtantalum system as a function of the ratio of core tantalum to plutonium.





Plutonium-tantalum Systems: Nuclear Parameters. The predominant effect of increasing the tantalum capture fraction in the core is to reduce the number of blanket injection neutrons per core fuel absorption.

These parameters were determined from a series of multigroup calculations. It should be noted that the blanket parameters are only slightly affected by an increase in the ratio $N(Ta)/N(Pu^{239})$, but the number of injection neutrons available from the core is sharply decreased by a similar variation. The blanket multiplication was determined from a computation involving a 45-cm thick blanket; the effective neutron multiplication was determined from the computed asymptotic, rather than the equilibrium, neutron spectrum. Figure 3 summarizes the breeding parameters for this system study; the dashed line indicates the loss of potential production ratio due to the use of a 45-cm blanket. This blanket thickness was chosen, even though it is far too thin, to a) allow a comparison with the work of Loewenstein and Okrent,⁽²⁾ and b) to demonstrate the importance of proper blanket design by indicating the substantial loss in potential production ratio incurred by too thin a blanket. For external breeders, proper design of the blanket is of most importance.

Another breeding parameter of interest is the so-called system "doubling time," which is inversely proportional to the product of production gain times specific power. This product is presented as the parabolic curve in Fig. 3, which demonstrates that on a doubling-time basis there is

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an optimum ratio of $N(Ta)/N(Pu^{239})$ equal to about 2. The further plutonium-tantalum system studies to be presented fix the ratio of $N(Ta)/N(Pu^{239})$ as 2.0.





The lower dashed line of Fig. 3 portrays the effect of a 3 to 1 increase in the $N(Na)/N(Pu^{239})$ ratio, indicating as expected, that the production ratio is relatively insensitive to this portion of the core design.

The two points in Fig. 3 are for infinite slab configurations and indicate that only a small penalty is paid for geometrical distortion. In these distorted configurations the core spectrum is more closely coupled to the blanket spectrum and is degraded slightly; this core spectrum degradation accounts for the slight loss in production ratio.

Another breeding parameter, although of little or no consequence in doubling-time considerations, but of large significance in economic studies, is the fraction of fissions produced directly in fertile material. In the $Pu^{239}-U^{238}$ cycle, the direct fissions in U^{238} can be a significant fraction of the total assembly fissions. These fissions are extremely cheap, in that they dodge the expense of high fuel costs and expensive processing. The fraction of total fissions occurring in the natural uranium blanket is also given in Fig. 3, and about 56% of this fraction is directly due to fissions in U^{238} .

Effects of Internal Breeding

The next study presented, and again this is based on the plutoniumtantalum system and spectrum averaged parameters, investigates the nuclear effects of combined internal and external breeding. For these hypothetical studies, the core material was volumetrically displaced with U^{238} . Figure 4 displays the effect of varying the core ratio $N(U^{238})/N(Pu^{239})$ on the nuclear parameters of the system for a fixed ratio $N(Ta)/N(Pu^{239}) = 2.0$.





Plutonium-tantalum Systems: Effects of Internal Breeding; <u>Nuclear Parameters</u>. The addition of U²³⁸ to the core reduces the number of available blanket injection neutrons.

It should be noted that the blanket parameters change only slightly as the spectrum shifts to slightly lower average values with addition of U^{238} to the core. The number of injection neutrons decreases at rate approximately commensurate with the build up of internal breeding. The net effect on potential production ratio is nil, as is made evident in Fig. 5. It should be mentioned that for other types of systems, such as the PuCl₃-UCl₄ system, the effects of internal breeding are somewhat more drastic.⁽³⁾ In these systems the spectrum is more strongly dependent upon the U²³⁸ concentration because of the necessary addition of neutron spectrum degraders along with the fertile uranium.





Plutonium-tantalum Systems: Effects of Internal Breeding; Breeding Parameters. The loss of external production ratio is compensated by the buildup of internal production ratio.

The dashed curve of Fig. 5 again indicates the effect of a 45-cm blanket; as internal breeding becomes predominant, the effects of the blanket size are diminished. It is to be noted in Fig. 5 that the fraction of total fissions due to U^{238} increases significantly as the fertile material becomes more intimately mixed with fuel.

For the molten plutonium-tantalum system of interest, internal breeding does not significantly affect the potential production ratio. If the uranium were to be added to the fuel alloy to accomplish internal breeding, a penalty would be paid, due to the increased melting temperature of the alloy. Hence, in this sense, external breeding is more advantageous for these systems. On the other hand, it is recognized that dilute fuels are more attractive from heat transfer and mechanical viewpoints, and if the diluting material is also a fertile material, then an inherent nuclear advantage is retained. In an engineered external breeding system, care must be taken to insure that all leakage neutrons are effectively utilized in fertile material. Streaming through coolant inlet ducts, thimbles, etc., must be minimized; otherwise severe breeding losses may be encountered.

Effects of Blanket Enrichment

The simple formulas and their applications given above indicate that the effects of blanket multiplication can be extremely helpful to breeder design By judiciously using the properties of a multiplying blanket it is possible to obtain significant production gains for systems having less than one injection neutron percore fuel absorption These concepts should then be of particular interest to thermal breeder designers, where the η of the core fuel may be of the order of 2.0, or less.

To investigate the effects of blanket enrichment (and therefore of increased blanket multiplication), a plutonium-tantalum system core $[N(Ta)/(Pu^{239}) = 2.0]$ was coupled to a dilute 55-cm blanket containing various ratios of $N(Pu^{239})/N(U^{238})$. The core and blanket nuclear parameters obtained from this study are given in Fig. 6. The blanket parameters are of interest It may be noted that η_b of the dilute blanket exceeds unity for a ratio of $N(Pu^{239})/N(U^{238}) \simeq 0.045$ The simple formulas above do not apply to blankets for which $\eta_b > 1.0$



F1g. 6

Plutonium-tantalum Systems: Blanket Enrichment Effects; Nuclear Parameter. The blanket multiplication is strongly dependent upon blanket enrichment. As the blanket multiplication increases, the system potential production ratio rises to a value equal to the production ratio of the infinite self-sustaining blanket, or PR ≈ 2.05 (see Fig. 7). The dashed curve indicates the detrimental effects of having too small a blanket thickness. As blanket multiplication increases, the size of the blanket becomes very important; neutron leakage out of the blanket must be minimized.





Plutonium-tantalum Systems: Blanket Enrichment Effects; Breeding Parameters. The potential production ratio increases with blanket enrichment.

The blanket nuclear parameters given in Fig. 6 are approximately correct for a wide variation of core configurations. The value of η_b is probably the most sensitive to a lowering of the incident neutron spectrum. The presented values may be used to speculate about the effects of enriched blankets on the breeding performance of other systems. For example, using the nuclear blanket parameters of Fig.6, it may be calculated that a core region yield of only 0.75 injection neutron per fuel absorption has a potential production ratio of 1.5 if the blanket is enriched to 3.5% in Pu²³⁹.

Miscellaneous Blanket and Breeding Topics

The blankets considered have been characterized by an asymptotic, rather than an equilibrium, neutron spectrum. A very large thickness of

blanket is necessary to achieve an "equilibrium" blanket spectrum. For driven blankets, the "effective" spectrum may never reach the equilibrium spectrum, even for very large blankets. The effective region of the blanket is that portion having a significant volume flux integral, and is generally located near the core, where the neutron spectrum is the most energetic. It should be noted that lattice arrangements will preserve the asymptotic blanket spectrum. It is of some interest at this point to compare the multiplication properties of an equilibrium blanket; a dilute, slightly enriched, blanket which is critical in the infinite sense; and the asymptotic, driven blanket of the problems above. Such a comparison, indicating the degree of hardening of the neutron spectrum with blanket enrichment or driving spectrum, is given in Table I. A comparison of the equilibrium flux spectrum obtained with the ten-group set of cross sections utilized in this work is made to a similar spectrum obtained by Meneghetti <u>et al.</u>, (4) in Fig. 8.

TABLE I

	Natural Uranıum Equilibrium Spectrum	Dilute Infinite- Medium Spectrum	Dilute Blanket Asymptotic Spectrum
	0.183	1.0	0.406
MB	1.22	œ	1.68
$\sigma_{\rm f}^{28}/\sigma_{\rm C}^{28}$	0.032	0.18	0.14
$\sigma_{\rm C}^{28}$ (barns)	0.24	0.21	0.228
ā ⁴⁹	0 27	0.21	0.23
U ²³⁸ Fission Fraction	0.39	0.30	0.71

Spectrum Parameters



Fig. 8. Equilibrium Spectra for Natural Uranium. These curves are presented as a comparison of the ten-group cross-section set utilized in this work and the ten-group set used in Ref. 4.

To demonstrate the utility of blanket multiplication, a number of cell calculations were completed in plane geometry for a speculative system consisting of 40 cm of graphite, a plutonium fuel zone of low density fuel, and a depleted (and enriched) uranium blanket, 50 cm thick. The thickness of the fuel zone was adjusted for criticality. For these systems over 40 per cent of the fuel zone fissions were thermal; $\bar{\alpha}^{49}$ of the fuel zone was greater than 0.5, and yet a significant breeding gain was achieved for the enriched blanket systems. The nuclear and breeding parameters for two such cases are given in Table II. Similar results should also apply to a thermal thorium-uranium system; and although the self-multiplication of natural thorium will be much less than normal uranium, an enriched thorium blanket will be quite effective.

TABLE II

	Case I	Case II
Blanket enrichment $N(Pu^{239})/N(U^{238})$	0	0.04074
η	1.978	1.936
Injection neutrons/core fuel absorption	0.914	0.843
ηb	0.288	0.887
A	0.875	0.604
B	0	0.284
Potential production ratio	1.122	1.454
Fraction of fissions of U ²³⁸	0.184	0.257
Core thermal fission fraction	0.45	0.4

Blanket Enrichment Effects For A Hypothetical Cell System

The question may arise as to why not use a slightly enriched plutonium-uranium system as the breeding system itself? If the fuel inventory problem (several tons of plutonium per unit) is ignored, then these slightly enriched systems are possibilities. The use of a driver-enriched blanket system only allows the possibility of obtaining a breeding system with a relatively low critical inventory per unit. Fast reactors achieve this result by the use of high density, low critical mass cores. Thermal systems could achieve similar low mass results, but the thermal value of $\eta_{\rm C}$ in such systems is not large enough to permit large breeding gains. The use of unmoderated, slightly enriched blankets coupled to thermal cores may be a method of achieving relatively low inventories, a substantial breeding gain, and the heat transfer advantages associated with low-density thermal reactor fuels.

In practical blanket design it is desirable to decrease the blanket diffusion length to effect thinner blanket regions, less material holdup, and concentration of product. The use of moderator within the blanket to decrease the diffusion length is not always advantageous, because the blanket multiplication properties will be destroyed, lowering the potential breeding gain. Moderating reflectors outside of unenriched blankets prove to be effective. Figure 9 demonstrates the effect of blanket thickness on a hypothetical breeder system, with and without an external moderating reflector. A similar study for enriched blankets has not been completed.



Fig. 9. <u>The Effect of Blanket Thickness on External</u> <u>Breeding Ratio</u>. The use of an external moderating reflector reduces the required blanket thickness for a given breeding ratio by about one blanket diffusion length.

It should be mentioned that the use of blanket enrichment techniques to improve the production gain for certain systems does involve some new problems. For example, as the blanket multiplication increases, the inventory of fuel in the blanket increases. The power output of the system may be limited by the power density achievable in the core region. In this case the specific power of the total system, averaged over the core and blanket, probably will be significantly reduced below the value for the core region alone. Then, if economic charges for the unprocessed material in the blanket are assessed equal to the fuel charges in the core; the low average specific power may raise the power costs assigned to the system.

Furthermore, as the doubling time is inversely proportional to the product of specific power times production gain, for some systems it may not be advantageous to use enriched blankets. The production gain may be increased at the expense of average specific power to such an extent that the inverse doubling time product decreases.

Another point to consider in the U^{238} -Pu²³⁹ cycle is the effect of Pu^{240} on the production ratio. In a fast spectrum normally associated with external breeder designs, Pu²⁴⁰ has an effectiveness slightly less than that of Pu²³⁹. In these cases, then, the production ratio should be defined as new atoms of plutonium produced per effective plutonium atom destroyed. Fast spectrum systems will therefore have an additional advantage in production ratio over less energetic configurations. The effectiveness of Pu^{240} may be measured in terms of the isotopic value of η . However, little is known about the capture cross section of Pu²⁴⁰; hence in Table III two quantities are given: 1) the ratio of $\nu \sigma_f^{240} / \nu \sigma_f^{239}$, and 2) the ratio η^{240} / η^{239} , assuming that $\sigma_c^{239} \simeq \sigma_c^{240}$. These quantities are given for three spectra: 1) the equilibrium spectrum of natural uranium; 2) the infinite medium spectrum of the dilute enriched blanket; and 3) the representative fast spectrum of the plutonium-tantalum systems discussed above. These results demonstrate the advantages and inherent increase in production ratio obtained in a fast spectrum and due to the effective utilization of Pu^{240} . Qualitatively, the effective fuel α should be multiplied by $[1 - (\eta^{240}/\eta^{239})]$, increasing potential production ratios.

TABLE III

	Equilibrium Spectrum	Enriched Blanket Spectrum	Plutonium-tantalum Spectrum		
$\nu\sigma_{\rm f}^{240}/\nu\sigma_{\rm f}^{239}$	0.069	0.222	0.484		
η^{240}/η^{239}	0.267	0.675	0.930		

Parameters Relating to Pu²⁴⁰

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- Meneghetti, D. et al., <u>Ten-group-calculated Equilibrium Neutron</u> Spectrum and Diffusion Length in Natural Uranium, Nuclear Sci. and Eng., <u>3</u>, 151-160 (1958).

Discussion of Paper Presented by Mr. Kiehn

MR. MacCARTHY:

I might point out something in regard to the discussion of blankets being too thin. The Fermi blanket is two feet thick, radially. In our latest designs, we're going to thinner blankets, because the economics of the thicker blanket sometimes does not turn out to be worth while. It may be from the physics standpoint, but it certainly isn't from the investment standpoint. The fluxes are low and it takes a long time to use that amount of fabricated material.

MR. KIEHN:

What about buildup, for example, with the reflector moderator on the outside?

MR. MacCARTHY:

I have not looked at the moderator.

MR. GREEBLER:

You evaluated the effect on breeding ratio of the split, you might say, between internal and external breeding. Did you also consider the effect of this split on other physics properties, for example the sodium temperature coefficient of reactivity?

MR. KIEHN:

No, not in detail.

MR. CAMPISE:

Why did you take tantalum in your survey rather than, say, "moly"?

MR. KIEHN:

We are interested in a molten plutonium alloy system. Tantalum, as yet, is the only container we have found to hold the plutonium alloy at 650°C.

MR. OKRENT:

I might first note that we have built a reactor with an external graphite reflector, namely EBR-I. Fortunately, we have the graphite around it or our breeding ratio would have been down by quite a bit, since it was effectively a thin blanket of uranium. My question is: what would the core volume be for a simple spherical system having the ratio of plutonium to tantalum that you found optimum, if you didn't distort it?

MR. KIEHN:

If you didn't distort it, it would contain something like 200 kg of plutonium.

MR. KASTEN:

As you increase the tantalum amount in the core is the spectral shift important?

MR. KIEHN:

As you increase the tantalum in the core the spectrum does tend to soften, due to increased tantalum inelastic scattering, but I will have to think about that statement, as the increased absorption will have a tendency to harden it. I would have to look back at the figure again to just make sure. I believe I plotted on the figure, which you can see in the proceedings, the effective value of alpha 49 (as a function of the ratio N(Pu)/N(49)). As I remember these results, the 49 alpha value changes only slightly with the variation of tantalum in the core.

MR. KASTEN:

As you put U^{238} in the core does the spectrum shift produce much change ?

MR. KIEHN:

Again, not very much, on the basis that alpha 49 does not change very much.

PARAMETRIC ANALYSIS OF A PuO2-UO2 FUELED FAST REACTOR

Paul Greebler and Peter Aline General Electric, Atomic Power Equipment Department San Jose, California (Paper presented by Mr. Greebler)

The General Electric Company is engaged under AEC contract in a study on an oxide-fueled, plutonium fast breeder. In order to obtain physics information which is needed for the selection of a reference design, initial scoping studies were carried out to determine important physics characteristics over a range of design parameters. The physics properties investigated include the breeding ratios, doubling time, required plutonium fuel loading, the sodium temperature coefficient of reactivity, and the excess operating reactivity which must be held down by control rods as a function of the refueling interval.

Figure 1 shows the design parameters, which were varied, and the conditions, which were assumed fixed for these scoping studies. The diameter of the fuel rod determines the fuel specific power (kw/kg) and, for a given power rating, largely determines the core size and, consequently, the internal breeding ratio. The neutron leakage from the core and the

VARIABLE PARAMETERS

FUEL ROD DIAMETER - - - - 0.125 TO 0.280 INCH OXIDE FUEL DENSITY - - - 65 TO 90 % THEORETICAL STEEL CLAD THICKNESS - - 5 TO 20 % OF FUEL DIA. CORE CONFIGURATION - - -(1) CYLINDRICAL. (2) ANNULAR. (3) UNBLANKETED

FIXED PARAMETERS

BLANKET -- 18 INCH; 50 % UO2, 20 % STEEL, 30 % Na

Fig. 1.

parasitic capture are sensitive to the density of the oxide fuel. This parameter was included to evaluate the physics incentive for going to high-density fuel where the fabrication procedures might be more difficult than for low-density fuel. The steel clad thickness, or more specifically the ratio of the clad thickness to the diameter of the fuel rod, was varied over the indicated range in order to determine the amount of steel which could be tolerated without affecting the neutron economy too seriously.

Three core configurations were considered: (1) a conventionaltype cylindrical core completely surrounded by a fertile blanket; (2) an annular core with both central and outer blankets; and (3) an unblanketed internal breeder. It appeared that the annular core might have some potential advantages for radial power flattening, for the sustaining of reactivity with fuel burnup, and for increasing the effective delayed neutron fraction by promoting U^{238} fissions in the central blanket region. The unblanketed internal breeder has the advantage of design and operational simplicity. Its principal disadvantage is that it requires a very large core size with a resulting high inventory of plutonium.

The conditions held fixed are listed in the lower portion of Fig. 1. The power rating of the core was set at 812 megawatts thermal, corresponding with a 300-megawatt electrical plant. The fuel irradiation exposure was taken to be 100,000 MWD/T, which is about twelve per cent burnup of all of the fissionable and fertile atoms present. The ability to attain high burnup is perhaps the outstanding advantage of oxide fuel. The burnup value listed here is an extraplation of the KAPL irradiation tests which were carried out on oxide fuel to about one-half of this burnup value. The fuel is mixed plutonium-uranium oxide. The uranium portion of it is assumed to be entirely depleted, that is, just U²³⁸. The input isotopic plutonium composition was taken as that characteristic of the output of a high burnup thermal reactor: 60 per cent Pu^{239} , 25 per cent Pu^{240} , 12 and 3 per cent each of Pu^{241} and Pu^{242} . Some of the thermal properties of the fuel assumed in the scoping studies are as follows: a thermal conductivity of 1.0 BTU/hr-ft-°F, a temperature drop from the center to the outside of the fuel rod of 3500°F at the point of peak power, with a peak-to-average core power density of 2.5. The 3500°F temperature drop, which renders the peak fuel temperature slightly below the melting point, is the limiting condition on the fuel specific power. With these properties and the given core power rating, the core fuel volume is uniquely determined by the fuel rod diameter.

Having obtained the fuel volume, the steel clad volume is then determined from the ratio of clad thickness to the diameter of fuel rod. The sodium flow area is determined by allowing a 250°F sodium temperature rise in passing through the core and a flow velocity of twenty-three feet per second. Overall composition and dimensions are obtained by setting the height of the core equal to its diameter, allowing a two per cent void for possible control rod passages, and allowing ten per cent steel volume over and above clad for miscellaneous structure. Each of the conventionaltype cylindrical cores was completely surrounded by an 18 in. thick oxide blanket with composition as indicated at the bottom of Fig. 1. Unless otherwise stated, it is to be assumed that all of the results here are based upon a clean blanket, no plutonium, and an equilibrium or average fuel composition. By an equilibrium fuel composition we mean one that would be characterized by 50,000 MWD/T burnup - the average between the cleanest fuel in the core which has the indicated input isotopic composition and the most depleted fuel in the core which has been burned to 100,000 MWD/T and is ready for discharge.

In particular, we are removing the plutonium after an average fuel irradiation of 100,000 MWD/T and refueling with the initial isotopic composition. This may be termed an open cycle as opposed to a closed cycle (or recycle), which would return the discharged plutonium to the reactor after reprocessing and fabrication.

We do have a fast breeder, however. Also, we would like to point out that the physics of this system, with an equilibrium core composition, is not appreciably different from that which is assumed for a closed cycle, steady-state situation. We have about a two-to-one ratio of Pu^{239} and Pu^{241} to Pu^{240} and Pu^{242} , which is characteristic of a steady-state, closed cycle.

Since cross sections for Pu^{241} and Pu^{242} were not available to us, we assumed that Pu^{241} was identical with Pu^{239} , except for a higher ν value (3.01 below the Mev region). Also, we assumed that Pu^{242} was identical with Pu^{240} . Multigroup diffusion theory was used for most of the calculations with eleven energy groups and one space dimension. The eleven-group cross sections were obtained from a basic seventeen-group library, using a group condensation calculational program. The top ten energy groups for most of the materials were taken from the Lowenstein-Okrent status report on fast reactors. Isotopic fuel changes with burnup were obtained using a one-group burnup calculational program in which the one-group flux and cross sections for each geometrical region of the reactor were derived from the output of the multigroup calculation.

Breeding ratio is plotted against the fuel rod diameter in Fig. 2 for ninety and sixty-five per cent density fuel. When we use the term "per cent density," we mean per cent of the theoretical density of UO_2 , which is 10.9 gm/cm³. The solid lines are for the total breeding ratio. The breeding ratio here is defined as the rate at which Pu^{239} and Pu^{241} are being created to the rate at which these isotopes are being destroyed.



Fig. 2. Breeding Ratio vs Fuel Rod Diameter

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For all of the cases considered here we used 0.1 steel ratio. (By the steel ratio we mean the ratio of the steel clad thickness to the fuel rod diameter.) The numbers above the arrows indicate core diameters for three of the fuel rod diameters. The breeding ratio is appreciably higher for ninety per cent density fuel than for that of sixty-five per cent density. The internal breeding ratio is highly sensitive to the core size consequently, to the fuel rod diameter. The internal breeding ratio is a very important economic parameter, since plutonium which is made in the core can be burned without undergoing an additional reprocessing and fabrication step, as is required for plutonium made in the blanket. The total breeding ratio also goes up slightly with increasing fuel rod diameter and core size. The breeding in the blanket does not quite compensate for the loss in breeding in the core as the core size decreases. It involves factors such as the effect of core size on the plutonium alpha value and its sensitivity to the spectrum, the change in the U^{238} fission fraction, the total leakage, and parasitic capture. These factors will be discussed in more detail later.

The mass of Pu²³⁹ and Pu²⁴¹ in the equilibrium core (see Fig. 3) is plotted over the same range of parameters as in the preceding figure. (you may wonder why we use the term "plutonium loading" instead of critical mass. The difference really is very small. We assume for all of these cases a four-month refueling interval; that is, we put in enough excess fuel - enough excess reactivity in our batch loading procedure - so that we could operate for four months before we would have to supply some



Fig. 3. Mass of Plutonium vs Fuel Rod Diameter

additional fuel to the reactor. The fuel mass is just a little bit greater than the critical mass.) As would be expected for very large cores with low neutron leakage, the plutonium mass is less for sixty-five per cent density than it is for ninety per cent density. As the core size becomes smaller and leakage increases, fuel of sixty-five per cent density very quickly loses this advantage. For even smaller cores we would expect a crossover. If we increase the steel content, we would expect a crossover to occur at a larger core size because the

fuel of sixty-five percent density would be penalized more severely by the added parasitic neutron capture than would the fuel of ninety per cent density.

The doubling times shown in Fig. 4 are plotted over the same range of parameters as in the preceding figures. For these calculations a plant utilization factor (load factor) of 0.70 was assumed. In order to estimate



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Fig. 4. Doubling Times

the out-of-pile inventory, we calculated the fuel batch size that would be needed for a fourmonth refueling interval. The out-of-pile inventory was then set equal to two such batches. We note that if we go to very large fuel rod diameters, we are penalized in our doubling time because of the low specific power (kw/kg). For the fuel rods of small diameter we have very high specific power, but are penalized because the total breeding ratio is small and also the fractional out-of-pile inventory is large. As a result, we

obtain a minimum doubling time which, for the fuel of ninety per cent density, is in the neighborhood of seventeen years. With half as much steel and a plant utilization factor of 0.8, the minimum doubling time is reduced to 10 years.

The curves in Fig. 5 show the penalties incurred in breeding ratio for increasing the steel content. As you see, the steel penalty is quite appreciable. The smallest core, which would be associated with the smallest diameter of fuel rod, is the least sensitive to the core steel content. This core has the highest plutonium concentration, the highest ratio of plutonium to steel, and the highest leakage from the core into the blanket.



Two different bases for defining the "fuel loading" and "breeding ratio" are shown in Fig. 6. The total plutonium basis in the left hand column is just the definition proposed by Spinrad as the "chemist's breeding ratio." In the right-hand column we consider only the Pu²³⁹ and Pu²⁴¹ fraction in defining breeding ratio and inventory; the Pu²³⁹ and Pu²⁴¹ fraction

FUEL ROD DIAMETER = 0.20 INCH FUEL DENSITY = 90 % OF THEORETICAL STEEL CLAD THICKNESS = 10 % OF FUEL DIAMETER CORE DIAMETER = 4.94 FT.

	TOTAL Pu	Pu - 239 AND Pu - 241
CORE FUEL MASS - Kg.	1400	940
INTERNAL BREEDING RATIO	0.83	0,76
TOTAL BREEDING RATIO	1.43	1.26
DOUBLING TIME - YEARS	19,1	17.5

Fig. 6. Core Parameters

constitutes about two-thirds of the total plutonium. The breeding ratio is markedly higher for the total plutonium basis. This is due primarily to the nonfission capture in Pu^{239} . When we take the balance between the inventory and breeding ratio, we see that doubling times are not appreciably different. In fact, the Pu²³⁹ and Pu²⁴¹ mass is increasing at a slightly greater rate than is the total plutonium. If we had a steady-state closed cycle instead of an open cycle, the two doubling times would have to be identical.

Figure 7 compares the numerical results for rod size, clad thickness, and fuel density. In the upper set, the fuel rod diameter and core size have been varied, and the fuel density and steel ratio held constant. In the second set of results, only the steel ratio is varied, and in the third set only the fuel density.

FUEL Rod fuel			CORE	KW	MEAN NEUTRON	FISSION Fraction		
DIA. IN.	DENS. •/。	STEEL RATIO	DIA. FT.	Kg Pu	ENERGY KEV.	U-238	Pu - 240	<u>(</u> 239
0.125	90	0.10	4.15	1391	238	0.089	0.066	0.259
0.200	41	61	4.94	867	185	0.123	0.053	0. 283
0.280	ei.	н	5.77	530	176	0.136	0.047	0.287
0.200	90	0,05	4.75	914	231	0,144	0.054	0.271
"		0.10	4.94	867	185	0.123	0.053	0.283
"	19	0.20	5.31	780	137	0.091	0.050	0.303
0.200	90	0.10	4.94	867	185	0.123	0.053	0.283
**	65	M		950	193	0.098	0.058	0.276

Fig. 7. Dependence of Parameters on Core Size.

In the fifth column we have the fuel specific power in kilowatts per kilogram of Pu²³⁹ and Pu²⁴¹. Over this range of fuel rod diameters, an increase in the rod diameter decreases the specific power by a factor of about 2.5. In column six we show the mean neutron spectrum. This is the neutron energy in kev such that half of the fissions are caused by neutrons of higher energy and half by neutrons of lower energy. In the subsequent columns we have the fraction of the fissions due to U²³⁸, the fraction of fissions due to the Pu²⁴⁰, and the spectrum-averaged alpha value for Pu²³⁹. As the core size increases, the ratio of uranium to plutonium also increases. Consequently, we get an appreciable increase in the U^{238} fission fraction. For the same reason we get a decrease in the Pu^{240} fission fraction. The neutron energy is degraded as we increase the core size because we are decreasing the concentration of the fissionable material, and this is reflected in the increase of the plutonium alpha value. We feel that for an oxide-fueled core the spectrum degradation and increasing alpha value with increasing core size are not quite as severe as in a metallic core. The alpha values, even for the smallest of the cores, already are somewhat larger than for many of the metal systems.

The effect of the steel is quite appreciable. In going from case four to case six we have quadrupled the steel ratio. There is a very appreciable effect on the neutron spectrum, due largely to the inelastic scattering by the nickel and the iron atoms in the steel. This results in a corresponding increase of the alpha value for plutonium. The U^{238} fission fraction is very appreciably decreased; this is because of the direct competition for neutrons between the U^{238} fissions and the inelastic scattering of the steel. The Pu^{240} fission fraction is also reduced, but not as much as that of the U^{238} because of the lower fission threshold of the Pu^{240} , such that neutrons which are scattered by the steel atoms still get an opportunity to fission the Pu^{240} . The most interesting effect of increasing the fuel density is the increased U^{238} fission fraction because of the higher uranium to plutonium ratio.

A neutron balance for one case is given in Fig. 8. Perhaps what is most interesting here is that the parasitic capture due to the fission products is higher than the combined steel and sodium capture in the core. This fission product capture accounts for about a seven per cent loss of breeding ratio and is one of the penalties which is associated with attaining high burnup. This is more than compensated by other economic considerations, such as processing losses and fabrication costs. I might add that the fission product cross sections we used were the Greebler-Hurwitz values, except we anticipated that the cross sections at 100 kev would be higher if we took into account the p-wave effect, and hence arbitrarily doubled the fission product cross section at that energy and then replotted the fission product absorption curve. Another effect here, which may be of interest and which to a large extent accounts for the fact that the internal breeding ratio is going up with increasing core size, is a spectrum difference between the

	CORE	BLANKET	TOTAL		
U-238 FISSIONS	. 0410	.0083	.0493		
Pu-(239+241) "	. 2859	0	.2859		
Pu-(240+242) "	.0198	0	.0198		
SUM FISSIONS	.3467	.0083	.3550		
U-238 CAPTURES	. 2 5 7 0	.1874	.4444	Fig. 8.	Neutron Balance
Pu-(239+241) "	.0830	0	.0830		
Pu-(240+242) *	,0267	0	.0267		
F.P. "	.0299	ο	0299		
STEEL "	0242	.0106	.0348		
SODIUM "	.0018	.0004	.0022		
SUM CAPTURES	. 4226	. 1984	.6210		
LEAKAGE		. 0240	.0240		
			1.0000		

0.2" FUEL DIA., 90% FUEL DENSITY 0.10 STEEL FACTOR, 4.97 FT. CORE DIA.

core and the blanket. The ratio of U^{238} fissions to captures in the core is about 1 to 6. The ratio of fissions to captures in the blanket is less than 1 to 20, which is accounted for by the degradation of the neutron spectrum as we go out into the blanket. Thus, if we have a small core with a high ratio of plutonium to uranium in the core, we have a high neutron leakage into the blanket; but the neutrons which leak into the blanket really do not get the same opportunity to fission the U^{238} as do those in the core, even with the larger fertile material concentration in the blanket.

Figure 9 shows the excess operating reactivity which must be held down by control rods as a function of the refueling interval. As we go to the smaller fuel rod diameters, corresponding to the smaller core sizes, two things happen: first, we have a higher specific power, a faster burnup; and secondly, we get a lower internal breeding ratio, both of which give us a more rapid decrease of reactivity with operating time. Thus we see that if we pick a given refueling interval, say four months, and if we want such a refueling interval with the smallest of these cores, we must have about 7.5 dollars of excess operating reactivity. For the intermediate core size we need something like 2.5 dollars, whereas for the largest of the cores studied we can get by with about seventy cents of excess operating reactivity. If it were necessary to keep the operating reactivity in a fast reactor below one dollar, and I feel that this will not be the case, then we see that with the smallest of these cores we would have to shut down for refueling about every two weeks. For the core of intermediate size we could go about a month and a half, and there would be no problem in the largest of



Fig. 9. Operating Reactivities

the cores, as we could operate for about six months before refueling. Whether or not we limit ourselves to a dollar, the excess reactivity is an important safety consideration. It will be necessary to design a control system which will not permit the reactor to go prompt critical under any conceivable circumstance, and the ease with which such a control system could be designed would depend strongly on the amount of excess reactivity that must be controlled by rods.

The sodium temperature coefficient of reactivity (Fig. 10) is plotted over the same range of parameters as previously. The scale here is $\Delta \supset x \ 10^{-6}$ per °C. The temperature coefficient is negative over the entire range except for the very largest of the core sizes that were considered. As the neutron leakage increases, either as a result of decreasing the core size or of lowering the fuel density, the magnitude of the negative temperature coefficient increases. The numbers which are listed adjacent to some of the points are the ratios uranium to Pu²³⁹ plus Pu²⁴¹. The sodium temperature coefficient of reactivity is sensitive to the steel content. This is largely a spectrum effect. As we reduce the amount of steel in the core, the effect of the sodium on the spectrum becomes more important. For the 0.2-in. fuel rod diameter at 90 per cent fuel density, removing onehalf of the steel reduces the magnitude of the negative temperature coefficient by one-half. Also, as we allow plutonium to build up in the blanket, the temperature coefficient becomes slightly less negative. Here again we get a positive contribution because we are effectively increasing the core size.



Fig. 10. Sodium Temperature Coefficient of Reactivity

A comparison of physics properties for the annular core and the conventional cylindrical-type core, both having the same fuel rod diameters, fuel density and steel ratios, is shown in Fig. 11.



FUEL ROD DIAMETER = 0.20 INCH FUEL DENSITY = 90 % OF THEORETICAL STEEL CLAD THICKNESS = 10 % OF FUEL DIA.

	CORE	CORE
TOTAL BREEDING RATIO	I. 26	1.20
INTERNAL (CORE) BREEDING RATIO	0.76	0.66
CENTER BLANKET BREEDING RATIO		0.09
CORE MASS, Kg. Pu (239 + 241)	937	965
CORE MASS, Kg. Pu (TOTAL)	1400	1440
CORE RADIAL PEAK/AVERAGE POWER	1.64	1.28
AVERAGE FUEL POWER DENSITY, KW / Kg. Pu (239 + 241)	867	841
OPERATING REACTIVITY FOR 4 MONTH REFUELING INTERVAL, DOLLARS	2.4	1.9
SODIUM TEMP. COEFF. OF REACTIVITY, (Δρ/°C)	-6.18 X 10-6	-3.85 X 10-6

ON INDRICAL

Fig. 11. Comparison of Conventional and Cylindrical Cores

At the top of the figure is a cross-sectional sketch of the annular core configuration. The central blanket is nine inches in radius; the outer blanket was reduced from the eighteen inches used previously to a sixteen-inch thickness. The composition of the outer blanket is the same as that for the cylindrical core. The composition of the central blanket is the same as that of the core except that it is clean - there is no plutonium in the fuel. The annular core did not live up to many of the expectations that we had for it. The total breeding ratio is slightly less than for the cylindrical core. In spite of an improved radial power distribution, as indicated by the radial peak-to-average values shown, the fuel specific power was less for the annular core; that is, the plutonium loading was higher in the annular core due to a higher plutonium-to-uranium ratio which was required to override the higher core leakage. The excess operating reactivity required was lower for the annular core than for the cylindrical core, but the difference was much less than had been expected. The temperature coefficient was less negative for the annular core than for the cylindrical core. We believe that this is due to the central blanket effect, i.e., neutrons leaking into this blanket causing U²³⁸ fissions.

The other factor considered was the effective delayed neutron fraction. Here we have done only a very crude calculation, but again we have found that there is no appreciable difference in the effective delayed neutron fraction as between the annular configuration and the cylindrical core. On the basis of these results it appears that the advantages of the annular core, if any, do not warrant the added complexities of design. At the other extreme, going to an internal breeder, a nonblanketed system does have design simplicity; but if we desire both good fuel economy (i.e., low fuel cost) and a short doubling time, the core size must be very large and requires a high inventory of plutonium. We feel this is a reactor concept which may become very attractive sometime in the future when there is a demand for plants of considerably higher power rating than 300 megawatts electrical. As of now, it appears that a good design for a fast oxide breeder reactor at about 300 megawatts would utilize a cylindrical-type core with high-density fuel, a fuel rod diameter of intermediate size (~ 0.2 in.), and the minimum steel clad thickness consistent with the requirement for high burnup.

Discussion of Paper Presented by Mr. Greebler

MR. SAMPSON:

I would like to raise a discussion both on this paper and possibly Dr. Okrent's. First, I gather that your work essentially substantiates all former KAPL analyses of a few years ago as far as the matter went. It is much less detailed. I can't detect any great difference in appearance or concept. You have started with more accurate input information.

MR. GREEBLER:

Well, we had all the fine work of yours to lean on and I think we just carried it a little bit further and in a little bit more detail.

MR. SAMPSON:

What did you assume in your alpha curve? There were various references this afternoon to the standard curve and it is something we fooled around with, but I am not sure that these are one and the same. Is there any more experimental information in the last few years?

MR. GREEBLER:

I am not aware of any. We used the so-called "best fit" to the available experimental data, which yields the intermediate plutonium alpha values.

MR. SAMPSON:

That is the same old ---

MR. GREEBLER:

That you used, right.

MR. SAMPSON:

Well, the other point: I would certainly like to congratulate the General Electric Company for getting back to a good thing.

MR. MacCARTHY:

Are there any further questions?

MR. KASTEN:

I want to raise a question about the spectrum. You mentioned that when you add stainless steel to the core the spectrum went down; but you made the core larger, presumably, to keep the same fuel concentration. Is this right?

MR. GREEBLER:

That is correct - to keep the same fuel volume.

MR. KASTEN:

Now, presumably you could have kept the spectrum up by keeping the same core size and putting in more fuel.

MR. GREEBLER:

This would have hardened the spectrum some. If you notice the effect of the steel on the core size, considering the very appreciable range of core sizes that accompany the range of rod sizes covered, the change in core size as a result of increasing the steel content was a very small one.

MR. KASTEN:

Another point concerning the breeding ratio - it went up as you increased the rod diameter. What did you use to determine the rod diameter to use?

MR. GREEBLER:

We kept the core power fixed at 812 megawatts thermal.

MR. KASTEN:

Just increased the core diameter?

MR. GREEBLER:

The volume of fuel was determined from the rod size. The rod size essentially determines the specific power for a given peak temperature and peak-to-average power distribution. Dividing the specific fuel power into the total power required gives the fuel volume. We have this direct relationship between rod size and core size.

MR. KASTEN:

When you calculated doubling time did you use the fuel inventory alone or did you consider the whole fuel cycle?

MR. GREEBLER:

We used an out-of-pile inventory ratio which was based on our fuel batch size. We had the fuel batch sized for a four-month refueling interval and then we took two of these batches for the out-of-pile inventory. This is equivalent to saying that the cycle process line requires an eight-month period.

MR. HAEFELE:

I am sure you did take into account the fission product poisoning for the average breeding ratio?

MR. GREEBLER:

Yes, the fission product level was equivalent to that of 50,000 MWD/T burnup, the average core condition.

MR. HAEFELE:

Did you take into account the absorption cross section present due to the (n, α) effect in oxide?

MR. GREEBLER:

No.

MR. WIGNER:

What did you estimate about the efficiency of the chemical separation?

MR. GREEBLER:

We did not include processing losses into our breeding ratio evaluations.

MR. WIGNER:

What would you estimate they would be?

MR. GREEBLER:

This can easily be kept down to two per cent. Some people deliberately make it as high as four percent to throw out the Pu^{242} . This is commonly considered for plutonium recycle in thermal reactors. For a fast reactor one would not want to throw out the Pu^{242} , and the process loss would be of the order of two per cent. With burnups as high as 100,000 MWD/T, this represents an extremely small loss of breeding ratio.

MR. MacPHERSON:

I would like to ask about the relationship between the time that you showed between recycling and the assumed 100,000 MWD/T fuel burnup. Is there a relationship?

MR. GREEBLER:

Yes, there is a very direct relationship, but it depends on the specific power. You are asking essentially how many years one fuel element remains in the core. Taking a plant utilization factor of 0.70 and the largest of the rod sizes that we considered here, this would be as long as ten years. For the smallest of the rod sizes it would be about 2.5 years.

MR. MacPHERSON:

What is the relation of this to the variation in control rod that you had to show?

MR. GREEBLER:

What we stated here was that if we wanted a given refueling interval, say four months, then in the smallest of the cores which also has the smallest rod, we get the most rapid decrease of reactivity during the four month interval. We would get the greatest amount of negative Δk , because (1) we are burning at the highest specific power, and (2) our breeding is not as large in the core as it is out in the blanket. Therefore, for a four-month refueling interval, we must use a larger batch size with the smaller core to give us the higher required Δk .

MR. MacPHERSON:

After this four-month period you replace a portion of the rods. Is this correct ?

MR. GREEBLER:

That is right.

MR. VON HERRMANN:

How did you calculate these very small termperature coefficients? What confidence do you have in them?

MR. GREEBLER:

Actually, we used "brute force" multigroup methods, that is, we repeated a set of calculations using different densities of the sodium corresponding to a range of temperatures.

MR. VON HERRMANN:

In other words, you converged the problems down to these extremly small numbers.

MR. GREEBLER:

They iterated and iterated, yes, until we satisfied a very fine convergence criterion. In this connection, I would think that perturbation methods, for example, would be very desirable for an evaluation of the sodium temperature coefficient; and this is something we intend to do as soon as we can get a good program for calculating adjoint fluxes with inelastic scattering. With perturbation techniques one can clearly examine each effect that contributes to the overall sodium temperature coefficient separately. However, the sodium temperature coefficients given should be as accurate as if they were calculated with existing perturbation techniques.

MR. BARTELS:

On one of your figures you showed your critical masses, and they were lower for your lower density oxide. Could you show how your ground rules ended up with this rather unexpected result?

MR. GREEBLER:

Why were critical masses lower with low density oxide? Yes, you noted that this was particularly the case for large core sizes. For a very large core size, where the leakage is small, the principal competition for neutrons is between uranium and plutonium. Consequently, criticality is achieved with a given uranium-to-plutonium ratio, independent of the oxide fuel density. Given a fixed uranium-to-plutonium ratio, there is a larger quantity of both uranium and plutonium at ninety per cent fuel density than at sixty-five per cent fuel density.

MR. BARTELS:

Did you estimate internal gas pressures at your twelve per cent burnup for any of these?

MR. GREEBLER:

A rough calculation has been made. This is another reason why we are considering low-density fuel. If we get a one hundred per cent release of the fission product gases, which is possible for 100,000 MWD/T burnup, then at ninety per cent fuel density the clad would not withstand the pressure of the fission gases. At sixty-five per cent density we would just about make it. We are conducting irradiation tests now to determine whether or not we will get such high fission gas release from the oxide, and if we do, then we will have to find some solution, such as a void chamber above the fuel element, in order to use high density oxide.

MR. ZINN:

My question, I believe, was almost answered by the question and answer; but if I understood you correctly, your best performance reactor had a fuel element diameter of about 150 mils and a jacket thickness of about 15 mils. Then, the chance of containing the gas pressure is really nil.

MR. GREEBLER:

Actually, the important parameter is the ratio of the clad thickness to the fuel rod radius, not so much the absolute thickness.

MR. ZINN:

But the original concept was one in which the pin, so to speak, did have the strength to contain the fission gases at whatever burnup was assumed. You are not retaining that concept here.

MR. GREEBLER:

Our ratio of wall thickness to rod diameter is not appreciably different from what KAPL had assumed, if we take our intermediate value of one-tenth. If you are referring to the KAPL irradiation test, they never quite reached 100,000 MWD/T; only about half of this value. Furthermore, they were using sixty-five per cent density fuel, instead of ninety per cent, so they had a sizable void fraction in their fuel element. Our calculations indicated that their steel clad thickness was adequate to retain the fission gases even at one hundred per cent release. Consequently, that particular test did not prove the ability to retain fission gases for high-density fuels with 100,000 MWD/T burnup. MR. BAKER:

I would like to ask two short questions. The first concerns the temperature coefficients with sodium. Have you any figures for the effects of loss of sodium separately from the inner blanket of your inner blanketed core?

MR. GREEBLER:

No.

MR. BAKER:

From your figures could you tell me what would be the gain in reactivity if you lost the entire sodium?

MR. GREEBLER:

We did not calculate that.

MR. BAKER:

The second question - this is in your basic data which determined the size of your reactor - you said you chose the ratio of height to diameter to be equal. Now, there would seem to be some advantage in having a shorter core when you go to a five-foot diameter. Did you consider this?

MR. GREEBLER:

We did not include that in the initial scoping study, but we realized this would be the case. In an engineering design we very likely would use a value of H/D something less than 1. This should improve the core physics by reducing the sodium volume fraction, but otherwise would be equivalent to a smaller core with H/D = 1.

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SESSION III

Tuesday Morning, October 20, 1959

Chairman: D. B. Hall Secretary: G. J. Fischer

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BREEDING IN FAST PLUTONIUM METAL SYSTEMS WITH EBR-II-TYPE RECYCLE

Harry H. Hummel Argonne National Laboratory Lemont, Illinois

I shall discuss the effect of using a fuel processing system similar to that to be employed by the EBR-II reactor for a somewhat larger plutonium metal system. Attention will be focused on an 800-liter core reactor, considered by Loewenstein and Okrent in their parameter study.⁽¹⁾

This reactor has 25 volume per cent fuel plus fertile material, 25 per cent steel and 50 per cent sodium, and the ratio of fuel to diluent is 0.14. Arbitrarily it is assumed that the power density is 1.0 megawatt per liter.

In Fig. 1 is a description of the EBR-II fuel $cycle^{(2,3,4)}$ as it would be applied to such a reactor.



Discharged fuel from the reactor is first cooled for 15 days, after which the radiation level is low enough to permit the remote processing that it is planned to use. The fuel is then melted and an oxide slag forms, a number of the fission products being oxidized and tapped off. Volatile fission products are evolved as gases. Fission products which are noble elements are not removed by this process and part of these remain in the recycled fuel. The recycled fuel containing these elements has been given the name "fissium" by the chemical engineers. The most important elements from the standpoint of fission product poisoning are listed in Table I in the approximate amounts in which they would be present for 5 w/o total recycled fission products. The relative amounts would be about the same for 10 w/o recycle.

TABLE I

101 a 10(a) 01 9 w/ 0 /0					
Isotope		w/o	%		
Мо	95 97	0.3			
	98	0.3			
	100	0.3			
			1.2		
Ru	101	0.6			
	102	0.6			
	104	0.6			
	106	0.2			
			2.0		
\mathbf{Pd}	105	0.3			
	106	0.2			
	107	0.3			
	108	0.2			
			1.0		
Rh	103		0.4		
Tc	99		0.4		
			5.0		

Approximate Amounts of Important Fission Products in Recycled Fuel for a Total of 5 w/o %

The isotopic composition of Mo, Ru, and Pd has been based on the work of Burris and Dillon⁽⁵⁾ for fast fission in Pu^{239} , using their results for 135 days of irradiation and a 1-yr decay on a once-through basis. Fission product burnup is neglected. This of course does not correspond exactly to the

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present conditions, but the composition should be close enough for the purposes of the present analysis, especially in view of the uncertainty about the value of the capture cross section for any given isotope.

In addition to the slag, a certain amount of the fuel is tapped off every time in order to permit the removal of those fission products that cannot be separated out in the slag. This metal is subjected to a further processing step, the exact nature of which has not yet been determined, where it is cleaned up until everything except the plutonium isotopes is removed. Some pyrometallurgical process will be employed; liquid metal extraction using cadmium as a solvent is a possibility.

The purification process according to the chemical engineers will require two weeks at most. This is a short time compared to the reactor cycle which has been assumed in this example to be based on 2 per cent burnup of total fuel alloy atoms. The cycle length turns out to be of the order of 90 days, so that there is time for a number of fuel purification steps. The processing plant would be sized so that one could run the required number of batches through it in the time that a fuel charge is running in the reactor.

It should be made clear at this point that no plutonium metal or alloy fuel element yet exists capable of achieving 2 per cent burnup at satisfactory operating temperatures, and the conditions chosen for this example illustrate what one hopes to be able to do.

The amount of fission products that are sent back in the recycled fuel can be controlled by changing the fraction of fuel drawn off with the slag for further processing. This stream is referred to as "drag out." The drag out is set at about 10 per cent of the total fuel in order to obtain 5 w/o fission products in the recycled fuel and about half this to obtain 10 w/o. It turns out to be desirable from a metallurgical point of view to have these fission products present in the recycled fuel, as swelling from fission product gases is reduced. For uranium alloy fuels 5 w/o recycled fission products is sufficient to give rather satisfactory properties with regard to radiation damage.

Unfortunately, the picture is not so bright for plutonium alloy fuels. For fuels containing 20 per cent plutonium it appears that, even with 10 w/o fission products in the alloy, an operating temperature of no more than 350 to 400°C is possible before prohibitive swelling of the fuel occurs. Present experimental results suggest that somewhat higher operating temperatures would be permissible for 10 per cent plutonium. The whole problem of plutonium alloy fuels is still in an early stage of study, and it may be that additives will be found that will improve the situation. The present outlook is not encouraging, however.

In order to study the effect of this recycling operating on the breeding properties of such a reactor, the isotopic composition of the recycled fuel was examined first. In Table II are listed fission and capture cross sections
for the various plutonium isotopes of interest. In some cases these are based on measurements which have been averaged over the calculated spectrum of the 800-liter core reactor being considered. The effective neutron energy for this spectrum is of the order of 100 to 200 kev. In other cases, particularly for capture cross sections, measurements are not available, and estimates must be made from theoretical considerations.

TABLE II

Isotope	σ _f (barns)	σ _c (barns)	Equilibrium Isotopic Ratio for a Processing Loss per Cycle of	
			0%	2%
Pu ²³⁹	1.85 ^{a(6)}	0.39 <u>+</u> 0.10 ^c	1.00	1.00
Pu ²⁴⁰	0.36 ^{a(6)}	0.36 ^b	0.546	0.396
Pu^{241}	3.4 ^b	0.30b	0.053	0.036
Pu ²⁴²	0.31 ^{a(7)}	0.40^{b}	0.022	0.011

Fission and Capture Cross Sections of Plutonium Isotopes, and Equilibrium Isotopic Ratios in Recycled Fuel

^aBased on measured cross sections.

^bEstimated (see discussion following paper).

^CBased on EBR-I irradiations (see Ref. 1).

These uncertainties actually do not turn out to be serious insofar as prediction of the breeding properties of the type of system being discussed is concerned. In Table II are given the equilibrium ratios of Pu^{240} , Pu^{241} , and Pu^{242} to Pu^{239} in the recycled fuel based on these cross sections. Results have been given for 0 per cent and 2 per cent processing loss per cycle. The 2 per cent figure is believed by the chemical engineers to represent an upper limit, and they hope to stay well below it. It is seen that the relative amounts of Pu^{241} and Pu^{242} are small and it seems unlikely that reasonable alterations in the cross sections could make them much larger. The exact amount of Pu^{241} present is not very important, since it is a fuel at least as good as Pu^{239} .

The only isotope present in large concentration besides Pu^{239} is Pu^{240} , which behaves as a fertile material similar to U^{238} in a fast reactor. It is somewhat more valuable than U^{238} because of a larger fast fission effect due to a lower fission threshold. Loewenstein and $Okrent^{(1)}$ found for the 800-liter core reactor under consideration that a variation of the ratio of Pu^{240} to Pu^{239} from 0 to 1 caused a variation in breeding ratio from 1.61 to 1.70, while the critical mass was decreased from 468 kg Pu^{239} to 421 kg. Thus Pu^{240} has a slightly beneficial effect from a physics standpoint and acts principally to displace U^{238} as a fertile material, assuming the fuel alloy volume fraction held constant. This is unfortunate from a metallurgical point of view because of the considerations previously mentioned.

The effect of the recycled fission products on breeding must now be considered. In Table III are listed approximate breeding ratios and doubling times; fission product poisoning and processing loss have been considered. The work presented by Moldauer⁽⁸⁾ indicates that the fission product capture cross sections as calculated by Greebler, Hurwitz, and Storm⁽⁹⁾ give results too low by about a factor of 2, which was also suggested by those authors. Two different assumptions have been made in Table III: an "optimistic" one in which fission product capture has been based on 2 times the cross sections of Greebler et al. with 5 w/o recycled fission products, and a "pessimistic" one in which a factor of 3 has been applied to the cross sections of Greebler et al. and 10 w/o fission products assumed. Doubling times have been based on an average power density in the core of 1.0 Mw/liter and an 80 per cent operating factor.

TABLE III

	⁰ FP per Fuel Atom,	Breeding	Doubling Time, years ^a	
	barn	Katio	No Loss	2% Loss
No Fission Products		1.67	5.5	~
5 w/o Fission Products; 2 x σ _{FP} of GHS ⁽⁹⁾ 10 w/o Fission	0.2	1.59	6.3	8.0
Products; $3 \times \sigma_{FP}$ of GHS ⁽⁹⁾	0.6	1.47	7.8	10.6

Approximate Breeding Ratios and Doubling Times

^aBased on twice the reactor core inventory and an 80% operating factor.

It is seen that while, in the "pessimistic" case the effect of fission product poisoning is certainly beginning to be felt, the breeding characteristics are still very satisfactory. It is worth noting that the fission product poisoning from the recycled fission products is much larger than that from those freshly produced in the cycle. The ratio of the former to the latter is 10 for 5 w/o fission product recycle. The effect of the freshly produced fission products will always be very small in a fast metal-fueled system because of the severe limitation on burnup imposed by radiation damage.

The numbers in Table III are only approximate, as they represent estimated corrections to calculations made without poisoning. They are believed to be sufficiently accurate, however, to illustrate the point that the real problems of breeding in this type of system lie not in the physics but rather in the engineering.

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Discussion of Paper Presented by Mr. Hummel

MR. HALL:

Are there any questions on this paper?

MR. WIGNER:

There is one point I didn't quite catch, and perhaps you can elucidate. If you had a production ratio, as people now call it, of 1.67 and you burn up 5 per cent, then you have in one cycle a production of 3 per cent. If you have a chemical loss of 2 per cent, then two-thirds of this is lost. I am sure that there is some error in this calculation, but what is it ?

MR. HUMMEL:

The fuel burnt per cycle is about 16 per cent of the total and the breeding gain is about 0.60, so that the production per pass is about 10 per cent of the fuel. The internal breeding ratio is about 0.75 for a clean core but drops somewhat when fission products are present. Most of the fuel burnt is thus regenerated in the core, so that the plutonium inventory does not drop much in the cycle.

(MR. WIGNER inquired if it was possible to achieve 2 per cent burnup in the type of system under discussion.)

MR. HUMMEL:

I don't know whether any significant burnup could be achieved, since according to the metallurgists it looks as if one could go only to about 400° C, so that it is questionable whether anything like this is ever going to be built.

MR. CAMPISE:

What assumptions have you made about neutron leakage as far as calculating your breeding ratio?

MR. HUMMEL:

I haven't done any machine calculations myself; I drew all my strength from Loewenstein and Okrent.⁽¹⁾ These breeding ratios are approximate only but the error is probably not important for the purposes of the present discussion.

MR. OKRENT:

I think you can always design a blanket which will catch a reasonable number of the neutrons that get out. Leakage from the blanket is not going to be, I think, a problem in the end There are ways of moderating neutrons so that they are caught efficiently. It is fair to assume that the lion's share of the neutrons escaping the core are captured until you are at the point where you have good cost figures and have a basis for letting neutrons escape rather than catch them.

MR. HALL:

Just to join the argument, there is, however, the point that Mac brought up yesterday, in that if you go to the extremes of catching all of the neutrons by making your blanket essentially infinitely large, you may find yourself at an economic disadvantage.

MR. OKRENT:

Well, the particular blanket used here was probably about 18 inches thick. This is not a very thick blanket, in fact, so there was some leakage. I think one can do a variety of things to blankets, like putting moderator into the blanket, to get rid of the problem you just mentioned. You can concentrate your collection of uranium. One form of this idea is the coupled reactor which we will hear discussed this morning, but you can do this in a way so you don't couple the core and the moderator and still concentrate the collection of the uranium. So, this is an artificial problem that exists in the present design because there was no absolute need for solving it yet. I think we will have an adequate solution in the future reactors.

If you took a standard 18-in. thick uranium blanket and merely substituted graphite for the uranium in the central third, the blanket efficiency would be essentially unchanged, but the volume of uranium involved would be cut by one third. The plutonium production would be much more highly concentrated, and you would be rid of this edge effect. Also, one could put a graphite reflector just outside the blanket, simultaneously thinning the blanket somewhat, and again improve the overall economics with no cost to breeding ratio.

For this type of analysis it is not important to specify just how you are catching the neutrons as long as you allow for some parasitic absorption, which we have done here in the form of cladding and coolant in the blanket.

MR. MacPHERSON:

I wonder if you have taken into consideration in calculating the equilibrium ratio of the isotopes the net effect of the breeding ratio, such

as was pointed out by Spinrad yesterday. You have an effective removal rate which is in excess of this 2 per centas a result of generating excess material in the reactor. Is this right?

MR. HUMMEL:

All I have done in making this calculation is to equate the production and consumption rate -

MR. MacPHERSON:

Well, I think the point I am getting at is that in considering the longterm operation of a reactor like this, since it is a breeder, you will be taking out the fuel which will be, presumably, of some composition, and this will affect the build-up of the higher isotopes of plutonium just in the same way that your two per cent processing loss affects it.

MR. HUMMEL:

These ratios are based on assuming that the make up fuel is Pu^{239} taken from the blanket, which has about 0.8 per cent Pu^{240} . Is this what you are referring to?

MR. MacPHERSON:

And recycling the core?

MR. HUMMEL:

Yes, recycling the core plutonium -

MR. MacPHERSON:

It isn't exactly the same. I just think the process of breeding in itself will effect a removal rate which will probably limit the build up of the isotopes 240, 241 and 242.

MR. HUMMEL:

Well, I am afraid I have not taken into account whatever it is you are worried about.

MR. MacPHERSON:

Well, okay.

(MR. HUMMEL - WRITTEN DISCUSSION)

The difficulty with introducing moderator in the blanket caused by the high alpha of Pu^{239} at low energies would arise from interaction with bred Pu^{239} in the blanket. Studies on the coupled system have shown that a relatively thin blanket is sufficient effectively to exclude moderated neutrons from the core. Lowering of the breeding ratio because of blanket moderation could be minimized by rotating blanket material to maintain a low concentration of Pu^{239} adjacent to the moderator. Conceivably it could be necessary to process blanket material more frequently for a moderated than for an all-fast blanket. It was assumed that the bred plutonium was that made in the blanket. No core plutonium would ever be taken out, so that with this method of operation the effect Mr. MacPherson was referring to would not be present.

MR. GREEBLER:

I am interested in this Pu^{241} cross-section set. Was this calculated or measured?

MR. HUMMEL:

Mssrs. Moldauer and S. Yiftah of Argonne have been worrying about this - these are calculated numbers that Moldauer wrote on the back of an envelope, so to speak.

MR. OKRENT:

There exists one measurement in a small assembly at Los Alamos from which one can derive a value of $\nu \sigma_f$ fission for Pu^{241} . One can also, on theoretical considerations, make a guess as to what the fission cross section for plutonium-241 should be. The theoretical estimate that Moldauer made turned out to be higher than the upper limit that we could derive from the measurements, so we are using the upper limits of experiment in this case.

MR. KASTEN:

In calculating your doubling time, did you assume any fuel in the blanket?

MR. HUMMEL:

No. This was just based on the core inventory.

MR. KASTEN:

But under normal conditions you would build up fuel in the blanket and doubling time should be based on fuel in the blanket. MR. HUMMEL:

You would have to change the doubling time on this basis, as it has been based on the core inventory only.

MR. KASTEN:

You mentioned there are very little data available which indicate that the fuel elements have substantial life. Can you tell me anything about the effort in that direction?

MR. HUMMEL:

I really don't know anything about this. I am sure that the metallurgists are working hard on this point, but I had only brief conversations with them. Perhaps Howard Kittel in Metallurgy would be a good person to talk to. He is really the one who has the best collection of information on this.

MR. KASTEN:

Thank you.

MR. OKRENT:

Just to indicate how things have been going here, until recently there was not a facility available wherein one could make many plutonium samples for irradiation studies and so forth. So all the early work has been done on the uranium fissium alloy. I think it is believed that one can run uranium fissium alloy at temperatures up to about 600°C, and get one per cent burnup or more. That is total burnup now, not fissionable atoms.

The plutonium work is more recent. One expects it to behave more poorly because the melting point is lower for the plutonium-uranium mixture and, therefore, its high temperature strength should be poorer. Therefore, you expect the fission product gases to cause swelling at a lower temperature.

I think the degree of pessimism that Hummel has expressed reflects this. We are not going to claim we have fuel elements when we are inclined to doubt that we have a very good one at the present time. But we really don't know. This work is in its infancy.

MR. BARTELS:

If I understand your terms correctly, your breeding ratio and doubling times are defined as follows: your breeding ratio takes into account the breeding in both the core and the reflector, and your doubling time is the time required for the fissionable material bred in the core and breeder region to double the initial inventory in the core itself.

MR. HUMMEL:

The doubling time is that required to double twice the core inventory. This allows for holdup in the processing plant.

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MR. BARTELS:

Now that I understand the terms which I was a little bit shaky about if it is possible to surround the breeder region, by say an inexpensive graphite region, would you not increase your breeding ratio for this type of system significantly by going to a core of still higher leakage? I am speaking now of the average breeding ratio of a fuel cycle, since you would be, apparently, less penalized by your fission products.

MR. HUMMEL:

Well, of course in a plutonium system the alpha is rather high in the regions of lower energy, so that one has to be careful about introducing moderator into such systems.

Let's see. Your idea is that at the outside of the blanket you put a graphite region to moderate neutrons. How is this going to affect the problem of fission products?

MR. BARTELS:

Well, I am trying to do two things implicitly. Go to a higher leakage system and do more of your breeding in the blanket and still not get a thick, massive blanket that increases your chemical processing cost.

MR. HUMMEL:

Yes. Well, my off-hand opinion is that this might be better on the U^{233} -thorium cycle because of the high intermediate energy Pu^{239} alpha.

Pu-U AND U²³³-Th CERMET-TYPE FAST BREEDERS

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General

One of the most attractive fast reactor fuel types from the standpoint of achieving high breeding ratios is the cermet type, in which fissionable material in a ceramic form is dispersed in a matrix of fertile material. Cermet fuels are considered to be capable potentially of achieving high burnup and, of equal importance from a breeding standpoint, they permit high concentrations of fertile material in the core region. Future designs of fast breeder reactors will be strongly influenced by economic considerations. It appears that the effect of these economic considerations will be to influence the reactor designs in the direction of larger power outputs, lower power density, and higher burnup. Lowering the power density makes it possible to attain higher burnup by reducing fuel temperatures. Another means of achieving higher burnup is to increase the dilution of the fissionable material. These trends add up to larger, more dense cores. In such cores there is reduced neutron leakage to blanket regions where it is easily possible to achieve high concentrations of fertile material and thus high probability of productive neutron absorption. In other words, in future designs the core will become more significant as a breeding region, and the blanket less so. Therefore with advanced cores it becomes more important to obtain high concentrations of fertile material in the core region for breeding purposes. This is easily accomplished with a cermet-type fuel where the matrix is composed of fertile material. If, in the interest of achieving high burnup, the fissionable material is diluted with nonfertile materials, such as stainless steel, as in the case of a stainless steel cermet, even though burnup is significantly improved, capture in $U^{238} \label{eq:constraint}$ is seriously decreased to the point where the system probably no longer breeds. Thus, while steel cermets are attractive economically, they have no place in the advanced fast breeder program.

Aside from locating the fertile material where it gives the highest probability of productive absorption, there are other benefits that result from having large concentrations of fertile material in the core. For one. the effect of the fertile material on the spectrum is to cause it to peak more in the central part and drop at either extreme. Reducing the spectrum on the low-energy side improves breeding efficiency by reducing the effective value of alpha and also by decreasing other nonproductive absorptions in structure and alloying materials - in general, losses to materials whose capture cross-sections decrease rapidly with increasing energy. The most significant effect is the increase in fast fissions. Even though the increased inelastic scattering lowers the neutron flux above threshold fission energies, this is more than compensated by the higher concentration of fertile material. For this meeting a study was made of fast Pu-U and U^{233} -Th systems. The fuel consisted of PuC dispersed in a matrix of U-15 w/o Mo in one case, and of UC in a pure Th matrix in another. A general description of these reactors is given in Table I.

TABLE I

Reactor Description

Core Volume: 1750 liters Control Volume: 7%	
Active Core Composition	(volume percent)
Fuel	50
Structure and Clad	18
Coolant	32
Blanket Thickness (cm)	45
Blanket Composition	(volume percent)
Fertile Material	44 .
Structure and Clad	20
Coolant	36

The PuC-U-Mo cermet systems were recently considered in a parameter study performed at APDA under AEC contract AT (11-1)-800. In this study the effect of core size and composition on fuel cycle cost were considered. The core size and composition used here are a result of that study. This reactor would be capable of producing about 800 megawatts of heat.

This power obviously could be developed with a smaller core. The large size reflects the dependence of the radiation stability of the fuel on fuel temperature, and, hence, its effect on fuel cycle cost. For a given specified power it is possible with the larger core to operate at lower fuel temperatures, and therefore at higher burnup. This increase in burnup is more significant in reducing fuel cycle cost than reducing in-pile inventory. Although only the PuC-U-Mo systems were considered in detail as far as thermal performance is concerned, it was assumed that the UC-Th systems would be capable of the same power and burnup. The fuel temperatures would certainly be lower because of the higher thermal conductivity of the thorium, but the burnup characteristics of thorium cermets are poorly known. The average burnup of the fuel material was assumed in both cases to be 16×10^{20} fissions per cubic centimeter of fuel. This is equivalent to about 3 atom percent burnup of the PuC-U-Mo fuel and 5 atom percent of the UC-Th fuel.

For both fuel types a range in the isotopic composition of the primary fissionable material has been covered. In one extreme either the Pu²³⁹ or U²³³ is considered to be the only isotope present. This could be representative of the initial operation of these plants, depending upon the initial source of the fissionable material. The second case represents the composition of the fuel after many recycles in the course of which it has reached an equilibrium composition.

PuC-U-Mo Cermet

Some of the pertinent characteristics of the clean PuC-U-Mo system are given in Table II. In arriving at these results the composition of the core was adjusted to correspond to one-half the maximum burnup. The critical mass is slightly less than 1000 kg. Approximately 50 per cent of the neutrons are used in producing new fissionable material. Of the 8.5 per cent of the neutrons lost in nonproductive processes, about 2 per cent leak out of the blanket, about one-half of these losses are due to capture in the molybdenum alloying material, and most of the remaining losses are captures in steel. Only about 0.5 per cent of the neutrons are captured by fission products at equilibrium burnup.

TABLE II

PuC-U-15 w/o Mo Fuel				
u ²³⁹				
34.7%				
in Fuel 5.7				
Material 51.1				
8.5				
1.03				
0.50				
1.53				
1.48				
0.205				
21% of total fissions				

One of the interesting results is that the internal breeding ratio is slightly greater than one. By increasing the amount of U^{238} in the core it would be conceivable to adjust the core conversion ratio to compensate for reactivity losses associated with burnup, and thereby to reduce neutron losses connected with reactivity control.

INDUE II

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The core, blanket, and total breeding ratios as they are given here apply only to the plutonium produced and destroyed in the reactor. When external losses (in reprocessing and fabrication) are taken into account, the total breeding ratio decreases by about 0.05, assuming an external loss of 2 per cent, to give a system breeding ratio of 1.48. If the external losses doubled or if the achievable burnup was only half the assumed value, the reduction in the breeding ratio due to external losses would increase by a factor of two.

Similar results for the recycled PuC-U-Mo are shown in Table III. In this case there are about 0.4 atom of Pu^{240} and 0.06 atom of Pu^{241} for each Pu^{239} atom. Nonproductive losses and alpha are slightly higher than for the clean fuel, indicating that the spectrum is softer. This is probably the result of the increased amount of carbon. If the breeding ratio is based only on Pu^{239} , it is approximately the same as it was with the clean fuel. Both the U^{238} captures and the Pu^{239} fissions and captures are reduced in about the same proportion by the fission and capture in Pu^{240} . If the Pu^{241} that is formed is credited to the breeding ratio, then there is an improvement of about 0.05. In addition, the fast fission effect has increased from 20 to 24 per cent as a result of the Pu^{240} that was introduced.

TABLE III

Condition: Equilibrium 4	0/49 = 0.42;	41/49 = 0.063; 28/49 = 10.9
Critical Mass: 952 kg Pu	1^{239} and Pu ²⁴¹	
Neutron Economy Fission Absorptio Nonfission Captur Capture in Fertile	n e Material	34.9% 5.5 48.9 (50.8 including Pu ²⁴⁰ capture)
Other Breeding Ratio		10.7 (8.8 excluding Pu ^{***} capture)
Reactor - Core Blanket Total System	1.03 2.0.50 1.53	1.08 (including Pu ²⁴⁰ capture) 0.50 1.58 (including Pu ²⁴⁰ capture) 1.53
a ₄₉		0.208
Fissions in U^{238} and Pu^{240}		24% of total fissions

Recycled PuC-U-15 w/o Mo System

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A detailed breakdown of neutron absorption is shown in Table IV. The most significant thing to note is the substantial contribution of Pu^{240} to the fast fission effect. The fission product neutron absorptions amount to only $\frac{1}{2}$ per cent of the total. Even if these absorptions were underestimated by a factor of 2 or 3, the breeding ratio would not change drastically. Molybdenum accounts for the largest single component of unproductive absorptions. From what is known about this material from danger coefficient measurements in the ZPR-3 critical experiments, it is likely that captures in this material are overestimated. The leakage of neutrons from the blanket amount to 2 per cent of the total. Breeding could be improved by about 4 per cent by increasing blanket thickness or the uranium concentration in the blanket.

TABLE IV

Absorption Breakdown

		Core	Blanket	Total
Fissions	Pu ²³⁹ Pu ²⁴¹ Pu ²⁴⁰ U ²³⁸ U ²³⁵	0.2406 0.0152 0.0181 0.0564	0.0066 - 0.0083 0.0041	0.2472 0.0152 0.0181 0.0647 0.0041
Captures	Pu ²³⁹ Pu ²⁴¹ Pu ²⁴⁰ U ²³⁸ U ²³⁵ F.P. Mo Zr SS Na	0.0502 0.0032 0.0192 0.3287 	0.0017 - 0.1598 0.0010 0.0001 0.0028 - 0.0061 0.0005	0.0519 0.0032 0.4885 0.0010 0.0045 0.0404 0.0072 0.0144 0.0016
	SUM	0.7902	0.1910	0.9812
			Leakage	0.0178
			Total	1.0000

Fuel: Equilibrium PuC-U-15 w/o Mo

UC-Th Systems

As previously stated, it has been assumed that a UC_{-} Th system would be capable of achieving the same burnup, and from a thermal design standpoint the same core composition would be feasible as with the PuC-U-Mo system. Because of the higher ductility of thorium metal it is doubtful if the burnup characteristics would be as good. Perhaps with the addition of some alloying material it could be as good.

In general, the breeding ratios of the U^{233} -Th systems are not as large as those of the Pu- U^{238} system. This difference is due to the smaller effective ν of U^{233} , and to the smaller effective fission cross section and the lower density of thorium as compared to U^{238} . The difference in the fast fission effect is quite significant. Only 4 per cent of the fissions occur in the fertile material, as compared to 20 per cent in the Pu- U^{238} system. This difference is compensated somewhat by the smaller α of U^{233} , which is only one-half that of Pu²³⁹.

In Table V some of the features pertinent to breeding in a UC-Th system are shown.

TABLE V

U²³³ C-Th System Condition: Clean Critical Mass: 1010 kg U²³³

39.6%
3.3
50.2
6.9
0.76
0.45
1,21
1.17
0.088
4% of total fissions

A slightly higher percentage of neutrons producing fission are required. The nonfission captures in U^{233} are considerably less, only about 3 per cent as compared to approximately 6 per cent in the plutonium systems. Roughly the same percentage of neutrons are absorbed in fertile material. The losses to structure and leakage are somewhat smaller.

The total breeding ratio of the reactor is about 1.21. This does not allow for any losses due to neutron capture in protactinium. This loss is probably negligible in a fast system. If the protactinium capture cross section is not unusually large - say approximately 200-300 mb - then the reduction in the breeding ratio attributable to this effect is less than 1 per cent.

The core breeding ratio is less than one, so that the neutron losses to poison control rods, which have not been considered here, would be larger than for the plutonium system.

The system breeding ratio, taking into account external losses, would be about 0.04 less than the total, on the basis of the same losses as were previously used. Here, since the assumed burnup may be overly optimistic, a realistic system breeding ratio is probably somewhat less than 1.17.

The breeding characteristics of the equilibrium UC-Th fuel are shown in Table VI.

TABLE VI

Equilibrium U²³³ C-Th System

Condition: Equilibrium $24/23 = 0.27$;	25/23 = 0.04
Critical Mass: 1010 kg $U^{233} + U^{235}$	
Neutron Economy	
Fission Absorptions	39.4%
Nonfission Absorptions	3.4
Capture in Fertile Material	50.3
Other	6.9
Breeding Ratio	
Reactor - Core	0.82
Blanket	0.44
Total	1.26
System	1.21
a ₂₃	0.088
Fissions in Th^{232} and U^{234}	7.3% of total fissions

Here the uranium composition is 0.27 U^{234} atom and 0.04 U^{235} atom per U²³³ atom. The captures in fertile material are slightly higher, principally because U²³⁴ captures have been included. As with the plutonium system, the equilibrium fuel has a slightly higher breeding ratio than the clean. Here the reactor breeding ratio has increased from 1.21 to 1.26. Again considering external losses, this ratio is reduced to 1.21 for the system.

An absorption breakdown for the equilibrium UC-Th reactor is given in Table VII. The noticeable differences between the neutron absorptions for this fuel and the PuC-U-Mo are the thorium fission absorptions, which are about one-fourth of those in U^{238} . The fission absorptions in the U^{233} are

also much higher because of the smaller fast fission effect and the smaller effective value of ν of U²³³. There are no large parasitic neutron losses to alloy materials similar to the molybdenum captures in the plutonium system. However, the leakage of neutrons from the blanket is considerably larger than it is with the U²³⁸ blanket because of the relatively low density of the thorium metal. This leakage could be reduced considerably by increasing the volume percentage of thorium metal in the outer part of the blanket, where power density is low and cooling is not a problem. In addition, alpha is not changed appreciably, but the fast fissions have approximately doubled, although they are still considerably less than the 24 per cent of the equilibrium plutonium system.

TABLE VII

		Core	Blanket	Total
Fissions	U ²³³ U ²³⁴ U ²³⁵ Th	0.3428 0.0143 0.0110 0.0130	0.0113 - - 0.0019	0.3541 0.0143 0.0110 0.0149
Captures	U^{233} U^{234} U^{235} Th Zr SS Na F.P. SUM	0.0302 0.0136 0.0025 0.3148 0.0060 0.0077 0.0010 0.0039 0.7608	0.0011 - - 0.1750 - 0.0082 0.0009 0.0001 0.1985	0.0313 0.0136 0.0025 0.4898 0.0060 0.0159 0.0019 0.0040 0.9593
	<u>en alfen en en en e</u> gymeine de antaŭ		Leakage Total	$\frac{0.0407}{1.0000}$

Equilibrium UC-Th Absorption Breakdown

The core neutron spectra for the two fuel systems discussed here and also for the Enrico Fermi reactor are compared in Fig. 1. The Pu-U cermet system has a slightly more degraded spectrum than that of the Fermi reactor. The spectrum of the U^{233} -Th cermet peaks more and falls in between the other two on either extreme.

There is one aspect affecting breeding that has not been considered. This is the loss of neutrons to poison materials used in the control of the reactor. In advanced designs, infrequent shutdown for unloading is essential and burnup reactivity compensation may be substantial. This will not be a serious loss with the plutonium system because of the high internal breeding



Fig. 1. Comparison of Flux Spectra

ratio. The excess production in the core and in the blanket should very nearly compensate for fission product accumulation and other associated effects, such as radiation-induced swelling of the fuel. With the U^{233} -Th cermet, the ultimate internal breeding ratio is less than 1 and the long half-life of the protactinium makes this effective in-pile breeding ratio even less. Two effective, in-pile core breeding ratios are shown for a fuel subassembly group as a function of the length of time that this group has been in the reactor in the reactor in Figures 2 and 3. The purpose of these effective breeding ratios is

to account for the time delay between the formation of Np^{239} or Pa^{233} and the fissionable isotopes Pu^{239} or U^{233} . The first of these effective core-breeding ratios, termed the "instantaneous in-pile core breeding ratio," gives the ratio of rate of production to rate of destruction of fissionable material as a function of the length of time that the subassembly has been in the core at that particular time. The results are based on maximum core breeding ratios of 1.0 and 0.75 for the Pu-U and U^{233} -Th systems, respectively. As is to be expected, the breeding ratio of the Pu-U system rises to its maximum value within a few days, while approximately 100 days is required for the breeding ratio of the U^{233} -Th system to reach 90 per cent of its maximum value, and about 200 days to saturate.



Fig. 2. Long Term In-Pile Core Breeding Ratio



Fig. 3. Comparison of Flux Spectra

The second effective breeding ratio is defined as the "long-term in-pile core breeding ratio." This core breeding ratio, which is just the time integral of the first effective breeding ratio, gives the effective breeding ratio of a group of fuel subassemblies during the entire period that the subassemblies have been loaded as a function of in-pile time.

One possible method of operating this reactor is to unload approximately every three months and replace a fraction of the core. The life of these cermet cores is about two years for the burnups assumed and therefore approximately one-eighth of a core must be replaced at each unloading. For this set of conditions the effective core breeding ratio is 0.71, or about 0.04 less than the maximum value, because the rate of protactinium of the last group of subassemblies loaded has not reached saturation. The amount of poison control required to compensate for the net decrease in fissionable material only will reduce the breeding ratio by approximately 0.02.

The results given here indicate that Pu-U or U^{233} -Th cermet fast breeders are feasible and attractive from the standpoint of efficient utilization of raw materials. The breeding potential of the Pu-U system is much more secure than that of the fast U^{233} -Th system. The excess production of the U^{233} -Th system, being considerably smaller than that of the Pu-U system, can be much more severely affected by such things as not being able to achieve high burnup. But it still appears that the use of this material in fast reactors offers good prospects for the utilization of the thorium reserves.

Discussion of Paper Presented by Mr. Nims

MR. GREEBLER:

One question I had was on the U^{235} in the blanket material. I assume that this was depleted uranium?

MR. NIMS:

Yes, depleted to 0.36 per cent.

MR. GREEBLER:

How do you count the destruction of depleted U^{235} in the breeding ratio?

MR. NIMS:

It was not counted. If it were included, the breeding ratio would be reduced by about 1.5 per cent.

MR. GREEBLER:

What were the power densities in these cores and the specific powers in kilowatts per kilogram of total plutonium?

MR. NIMS:

The power densities are approximately 500 kilowatts per liter. The specific power for the plutonium systems are 800 and 500 kilowatts per kilogram for the clean and recycled fuel, respectively.

MR. CAMPISE:

I have one question as far as characterizing your uranium-thorium systems. Could you give me either the median fission energy or median flux or absorption energy to indicate what kind of spectrum you had?

MR. NIMS:

As may be seen from Fig. 1, which compares the spectra of the plutonium system, the U^{233} -Th system and the Fermi reactor, there is not a great deal of difference. The U^{233} -Th spectrum is peaked more in the middle and falls in between the other two on either extreme. The median fission energy is about 200 kev.

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MR. WIGNER:

May I ask the same question that I asked before ? What is the burnup you are calculating ?

MR. NIMS:

The maximum burnup of the Pu-U fuel is about 3 atom per cent. The burnup of the fissionable material is about 50 per cent.

MR. WIGNER:

About 50 per cent?

MR. NIMS:

That's right; that is why these external losses are so low.

MR. WIGNER:

I see. Thank you. How about the fuel element? Can it stand it?

MR. NIMS:

Well, that is a question to which we would also like the answer. We have some data for the Fermi reactor fuel, which is a U-10 w/o Mo alloy. We have also irradiated a few specimens of U-15 w/o Mo and the burnups that we have assumed here are intermediate between the two. From these data we have deduced an optimistic and a pessimistic burnup curve. We have taken something in the middle as a design basis. That's the situation with the Pu-U fuel. It is reasonably well substantiated. A lot more work is necessary to justify completely the burnups used, of course. In the case of the U²³³-Th system there are very little data that I know of and the indications are that the thorium metal, which is quite ductile as a cermet, would probably not stand the damage that the U-Mo would. If you can find some alloying material to increase its strength, you might improve it to a reasonable value. I think the only material for alloying thorium for which there is any experience is uranium. This would certainly be a reasonable type of fuel to use, but we just haven't looked at what happens when you have a U^{233} -Th system that also contains U^{238} and therefore plutonium isotopes.

MR. WIGNER:

You believe that indications are that it can stand the assumed burnup?

MR. NIMS:

It could work, yes, if its strength could be sufficiently increased by alloying the matrix material.

MR. WIGNER:

May I ask you another question? We heard yesterday from Dr. Greebler that the production ratio was reduced by the addition of two oxygen atoms - in other words, by the use of oxide to 1.25. Now one carbon atom is, of course, less than two oxygen atoms, but not terribly much less and the cross section is higher. Can you comment on this?

MR. NIMS:

Well, we have a much harder spectrum. We do not have all the fissionable and fertile material in ceramic form as you do with the oxide. Only the initial plutonium loading is in the form of a ceramic, so that the amount of carbon contained here is much smaller than it would be with a pure ceramic type of element. In addition we have much larger amounts of U^{238} present, which tends to reduce the flux at the low end of the spectrum as a result of strong neutron absorption.

MR. WIGNER:

Could you tell us what the composition of your core is? I don't think we saw that.

MR. NIMS:

No, I didn't show it in detail. I assume what you want is the relative volume of the ceramic and the matrix material. The plutonium carbide is about 12 volume per cent of the fuel, the U-Mo alloy making up the remainder. For the U^{233} -Th system, the UC accounts for 15 per cent of the fuel volume.

MR. WIGNER:

Could you give the doubling times which you anticipate?

MR. NIMS:

The doubling times for the plutonium systems are about 15 years. For the U^{233} -Th system it is more like 25 years.

VOICE FROM FLOOR:

How does molybdenum compare with steel in inelastic scattering and its effect on the spectrum?

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MR. NIMS:

Of course, molybdenum is a much stronger absorber. Its inelastic scattering cross section as compared to steel is somewhat larger.

MR. KASTEN:

In choosing these reactors, what were your criteria? Did you use some basis for choosing this particular composition?

MR. NIMS:

Recently at APDA we went through a Fast Reactor Parameter study under an AEC contract in which we examined different types of fast reactor fuels. One of these was the plutonium-carbide, U-moly cermet, and the size I picked here was what was indicated by this study as being a practical one. It lies in the most economical range as well as being practical, insofar as thermal performance is concerned. In other words, the design has been given some thought. The thermal analysis has been gone through. Burnup effects have been considered. It is a practical design; preliminary, but some attention has been given to the practical considerations of the design.

THE FAST MULTIPLICATION EFFECT DUE TO THE (n, 2n) REACTION IN BERYLLIUM AND BERYLLIUM OXIDE *

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The fast multiplication effect due to the (n, 2n) reaction in beryllium or beryllium oxide can be of considerable interest in thermal breeders, since in these cases every gain of reactivity may cause a remarkable improvement of the breeding gain.

In the past, therefore, several papers have been published in which investigations of the beryllium fast-neutron multiplication effect were reported, but unfortunately the cross-section information that was available for the investigations was not sufficient and there were large discrepancies among the results. These discrepancies have prompted this investigation, in which an attempt is made to establish upper and lower limits on the fast multiplication effect.

C. B. Mills and N. M. Smith, Jr.,⁽¹⁾ using only integral measurements of the beryllium cross sections made with Ra-Be and Po-Be sources, estimated that the order of magnitude of the additional fast multiplication effect is 10%. Some years later D. Dillenburg, G. Jacob, and P. S. $deToledo^{(2)}$ considered this problem. Although they used the best cross section information available at that time, the (n,α) values were wrong by a factor of about two and the (n,2n) values that they used are not in very good agreement with present information. Their final result is a reactivity increase of about 6.8 per cent. At the Second Geneva Conference this effect was discussed by A. \tilde{V} . Krasin et al.,⁽³⁾ who considered a critical assembly of enriched uranium (10 per cent) and beryllium in which it was required that there be a neutron balance. But this way of determining the (n,2n) effect is indirect, because all other contributing reactivity effects (e.g., η , e, p, f) have to be known exactly. They also made experiments studying the neutron density in a prism of light water in comparison with a prism of beryllium for both thermal and epithermal neutrons, using the same neutron sources for both water and beryllium. But these second measurements are rather unsure; the reported multiplication effect is 12 ± 9 per cent for thermal and 16 + 9 per cent for epithermal neutrons. The critical experiment gives 10 ± 5.5 per cent and 12 ± 5 per cent for a somewhat other configuration.

^{*}W. Häfele, The Fast-Neutron Multiplication Effect of Beryllium in Reactors, ORNL-2779 (1959); see also W. Häfele and M. Tsagaris, The Fast Multiplication Effect of Beryllium Oxide in Reactors, ORNL-2849 (to be published).

^{**}On leave from Kernreaktor, Karlsruhe, Germany.

P. Benoist <u>et al.</u>,⁽⁴⁾ were concerned with critical and subcritical experiments on U-BeO lattices, and they carefully investigated the fast multiplication with beryllium and beryllium oxide. They used the latest cross section data available at that time. For example, they used the correct (n, α) cross section and also calculated the spectrum rather carefully, but taking into account only one angular distribution of elastically scattered neutrons in the most important region of 2.9 Mev.

In addition consideration should be given to the available information on the relevant cross sections. In May 1957 H. Goldstein⁽⁵⁾ considered very carefully the available information on the (n,2n) and (n,α) cross section and we refer especially to this report. One year later he used this information for calculating the penetration of neutrons from a point fission source through beryllium and beryllium oxide. In December 1958 J. B. Marion et al.,⁽⁶⁾ presented measurements of the total and the elastic cross sections; they also gave nonelastic cross sections obtained by subtraction of the elastic from the total cross section. From comparison of these total cross sections with the data given by J. L. Fowler and H. O. Cohn,⁽⁷⁾ it may be concluded that Marion's data are a little bit too low, because Fowler and Cohn agree with the previous measurements of C. K. Bockelman <u>et al.</u> (8) Using these too low total cross sections, Marion et al. get correspondingly low nonelastic cross sections. To compare the several cross-section data we made the assumption that the (n,α) cross section is precisely known, using Stelson and Campbell's (9) data. The subtraction can be done because the (n, α) data lie on a gentle and smooth curve going from threshold to 4.4 Mev. Above 4.4 Mev the (n, α) data are not so well known, but we follow H. Goldstein, who extrapolated this curve quite safely to the 14.0-Mev point. So we subtract this (n, α) cross section in the case where only the difference between total and elastic cross section is given. If one then considers Fischer's⁽¹⁰⁾ direct measurements of the (n, 2n) cross section, one sees that Fischer's (n, 2n) cross section above 2.9 Mev is much higher than the data of Marion. (Below 2.9 Mev Fischer's data are lower.) However, nearly all other presented values are higher except the one point given by J. M. Fowler, et al. (11) On the other hand, the values of Fischer are relatively unsure and Fischer himself offers them as upper limits. To judge between these two extreme sets of data it is useful to consider the values at 4.07 and 7.0 Mev given by Beyster, et al. (12,13) They use the sphere method and have relatively small errors, so that these data are supposed to be comparatively exact. This has the consequence that a plot between Fischer and Marion is presumed to be correct. We also refer to the values of Ball, et al., (14) for the region from 7 to 14 Mev. These authors also used the sphere method. Finally a value at 14 Mev is given by N. N. Fl'orov and V. M. Talyzin.⁽¹⁵⁾ This value is a little bit higher than the value of Ball.⁽¹⁴⁾ The data considered are plotted in Fig. 1.

Since it is our aim here to determine an upper and lower limit for the fast multiplication effect, it is necessary to obtain upper and lower limits for the (n, 2n) curve, with special attention given to the region between



Fig. 1. Beryllium (n,2n) Cross Section as a Function of Energy

2 and 4 Mev. To obtain the lower plot (see Fig. 2) we go close to Fischer's data between 2.5 and 2.7 Mev, while between 3 and 6 Mev we suppose the curve to be a little bit larger than the extremely low values of Marion; above 6 Mev we finally approach nearly the NDA curve of Fig. 1. To obtain an upper limit we first approach the NDA curve and then the first Marion value (2.6 Mev). Then we come close to the Fischer value of 3.2 Mev, and above 7 Mev we take the Ball values and reach the 14 Mev point at the Fl'orov and Talyzin value. The biggest difference between these two curves, about 190 mb, is at 3.0 Mev. Of course, the chosen curves are still a bit arbitrary. The curve as given by NDA lies between our upper and lower limits.

In calculating the neutron spectrum in an infinite block of beryllium or of beryllium oxide the nonisotropic scattering law must be considered. To this end we have used the angular distributions as presented by Marion et al.,⁽⁶⁾ and by J. L. Fowler and H. O. Cohn.⁽⁷⁾ Together with the previously known angular distribution one has quite good information over the whole energy range of interest. In the case of beryllium oxide, the oxide cross sections are needed. Fortunately the oxygen cross sections are



Fig. 2 Upper and Lower Estimates of the (n,2n) Cross Section and the (n,α) Cross Section of Beryllium

much more sure. The data of H. Lustig, H. Goldstein, and M. H. Kalos⁽¹⁶⁾ were employed. The angular distributions were taken from the paper of J. L. Fowler and H. O. Cohn ⁽¹⁷⁾ on the one hand and the report of N. A. Bostrom <u>et al.</u>,⁽¹⁸⁾ on the other hand. From here on the calculation is straightforward.

Parallel and independent of these investigations, J. Chernick and S. Oleska Moore⁽¹⁹⁾ recently have treated the same question for beryllium. They also employed two guesses of the (n,2n) cross-section behavior, but they employed on the one hand essentially the Fischer data and on the other the Marion values. In calculating the spectrum they assumed, however, isotropic scattering. As pointed out later they get essentially the same result.

The fast multiplication effect is described by the following expression:

$$\epsilon = 1 + \int_{E_0}^{\infty} S \frac{\sigma_{2n} - \sigma_{\alpha}}{\sigma_T} dE$$

If we define

$$P_{2n} = \int_{E_0}^{\infty} S \frac{\sigma_{2n}}{\sigma_T} dE$$
$$P_{\alpha} = \int_{E_0}^{\infty} S \frac{\sigma_{\alpha}}{\sigma_T} dE$$

then

$$\epsilon = 1 + P_{2n} - P_{\alpha}$$

Here σ_{2n} is the (n, 2n) cross section, σ_{α} is the (n, α) cross section, and σ_{T} is the total cross section. For beryllium oxide σ_{α} and σ_{T} have two components: one due to beryllium and the other due to oxygen. The quantity S is the spectrum of neutrons in the infinite block. In the stationary state all types of neutrons are present: those which have undergone zero, one, two, three, etc., elastic collisions. Therefore S is an infinite sum:

$$s = \sum_{m=0}^{\infty} s_m$$

where $S_{\mathbf{m}}$ is the spectrum after the $\mathbf{m'}^{th}$ elastic collision. The energy E_0 is a threshold for which

$$\sigma_{T} = \sigma_{n}; E < E_{0}$$

 $(\sigma_n \text{ is the elastic cross section}).$

We also have assumed that the neutrons which are produced by the (n, 2n) reaction have energies less than E_0 , so they do not contribute to further multiplication.

By far the greatest part of the work is the calculation of the spectrum S. We start with fission neutrons and follow the elastic nonisotropic scattering processes step by step. In the metal case one has to take into account 12 collisions before the partial sum

$$S^{(n)} = \sum_{m=0}^{n} S_{m}$$

converges reasonably. In the oxide case the number of collisions to take into account is 15. The numerical procedure was as careful as possible, all integrations being carried out by Simpson's Rule, with a very small step width.

Here are the results: For the metal case: $P_{n,2n}^+ = 11.81\%$; $P_{\alpha} = 4.22\%$; $P_{n,2n}^- = 9.37\%$,

which means that

 $\epsilon^{+} = 1.076$

 and

€[−]= 1.051 .

For the oxide case

$$P_{n,2n}^{+} = 7.94\% ;$$

$$(P_{\alpha})_{Be} = 2.91\% ;$$

$$P_{n,2n}^{-} = 6.03\% ;$$

$$(P_{\alpha})_{0} = 0.44\% ;$$

which means that

$$\epsilon^+ = 1.046$$

and

$$\epsilon^{-} = 1.027$$

In accordance with the ideas of Horowitz, (20) attention should be called to the poisoning effect due to the daughter products of the (n,α) reaction. We have

;

$$n + Be_4^9 \longrightarrow He_2^6 + He_2^4$$
$$He_2^6 \longrightarrow Li_3^6 + \beta^{-}$$

and

$$n + Li_3^6 \rightarrow He_2^4 + H_1^3$$

In the saturation case, the Li⁶₃ poisoning is nearly exactly equal to the value of P_{α} . So it is of interest to consider also the Li⁶₃ poisoned effects.

For the metal case,

$$\epsilon^+$$
 - P_{α} = 1.034

and

 $\epsilon^{-} - P_{\alpha} = 1.009$

For the oxide case,

$$\epsilon^+$$
 - $(P_\alpha)_{Be}$ = 1.017

and

 $\epsilon^{-} - (P_{\alpha})_{Be} = 0.998$

In principle there is also a nonsaturating He_2^3 poisoning, but the time for a reasonable buildup is several years, and it is fair to assume that over these years the He_2^3 might escape from the beryllium because it is a volatile gas.

It is interesting to compare the unpoisoned results with previous results:

Metal case:

	Hafele	Chernick	<u>Benoist</u>
∈-1	7.6%	6.1%	5.8%
	5.1%	6.6%	

Oxide case:

 ϵ - 1 4.6% 6.0% 2.7%

In the metal case the nonisotropic scattering is fully taken into account in this paper; in the paper by Benoist et al., (4) the 2.9-Mev distribution is taken for all energies; in the Chernick paper only isotropic scattering is taken into account. Apparently the result in the metal case is quite insensitive to the type of nonisotropic scattering; in the oxide case, however, the difference is quite remarkable. In this paper the nonisotropic scattering, especially of the oxygen component, is carefully taken into account, whereas Benoist <u>et al</u>., again take the 2.9-Mev distribution for all energies, which apparently causes a remarkable deviation from the correct value.

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Discussion of Paper Presented by Mr. Häfele

MR. HALL:

I think we might take time for one or two questions.

MR. REYNOLDS:

I had a remark about the cross sections which maybe Dr. Cranberg might want to comment on as well as Dr. Hafele. It seemed to me that your maximum cross-section curve is somewhat pessimistic. You could have drawn a curve which is more optimistic, especially between threshold and 3 Mev. Another thing is this: if one subtracts Marions's elastic scattering data from Fowler's total scattering instead of Marion's total cross sections (and Fowler and Cohn's total scattering agrees with the old BNL numbers), then the Marion values are consistently higher and your guesses must be more optimistic. The fast multiplication effect is influenced very greatly by the cross-section curve in the range from threshold to 3 Mev, and you can get or calculate reactivity gains of 12 to 15%. (Dr. Cranberg was not present at the time of this remark.)

MR. HAFELE:

Perhaps I may answer in this way. It is true that the information on cross sections up to now is not really sure, and one should emphasize the point that the greatest difficulty in establishing the fast multiplication effects is the calculation of the spectrum; if the spectrum is available, it is a matter of two hours to produce newer numbers with a new crosssection guess. But it takes 8 or 10 weeks to calculate the spectrum. As long as the basic experimental cross-section information is not more sure than it is now, you may always have different guesses.

MR. WOLFE:

I would like to emphasize somewhat the last remark. We have been doing the same type of calculations using the multigroup technique, which is a little easier to calculate, but possibly not quite as exact, and we get numbers very close to yours. However, we have been looking at the effect of varying the assumed (n, 2n) cross sections and we find, drawing curves through reasonable points and interpreting the data in a reasonable way, that multiplication values vary from 7 per cent up to as high as 15 per cent; we get (n, 2n) to (n, α) ratios, which tells how long one is going to keep this effect, that go from $2\frac{1}{2}$ to 4. In looking over the data we have been impressed by the fact that below the 4-Mev point from Beyster, there is one point by Fowler. I have not been able to find that paper and can't talk about it, but the other points, the ones by Fischer are unsure, the ones by Marion and Levin and Cranberg, are in serious doubt. If you do interpret them according to Fowler's total cross section, you end up with a 13 or 14 per cent effect, as a matter of fact. So really you should not consider these points at all if you are trying to get an optimistic value of the (n, 2n) reaction. At least you shouldn't interpret them directly as Marion, Cranberg, and Levin do, with their total cross sections.

MR. HÄFELE:

Right. You have to distinguish between nonelastic and (n, 2n) cross sections and that causes, of course, some difficulties.

MR. WOLFE:

Well, the point I wanted to make was that it seems to me we have a serious lack of experimental data, and it is a very important effect and one just does not know, right now, how big the effect is or how long it will last.

MR. HAFELE:

Well, perhaps we can see the first slide again, where the experimental data are plotted. So, I am sure, if you believe in these two points you can produce 12 per cent and more and I cannot exclude that. On the other hand all these values, especially the Beyster values, should be used as a guidance to these informations. The large question is the behavior of the data in this region, because there is maximum of the spectrum.

In reading Fischer's paper and observing these large possible errors which he has, I think, I came to the conclusion that I do not worry too much about these two points, but in principle, of course, it cannot be excluded.

(QUESTION FROM THE FLOOR WITHOUT MICROPHONE)

ANSWER:

Well, remember, that is the NDA guess; my own upper guess is somewhat more optimistic than that curve. I think these questions can be settled finally only if you have more sure experimental data.

MR. FISCHER:

One of the things which I think my thesis tended to point out was that the reaction apparently goes through the 2.43-Mev level in beryllium-9, so that it indicated that the threshold of this reaction should be at 2.7-Mev, if you can believe my unreliable data. I wonder if you have made any calculations assuming that this part of my thesis is correct; that is, rather than take a combination of things for an optimistic curve, test the consequence of this prediction of my paper.

MR. HÄFELE:

No. As I pointed out, I emphasized the spectrum calculation and now these spectra are available in these two ORNL reports. They are now available to produce newer values with a given spectrum.

MR. HALL:

I think this paper has raised a number of very interesting questions which point to the need for more experimental data. I think it would be of interest to a number of people here to continue the discussion, and Dr. Häfele has agreed to answer more questions.

MR. BOGART

Isn't it quite important to first establish the mechanism of the (n, 2n) reaction? From Mr. Fischer's thesis, it seems that the reaction would be first an excitation of the first level in beryllium-9, and that would give a clear-cut threshold of 2.7-Mev, and then that would give you the first N', appearing just as a result of a direct two-body interaction; the excited beryllium-9 has been shown to go by an N" and two α 's, and this is the reaction that goes 90 per cent of the time. The two α 's are really due to the beryllium-8, so again you have another two-body break-up by which you can describe the emergent N', N" quite straightforwardly. Is this not the mechanism for the break-up?

MR. HÄFELE:

Well, I agree with you that if one really has a model for this nuclear reaction, then the threshold would be much more serious. All I can do is to refer to the conclusion of Marion's paper where he points out that the mechanism is not as easy as described and supposed heretofore; he gives reasons to believe that there is a 3 and 4-body process, not simply inelastic scattering in advance of the emission of the second neutron. I also refer to the survey paper of Goldstein, where he emphasizes that the situation is unfortunately not as easy as you pointed out. According to the literature, the situation is not yet really clear in that respect. The paper by Marion emphasizes that it is not a clear process and therefore the threshold is somewhat doubtful in its importance. So I think that is also a question for further experimental investigations. I think it may be of interest to mention some integral experiments which have been done at Harwell. These were done some time ago; I think they were written up finally by Dr. Robert Richmond. There were two sets of experiments. First, the multiplication effect of a beryllium sphere was measured by a series of techniques, both with a long counter and with a manganese bath technique, the source of neutrons being a small sample of uranium-235 irradiated by a beam of fission neutrons, and he was able to measure the multiplication of this sphere to a fair degree of accuracy.

The second experiment he did was to irradiate some samples of beryllium in a fast slug in BEPO, and measure the helium production in this small sample of beryllium. From this one can get this combination of the (n, 2n) and (n, α) reactions. Then, knowing the (n, α) reaction from the Campbell and Stelson results, one can subtract and get the (n, 2n) reaction averaged over a fission spectrum. I have the data on the averaged (n, 2n) cross section here with me, but I haven't got the data on multiplication measured in the beryllium spheres; but I'm quite sure that a letter to Dr. Richmond would extract this data. As far as I remember, the beryllium sphere he used had a diameter of 11.4 cm, and the multiplication that he measured was around 4 or 5 per cent. This is the net effect of the (n, 2n) and (n, α) reactions. I'm not sure how this ties in with the calculations of the infinite medium that we're talking about here. For the averaged cross section over a fission spectrum of the (n, 2n) cross section, the numbers that he quotes are of the order of 125 mb. This is the average cross section of the (n, 2n) cross section in an essentially unmoderated fission neutron spectrum. This work has been done some time ago and I don't know when it's been re-evaluated and compared with the results averaging Fischer's data, but we'd be quite pleased to furnish any further information on this point.

MR. MENEGHETTI:

I was wondering whether you would comment on the type of scattering matrix you use, that is, the inelastic scattering matrix in the (n, 2n) process, in order to determine the degree of degeneration of both the primary inelastically scattered neutron and also that which was emergent from the excited state of the remaining nucleus.

MR. HÄFELE:

Excuse me, I didn't get the question.

MR. MENEGHETTI:

I'm sorry. Basically what I would like to know is in what manner you determine the energy degradation due to the inelastically scattered neutrons in the (n, 2n) process?

MR. HÄFELE:

I see what you mean. Well, there was not very much to determine. I only made the assumption that both of these neutrons produced in the (n, 2n) reaction are slower than 1 Mev, because all the reactions considered are important only above 1 Mev. All that I assumed, and that of course was an assumption, is that both of these neutrons are slower than 1 Mev.

MR. MENEGHETTI:

That means in your calculation both of the two neutrons from the (n, 2n) process are in their scattering degraded to below the (n, 2n) reaction threshold.

MR. HÄFELE:

Right, also below the possible (n, α) reaction.

MR. MENEGHETTI:

In this regard I would like to comment on a rather crude paper which appeared in the July issue of Nuclear Science and Engineering by Meneghetti and Hummel, in which the detailed scattering matrix, that is, for the elastic scattering matrix, was not utilized. It was a rather simpleminded multigroup calculation. However, we did worry about the fact that it is altogether possible that especially the first inelastically scattered neutron can indeed be re-scattered by an (n, 2n) process. It indicates that of the neutrons that have undergone an (n, 2n) reaction, possibly 22 per cent of them, could emerge with energies higher than 1.35 Mev. Now to be sure 1.35 Mev is below the threshold of the (n, 2n) reaction, yet there is no doubt that there are neutrons which undergo another (n, 2n) process nevertheless.

MR. HÄFELE:

Well, if they are at about 1.35 Mev it would lower the net effect, because then they only contribute to possible (n, α) reactions.

MR. WILKINS:

I just wanted to say that the earlier work of Dr. Goldstein which you referred to is now under review at NDA. I don't have anything to report, because this has just essentially gotten started. But I'm sure that before long Goldstein will have some things to say about our moments-method calculations.
MR. HÄFELE:

Yes. Perhaps I may mention that these are the two papers I talked about. The first is metal case, and the second is the oxide case.

MR. BELL:

I would just like to comment on some more experiments which have been done at Los Alamos on this subject, namely, in the fast critical assemblies Topsy, Godiva, and Popsy. There have been replacement measurements made with a large number of elements, including beryllium. From a fairly careful study of essentially the danger coefficient of the central sample, and by comparing the beryllium and carbon values to try to eliminate the effect of the elastic scattering, Gordon Hansen has arrived at an average value of the (n, 2n) minus the (n, α) cross section averaged over the fission spectrum. I don't have the results with me, but it's of the order of 100 mb. This is another bit of information which is available.

BREEDING IN COUPLED FAST-THERMAL SYSTEMS

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A breeding study of a coupled fast thermal system can be made by way of a comparison with a prototype all-fast system. The reason for this is that the motivation for a fast thermal system is primarily the same as for an all-fast system i.e., to attain a high breeding ratio. The reason one considers the coupled system rather than the all-fast system is in order to obtain more desirable kinetic or safety characteristics. These characteristics are obtained at an admitted loss in breeding ratio, so that the extent to which one does lose breeding ratio is of extreme importance. A primary design consideration in the coupled systems would always be to minimize this loss.

We consider here two different types of coupled systems. One is cooled with sodium and consists basically of a thermal system surrounding the fast core. The calculations refer to a heterogeneous metal fast core, but this is in no way of essential importance; it could be similar to any of the types that have been previously discussed here. The other type of coupled system considered is cooled with water and steam. Geometrically it will be quite similar to the sodium-cooled system. Now, however, the



Fig. 1. Schematic Drawing of Sodium-cooled Coupled Reactor fast core will be acting as a superheater and will be steam cooled, while the thermal part will be cooled by water of approximately normal density.

The two systems will be quite similar in some ways. Approximately 5 per cent reactivity will be tied up in the thermal system in each case. This means that the fast system is approximately 5 per cent subcritical without the contribution of thermal fissions. This will result in the kinetic characteristics being similar to those of thermal systems. Approximately 20 per cent of the power will arise from thermal fissions. In both cases there will be a loss of approximately 10 per cent in the breeding ratio compared to an all fast system.

We consider the sodium-cooled system first. A schematic drawing is given in Fig. 1. The system consists of a fast core surrounded by an inner blanket region, which consists primarily of natural uranium and is sodium cooled. This natural uranium region is the location of the thermal fissions. The thermal neutrons arise from neutrons that are thermalized in the beryllium moderator region. The natural uranium also serves as a filter, preventing low-energy neutons from entering the fast core. This, of course, is necessary in order not to degrade excessively the spectrum in the fast core, thus retaining the breeding characteristics of the fast system. The outer blanket captures neutrons that have diffused outwards. Table I gives the volume composition of the various regions. The volume of the fast core is 400 liters.

TABLE I

Region	Volume Fractions							
	Pu ²³⁹	U ²³⁸	U ²³⁵	Zr	Be	Na ^a		
Core	0.0354	0.4637	0.0009	0.1000	0	0.4000		
Inner Blanket	0	0.6453	0.0047	0.1000	0	0.2500		
Moderator	0	0	0	0.1000	0.8800	0.0200		
High Density								
Blanket	0	0.7984	0.0016	0.1000	0	0.1000		
Core Blanket	0	0.4990	0.0010	0.1000	0	0.4000		

Composition of Various Regions of Sodium Cooled Coupled Reactor

^a480°C

Zirconium is used as the clad material. Its selection is not of great importance in the fast core; however, a structural material of low capture cross section is needed in the thermal region. The thickness of the inner blanket is of the order of 10 cm. This is a rather sensitive dimension, since it largely determines the coupling between the two systems. The moderator region has a thickness of the order of 30 cm.

Table II gives a neutron inventory of the clean system. The resulting breeding ratio is 1.59. The calculations were done on a four-group model, but they have been checked against calculations using a larger number of groups, which yielded essentially the same results. A small number of groups were used to allow two-dimensional calculations which would determine more accurately the leakages from the various regions. The cross sections were used with some confidence, since they lead to good agreement with experiment on a similar system mocked up on the Argonne Fast Critical Facility, ZPR-III.

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TABLE II

	Region						
Event*	Core	Inner Blanket Moderator		Moderator Blanket	Core Blanket		
Pu ²³⁹ Fissions	0.261	-	-	-	-		
Pu ²³⁹ Captures	0,081	-	-	-	-		
U ²³⁵ Fissions	0.006	0.066	-	0.026	0.002		
U ²³⁵ Captures	0.002	0.021	-	0.0 0 6	0.0009		
U ²³⁸ Fissions	0.083	0.022	-	0.008	0.007		
U ²³⁸ Captures	0.299	0.177	-	0.132	0.074		
Na Captures	0.0008	0.002	0.004	0.0 00 6	0.0004		
Zr Captures	0.007	0.002	0.016	0.001	0.001		
Be Captures	-	-	0.006	-	-		

Neutron Inventory for Sodium Cooled Coupled Reactor

Neutrons Leaking = 0.025

*All numbers normalized to one fission neutron in the core.

A neutron inventory was also obtained for a prototype all-fast system obtained by replacing the beryllium by a depleted uranium region. The resulting breeding ratio is 1.71, so one has reduced the breeding ratio from 1.71 to 1.59 in going to the coupled system.

We have also looked at a 50-liter core, since there is a possibility that one might want to put a coupled loading into EBR-II. The thicknesses of the inner blanket and moderator are the same as in the previous case. The neutron inventory is given in Table III. The breeding ratio for this system is 1.61.

All of the previous calculations were done on the clean system. Calculations have also been made on the more realistic situation representing some sort of equilibrium condition. One in general finds that the losses associated with burnup of the system are quite similar to what they would be in the all-fast system. The calculations were made for a burnup of 2 per cent of the total fuel atoms, which corresponds to a burnup of the order of 15 or 20 per cent of the plutonium atoms. During this cycle the total change in breeding ratio was rather small, of the order of a few per cent. One now also has a loss in breeding which is related to the xenon effect in the thermal system. This again is rather small, of the order of one per cent. If one goes to higher burnups these effects will increase. The long-term losses to fission products and higher isotopes will become larger, but again will probably be of the same order of magnitude as in the all-fast system.

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	Region						
Event*	Core	Inner Blanket	Moderator	Moderator Blanket	Core Blanket		
Pu ²³⁹ Fissions	0.289	-	_	-	-		
Pu ²³⁹ Captures	0.084	-	-	-	-		
U ²³⁵ Fissions	0.003	0.093	-	0.050	0.003		
U ²³⁵ Captures	0.0008	0,030	-	0.013	0.001		
U ²³⁸ Fissions	0.061	0.039	-	0.016	0.014		
U ²³⁸ Captures	0.142	0.263	-	0.298	0.096		
Na Captures	0.0004	0.003	0.005	0.001	0.0007		
Zr Captures	0.004	0.003	0.021	0.002	0.002		
Be Captures	-	-	0.003	-	-		

Neutron Inventory for 50 Liter Sodium Cooled Coupled Reactor

Neutrons Leaking = 0.030

*All numbers normalized to one fission neutron in the core.

The motivation for the water and steam-cooled system can be given in either of two ways. One can compare it with other thermal water systems. The primary advantage would then be the very high breeding ratio, which we will see is of the order of 1.5. A further advantage is that the fast core acts as a superheater. Naturally, this is not the only way to superheat. There are many proposed thermal designs under consideration.

Alternatively, one can compare the coupled superheater with the sodium-cooled fast reactor (or the sodium-cooled coupled system). The main difference is that one is dealing with a water system. I will not go into whether this is an advantage or not, but at least many people have had more experience with water. There is one other advantage or possible advantage in that one now has a safety mechanism in the loss of reactivity associated with expulsion of water from the thermal region. It is only fair to add that one has also the potential hazard of water entering the fast core, though from what we have looked at so far it does not seem too difficult an engineering problem to obtain a design that effectively guarantees against this possibility.

A major problem associated with the system relates to the power density that might be attainable in the steam-cooled fast core. It is unlikely that one can obtain the power densities in the fast core that one might be able to obtain in the sodium-cooled system. On the other hand, it may not be too much lower, perhaps by a factor of 2 or 3.

One finds, from general considerations on the thermodynamics of water, that to obtain desirable steam outlet conditions only 20 or 25 per cent of the total energy is put into superheating. However, in order to obtain a high breeding ratio one would like to have most of the power, perhaps 80 per cent, generated in the fast core, a region where only superheating is accomplished. This difficulty can be easily resolved by having auxiliary boilers which use some of the superheated steam to boil water.



REGION I: STEAM COOLED PLUTONIUM FAST CORE REGION 2: STEAM COOLED DEPLETED URANIUM BLANKET REGION 3: STAINLESS STEEL WATER BARRIER REGION 4: WATER COOLED NATURAL URANIUM ANNULUS REGION 5: WATER COOLED DEPLETED URANIUM BLANKET

Fig. 2. Schematic Drawing of Nonboiling Coupled Steam Superheater

Figure 2 is a schematic drawing of the system under consideration. It is quite similar to the sodium-cooled concept. Most of the calculations to date have been with D_2O . The moderating effect of the D_2O steam in the fast core, region l, is of an order, perhaps, somewhat less than that associated with sodium in the normal fast core. Region 2 serves as a blanket above and below the core. In this design there is a stainless steel tank (region 3), one and one half cm thick, separating the thermal and fast systems. On the inside of the tank there is a region of depleted uranium in order to cut down the power peak which would occur if the fast core were adjacent to the tank. Region 4 consists of a natural uranium, heavy water lattice with zirconium the cladding material. Region 5 consists of a depleted uranium, heavy water lattice and serves as a blan-

ket. We have found that there is no necessity to cluster the D_2O lattice since there is no need to have this region very reactive. It is very much subcritical on its own. In some further studies we have tried either to eliminate the separating tank or to use it in conjunction with a tube arrangement where the tubes which hold the assemblies outside the fast region would individually contain the system pressure. In case of rupture, probably only one tube would break with only a small amount of water released.

Table IV gives the composition of the various regions.

TABLE IV

Composition of Various Regions of Coupled Steam Superheater

Destau	Volume Fractions							
Region	Pu ²³⁹	U ²³⁸	U ²³⁵	Fe	Zr	D ₂ O ^a		
Core	0.0531	0.4469	0	0.15	0	0.0114		
Core Blanket	0	0.4990	0.0010	0.15	0	0.0114		
Water Annulus	0	0.0993	0.0007	0	0.0102	0.8898		
Water Blanket	0	0.0998	0.0002	0	0.0102	0.8898		
		1			1	1		

a300°C

Table V gives the neutron inventory. The breeding ratio for the clean system is 1.53.

TABLE V

Neutron Inventory for Coupled Steam Superheater

	Region						
Event*	Core	Core Blanket	Stainless Steel Tank	Water Annulus	Annulus Blanket		
Pu ²³⁹ Fissions	0.277	-	-	-	-		
Pu ²³⁹ Captures	0.073	-	-	-	-		
U ²³⁵ Fissions	-	0.005	-	0.066	0.036		
U ²³⁵ Captures	-	0.002	-	0.019	0.009		
U ²³⁸ Fissions	0.074	0.013	-	0.002	0.001		
U ²³⁸ Captures	0.238	0.132	-	0.126	0.169		
D ₂ O Captures	0	0	-	0.0001	0.0003		
Zr Captures	-	-	-	0.0004	0.0007		
Fe Captures	0.005	0.006	0.031	-	-		
	1	•			1		

Neutrons Leaking = 0.044

*All numbers normalized to one fission neutron in the core.

All of these calculations have been on D_2O systems. D_2O was selected on the reasoning that it would be better than H_2O , because it would have less moderating effect in the fast core and also less parasitic capture in the thermal system. Some preliminary calculations with H_2O indicate that the difference in breeding ratio between D_2O and H_2O systems may be quite small. This comes about because even with H_2O there is not too much moderation in the fast core and the outer region is so heavily loaded that the parasitic capture due to H_2O is small.

Much of what I have reported here is based on the work of G. J. Fischer and B. J. Toppel, both at Argonne.

Discussion of Paper Presented by Mr. Avery

MR. HALL:

This paper is open for questions.

MR. NESTOR:

I was interested in learning what reactor code you used for your calculations.

MR. AVERY:

For the two-dimensional calculations we used the PDQ code. For the one-dimensional calculations, which usually had a much larger number of groups, we used a diffusion theory code which permits inelastic scattering and we also used the S_n code.

MR. NESTOR:

You used PDQ in R, Z geometry?

MR. AVERY:

Yes.

MR. NESTOR:

Were you able to get a neutron balance?

MR. AVERY:

We thought we did. We have heard since that PDQ in R, Z geometry does not quite give an accurate numerical neutron balance. We have not made any corrections for it.

MR. NESTOR:

We ran some PDQ cases with a rather coarse mesh spacing and we got pretty bad neutron balances. I wondered if you had run across this.

MR. AVERY:

I am sure the discrepancy cannot be more than of the order of one per cent, both because of checks we have against one-dimensional calculations and from other general considerations.

MR. WOLFE:

Can you tell us what power density you expect in the superheater core and, on the same line, do you have any idea what sort of inventory costs you would get as a result?

MR. AVERY:

I should qualify my answer as being very tentative. Most of our calculations were based on thoughts that we would be able to get the order of a hundred megawatts thermal in the 400-liter core. This is approximately one quarter of the power density expected in sodium-cooled systems. Some more recent considerations indicate that our initial power density estimates may possibly be increased by a factor of two.

MR. SANDERS:

Could Dr. Avery be a bit more specific about the improved kinetic performance available from the longer lifetime in these coupled systems. In particular, have there been any quantitative calculations on the effect of the increased lifetime in the sodium-cooled system ?

MR. AVERY:

I would not want to be too specific, but we have done quite a few calculations using various models and explosion codes with the lifetime as a variable. At this point the picture is not clear in all respects. It seems that in certain categories of accidents, characterized by assumed reactivity insertion rates, there are pronounced advantages to the longer lifetime. Under other conditions there may be a little difference.

MR. OKRENT:

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In this respect it might be worth noting that some of the fuel elements that people have been discussing here for fast reactors, like cermets, carbides, and oxides, may turn out to have safety features which might not be the most desirable, but which might be made palatable if one could attain with these the longer lifetime. Such considerations are only relevant in discussion of events of very low probability, but nevertheless the longer lifetime might be just the difference.

ADVANCED EPITHERMAL THORIUM REACTOR CONCEPT

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 A Division of North American Aviation, Inc.* Canoga Park, California (Paper Presented by Mr. Campise)

Introduction

Atomics International (AI) is under contract to Southwest Atomic Energy Associates to investigate the economical potential of a reactor concept utilizing U^{233} and thorium in a neutron energy spectrum considerably above thermal. This system is called the Advanced Epithermal Thorium Reactor (AETR). This reactor system will probably incorporate fixed fuel elements cooled with liquid sodium and may be moderated by graphite or beryllium metal.

The AETR concept represents a natural extension of the sodium technology gained at Atomics International. Operating in the epithermalintermediate energy range, the AETR concept will improve the neutron economy of thermal sodium systems. Smaller sizes of reactors will be obtainable in this range of neutron energies.

The main assumption that generated the AETR concept was that the value of alpha for U^{233} is probably small in the epithermal intermediate energy range and much smaller than that of Pu^{239} . The alpha data for Pu^{239} as reported in KAPL-1793 and the estimated alpha values for U^{233} are depicted in Fig. 1. If the depicted values for alpha are indicative of the two fuels, a Th- U^{233} cycle would have greater merit in a power reactor than the U^{238} - Pu^{239} cycle in this range of neutron energies. Thus, the AETR concept will fill a gap in our power breeder program, where the Th²³²- U^{233} cycle appears to be superior to the U^{238} - Pu^{239} cycle.



*Under Contract with Southwest Atomic Energy Associates, Little Rock, Arkansas.

The economics of the AETR system depends to some extent on the breeding potential and fuel utilization, but to an even larger extent on the power potential of the final reactor design.

During the last year and a half, a research program has been conducted to determine to what extent nuclear data are available. Nuclear cross sections and resonance parameters in the epithermal-intermediate neutron energy range are limited. In addition, an effort was made to adapt the latest calculational methods available for the evaluation of such a reactor system. Calculational methods used for the parameter survey studies consist of the GNU-II (multigroup, multiregion diffusion) code and the SNG (transport) code.

During the second phase of the program, AI will continue the core evaluation program and supplement the nuclear data by conducting critical experiments in an Epithermal Critical Assembly, which is currently being designed.

Nuclear Data

The cross-section data of primary concern to the AETR project are those for U^{233} . At present, the value of η for U^{233} , even in a thermal spectrum, is not known to better than a few per cent. There is as much as 5 per cent variation between the extreme thermal values quoted for the ratio of η^{233}/η^{235} (a value of 1.057 is reported from the British Atomic Energy Research Establishment, and a value of 1.114 is reported by Russian scientists). Superimposed on this uncertainty in the ratio of the two value of η is the uncertainty in η^{235} . This problem has been assigned a high priority by the Atomic Energy Commission, and the thermal neutron cross sections of U^{233} are being actively investigated at several national laboratories.

There exist many different sets of cross-sectional data for U^{233} that have been used in the reactor field. The following three sets of data were considered in our studies:

- 1) those published in ANL-5800;
- ORNL data presented at the 1958 Detroit meeting of the American Nuclear Society and also obtained through private communication with A. M Perry of ORNL; and
- 3) a so-called GNU-II set, compiled at Atomics International, consisting of original data from the MTR Fast Chopper, Russian eta data, Los Alamos fast cross sections, and selected points from BNL-325 (see Figure 2).





These sets of data are further characterized by the eta values as a function of neutron energy group listed in Table I and depicted in Fig. 3.

TABLE I

Group*	EL	AI η(g)	ANL-5800 $\eta(g)$	$ORNL \eta(g)$	Group*	EL	$_{\eta(g)}^{\rm AI}$	ANL-5800 η(g)	ORNL η(g)
1	6.065 Mev	2.95	2.95	2.95	18	0.372	2.25	2.20	2.18
2	3.679	2.95	2.95	2.95	19	0.304	2.25	2.19	2.16
3	2.231	2.61	2.61	2.61	20	0.249	2.25	2.19	2.16
4	1.353	2.61	2.61	2.61	21	0.204	2.25	2.19	2.16
5	0.821	0.50	2.50	2.40	22	0.167	2.25	2.19	2.16
6	0.498	2.43	2.43	2.43	23	0.112	2.28	2.19	1.79
7	0.302	2.43	2.47	2.51	24	61.44 ev	2.28	2.20	1.79
8	0.183	2.37	2.47	2.48	25	33.72	2.28	2.20	1.85
9	0.111	2.37	2.25	2.19	26	22.6	2.28	2.12	1.85
10	67.38 kev	2.25	2.25	2.18	27	8.32	2.28	2.12	1.93
11	40.87	2.25	2.21	2.18	28	3.06	2.20	2.07	1.93
12	24.79	2.25	2.21	2.18	29	1.125	2.20	2.07	1.84
13	15.03	2.25	2.20	2.18	30	0.414	2.30	2.07	2.28
14	9.119	2.25	2.20	2.18	31	0.152	2.30	2.29	2.20
15	3.355	2.25	2.19	2.18	32	0.0252	2.30	2.30	2.28
16	1.234	2.25	2.19	2.18	Thermal	0	2.28	2.24	2.28
17	0.454	2.25	2.20	2.18					

Eta Values from AI, ANL, ORNL

*For the fast groups one through four, all three sets of data utilize information obtained from Los Alamos Report N-2-731.



The above variations result in uncertain prospects for a U^{233} breeder. However, the reported uncertainties in cross-section data to some extent exaggerate the seriousness of the problem. Much of the reported variation may be due, at least in some measure, to variations in the physical model and to the methods used for extracting cross-section data from the raw experimental results. In spite of current uncertainties, it would appear that there is a good probability that a useful U^{233} power reactor can be built with an appropriately chosen neutron energy spectrum.

A Reference Core

An attempt was made to evaluate the three different sets of crosssection data available for U^{233} . The reference core for this calculation was chosen so that a large percentage of the neutron flux in the system was located in the energy range of most uncertainty. The reactor compositions are listed in Table II.

TABLE II

Core Compositions for 50% Carbon-moderated Reactor Core (Infinite cylindrical geomerty)

Compositions					
Region 1 (core)	Region 2 (blanket)				
Carbon = $0.0401 \times 10^{24} \text{ atoms/cm}^3$ Sodium = 0.00765 Stainless Steel = 0.0022611 Thorium = 0.003275 Uranium-233 = variable	Carbon = $0.01849 \times 10^{24} \text{ atoms/cm}^3$ Sodium = 0.00543 Stainless Steel = 0.0034885 Thorium = 0.01407				

The median fission energy (MFE) and net breeding ratios for the different cross-section sets of data are listed in Table III.

TABLE III

Median Fission Energy and Net Breeding Ratio for 50% Carbon-moderated Core (For different cross sections)

Cross-section Set	Critical Mass (kg)	MFE (kev)	Breeding Ratio
AI	1242*	1.25	1.112
ANL-5800	1067	0.98	1.061
Oak Ridge	1380	2.37	0.980

*1242 kg corresponds to 0.00037098 x 10^{24} atoms/cm³

The neutron balance and the averaged properties of U^{233} in this 50 per cent graphite-moderated core are given in Table IV and Table V, respectively.

TABLE IV

	AI	ANL-5800	Oak Ridge
Absorptions in Core (%)		5	n pina ny mandri dia 1940 mila mila mila mila mila mila mila mila
Carbon	0.0056	0.0044	0.0044
Sodium	0.4951	0.4746	0.4532
Chromium	0.4853	0.4790	0.4729
Iron	0.4493	0.4232	0.4117
Nickel	0.8496	0.8530	0.8344
Thorium	42.2863	41.0643	39.3519
Uranium-233	46.0111	47.3371	49.2923
Absorptions in Blanket (%)			*
Carbon	0.0001	0.0001	0.0001
Sodium	0.0140	0.0142	0.0139
Chromium	0.0520	0.0515	, 0.0514
Iron	0.0459	0.0439	0.0435
Nickel	0.0976	0.0979	0.0972
Thorium	9.2139	9.1560	8.9711
Contribution by Region to			
the Breeding Ratio			'
Core	0.9119	0.8674	0.7983
Blanket	0.2002	0.1934	0.1820
Total	1.1121	1.0608	0.9803

Neutron Balance for 50% Graphite Core

TABLE V

Case	Critical Mass (kg)	MFE (kev)	η	$\overline{\nu}$	ā
AI	1242	1 2 5 0	2.30	2.524	0.0956
ANL-5800	1076	0.98	2.241	2.518	0.1237
Oak Ridge	1380	2.370	2.173	2.523	0.1610

Averaged Properties of U²³³ in 50% Graphite-moderated Core

As is obvious from Table IV, the GNU cross-section data yield the most optimistic breeding ratio, and the ORNL data yield the least optimistic value for this reference reactor with 50 vol per cent graphite core. The variation in the calculated breeding ratio for this core is about 13 per cent.

The uncertainty in the eta data for U^{233} is a reflection of the uncertainty in alpha for U^{233} . The values of $\overline{\nu}$ are fairly well known over the entire neutron spectrum for U^{233} . B. C. Diven, et al., (1) reported $\overline{\nu}$ (U^{233}) = 2.585 \pm 0.062 at 80 kev, and Saunders⁽²⁾ reported a slope of $\overline{\nu}$ with energy to be 0.125 (Mev)⁻¹. The three sets of cross-section data are reasonably consistent with regard to the value of $\overline{\nu}$ in the kilovolt region; the major differences are in the values of α (or η) which are used. J. J. Devaney of Los Alamos Scientific Laboratory (private communication) indicated that a value of $\alpha = 0.1$ represented a good guess; however, this is only a guess. Since the value of alpha for U^{233} will more or less determine the optimum neutron energy spectrum in which the AETR will operate, a survey using the AI cross section set was made. Assemblies with 23 to 70 vol per cent



Fig. 4 Median Fission Energy vs Maximum Breeding Ratio and Moderator Energy

graphite in the core were studied. The probable design of the AETR core characteristics should lie between these limits. Infinite and finite cylindrical reactor models were used to determine the possible range of breeding ratios for the above systems. The effect of end blankets is relevant to design of the fuel element and reactor core. The results of these surveys are depicted in Fig. 4.

As indicated in Fig. 4, it appears that a reasonably high breeding ratio is obtainable with a graphite-moderated reactor operating in the above energy interval. The energy range of interest to the AETR project is between 100 ev and 1 Mev. In this range the uncertainties are even greater than the uncertainties in either the lower or the higher energy ranges. Experimentally, this energy region is the most difficult one in which to work, and those research groups which are active in cross-section measurements see no immediate prospects for large increases in accuracy. Thus, a real need exists for a critical experiment to obtain integral data in this neutron energy range. These data would be needed in reference to the above circumstances and to evaluate the relative merit of the parameters survey and future studies along these lines. A critical experiment program is planned to furnish useful data in this energy range. A brief description of this critical assembly is presented in the following section.

Critical Experiment

Since the available quantity of U^{233} (25 kg) is only about 2 per cent of the estimated critical mass of the full scale AETR core, a multiregion critical assembly has been designed (see Fig. 5). It is of the fast-slow type with a central test region of thorium and U^{233} , a buffer region of thorium and U^{235} , a thermal driver containing U^{235} and polyethylene, and a polyethylene reflector. The use of a thermal driver reduces the total critical mass and makes the neutron generation time approximately that of a thermal reactor.



Fig. 5 Conceptual Design of Critical Assembly

In the preliminary design of the assembly a physics survey has been made to develop a central test region which has the same neutron spectrum and importance functions as the full-scale AETR over as much of the test region as possible. A series of calculations have been used to optimize the critical assembly design.

The first part of the experimental program will compare the neutron spectra and importance functions of the test regions containing various fuelto-moderator ratios with those calculated for the systems. Energy average flux distributions will be determined. Experimental techniques need to be developed for these measurements. Foil and fission counter methods are proposed. To increase the sensitivity, oscillator measurements will be tried.

Other parameters of particular interest to the AETR program (such as breeding ratios, Doppler coefficients, and temperature coefficients) will be measured in the test region of the critical assembly.

In conclusion, if the critical experimental program provides integral data that indicates that eta is larger than 2.2, then over a wide energy interval a useful power breeder system using the $Th-U^{233}$ cycle can be designed to operate in an epithermal-intermediate neutron energy range.

References

1. B. C. Diven et al., Phys. Rev. 101, 1012 (1956).

2. J. E. Saunders, NDRC-94 (May 1956).

Discussion of Paper Presented by Mr. Campise

MR. HALL:

Are there questions on this paper?

MR. GREEBLER:

Did you allow for any burnup in your calculations?

MR. CAMPISE:

In our first calculations, we tried to prove the feasibility of the epithermal energy range. We are presently looking at burnup. It doesn't look too bad. We estimate that 12,000 MWD/T for uranium-thorium fuel is probably a reasonable number. Assuming that it is, our effective breeding ratio, using our current cross-section tables, indicates a breeding ratio of about 1.2 (assuming a 1 per cent reprocessing loss).

MR. GREEBLER:

I see; thank you.

MR. SAMPSON:

In view of the great difficulties in tying down nuclear data of eta, nu, alpha, and so on, say in U^{235} , even with critical assemblies, I wonder how sure you can be of getting good answers to your questions with an integral experiment. Have you thought this through or is it possible to say anything further about that point at this time ?

MR. CAMPISE:

We hope that we have developed our programs to the point where we might use these as tools to obtain integral data by making reasonable assumptions about the cross sections and assuming that one of the biggest problems we have to contend with is that of alpha. We will take off from this point.

MR. PATTENDEN:

I am not familiar with the eta curve which you gave us as being the Oak Ridge data. Where did it come from? Is it a measured curve or a calculation?

MR. CAMPISE:

The curve embodies the cross sections they have been using at Los Alamos; these were presented at the 1958 Detroit meeting and in communications I had with A. M. Perry from Oak Ridge. He sent me a table of 32-group cross sections that they were presently using at Oak Ridge.

MR. PATTENDEN:

I see. I can't think of what these cross sections are because I don't think the fission cross section has been measured at Oak Ridge. Maybe they have used some other cross section for this.

MR. CAMPISE:

This is the big difficulty. We have total cross-section measurements up to a kilovolt and fission cross sections up to a kilovolt in the MTR. We have fast cross sections that have been tested and reworked at Los Alamos and seem to calculate fast critical assemblies. So we have a range of from about a kilovolt to a 0.1 MEV, where we have to guess what the cross-section variations are.

In addition to this, the alpha value is one of our problems. Another problem is the scattering cross section inside the resonance. This makes a big difference as to how you average your cross section from the raw data. I plan to talk to Jack Chernick, who has looked into this problem.

MR. PATTENDEN:

I think that, probably, this afternoon the more recent data on total cross-section measurements of uranium-233 will be talked about.

MR. CAMPISE:

Yes, but our thermal flux is down to pretty close to zero, so the thermal values are of no immediate interest to us.

MR. PATTENDEN:

We have continued our results out to thousands of electron volts, where the spectrometer is very poor.

MR. SHAFTMAN:

What are these ANL-5800 values you are quoting? Is that the multigroup set based on the 30-group set in section 3 of ANL-5800? 196

MR. CAMPISE:

Yes.

MR. SHAFTMAN:

Well, that came from Oak Ridge.

MR. CAMPISE:

I think we had better get together because I have a carbon copy of the data, which is a little bit different, from A. M. Perry.

MR. NESTOR:

I think I can clarify this situation a little bit. The ANL-5800 numbers are some which are quite ancient. When the Eyewash Code was first running, we needed a set of U^{233} cross sections. We put on a tall conical hat, and made some cross sections. They were put on the tape, and they acquired a certain, shall we say, sanctity, and it was accepted as the Word. But we never intended this! The newer cross sections which you got from Bud Perry are based primarily on MTR data, and that is a newer set of cross sections.

MR. CAMPISE:

Right. Of more general interest is the fact that we are trying to prove feasibility in that respect. Having three different sets, whether they are indicative of nature or not, serves the purpose of comparing the three sets of data that have different alpha values. In this respect they were well worth the time to use it.

However, one of the important points that has not been resolved, in my estimation, is the eta values that have been published by the Russians. It would have been nice if they were invited to a meeting like this to justify them.

MR. OKRENT:

We did extend an invitation. Unfortunately, there were delays in getting permission to extend the invitation and so we did not have their representation here.

MR. VonHERRMANN:

Are those the sets of cross sections that come with the GNU code ?

MR. CAMPISE:

I pointed out that this GNU set was compiled at AI.

MR. VonHERRMANN:

The reason I am asking is because I have been connected with putting certain values for eta from Russian data into this code. This was several years ago.

MR. CAMPISE:

We did not use the raw data that came with the GNU program.

MR. VonHERRMANN:

Then there is a basis for the value of eta that you have in the code? I just wanted to check that.

MR. CAMPISE:

Right.

MR. SANDERS:

The neutron spectrum in this system is in the region where it is extremely difficult to make experimental measurements. I wonder, in your critical facility, whether you have in mind any exotic technics for elucidating the neutron energy spectrum.

MR. CAMPISE:

We plan to have medium fission energies that will cover this intermediate energy range in the test region of the critical assemblies. From this we hope to make integral measurements and determine what the crosssection variations might be.

MR. SANDERS:

These will be entirely integral measurements, will they?

MR. CAMPISE:

Yes, they will.

MR. WOLFE:

How do the economics and prospects for this reactor compare to the prospects of the other sodium reactors you have been working on at AI?

MR. CAMPISE:

I can give you an off-the-cuff answer. We have not looked into this in detail yet, mainly because of the cross-section difficulties. I would say that the neutron economy alone should help us. But I may not say more than that.

MR. OKRENT:

Why don't you use beryllium instead of graphite, since one can get this bonus from breeding?

MR. CAMPISE:

We have investigated beryllium but, from an engineering standpoint, it is fairly expensive as a reactor material.

MR. OKRENT:

Is it ruled out, however?

MR. CAMPISE:

No, it is not. As I pointed out in my introduction, the system will be moderated by either graphite or beryllium metal.

FISSION PRODUCT POISONING DATA

N. J. Pattenden Oak Ridge National Laboratory Oak Ridge, Tennessee

Introduction

To estimate the poisoning due to fission products in a reactor as a function of neutron flux and integrated irradiation, in general one needs to know the half-lives, yields and cross sections of all the fission product nuclides. The reactor neutron energy distribution extends from fission energies down to thermal energies (about 10^6 to 10^{-2} ev); hence, in principle, the cross sections are required over this range. If the yields vary with neutron energy (and there is evidence that those in the valley of the mass-yield curve do), then it might also be necessary to measure them as a function of energy. This represents a formidable quantity of nuclear data. Fortunately, the practical requirements are less rigorous, and reasonable criteria of poisoning significance may be set to limit the experimental work necessary, which depend on the properties of the fission products themselves and on the particular reactor specification.

Poisoning Definition

Let us define the poisoning as the ratio of the absorption rate of neutrons by a fission product nuclide to the fission rate in the reactor. For simplification we make the following assumptions. (1) The fission rate remains constant during operation. (2) No second-order production of fission products occurs (i.e., formation by neutron capture from another fission product), except in the case of high cross-section fission products. (3) A particular mass chain β -decays straight to fission product nuclide i without loss through neutron capture.

Then the poisoning may readily be shown to be given by

$$\frac{\mathbf{y}_{i}\phi\sigma_{i}}{\lambda_{i}+\phi\sigma_{i}}\left[1-e^{-(\lambda_{i}+\phi\sigma_{i})t}\right]$$

where y_i is the fission yield, ϕ is the neutron flux, σ_i is the effective neutron absorption cross section, λ_i is the decay constant, and t is the irradiation time. The subscript i refers to a particular fission product nuclide.

For a stable poison, or one with $\phi \sigma_i \gg \lambda_i$, the poisoning is $y_i(1-e^{-\phi \sigma_i t})$. In this case, at saturation, the poisoning is y_i but if $\phi \sigma_i t$ is small (i.e., far from saturation), the poisoning is $y_i \phi \sigma_i t$. Of the known cross sections of fission products, those of six (Cd¹¹³, Xe¹³⁵, Sm¹⁴⁹, Sm¹⁵¹, Eu¹⁵⁵ and Gd¹⁵⁷) are above about 10,000 barns, and the rest are below about 500 barns. Hence we may usefully divide fission products into groups of high and low cross sections. For small irradiations, only the first group may be considered, but for large irradiations (greater than about 10^{20} neutrons/cm²), their poisoning saturates at a value of about 0.075 for U²³⁵ fission. (For U²³³ fission the figure is about 0.059, and for Pu²³⁹ fission it is about 0.083.) In the case of the second group, their poisoning is proportional to irradiation until very large irradiations are reached. At 10^{21} neutrons/cm² the known cross sections of the second group have a poisoning value of about 0.06, increasing nearly linearly with irradiation.

Effective Cross Sections in Reactor Spectra

Thermal reactors have a neutron flux distribution which is approximately represented by

 $\phi(\mathbf{E}) = \mathbf{M}(\mathbf{E}) + \lambda \mathbf{F}(\mathbf{E})$

where M(E) represents the normalized Maxwellian flux distribution, usually written in the form

$$M(E)dE = [E/(kT)^{2}]exp(-E/kT)dE$$

The quantity F(E) represents the slowing-down flux, proportional to E^{-1} per unit energy interval at energy E. The ratio of the slowing down flux per log e energy interval to the integrated Maxwellian flux is given by λ .

To determine an effective absorption cross section in such a spectrum, it is necessary to know details of the variation of cross section with energy in the Maxwellian region, but not in the slowing-down region, where an integral cross section is sufficient. The latter may be obtained from known resonance parameters or from a resonance integral experiment. The effective cross section may then be derived for any thermal reactor knowing the temperature and the hardness of spectrum (λ). The notation of Westcott, (1) at Chalk River, and Campbell and Freemantle, (2) at Harwell, is convenient for this purpose. A nuclide is considered to have an effective cross section in a thermal reactor spectrum defined by $\sigma_{eff} = \sigma_{2200}$ (g+rs), where σ_{2200} is the absorption cross section at 2200/m/sec, and g and s characterize the departure of the cross section from a 1/v dependence in the Maxwellian and slowing-down regions, respectively. Thus the thermal cross section is

$$\sigma_{\mathbf{M}} = g\sigma_{2200} \int_{0}^{\infty} M(\mathbf{E})\sigma(\mathbf{E})d\mathbf{E}$$
$$\int_{0}^{\infty} M(\mathbf{E})\left(\frac{0.0253}{\mathbf{E}}\right)^{\frac{1}{2}} d\mathbf{E}$$

The epithermal cross section is

$$\sigma_{epi} = s\sigma_{2200} = \frac{\sigma_{r,i.}}{\int_{0}^{\infty} M(E) \left(\frac{0.0253}{E}\right)^{\frac{1}{2}} dE}$$

and $\sigma_{r,i}$ is the resonance integral in excess of a 1/v integral:

$$\sigma_{\mathbf{r.i.}} = \int_{\mu \mathbf{kT}}^{\infty} \left[\sigma(\mathbf{E}) - \sigma_{\mathbf{M}} \left(\frac{0.0253}{\mathbf{E}} \right)^{\frac{1}{2}} \right] \frac{d\mathbf{E}}{\mathbf{E}}$$

It should be noted that

$$\int_{0}^{\infty} M(E) \left(\frac{0.0253}{E} \right)^{\frac{1}{2}} dE = \left(\frac{0.0253\pi}{4kT} \right)^{\frac{1}{2}}$$

,

The quantity r depends on the ratio of the slowing-down to the thermal flux and is defined as

$$\mathbf{r} = \frac{\lambda}{1 + 4\lambda / (\mu \pi)^{\frac{1}{2}}}$$

where the slowing-down flux is assumed to have a low energy limit of μkT . Westcott takes μ to be 5, and Campbell and Freemantle use an arbitrary smooth function equivalent to a μ of 2.81. The value of r is close to λ when λ is small, and is not likely to exceed 0.1 in the fuel in most practical cases.

For a cross section with a 1/v dependence, g = 1 and s = 0; otherwise g and s are functions of neutron temperature T. The quantity s is effectively independent of μ unless there happens to be a resonance in the cross section in the neighborhood of μ kT (unfortunately this is often the case with nuclides of high cross section).

Data on the six known high cross-section poisons and the known low cross-section poisons are shown in Tables I and II, respectively. The effec-

н	GH CROSS SEC	TION FISSION	PRODUCT POIS	ONS
FISSION PRODUCT	U ²³⁵ YIELD (%) (KATCOFF)	DECAY CONSTANT λ (sec ⁻¹)	^{g o} 2200 (barns) (T ~ 300°K)	$ \begin{matrix} \sigma_{eff} \\ (barns) \\ \left(T \sim 300^{\circ} K \\ r = 0.10 \end{matrix} \right) $
C9 ₁₁₃	0.010	0	26,600±300	27,800
Xe ¹³⁵	6.3	2.1 × 10 ⁻⁵	3.0 × 10 ⁶	
5m ¹⁴⁹	1.13	0	68,500 ± 1000	67,000
Sm ¹⁵¹	0.45	3.0 × 10 ⁻¹⁰	11,500±1500	
Eu ¹⁵⁵	0.03	1.3 × 10 ⁻⁸	14,000±4000	
Gd ¹⁵⁷	0.0078	0	220,000±5000	197,000

tive cross sections have been estimated for a temperature of about 300°K and an r value of 0.10. The yield values are for U^{235} fission from the tabulation by Katcoff.⁽³⁾ In Table II, the criterion of significance is that $y\sigma_{eff} > 0.1$ barn/fission. For comparison, the thermal cross sections of $g\sigma_{2200}$ are included; it may be seen that in the low



cross-section group, the total fission product cross section is increased by 47 per cent if the effective cross sections (including resonance contributions) are used, rather than the thermal cross sections.

LOW CROSS SECTION FISSION PRODUCT POISONS						
FISSION PRODUCT	U ²³⁵ YIELD (%) (KATCOFF)	^{g σ} 2200 (barns) (T ~ 300°K)	^σ eff (barns) (T ~ 300°K) r = 0, 10	^{y × σ} eff (barns/fission)	^{y × g σ} 2200 (barns/fission)	
Kr ⁸³	0.54	220 ± 40	288	1.55	1.2	
Zr ⁹¹	5.84	1.58 ± 0.12	2.1	0.12	0.09	
M0 ⁹⁵	6.27	14.7 ± 0.5	29	1.82	0.92	
M0 ⁹⁷	6.09	2.2 ± 0.7	3.6	0.22	0.13	
Tc ⁹⁹	6.06	25 ± 2	39	2.36	1.5	
Rh ¹⁰³	3.0	146 ± 5	261	7.83	4.4	
1 ¹²⁹	0.9	32 ± 5	36	0.32	0.29	Α
Xe ¹³¹	2.93	120 ± 15	199	5.34	3.5	(TABLE II
Cs ¹³³	6.59	28 ± 1	84	5.54	1.8	4
La ¹³⁹	6.55	8.9 ± 0.3	10	0.66	0.58	
Pr ¹⁴¹	6.0	11.5 ± 0.3	18	0,78	0.69	
Nd ¹⁴³	5.98	339 ± 20	341	20.8	20.2	
Nd ¹⁴⁴	5.67	5.0 ± 0.6		(0.28)	0.28	
Nd ¹⁴⁵	3.95	54 ± 3	82	3.24	2.1	
Nd ¹⁴⁶	3.07	10 ± 1		(0.31)	0.31	
Pm ¹⁴⁷	2.7	180 ± 50	408	11.0	4.9	
Sm ¹⁵⁰	1,13*	105 ± 5		(1.2)	1.2	
Sm ¹⁵²	0.74*	210 ± 10	535	3.96	1.6	
_{Eu} 153	0.15	458 ± 16	604	0.91	0.69	
			TOTAL	68.2	46.4	

*INCLUDE SECOND ORDER PRODUCT.

Figure 1 shows the poisoning of Xe^{135} , the rest of the high crosssection group, and the low cross-section group as a function of irradiation time for U^{235} fission in a flux of 10^{14} neutrons/cm²/sec.



Discussion

A summation of individual nuclide cross-section measurements can only give a lower limit to the total fission product cross section until all nuclides of high yield have been measured. In the low cross-section group, better values are required for the thermal cross section and resonance parameters of Pm^{147} , and for the Ru and Pd isotopes (including active Ru^{106} and Pd^{107}). Resonance parameters are required for the Kr, Te and Xe isotopes (including active Kr^{85}), and for Sr^{90} .

In the high cross-section group, better values are required for the thermal cross sections and resonance parameters of Sm^{151} and Eu^{155} , and the spin assignment of the low-energy Xe^{135} resonance is required.

Several papers presented at the 1958 Geneva Conference gave data on fission yields. If these are compared amongst themselves and with the Katcoff compilation, large discrepancies may be seen to exist. Errors in yield values may not be important in the low cross-section group since they may tend to cancel out. However, in the high cross-section group this is not likely to occur. Table III lists some high and low yield values which have been published for some members of this group. It is clear that these discrepancies must be removed as part of the program for improving the accuracy of poisoning estimates.

TABLE III

Fission Products	Yields (%)					
	U ²³³	U ²³⁵	Pu ²³⁹			
Xe ¹³⁵	5.1 (a) - 6.0 (b)	6.41(b)	5.8 (a) - 7.27(b)			
Sm ¹⁴⁹	0.61(a) - 0.8 (b)	1.13(b) - 1.50(c)	1.32(b) - 1.89(a)			
Sm ¹⁵¹	0.26(a) - 0.33(c)	0.45(b)	0.79(b) - 1.17(a)			

Discrepancies in Yield Measurements

(a) S. Katcoff, Nucleonics 16, No. 4, 78 (1958).

(b) Bidinosti, et al., Proc. 2nd UN Intnl. Conf. on Peaceful Uses of Atomic Energy, United Nations, Geneva (1958), Vol. 15, p. 459
(c) Anikina, et al., ibid., Vol. 15, p. 446

References

- 1. C. H. Westcott, AECL, CRRP-680 (1957) and CRRP-787 (1958).
- 2. C. G. Campbell and R. G. Freemantle, AERE, RP/R2031 (1956).
- 3. S. Katcoff, Nucleonics <u>16</u>, 78 (1958)

Discussion of Paper Presented by Mr. Pattenden

MR. HALL:

Are there any questions on this paper?

MR. SAMPSON:

Do you have available a resonance integral over all the low crosssection poisons above some stated energy? I don't remember you giving your results in quite that form.

MR. PATTENDEN:

Yes.

MR. SAMPSON:

Is r in the Westcott relation just σ_a/σ_s , and are these results for a specific r?

MR. PATTENDEN:

These results are for a specific r, but r is simply the ratio of the epithermal to the thermal parts of the reactor spectrum -

MR. SAMPSON:

Right.

MR. PATTENDEN:

- without any bearing on the cross sections.

MR. SAMPSON:

Are all of these for r equals 0.1?

MR. PATTENDEN:

Right.

MR. GREEBLER:

Are these ratios of fission product captures to fission rates that you listed on the board for 2200 meters per second? Or are they for 300°K?

MR. PATTENDEN:

The low cross-section ones refer to a neutron temperature of 300° K and an r value of 0.1.

MR. GREEBLER:

I see. If we go to higher temperatures, how does the g factor change for the fission products as compared to the change for the g factor of U^{235} ?

MR. PATTENDEN:

If the cross section is l/v, the g factor is one, irrespective of the temperature.

MR. GREEBLER:

Are you saying that the low cross-section fission products are characterized by a 1/v cross section?

MR. PATTENDEN:

In general, yes.

MR. GREEBLER:

Thank you.

MR. SHAFTMAN:

If I may quote from your own paper (Table IV, ORNL-2778) with regard to the question of the temperature variation of the g factor for the total fission products, from 300 to 2000° K, g changes from 1.0 to 1.06 in a roughly linear fashion after 1000° K. I think that the more important variation is the change in the s factor. Again referring to your paper (ORNL-2778), at 300°K, s is 3.43. This is for the total fission products. The factor s rises to 9.36 at 2000°K, and this brings me to the point that I should like to raise here.

In your presentation you didn't emphasize the change of effective cross section because of the contribution of the resonances. I think you didn't emphasize that enough. You do so properly in your paper. However, it was indicated that there is about a 50 per cent increase in the cross section, because of the contribution of resonances.

What was emphasized in Dr. Pattenden's work and in the work that I have done, for example, is that the important uncertainties in fission product poisoning arise perhaps largely from unknown resonances in the fission products, and also from the fission yield, and not so much from the small uncertainties in thermal cross sections.

MR. PATTENDEN:

I will just say that in the report that was mentioned, I said the resonance effect increased the effective poisoning by about 33 per cent. In fact, I have managed to put this up to 47 per cent now by considering more resonances.

MR. LEVENE:

Would you give a number for the resonance integral in barns per fission for the low cross-section poisons?

MR. PATTENDEN:

The effective cross section came out to be about 68 barns per fission. The thermal value is about 46 barns per fission. Hence the excess resonance integral is about 190 barns per fission.

SESSION IV

Tuesday Afternoon, October 20, 1959

Chairman: L. Bollinger Secretary: P. Persiani

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Mr. BOLLINGER, CHAIRMAN:

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Approximately a year ago it seemed advisable to look into the question of the correct values of the cross sections of the three common fissionable materials, more closely than it had been previously. The reason for this was that it seemed to us who measure cross sections that suddenly the reactor people realized that some of these cross sections are in doubt.

The first three papers are related to the extent that they all concern fissionable materials, but also in that they all in a sense are an outgrowth of this sudden stir three years ago.

Now, I think it should be obvious to all of you that when one has data which are in conflict, there is no man alive who can tell you what the correct answer is. Moreover, there is not even a way in which one can find the single best answer. The most that we can do is to set down in a concise way what the different answers are, to find out where there are discrepancies, to cross-examine what possible sources of discrepancies might be, and then make an educated guess as to what is the best answer.

The assignment of doing these things was given to the first three speakers today by the Nuclear Cross-section Advisory Group, and at least a part of what they say today is an outgrowth of studies that they have made. If you find what they say interesting, their papers are also just at the point of being published, so that you could examine the data in more detail.

Now let me call on the first speaker of the afternoon, Mr. G. Safford, Columbia University, who will review fission parameters of U^{235} , and in particular the low-energy portion of it.

REVIEW OF THE LOW ENERGY CROSS SECTIONS AND FISSION PARAMETERS OF U²³⁵

George J. Safford Columbia University New York, N.Y.

I think it might be well to open with a reminder that these papers could be said to concern mainly doubt and quite a bit of it. In recent years improvements in reactor theory and knowledge of the fission process have led to a demand for greater accuracy in the experimentally determined cross sections and fission parameters of U^{235} . Despite the time and effort placed on measurements of the parameters of this isotope, marked discrepancies have existed in the experimental values obtained.

Therefore, it is desirable to examine the more recent measurements of the parameters commonly used to characterize the fission process, their accuracy and the sources of uncertainty. These parameters and their relationships are summarized in Fig. 1.

Fig. 1

PARAMETERS CHARACTERIZING SLOW NEUTRON FISSION

(1)
$$\sigma_{nT}$$
, the total cross section $\sigma_{nT} = \sigma_{nn} + \sigma_{nf} + \sigma_{n\gamma}$ (1)
(2) σ_{nn} , the scattering cross section
(3) σ_{na} , the absorption cross section $\sigma_{na} = \sigma_{nf} + \sigma_{n\gamma} = \sigma_{nT} - \sigma_{nn}$ (2)
(4) σ_{nf} , the fission cross section $\alpha = \frac{\sigma_{n\gamma}}{\sigma_{nf}}$ (3)
(5) $\sigma_{n\gamma}$, the capture cross section $\alpha = \frac{\sigma_{n\gamma}}{\sigma_{nf}}$ (3)
(6) η , the average number of neutrons emitted per neutron absorbed (7) α , the ratio of the capture to fission cross section (8) ν , the average number of neutrons $\eta = \nu \frac{\sigma_{nf}}{\sigma_{na}} = \nu \frac{1}{(1 + \alpha)}$ (5)

One absolute measurement must be made in each of the following groups:

- (1) σ_{nT} (3) σ_{nf} , $\sigma_{n\gamma}$, or α
- (2) σ_{nn} or σ_{na} (4) η or ν

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Because of these relationships, α , σ_a , σ_f , η and ν are overdetermined by the experimental data. Therefore, to obtain an average set of the eight parameters, one must require that the measured variations with energy and the measured absolute values at thermal energy are consistent within error in keeping with the above relationships.

σ_t

The total cross section of U^{235} is generally obtained by measuring the neutron transmission of a sample of known thickness and composition, using monochromatic neutrons from a crystal spectrometer or a chopper and time-of-flight technique. A summary of the measured values of σ_t at 2200 m/sec are shown in Table I.

Source	$\sigma_{nT}^{}(barns)$	Comments
L. M. Bollinger, private communication, August 22, 1957	694 ± 2	Measured using ANL fast chopper
Palevsky, Carter, Eisberg and Hughes, Phys.Rev. <u>94</u> ,1088 (1954)	700 ± 5	Measured using BNL slow chopper
Melkonian, Havens and Levin, Report CU-115, February 1953 (unpublished)	691 ± 5	
B. R. Leonard, Jr., Report HW 33384 (unpublished) and private communication	701.4±4	Value listed is single datum from a $\sigma_{nT}(E)$ curve. Possibly a least square analysis of the whole curve would yield an improved value
P. A. Egelstaff, J. Nucl. Energy 1, <u>92</u> (1954)	739 ± 15	This value was measured in 1951 and was revised in 1955 to correct for error in velocity scale
Nikitin, Galanina, Ignatiew, Okorakow and Suchorutchkin, Proc.Int.Conf. on the Peaceful Uses of Atomic Energy, Vol. 4, 224 (1955)	710 ± 20	
J. E. Lynn and N. J. Pattenden, Proc. Int. Conf. on the Peaceful Uses of Atomic Energy, Vol. 4, 210 (1955)	729 ± 15	
J. A. Harvey and R. Block, Oak Ridge National Laboratory (Private Communication to G. J. Safford, September 1959)	693.0± 5	Was measured using rolled metallic U ²³⁵ foils
O. D. Simpson and M. S. Moore, M.T.R. Idaho Falls, (Private Communication, September,1959)	690 ± 10	
G. J. Safford, W. W. Havens, Jr., B. M. Rustad, Columbia Univer- sity 1959(to be published)	695 ± 2	This was measured using both liquid samples and metallic foils of U ²³⁵

TABLE	Ι	

Total Cross Section at 0.0253 ev

The four most recent measurements of σ_t which are in excellent agreement at 0.0253 ev illustrate the precision that has been reached:

σ_t = 693.0 ± 5 Harvey and Block (Oak Ridge) = 694 ± 2 Bollinger (Argonne) = 690 ± 10 Simpson and Moore (MTR) = 695 ± 1.8 Safford, Havens and Rustad (Columbia University)

The weighted average gives $\sigma_{t} = 694.3 \pm 1.3$ barns.

In addition, the variation of σ_t with energy for these four crosssection curves is in very good agreement between 0 and 0.1 ev, and, in particular, below 0.0200 ev, where previous results have differed by as much as 3 per cent.

The major sources of error in this type of experiment are:

- a. Uncertainty in sample thickness and in the isotopic and chemical composition. It is necessary to make sure that metallic samples are free from oxide and that oxidation had not occurred during the measurement.
- b. Contamination by neutrons from Bragg reflections of higher order when a crystal spectrometer is used below the peak of the pile spectrum. The presence of neutrons of higher order will tend to yield a low value of the cross section.
- c. Necessity for using very thin samples below 0.02 ev, which makes accurate thickness measurement difficult.

These recent measurements mainly reflect improved techniques in sample preparation and in producing thin samples whose thickness is accurately known.

To obtain σ_a from σ_t at thermal energy, it is necessary to know σ_n . As no measurements of σ_n have been made below 0.27 ev, it is necessary to infer the values in the thermal region from σ_n vs E. The data indicate a rise in σ_n with decreasing energy in the thermal region due to the presence of a negative energy resonance. Vogt's fit to the resonance data of Sailor and Shore extrapolates to $\sigma_n = 17$ barns at thermal energy, while Foote's data yield 14.7 ± 1 (see Table II). However, to obtain the absorption cross section from the total cross section measured with metallic foils, Bragg crystal effects should be considered in the thermal region. These are difficult to calcuate accurately. In addition, these effects will be peculiar to the individual foil used. To allow for these effects and to take an average for the above cross section, we used the value of 15 ± 2 barns and then
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Source	Value	Comments
H. L. Foote, Jr. σ _{nn} (0.0253 ev) Phys. Rev. <u>109</u> , 1641 (1958)	14.7± 1 barns	This value has been obtained from extrapola- tion of data in the range 0.27 to 7.7 ev. These data are incomplete but it appears that σ_{nn} at 0.0253 ev is 4 to 6 barns greater than the potential scattering estimated from nuclear radius.
E Melkonian σ _{nn} (> 1 ev) Unpublished, reported by V. L. Sailor, Proc. Int. Conf. on the Peaceful Uses of Atomic Energy, <u>4</u> , 199 (1955)	11.3 barns	These measurements give the average scattering cross section above 0.6 ev. The data were taken on the Columbia University neutron velocity selector. Vogt (E. Vogt, Phys. Rev. <u>112</u> , 203 (1958)) has obtained a theoretical curve for q_{nn} between $E = 0$ and $E = 1.8$ ev which is based upon the resonance parameters used to fit the fission and absorption cross section data of Shore and Sailor. In view of Vogt's excellent fit to the absorption and fission cross section data and the fact that the σ_{nn} vs E curve fits well the most recent data of Foote in the region between 0.2 and 1.8 ev, it is felt that the σ_{nn} . Vogt found the potential scattering to to be given by $4\pi a^2$ where $a = [1.32 \times (A^{1/3} + 1) \times 10^{-13} \text{ cm}]$.

Scattering Cross Section Data

simply broaden the error to allow for extinction effects in the region of thermal energy. This yields a value of $\sigma_a = 679.3 \pm 3$ barns at 2200 m/sec. Figure 2 shows the variation of $\sigma E^{1/2}$ in the energy region between 0 and 0.6 ev.

The solid curve is the best smoothed line through the data, except below 0.07 ev, where it is a least square fit to the data of Safford, Havens and Rustad. Also shown are the results of Vogt's fit (arbitrary number of channels but only nearby resonances interfere) and the Reich-Moore fit (only one exit channel but all levels of the same spin state interfere) to the data of Sailor and Shore in the region of the 0.282 and 1.138-ev resonances. While these fits have contributed greatly in explaining the asymmetries in the fission resonances, they are not to be interpreted as an accurate quantitative fit to the data in the thermal region. Figure 3 shows $\sigma_a E^{1/2}$ in region 0 to 0.15 ev. Note that below 0.02 ev the data of Leonard appear to be low.





Fig. 3. Absorption Cross Section between 0 and 0.15 ev

 $\frac{\sigma_{f} \text{ and } \alpha}{f}$

The variation of σ_f with E is normalized to an absolute value of σ_f at 2200 m/sec, which has been the subject of many measurements. However, large discrepancies still exist in the direct measurements of σ_f at 2200 m/sec. Tables IIIa and IIIb show a summary of such measurements for direct and pile-average measurements. It should be mentioned here that the value by Safford and Melkonian is not really a direct measurement. It is based on a value of $(1 + \alpha)$ measured at 0.00291 ev combined with the total cross-section measurement; the measured energy variation of the fission cross section has been used to bring it from 0.00291 ev to thermal.

The English values in Table IIIa are probably outdated now and the only intention in showing data that has been revised is simply to illustrate the kind of values that have been obtained in the past. These values have now been changed and the presently stated U.K. average value is 583 ± 10 barns. Even the most recent values vary between 552 and 605 barns, each claiming an error of about one per cent.

TABLE I	II-a
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Source	$\sigma_{f}^{}(barns)$	Comments
Saplakoglu, Bollinger, Thomas and Coceva, Second UN Conf. on the Peaceful Uses of Atomic Energy, A/Conf 15/P/1599	605± 6	Value revised from 614 ± 3 barns in July 1957 and confirmed August 22, 1957. Data comprising 7 runs were used to ob- tain quoted value by weighted average Not all 7 runs are consistent with quoted errors and fail X^2 test.
B R. Leonard, Jr., Proc. of Tri- partite Conf. on Cross Sections of Fissile Nuclei, AERE Harwell Report NP/R 2076 TNCC(UK)-9 (unpublished)	552± 6	Value revised from 556 ± 6 barns as reported in Bull Am Phys Soc Ser II, <u>1</u> , 249 (1956) by Friesen, Leonard, Seppi and White. This value was actually measured at $E = 0$ l ev and extrapolated to $E = 0.0253$ ev on basis of σ_{nf} vs E curve.
J. F. Raffle, Proc of Tripartite Conf. on Cross Sections of Fissile Nuclei, AERE, Harwell Report NP/R 2104 TNCC(UK)-9 (unpublished)	590± 12(*)	Discussed by P. A. Egelstaff in Proc. of Tripartite Conf. on Cross Sections of Fis- sile Nuclei, AERE, Harwell Report NP/R 2104, TNCC(UK)-9 (unpublished). (*) J. F. Raffle, European Physical Society meeting 1958. This tentative value was revised from 638 ± 20 barns by Raffle. The previous value had been obtained by using a thick mask with a circular open- ing over the fission foil and assuming a 2π geometry in the determination of the foil thickness by α -counting. However, the edges of the foil blocked off - particles giving an estimated 8% increase in the value of $\sigma_{\rm f}$.
G.J. Safford and E. Melkonian Phys. Rev. <u>113,</u> 1285 (1959)	591 ± 5	This measurement yielded $(1 + \alpha) = 1.171 \pm 0.009$ at 0.00291 ev $(\lambda = 5.3 \text{ Å})$ and was combined with a weighted value of σ_a obtained from $\sigma_t = 2147.6 \pm 5.7$ barns at 0.00291 ev as measured by the author together with a value of $\sigma_n = 15 \pm 2$ barns. The ratio of the fission cross section at 5 3 Å to 0.0253 ev was obtained from (a) Leonard's H.L.O. relative fission cross section data (unpublished), (b) curve in BNL-325 of $\sigma_f \sqrt{E}$ vs. E; (c) extrap- olation between authors' own relative measurements at 0 065 ev and 0.00464 ev were used to obtain σ_f at 0.0253 ev.

Source	$\sigma_{f}^{(barns)}$	Comments
J F. Raffle, Proc of Tripartite Conf. on Cross Sections of Fis- sile Nuclei, AERE, Harwell Report NP/R 2104 TNCC(UK)-9 (unpublished)	591 ± 18	Discussed by P A Egelstaff in Proc. of Tripartite Conf. on Cross Sections of Fissile Nuclei, AERE, Harwell Report NP/R 2104 TNCC(UK)-9 (unpublished). Data taken in pile beam This value will decrease when Raffle's new value of σ_f is used in this calculation.
J. F. Raffle, Proc. of Tripartite Conf. on Cross Sections of Fis- sile Nuclei, AERE, Harwell Report NP/R 2104 TNCC(UK)-9 (unpublished)	623 ± 20	Discussed by P. A. Egelstaff in Proc. of Tripartite Conf. on Cross Sections of Fissile Nuclei, AERE, Harwell Report NP/R 2104 TNCC(UK)-9 (unpublished). Data taken in thermal column at 30°C. This value will de- crease when Raffle's new value of σ_{f} is used in this calculation.
Bigham, Hanna, Tunnicliffe, Campion, Lounsbury and MacKenzie, Second UN Int. Conf. on the Peaceful Uses of Atomic Energy, A/Conf. 15/P/204.	569 ± 6	This value was revised from 575 ± 6 , reported at the International Conference on Neutron Interactions with the Nucleus, Columbia Uni- versity, September 9-13, 1957. The change of 1.3% was due to a reassessment of the flux depression in the gold foils used to meas- ure the thermal flux. The method consists of a determination of $\sigma_{\rm f}(U^{233})/\sigma_{\rm f}(U^{235})$ and an absolute determination of $\sigma_{\rm nf}(U^{233})$ The abso- lute value of $\sigma_{\rm nf}(U^{233})$ is based upon a value of $T_{1/2}(U^{233}) = 1 \ 611 \pm 0.008 \times 10^5$ years as meas- ured by the authors and the gold activation cross section of $\sigma_{\rm act}$ 2200 meters/sec = 98 8 \pm 0.3 barns, the value is $\sigma_{\rm nf}(U^{233})$ 20°C Max = 518 \pm 4 barns The value of the fission cross section ratio $\sigma_{\rm f}(U^{233})/\sigma_{\rm f}(U^{235})$ obtained is 0 9319 \pm 0 0013

Thermal Values of $\sigma_{\!f}$ Derived from Pile Measurements and Corrected to 0.0253 ev

Table IV shows the measurements of α at 0.0253 ev. The old Williams and Yuster measurement has been re-evaluated by Neuman and also by Bollinger. The most recent measurements do tend to lie below 0.180, but the errors are still fairly broad. The comment at the bottom of the table was based upon Palevsky's data on the direct measurement of η . The comment as stated is somewhat in doubt at the moment.

The existing measurements of $\boldsymbol{\sigma}_{f}$ (0.0235 ev) are in general of two types:

- (1) a measurement of $(1 + \alpha)$ combined with a determination of the absorption cross section; and
- (2) the direct measurement of the fission rate of a foil of a known composition and mass in a known flux.

TABLE IV

Values of α at 0.0253 ev

Source	Value	Comments
Williams and Yuster, Los Alamos Report LA-512 (1946) (unpublished)	α = 0.183± 0.006	This value is based upon a mass spectrometer determination of the amount of U^{236} created relative to the amount of U^{235} destroyed. Two difficulties were encountered (a) The correction from the neutron spectrum in which the irradiation was made. No Cd ratio measurements were made and the conditions of irradiation were vague. (b) The determination of the mass ratio, as the U^{236} content was very low and hence difficult to determine. This value was the basis of the U.S. "best value" of the σ_f (0.0253). These results have been recently re-evaluated to take into account the fact that the pile spectrum is non-maxwellian. The difference between the $(1 + \alpha)$ measured with the pile spectrum and the $(1 + \alpha)$ at 0.0253 ev has been determined from the curves by Westcott (C. H. Westcott, Atomic Energy of Canada, Ltd., Chalk River, Ontario, Report CRRP-680) and the results are not critically dependent on the shape of the pile spectrum. However, different assumptions about the correct factors can cause the value of α to vary as much as 2%.
Kanne, Stewart and White, Proc. Int. Conf. on the Peace- ful Uses of Atomic Energy, Vol. 4, 315 (1955)	α = 0.174 ± 20%	This value involved similar techniques to the above experiment. Thin samples of U^{235} having adequate isotopic impurities were exposed to the Hanford neu- tron flux filtered through appropriate shields The small amount of U^{236} formed less than 0.01% of the U^{235} in the sample and was measured using a two- stage spectrometer having a higher resolution of adjacent isotopes than previous machines
S. J. Cocking, AERE, Harwell Report TNCC(UK)-14 April 1957 (unpublished)	α = 0.172 ± 0.022	This value was obtained using a beam of cold neutrons obtained through the use of filters of Pb and Bi The filters allowed only neutrons of wavelengths greater than the 6.5 Å Bragg cut-off of the Bi cut-off to be transmitted The value of $(1 + \alpha)$ was obtained at 8 5 Å or (0.0011 ev) corresponding to the mean energy of the spectrum Palevsky et al show the value of $(1 + \alpha)$ decreases by about 2% from 0.1 ev to 0.02 ev. However, the decrease from 0.02 ev to 0.01 ev is not appreciable and the value obtained at 0 0253 ev by using Palevsky's relative value would be within the error of this value of $(1 + \alpha)$. The method used is essentially a comparison of the total cross section of the fissile isotope with that of gold together with a measurement of the fission rate in a neutron beam for which the flux was known in terms of the gold cross section The value of $(1 + \alpha)$
G. J. Safford and E. Melkonian, Phys. Rev. 113, 1285 (1959)	$\alpha = 0.171 \pm 0.009$	obtained is independent of the gold cross section. This value was obtained at 5 3 Å $(*)$ (For description of this experiment, see comment on Table III-a)
F. H. Tingey and F P Vance (MTR), TID 2019 P/405 (1955)	$\alpha = 0.175 \pm 0.025$	The value was obtained by correcting to 0.0235 ev a value of $\alpha = 0.184\pm 0.012$ for the MTR average core spectrum

^(*) H. Palevsky (Proc of Int. Conf. on the Peaceful Uses of Atomic Energy, U N, New York, 1956, Vol. 4, P/587) has determined that the values of α lie on a curve with slope (2.5 ± 2)% between 0 and 0.2 ev. This result shows that the values obtained at 8 5 Å and 5.3 Å, respectively, will not change significantly from the value extrapolated to 0.0253 ev.

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Some of the major experimental difficulties are:

- (a) The determination of thickness, composition and uniformity of the foils used, which are necessarily thin when fission fragments are counted. A recent uncertainty of this type resulted in a change of 8% in the U.K. value of σ_f at 2200 m/sec.
- (b) An accurate determination of both the absolute intensity and energy spectrum of the neutron flux. The intensity measurement generally involves a determination of the efficiency of the counter used in the flux measurement, or the activation measurement of a standard material such as gold that has been irradiated in the flux.
- (c) The older values of $1 + \alpha$ and σ_f , which were obtained from pile flux irradiations, suffered from uncertainties in the magnitude of the resonance flux, and from uncertainties in the energy dependence of α and the flux in the Maxwellian region. These uncertainties in some of the older measurements have caused errors of about 2 per cent in α in converting from the pile spectrum value to the 2200-m/sec value.

However, a recent method of measuring σ_{f} used by Bollinger, which counts the fission neutrons rather than fragments, seems capable of overcoming some of these difficulties as it allows thicker foils to be used.

It appears that measurements which have the following features would in principle be freer from systematic errors:

- (a) The use of monochromatic neutron sources. This eliminates the necessity for measuring and correcting for the shape of the pile spectrum in which the fission foil is irradiated, as well as the possible perturbation of the spectrum by the foil itself.
- (b) Flux and sample thickness determinations that do not require many separate steps tending to compound errors and uncertainties.

The relative fission cross section as a function of energy between 0 and 0.15 ev is shown in Fig. 4. The relative cross section has been normalized to a value of 580 barns at thermal energy. It will be noted that there is excellent agreement between the data of Leonard and Bollinger down to 0.02 ev. (Below 0.02 ev, the data of Bollinger indicate a pronounced rise but has large statistical errors, while the data of Leonard and Safford in this region, which have better statistical accuracy, show no such rise.) The solid line has been smoothed through the data with regard to the assigned errors.



 η

The measured values of η at 2200 m/sec are shown in Table V. It is seen that while the older measurements are consistent, the errors are rather large, being 2 - 3 per cent. Most direct measurements have been made using a pile-oscillator technique, or utilizing a control rod system designed to measure small changes in reactivity. However, values of η from pile-oscillator measurements depend upon a prior knowledge of the absorption cross section of the fissile nucleus for the reactor spectrum, as well as the fission neutron to thermal neutron importance, which is calculated from the lattice parameters of the reactor. Both of these quantities contain uncertainties which are reflected in η .

TABLE	v
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η (0 0253 ev)

Source	η	Comments
W H Zinn and H Kanner, Report CF-3651 (unpublished)	2 07 ± 0 05	
C O Muehlhause, Report ORNL-52-9-9 (unpublished)	208 ± 003	Reported value was modified to correct the σ absorption from 650 to 680 barns
D J Littler and E E Lockett (unpublished) cited in AERE, Harwell Report NP/R 2104 (1957) (unpublished) by P A Egelstaff	2 05 ± 0 05	Pile oscillator measurement
T M Snyder and R W Williams, Los Alamos Report LA-102 (1944) (unpublished)	216 ± 010	This value is based upon a σ absorption (0 0253 ev) = 644 barns and (1 + α) = 1 13 There also exists some doubt as to the "F" fac- tor used to obtain this value of η at (0 0253 ev) from the measured values using the pile spectrum
P E Spivak and B G Yerozolimsky,First U N Conf on the Peace- ful Uses of Atomic Energy, Geneva, 1955, P/657	2 065 ± 0 025	
A J Alichanov <u>et al</u> , First U N Conf on the Peaceful Uses of Atomic Energy, Geneva, 1955, P/658	212 ±005	
E Fermi <u>et al</u> (unpublished) 1944 Quoted in AERE, Harwell Report NRDC 84 by P A Egelstaff et al	2 10 ± 0 04	
H Rose, W A Cooper and R B Tattersall, (Harwell 1959), Private Communication	2 10 ± 0 05	
G de Saussure, J D Kington, R L Macklin, W S Lyon, Oak Ridge National Laboratory (Private communication) September 3, 1959	2 07 ± 0 02	A thermal beam of neutrons activates a man- ganous sulfate solution The beam then is totally absorbed in U^{235} and the fission neutrons activate the solution The ratio of the two activ- ities gives η except for small corrections This method appears capable of yielding η to an accu- racy of \pm 015

Note that all the measurements in this table are average thermal neutron spectrum values corrected to 2200 m/sec. This correction is generally <1% for U^{235} as η does not vary rapidly in the thermal region

The most recent measurement and that with the smallest quoted error is by de Saussure, Kingston, Macklin and Lyon, yielding

$$\eta = 2.07 \pm 0.02$$
 at 0.0253 ev.

This latter measurement utilizes a technique which is estimated to be capable of giving η to a precision of three-fourths percent or better. A thermal beam is utilized to activate a manganese bath. The beam then is totally absorbed in a sample of the isotope in question and the fission neutrons then activate the bath. The ratio of the two activities then yields η , after a number of small corrections of ~ 1 per cent were made. This measurement will be discussed in more detail in the paper by Dr. de Saussure.

In all of the above types of measurements it is necessary to correct the value of η measured over a particular neutron spectrum to a value at 2200 m/sec, which again requires an accurate knowledge of the spectrum.

The variation of η with energy is usually measured by the absorption of a monoenergetic neutron beam on a sample of fissile material black to the incident beam. The number of fission neutrons emitted is measured and the neutron beam is monitored by a $1/\nu$ detector.

Two corrections of about one per cent must be made to this type of measurement: one for neutron scattering, and one for multiplication in the sample. The variation of η with energy is shown in Fig. 5. The relative values of η were normalized to $\eta = 2.07 \pm 0.02$ at 2200 m/sec. The solid line has been smoothed through the direct measurements of η . The dashed line was obtained from the best lines through σ_f and σ_a as shown in the previous graphs and assuming ν constant. The η 's obtained from the crosssectional data of Leonard and of Bollinger are also shown. Both these η 's lie high below 0.02 ev in comparison with the dashed line. This can be attributed to Bollinger's rather high value of the fission cross section and to Leonard's rather low value of the absorption cross section in the region. In the region between 0.04 and 0.2 ev the cross-section data appear to lie lower than the direct η measurements, the latter showing a considerable scatter of data.

ν

The measured absolute values of ν at 0.0253 ev from several sources are shown in Table VI. While the older values of ν are consistent within the stated errors, the number of such measurements is small and the quoted errors are ~4 per cent. The three most recent values which also have the smallest quoted errors tend to be lower than the older values by more than 1 per cent. These values are:

Kenward and Sanders	ν	=	$2.420 \pm$	0.037
Colvin and M.G. Sowerby	ν	=	2.418±	0.039
Moat and McTaggart	ν	=	2.331 ±	0.053.



Fig. 5. Variation of η with Energy

TABLE VI

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 ν (0.0253 ev)

Source	ν	Comments
J. E. Sanders, J. Nucl. Energy <u>2</u> , 247 (1956)	2.45 ± 0.15	
J.E.Snyder and R.W. Williams, Los Alamos Report LA-102 (1944) (unpublished)	2.44 ± 0.12	This value was based on the absolute strength of a Ra-Be source Q (44). A re-evaluation was needed to correct for more recent recalibra- tions of this source.
V. I. Kalashnikova et al., Conf. Acad. Sci., U.S.S.R., On the Peaceful Uses of Atomic Energy, Proc. Phys. and Math. Sec. p. 156 (1955)	2.54 ± 0.10	
Kenward, Richmond and Sanders, AERE, Harwell Report R/R 2212 (revised) 1958 (unpublished)	2.420± 0.037	The number of prompt neutrons per fission $\equiv \nu p$ has been determined by the coincidence technique described in the text. The improved accuracy of this experiment is basically due to an improved calibration of the fission neutron de- tector using an accurately standardized Pu ²⁴⁰ spontaneous fission source. The Pu ²⁴⁰ source was calibrated using various photoneutron and Ra- α -Be sources as described by Richmond and Gardner, AERE, Harwell Report R/R 2097 (1957) (unpublished). A value of $\nu p = 2.405 \pm 0.037$ was obtained and was corrected for the delayed neu- tron fraction β according to $\nu = \nu p/(1 - \beta)$. β was taken as 0.0064 ± 0.0003 as given by Keepin, Wimett and Zeigler, Phys. Rev. 107, 1044 (1957).
D. W. Colvin and M. G. Sowerby, TNCC(UK), August 1959	2.418± 0.039	The fission neutrons were detected in a boron pile and counted in coincidence with the fission events. The improved accuracy of this experi- ment is a result of improved accuracy in deter- mination of the efficiency of the pile in neutron detection. Corrections were made for delayed neutrons.
A. Moat, M. H. McTaggart and D. S. Mather, Harwell, 1959, Report No. NR/P-1/59.	2.331 ± 0.053	This value was obtained from a measurement of (Cf^{252}) and ratio of the ν^{235} to $\nu(Cf^{252})$ at 75 kev. The resulting value of ν^{235} was then corrected to thermal energy.

It is questionable to consider the Kenward, Richmond, Sanders and the Colvin and Sowerby measurements as independent, since both used the Harwell spontaneous fission source in determining the efficiency of the fission neutron detector.

 ν is generally measured by placing a fission chamber in a neutron beam and measuring the coincidence rate between the fission events and the fission neutrons, as well as the fission singles rate. The prompt neutron yield per fission event is then simply proportional to the ratio of coincidence counts to fission events. To obtain an absolute value of ν , one must know the efficiency of the fission neutron detector, which is generally calibrated with a standard source. In the past, this has enabled relative ν measurements to be made to an accuracy of 1 per cent, while absolute values have been limited to an accuracy of 5 per cent. The smaller errors on the three most recent values reflect more precise standard neutron source calibration (in this case, the Harwell spontaneous fission source). The efficiency of the fission neutron detector used in the second of the above measurements was also measured for 265-kev neutrons by using Na²⁴ gamma rays to photodisintegrate deuterium and counting the coincidences between the photoprotons and the neutrons, and it was provisionally found that $\overline{\nu}$ is unlikely to be less than 2.40.

The above coincidence technique gives a value of ν for the prompt neutrons only, and a complicated correction of about 0.6 per cent must be made from other data for the effects of the delayed neutrons, and also a correction for fission events where a neutron is not emitted.

In conclusion, the set of average parameters shown in Table VII was obtained from weighted averages of the measured values in the above tables. In certain cases, older data or values having large errors have been omitted. The direct measurements of σ_f are simply listed to emphasize the discrepancies that are present, and that makes it difficult to obtain a meaningful weighted average.

The average of the direct measurements of η and ν yield a value of $1 + \alpha + 1.174 \pm 0.014$.

To obtain an average value of $1 + \alpha$ from the direct measurements, it is necessary to know the shape of $1 + \alpha$ in the thermal region. It is possible to extrapolate the values of $1 + \alpha$ of Safford and Cocking from 0.00291 ev and 0.001 ev, respectively, to 0.0253 ev. While $1 + \alpha$ is almost constant in this region, direct measurements of the variation of η and the shape of $1 + \alpha$ obtained from cross-section measurements do not rule out changes of the order of 1 per cent. Indeed, the measurements of Palevsky and recent cross-section measurements indicate $1 + \alpha$ decreases in going from 0.01 to 0.1 ev by about 1 per cent (as do recent cross-section measurements). If subthermal values of $1 + \alpha$ have been extrapolated to thermal

Average values of U ²³⁵ parameters from measured values listed in the previous tables (0.0253 ev)		Computed values (0.0253 ev)
$\overline{\sigma_{T}} = 694.3 \pm 1.3$ barns $\overline{\sigma_{n}} = 15 \pm 2$ barns		$\sigma_a = 679.3 \pm 3$ barns
$1 + \alpha$	$\frac{\Delta(1+\alpha)}{\Delta \mathbf{E}}$	$\frac{1}{1+\alpha}$ (0.0253 ev)
1.171 ± 0.009 at 0.00291 ev	± 0.05	
1.172 ± 0.022 at 8.5 Å		1.172 ± 0.012
1.175 ± 0.025 at 0.0253 ev		
$\frac{\eta}{\nu} = \frac{2.07 \pm 0.02}{2.43 \pm 0.017}$ at 0.0253 ev	,	1.174 ± 0.014
Avg. 1+	a (0.0253)	1.173 ± 0.009
	$\overline{\sigma}_{f} = \frac{\sigma_{a}}{1 + \alpha}$	$- = 579 \pm 5$ barns
	$^{\circ}\gamma = ^{\circ}a - ^{\circ}f$	$= 100 \pm 6$ barns
$\frac{\sigma_{f}}{\sigma_{f}}$		
Due variations for outside quoted measured values is given. Most r	errors no averag recent values are	ge value for σ_{f} from the

*	
$583 \pm 10 \text{ barns}$	Best U.K. value
569 ± 6	Canada
605 ± 6	A.N.L.
552 ± 6	Hanford

energy following the practice of Hughes and Westcott by assuming $1 + \alpha$ constant between 0 and 0.0253 ev with an error of $\Delta (1 + \alpha) / \Delta E = \pm 0.5 \text{ ev}^{-1}$, a value is obtained from the direct measurements of

 $1 + \alpha = 1.172 \pm 0.012$,

which, when averaged with the value of $1 + \alpha$ from η and ν , yields

$$1 + \alpha = 1.173 \pm 0.009$$
.

This with $\overline{\sigma_a} = 679.3$ barns leads to a value of $\sigma_f = 579 \pm 5$ barns. However, if the indicated slope

$$\frac{\Delta (1 + \alpha)}{\Delta E} = -0.57 \pm 0.23$$

is used, the above direct values of $1 + \alpha$ yield 1.161 ± 0.009 which, averaged with $1 + \alpha = 1.174 \pm 0.012$, yields $1 + \alpha = 1.165 \pm 0.007$ or $\sigma_f = 583 \pm 5$ barns, which is in good agreement with the recently revised U.K. value of 583 ± 10 barns.

TABLE VII

Discussion of Paper Presented by Mr. Safford

MR. BOLLINGER:

Are there some questions concerning this paper?

MR. RADKOWSKY:

Is there any knowledge of the variation of ν with energy?

MR. SAFFORD:

Yes, I believe, if I am correct, that Dr. Leonard of Hanford has measured this. I believe it is found to be constant within, if my memory is correct, to about 1 per cent throughout the thermal and subthermal region, although perhaps Dr. Leonard might want to comment on this.

MR. BOLLINGER:

For my benefit, would you mind reviewing what the steps are in getting at some of these average parameters, not in detail but just in general outline.

MR. SAFFORD:

Yes. The Tingey and Vance value of $1 + \alpha = 1.175$ was obtained from a measurement over the MTR core and then corrected to 2200 meters per second. The Cocking value, 1.172, was measured with neutrons at 8.5 angstroms, that is roughly 1 mv. The Safford-Melkonian value of 1.171 was measured at about 3 mv using a crystal spectrometer and monoenergetic neutrons.

The last two had to be extrapolated to thermal energy. We have tried to estimate the uncertainty in this slope, and this I do not claim to be too meaningful at all, on the basis of the Palevsky data, that is, the direct η measurement, and the dashed line in Fig. 2 based upon recent cross-section measurements which also shows a decrease in $1 + \alpha$ in the subthermal to thermal region. Using this estimate of the uncertainty in slope and correcting the Safford and Cocking value with it and averaging in the MTR value of Tingey and Vance, we get the first value: $1 + \alpha =$ 1.172. The best averaged value of $1 + \alpha$ obtained from η and ν measurements is 1.174. The two values are then averaged to give $1 + \alpha = 1.173$ $\frac{1}{2}$ 0.009. From the absorption cross section one then gets $\overline{\sigma}_{f} = 579 \pm 5$ barns.

STATUS OF THE LOW-ENERGY CROSS SECTIONS OF PLUTONIUM

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Dr. Okrent suggested that it might be of interest to include in this talk not only the review of neutron cross-section information on plutonium-239, but also that for the other major isotopes of plutonium. He also suggested that there was a recent article by Stoughton and Halperin in the August issue of Nuclear Science and Engineering which listed a set of recommended cross-section values, and that it might be of interest to comment upon these.

I must confess that I have enough difficulty deciding when I can on the values of mono-energetic cross sections; as regards reactor cross sections, I find it almost impossible to make a decision. If I do happen to make a seemingly irrelevant remark about Stoughton and Halperin's numbers, this is the source of it.

I will review the neutron cross-section information as it exists for the three major isotopes of plutonium in order to help to define the precision with which these quantities are known. The emphasis on the material that is presented here will deal with those aspects of the cross section where the largest uncertainties have existed in the past and to present some information which is either new or has not been previously widely circulated.

Pu²³⁹

The available low-energy neutron cross-section data for Pu^{239} has recently been reviewed at Hanford to evaluate the extent of agreement between the results of experiments that directly measured the variation of η with neutron energy and the variation of η deduced from the ratio of measured fission and total cross sections. The controversy which existed for some time over whether or not these results were in agreement was not well founded. The cross sections which were used to deduce the variation of η were not carefully analyzed to determine the precision of the resulting variation. Even worse, the results of total and fission cross-section data obtained with different instruments were used to obtain the ratio. It should be noted that the ratio of total (or fission) cross sections measured on different instruments with perhaps slight variations in energy scale and resolution might produce large variations in a narrow resonance. In short, the proper experiment to compare these η variations had not been done. In the last three or four years, however, both Bollinger's group at ANL and the group at Hanford have performed experiments designed to demonstrate the extent of agreement of the η variations.

The first step in the evaluation of the η variation with energy was to obtain a fitted curve to each of the total and fission cross-section variations. All of the known data on these quantities were assembled and subsequently adjusted in absolute value to agree best in shape. A primary basis which was used to weight the data to which a curve was to be fitted was to determine the precision with which each set of data could be fitted by an appropriate smooth curve. With this as an important aspect of the weighting, smooth curves were constructed to fit the absorption and fission cross-section data. These fitted curves were then forced to smoothness over the interval from zero neutron energy to about the half-maximum value of the 0.3-ev resonance by specifying a monotone increasing first derivative. Part of this region is shown in Fig. 1.



Fig. 1. Low-Energy Cross Sections for Pu²³⁹

A measure of the uncertainty of shape of the absorption cross section curve is the fact that three data sets (BNL, Han, Har) have dispersions of ± 0.7 to ± 0.9 per cent from the smooth curve from 0.03 to 0.12 ev. Below 0.03 ev the curve is determined entirely by the BNL data, which is the only set of data with a reasonably low dispersion. The low-energy fission curve is fitted to ± 1.0 per cent by the Hanford data (to 0.2 ev) and to ± 1.5 per cent by the ANL data (to 0.1 ev).

The problems previously discussed which arise in using the data from different instruments in the resonance were avoided by using the valuable data obtained by Bollinger <u>et al</u>. at ANL on the total and fission cross sections. These data obtained on the same instrument were analyzed to give a constant value of the ratio of fission to absorption cross sections from 0.2 ev to 0.35 ev with a RMS devi-

ation of ± 2 per cent. The resonance cross-section curves were then constructed to have the same shape and in general to fit the data with the least dispersion. These results are shown in Figs. 2 and 3. Although the relative shapes of the fission and absorption cross-section curves have been determined to a precision of about 2 per cent, it is probably apparent that the uncertainty in shape of each cross section in the resonance is probably twice that large. Different sets of data with relatively small dispersions differ systematically in shape through the resonance by several per cent. The curve which has been constructed is considered by the author to be the most probable shape, based on his evaluation of the probability of systematic effects in individual measurements.

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Fig. 2. Low-energy Fission Cross Sections for Pu²³⁹



Fig. 3. Low-energy Total Cross Sections for Pu^{239}

These fitted curves have been constructed for neutron energies up to 1 ev. It should be mentioned that the confidence of the fit becomes progressively worse and is only about ± 10 per cent for the absorption cross section above 0.5 ev because of the effects of scattering cross section and corrections for Pu²⁴⁰ impurity.



The ratio of the fitted cross-section curves is shown in Fig. 4.

Fig. 4. Ratio of Low-energy Cross Sections for Pu²³⁹

This curve has been compared with the seven reported experiments to measure directly the energy variation of η . Only one of the experiments, that was done at Brookhaven, shows a shape of the η variation which is statistically different from that predicted by the ratio of cross sections. The three other experiments which have the smallest dispersions from a smooth curve are compared with the ratio of cross sections on Fig. 5. The values obtained for the dispersion of the data are shown for each data set. These values are in good agreement with statistical expectations. The extent of the agreement of these data sets indicates that the <u>ratio</u> of cross-section variations is probably known to within ± 1 per cent over this energy interval. It is of interest to point out that the results of four separate experiments to determine the energy variation of ν have determined that ν is a constant over about the same energy interval to a precision of about ± 0.5 per cent.

Some comment must also be made on the absolute values of the cross sections. The fitted curve to the absorption cross section gives a value of 1008 barns for $\sigma_0(2200 \text{ m/s})$. A precision measure of ±20 barns would seem to cover the range of values given by the primary sets of data (ANL, Col, Han, and BNL). This value is somewhat lower than the value of 1030 ±40 barns





recommended by Stoughton and Halperin in their August Nuclear Science and Engineering article. I can add little to the absolute value of the fission cross section. A mono-energetic comparison with U^{235} done at Hanford based on the fitted cross section curve and the best value for U^{235} of 580 barns gives σ_0 of 717 ± 23 barns, while thermal neutron flux comparisons seem to give about 750 barns (Chalk River experiments).

Pu^{240}

The status of cross-section information for Pu²⁴⁰ has improved remarkably in the past couple of years. Three different measurements of the 1.06-ev resonance have been made; these are in quite good agreement. In addition, there have been two mono-energetic measurements of the total cross section in the thermal neutron energy range. These measurements agree quite well with the value of

 σ_0 expected from the 1-ev resonance parameters. Stoughton and Halperin list three thermal cross-section values obtained with pile neutrons, which also agree quite well with the mono-energetic results. The results for the 2200 m/s cross section are summarized in Table I.

The first line in Table I gives an evaluation of the 2200 -m/s absorption cross section deduced from the 1.056-ev resonance parameters. The second set of numbers are the results of mono-energetic measurements. The difference between these two values is somewhat larger than would be expected from the quoted errors. The last set of values are the results obtained from measurements with pile neutrons. The average value obtained from the three different types of measurements is (277 ± 7) barns, which agrees quite well with all but one single experimental value.

TABLE I

2200 m/s Absorption Cross Section of Pu^{240}

Deduced from Resonance Parameters

275 ± 20 Leonard, Seppi, and Friesen(a) [$\Gamma = (32.3 \pm 1.5)$ mv, $\sigma_{oa} = 1.87 \times 10^5$ barns]

Mono-energetic

290 ± 8	Block, Slaughter, and Harvey ^(b)
	\cdot ($\sigma_s = 2 \text{ barns}$)
271 ± 8	Pattenden and $Rainey(c)$
	$(\sigma_s = 2 \text{ barns})$

 $Avg = 280.5 \pm 9.5$

Pile Neutrons

270 ± 17	Westcott and $Walker(d)$
278 ± 15	Halperin, Oliver, and Pomerance (e)
274 ± 36	Cornish and Lounsbury $^{(f)}$

 $Avg = 274 \pm 10$

- (a) Nuclear Sci. and Eng. <u>5</u>, 32-35 (1959)
- (b) Bull. Am. Phys. Soc. II, <u>4</u> (1), 34 (1959), Private Communication
- (c) AERE NP/GEN 9 (1959)
- (d) To be published in Can. J. Phys., Private Communication
- (e) J. Inorg. and Nuclear Chem., 9 (1), 1 (1959)
- (f) CRC-633 (1956)

Pu^{241}

Two years ago at Hanford we measured the low-energy fission cross section of Pu^{241} through the first resonance with good statistical precision. The plutonium sample available at that time had about 19 per cent Pu^{241} and 6 per cent Pu^{239} . We have recently repeated most of these measurements using plutonium with a 96.6 per cent Pu^{241} content. Figure 6 shows the results



obtained with both sets of data. The two experiments are seen to be in excellent agreement, except near 0.35 ev. The deviations here were determined to be due probably to foil nonuniformities. The solid curve is a single-level formula fit to the high-energy side and peak of the resonance at about 0.255 ev. This resonance curve illustrates the difficulties involved in attempting to fit the data to a sum of single-level formulae. The excess theoretical cross section on the high-energy side of the resonance makes it virtually impossible to obtain single-level parameters which will fit the data to better than about 5 per cent.

The total cross section of Pu^{241} was measured by Schwartz <u>et al</u>. at Brookhaven. The isotopic purity of the sample used was not very good, containing some 11.6 per cent Pu^{239} and 23.7 per cent Pu^{240} . In addition, Am^{241} is a troublesome correction in total cross-section data. Much better samples of Pu^{241} are rapidly becoming available and better total crosssection information will be obtained soon. Nevertheless, these are the only total cross-section data known at present and, as will be seen, they are quite good in many respects.

The fission cross-section measurements have been compared with the total (minus scattering) cross section in Fig. 7. A smooth curve through the fission data was used in this comparison, which shows then the predicted variation in η . Although the dispersion of the low-energy data is several per cent, there seems to be a definite increase in η in the 0.255-ev resonance of about 18 per cent over the thermal value.



Fig. 7. Ratio of Low-energy Cross Sections for Pu²⁴¹

The fission cross-section measurements have also been extended into the resonance region. These results are shown in Fig. 8, which also includes the total cross-section data in this region. The resolution which



Fig. 8. Fission and Total Cross Sections for Pu²⁴¹

was used in the fission measurements was not good enough to determine the resonance parameters from the shape of the resonances. However, all of the resonances seen below 9 ev in the total cross section are reproduced in these fission cross-section measurements, including the doubtful resonance at 5.35 ev. The parameters of these fission resonances have been determined by area analysis. These results are shown in Table II compared with the total cross section parameters. There seems to be some systematic difference in the manner in which the total and

fission cross-section resonances have been analyzed, which result in values of alpha which are obviously incorrect. I have, therefore, also listed the maximum value of alpha as obtained from the observed fission and total cross-section peak cross sections.

TABLE II

$E_0(ev)$	σ ₀ Γ(b-ev) (BNL-325)	σ ₀ Γ _f (b-ev)	$\begin{pmatrix} \alpha \\ \hline \sigma_0 \Gamma \\ \hline \sigma_0 \Gamma_+ \end{pmatrix}$	amax
4.32	273	132	1.06	0.58
4.65	80	44	0.82	0.41
5.35	17	17	0	0.10
6.03	540	290	0.86	0.17
6.94	147	194	0	0.06
8.61	180	150	0.20	2.0

Pu²⁴¹ Resonance Parameters

The absolute fission cross-section value of Pu^{241} is in much the same status as that of Pu^{239} . We have made mono-energetic measurements of the ratio of the Pu^{241} fission cross section to that of U^{235} . The older measurement using the 19 per cent Pu^{241} sample gave a 2200-m/sec value of (941 ± 46) barns when based on the U^{235} value of (590.8 ± 5.4) barns recently recommended by Safford, Havens, and Rustad. Somewhat better measurements made with the 96.6 per cent Pu^{241} sample, for which the Pu^{239} correction was less than one per cent, gave a value of (956 ± 43) barns. This value is inconsistent with the value of (1013 ± 8) barns obtained by Bigham <u>et al</u>. with thermal neutrons, since Bigham's measurements also gave a U^{235} value of 569 barns.

Pu^{242}

The Pu²⁴² cross sections have been measured by the Argonne group and are fully presented in an April 15, 1959 Physical Review article. There is no need to summarize that excellent presentation here.

Discussion of Paper Presented by Mr. Leonard

MR. BOLLINGER:

Are there some questions concerning this paper?

MR. RADKOWSKY:

According to your curves, there are apparently no parasitic absorptions in the thermal region for Pu^{241} .

MR. LEONARD:

The curve was normalized to unity just to show the deviation from a constant. It depends on the cross-section values that you take. I think the Chalk River value for the fission cross section is about 1000 barns and the Brookhaven total cross section is about 1400 barns.

MR. BOLLINGER:

I have a question on a related subject. As I remember, the total cross-section measurements on Pu^{241} were plagued by the presence of large impurities of Pu^{239} and Am^{241} . I wonder if this energy dependence of eta of Pu^{241} in the region of 0.250 ev is real or whether it might not be caused by an error in the total cross-section measurements because of this difficulty.

MR. LEONARD:

Well, it certainly could be the cause of the difficulty in the total cross-section measurements, and I think there will be new and much better total cross-section information very soon. I showed this result primarily because I have never seen it listed in the literature anywhere, and I was very surprised to find out that here we have a thermally fissionable isotope of which the indication was that η increased in the resonance. It is the first case in history that I know of.

MR. BOLLINGER:

Are there other questions?

MR. RADKOWSKY:

How about the value of ν for Pu²⁴¹?

MR. LEONARD:

I don't know.

MR. BOLLINGER:

There is probably someone here that does.

MR. JAFFEY:

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We have made some comparative measurements. The ν of Pu²⁴¹ is only slightly larger than that of Pu²³⁹, whatever that is.

EVALUATION OF LOW-ENERGY CROSS-SECTION DATA FOR U^{233}

J. E. Evans and R. G. Fluharty Phillips Petroleum Company Idaho Falls, Idaho (Paper presented by Mr. Evans)

The text of a more detailed paper is being submitted to Nuclear Science and Engineering for publication and I hope you can find the details there. We are also publishing it as an IDO report, number 16554.

I will break this paper up into three parts. The first part will deal with the philosophy we have used in attacking this problem. Second, I will discuss the cross sections as a function of energy in the Maxwellian region and the determination of 2200-m/sec values. In the third part I will discuss the data on η , ν and α for a Maxwellian distribution.

Introduction

Critical evaluations of low-energy cross-section data for the fissile nuclei have been requested by the USAEC Nuclear Cross Section Advisory Group (NCSAG). About a year ago at a joint session of the NCSAG and the USAEC Advisory Committee on Reactor Physics, it became evident that problems existed not only in measurements of the absolute values of two of the basic parameters for reactors, i.e., the fission cross sections and eta values, but also in the communication to the reactor people of estimates of the validity of the assumptions used in obtaining the pertinent values, which are widely separated. As a first step in clarifying the situation, it was decided that the experimental measurements giving information on the cross-section parameters pertinent to thermal reactors (in the energy region below 1 ev) should be critically reviewed by some person or group with experience in making such measurements. Dr. R. G. Fluharty and I were asked to evaluate the existing measurements on U^{233} . Any evaluation of this nature must necessarily be progress report, since further study will bring additional measurements to light and will increase understanding of the measurements now being used. Also, new measurements will necessitate revisions. We hope that experimenters will write us when they feel that the use made (or not made) of their measurements in this evaluation could be more realistic.

One of the difficult problems in such an evaluation is to reduce personal bias insofar as it is possible to do so. The best way to remove personal bias is probably by a least-squares analysis as used for the fundamental physical constants as outlined by Cohen, Crowe and Dumond.(1) Although least-squares-weighted averages have been used in this report for the individual parameters, a careful study of the proper evaluation of the weights of related measurements has not been made. Such a study should be undertaken. However, the amount of study and correspondence required to learn the details of each measurement makes it an undertaking of considerable magnitude. Another deterrent to such an approach is the long delay between the time a new experimental result is obtained and is published. The result must then be understood in detail by an experienced evaluator, and the evaluation carried out with respect to all other pertinent measurements.

Since many precise ratio measurements of the fission cross sections, η , and $\overline{\nu}$ for U^{233} and U^{235} exist, U^{235} measurements serve as a valuable source of information for U^{233} values. The June, 1959 recommended values of Safford and Havens⁽²⁾ for U^{235} have been used mainly to show the consistency of U^{233} and U^{235} data, but in a few cases they have been used as input data for obtaining U^{233} results through the use of these ratios. The final values obtained through this process are not independent results, and when nonindependent results are being combined, complex weighting techniques should be applied. However, throughout this report the data are treated as if they are independent.

In the preliminary report submitted to the NCSAG in June, 1959, we derived a set of most probable values for U²³⁵ independently, since the final U²³⁵ evaluation was not available at that time and since there are more measurements, made with a wider range of experimental techniques, available for study of U^{235} than for U^{233} . The authors' 2200-m/sec values for U^{235} are shown with the June, 1959 values of Safford and Havens in Table I. The errors are expressed as standard deviations throughout this report. Several unpublished results were used in obtaining these values, which are in good agreement with those of Havens and Safford. However, it seems of interest to mention a major difference in approach. No direct fission crosssection measurement has been accepted as input data in our $U^{235} \mbox{ or } U^{233}$ evaluations. The authors believe that the most effective approach at the present time in arriving at accurate 2200-m/sec fission cross section values for U^{233} and U^{235} is to derive them from total and scattering cross-section and alpha (capture to fission ratio) values. The variations in the values of fission cross sections obtained by experienced investigators from direct measurements have demonstrated that the reliability of absolute fission counting is questionable. The primary result of not including the direct fission cross-section results in the evaluation of U^{235} data is to lower the weighted standard deviations in σ_{nf} , α and σ_{c} .

A network has been set up with effective values for a Maxwellian spectrum for η , α and $\overline{\nu}$ values and their ratios for U²³³ and U²³⁵, since most existing measurements have been made in reactor spectra. Then the 2200-m/sec value for each constant can be obtained by one conversion factor with its associated error. The validity of this procedure involves the assumption that many fine points of spectral and temperature differences can be neglected. Improvements in precision and confidence limits will require careful appraisal of these factors. The reason that these factors

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TABLE I

	Safford, Havens	Evans, Fluharty
$ \begin{array}{c} \sigma_{nT} \\ \sigma_{nn} \\ \sigma_{nX} \\ \alpha \\ \sigma_{nf} \\ \sigma_{c} \\ \eta \\ \overline{\nu} \end{array} $	$697 \pm 7b$ $16 \pm 2b$ $681 \pm 7b$ 0.178 ± 0.01 $580 \pm 12b$ $102 \pm 14b$ 2.07 ± 0.02 2.44 ± 0.03	694 ± 4b 15 ± 3b 679 ± 5b 0.172 ± 0.012 579 ± 7b 100 ± 9b

 U^{235} 2200 m/sec Values - June, 1959

are neglected here is based on the additional assumption that the uncertainty in the energy dependence of the cross sections is a determining factor in the errors involved in converting from thermal reactor spectra to 2200-m/sec values.

In general, we have limited our consideration to published measurements. The exception is in the case of total cross-section measurements, where the main errors are the purity and uniformity of the metal samples, and where the concentrated efforts of large laboratories in the US have produced major advances recently. Since this is a key measurement for much of our present study, two carefully made, but thus far unpublished, recent measurements (3,4) have been used.

Low-Energy Cross-Section Data in the Maxwellian Region

Two recent σ_{nT} measurements of U^{233} have been made, the first shown in Fig. 1, by O. D. Simpson, M. S. Moore and F. B. Simpson⁽³⁾ on the MTR fast chopper and the second by J. A. Harvey and R. C. Block⁽⁴⁾ on the ORNL fast chopper. The MTR 2200-m/sec value, obtained by a straight-line least squares fit to the data from 0.02 to 0.05 ev, is 587 ± 6 barns.

Note the U^{233} total cross section is 1/v, as shown by the zero slope on a $\sigma E^{1/2}$ plot. It continues to be 1/v up to about 0.12 ev, where the effect of the small resonance at 0.165 ev becomes noticeable. The ORNL 2200-m/sec value for U^{233} is 586 ± 3 barns.

The U^{235} data are the data from which the MTR value of 690 ± 10 barns by Simpson, Moore and Simpson⁽³⁾ was obtained, as quoted by Safford and Havens. The U^{235} data are presented in Fig. 1 because it is part of a paper that has already been submitted to Nuclear Science and Engineering for publication on the low-energy cross sections.

The Au data are taken as a standard along with each of the runs on U^{235} and U^{233} . We use this to keep constant check on the various things that can go wrong with thousands of vacuum tubes.

The principal uncertainties in both measurements are in sample thickness and uniformity, and more effort should be applied toward obtaining better samples for additional σ_{nT} measurements. The assigned 2200-m/sec value for U²³³ is <u>586 ± 3 barns</u>. All errors quoted in this report are standard deviations.

Oleksa⁽⁵⁾ has measured σ_{nn} for U^{233} at several energies, and theoretical fits to these data have been made by Moore and Reich⁽⁶⁾ and by Vogt.⁽⁷⁾ The data and a recent fit by Moore and Reich with multilevel parameters are shown in Fig. 2. Since possible coherence effects cannot be calculated and fairly large errors exist in scattering measurements, the error is estimated to be ± 3 barns. The assigned 2200-m/sec value for σ_{nn} is $\underline{13 \pm 3}$ barns.



Fig. 2. U²³³ Scattering Cross Section



Fig. 1. Total Low-Energy Total Cross Sections for U²³⁵, U²³³, and Au¹⁹⁷.

Figure 3 is a composite data plot as a function of energy showing the absorption cross section of U²³³ from various laboratories as reported by Hughes and Schwartz⁽⁸⁾ plus MTR</sup> data reported by Simpson, Moore and Simpson. (3)These low-energy MTR data are in excellent agreement with those of Harvey and $Block^{(4)}$ at ORNL, who have a preliminary curve of comparable statistics. The MTR curves should supersede



the older data, until the Harvey and Block results can be included. The 2200-m/sec σ_{nX} (absorption cross section) value resulting from this approach is 573 ± 4 barns. It is to be noted that this value is considerably lower than the value of 581 ± 7 (World Consistent Set) given in BNL-325(8) and the value of 585 ± 10 given by Hughes in his 1958 Geneva Conference Paper.(9)

As was stated above, we believe the most accurate value of σ_{nf} can be obtained at present by dividing σ_{nX} by $1 + \alpha$. In assigning a value for alpha, two distinct factors are involved here which are often not included in discussions or reactor constants. In the first place, it appears that the values of alpha obtained from reactor irradiations have not been corrected for the resonance-flux contributions. This correction tends to lower the value for U²³³ and also for U²³⁵ by the order of a few per cent. The other effect which is believed to be important is that of the necessity of relaxing errors on 2200-m/sec values for alpha derived from pile flux irradiations. These increases in error for the 2200-m/sec values are required to allow for the large uncertainties in the magnitude of the resonance flux under high-power reactor operating conditions and for the uncertainties in the neutron spectral shapes and the energy dependence of alpha in the Maxwellian region. Table II gives an outline of the derivation of the 2200-m/sec σ_{nf} value.

TABLE II

C	U	t r235	
Source	Inghram, <u>et</u> al	Kukavadse, <u>et al</u>	0
α (Reported) α (Maxwellian) α (2200 m/sec) α (Wt av)	0.0976 ± 0.0018 0.093 ± 0.0036 0.093 ± 0.0053 0.0935 ±	$\begin{array}{r} 0.0953 \pm 0.0034 \\ 0.094 \pm 0.0044 \\ 0.094 \pm 0.0055 \\ 0.0038 \end{array}$	
σ_{a} (2200 m/sec)	573	4 b	
σ_f (Maxwellian)	527	$\begin{array}{c} \pm 14.5 \text{ b} \\ + 14.5 \text{ b} \\ + \\ 0 \text{ b} \rightarrow 0.9319 \pm 0.0013 \rightarrow \end{array}$	580 ± 12 b *
$\sigma_{\rm f}$ 2200 m/sec wt av	- 524	±4b	

Derivation of U^{233} σ_{nf} 2200 m/sec Value

*Error increased 1.81% total for the two steps.

A direct measurement of alpha for U^{233} has been made by Inghram <u>et al.</u> (10) by measuring the ratio of U^{234} produced to the loss of U^{233} from a sample irradiated in the NRX reactor at Chalk River. The value reported is 0.0976 ± 0.0018. This yields a 2200-m/sec value of $\alpha = 0.093 \pm 0.0053$. A second measurement which has been used is that of Kukavadse <u>et al.</u> (11) who report a value of $\alpha/(1+\alpha)$ of 0.087 ± 0.003 for a reactor spectrum. After estimating various corrections (see IDO-16554), the 2200-m/sec value for α is 0.094 ± 0.0055. Combining these results, the recommended value of α for 2200-m/sec neutrons is 0.0935 ± 0.0038.

Two independent sources of information are now available to determine the U^{233} fission cross section at 0.0253 ev. The first value can be derived from the absorption cross section and $1 \pm \alpha$ values given above, yielding the value of 524 ± 4 barns. The second value is derived from the average for U^{235} fission cross section of 580 ± 12 barns arrived at by Safford and Havens⁽²⁾ by multiplying by the U^{233}/U^{235} fission cross-section ratio value of 0.9319 ± 0.0013 determined by Bigham et al.⁽¹²⁾ In using this ratio the 2200-m/sec U²³⁵ value is converted to a 20°C Maxwellian value before multiplication and then converted from the U²³³ Maxwellian value to a 2200-m/sec value. This procedure yields a value of 527 ± 14.5 barns. The weighted average of the two values is 524 ± 4 barns, and this is the recommended 2200-m/sec value for the fission cross section for U²³³.

Bigham et al.⁽¹²⁾ have made a direct measurement by fission counting and report a value of 518 ± 4 barns for the 2200-m/sec value. Although the agreement with our derived value is fair, the result has not been included in the average. This decision is based upon the general disagreement among direct fission measurements. Actually some effect of this direct measurement is included in our treatment, since a value for the fission cross section of U²³⁵ derived from the U²³³ fission measurement and the U²³³/U²³⁵ fission ratio is included in the average of Safford and Havens.⁽²⁾

I would like to emphasize some of our reasons for not using direct fission measurements; for, in the words of R. R. Wilson with reference to authorship on papers, [An Introduction to Scientific Research, McGraw Hill, p. 358 (1952)] "Here is a golden opportunity for losing friends." A somewhat analogous situation has been faced by Cohen, Crowe and Dumond⁽¹⁾ (pp. 260-263) in adjustment of fundamental atomic constants of physics, where they say "The determination of the short wavelength limit of the continuous X-ray spectrum are all in disagreement with the indirect value. Hence, we may tend to doubt the direct data. There are certain valid experimental reasons to believe that we should; these have been thoroughly discussed in Section 6.8." We are not criticizing the experimenters who have published direct fission measurements. In fact, we have the highest regard for their abilities as experimentalists and for the many ingenious techniques they have developed and used. I hope they will continue to try to resolve the discrepancies, and that other experimenters will participate. However, realizing the difficulties that have arisen in these complex measurements, we feel that in the evaluation the 2200-m/sec fission cross-section direct fission measurements cannot be used until experienced workers in different laboratories can agree within their experimental errors with fair consistency.

The derivation of capture cross section is shown in Table III. The first method is more direct and yields a much smaller standard deviation than does the third method which is often used.

TABLE III

Derivation of U²³³ Capture Cross Section

$$\sigma_{c} = \frac{\sigma_{c}}{\sigma_{c} + \sigma_{f}} \sigma_{abs} = \frac{\alpha}{1 + \alpha} \sigma_{abs}$$
$$= \frac{0.0935 \pm 0.0038}{1.0935 \pm 0.0038} (573 \pm 4 b) = 49 \pm 2 b$$
$$\sigma_{c} = \alpha \sigma_{f} = (0.0935 \pm 0.0038) (524 \pm 4 b)$$
$$= 49 \pm 2.1 b$$
$$\sigma_{c} = \sigma_{a} - \sigma_{f} = (573 \pm 4 b) - (524 \pm 4 b)$$
$$= 49 \pm 6 b$$

The measured values of η for Maxwellian spectra at 20°C are given in Table IV. The weighted mean of the four values used in 2.284 ± 0.015. The fourth significant figure is retained to prevent the introduction of sizeable errors by the "rounding off" process when this value is used to derive values for other constants. The values are listed in the order of their magnitudes. The weights for the individual measurements in the average are given in the last column. Details of the evaluation of the measurements listed in this and the following tables are to be found in IDO-16554.

The measured values for the ratio η_{23}/η_{25} for Maxwellian spectra are given in Table V. The weighted mean of the five values used is 1.105 ± 0.006. A disturbing point here is that the two values with the greatest weight are over a standard deviation of the average apart.

The experimental values for α_M for U^{233} for a Maxwellian spectrum are listed in Table VI and details of the treatment for the experimental data are given in IDO-16554. The weighted mean of the two accepted values

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TABLE IV

	x _i ± σ _i	x_i/σ_i^2
Muehlhause '59 Spivak, Yerozolimsky '56 Macklin, deSaussure '59 Magnuson, Gwin, '59 Alichanov, et al. '56	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	2522 2533 5725 787
Least squares wt av	2.284 ± 0.015	11567

U²³³ Absolute Eta - Maxwellian Spectra

*Not used in average.

TABLE V

η_{23}/η_{25} -	•	Maxwellian	Spectra
-------------------------	---	------------	---------

	$x_i \pm \sigma_i$	x_i / σ_i^2
Rose, <u>et al.</u> , Dimple '59 Gaerttner, <u>et al.</u> '58 Rose, <u>et al.</u> , Gleep '59 Muehlhause '59 Spivak, Yerozolimsky '56 Magnuson, Gwin '59 Richmond '56 Thomas, <u>et al.</u> '56 Alichanov, et al. '56 Zinn, Kanner '52	$1.057 \pm 0.015*$ $1.078 \pm 0.019*$ $1.091 \pm 0.015*$ 1.097 ± 0.023 1.104 ± 0.008 1.105 ± 0.025 1.108 ± 0.020 1.111 ± 0.015 $1.113 \pm 0.022*$ $1.114 \pm 0.010*$	2074 17250 1768 2770 4938
Least squares wt av	1.105 ± 0.0062	28799

*Not used in average.

TABLE VI

U²³³ Alpha - Maxwellian Spectra

	x _i ±σ _i	x_i / σ_i^2
Inghram, <u>et al</u> . '56 Kukavadse, <u>et al</u> . '56 Cocking '58	0.093 ±0.0036 0.094 ±0.0044 0.113 ±0.018*	7170 4855
Least squares wt av.	0.0934 ± 0.0028	12025

*Not used in average.

for a Maxwellian spectrum is 0.0934 ± 0.0028 , Another value has been derived and is given for comparison purposes. From the Safford and Havens' recommended value of $1 + \alpha$ for U^{235} , 1.178 ± 0.010 , with its error increased by 1.78% in $1 + \alpha$ in converting the 2200-m/sec value to a Maxwellian value, and the ratio

$$\frac{(1+\alpha)_{23}}{(1+\alpha)_{25}} = \frac{\overline{\nu}_{23}}{\overline{\nu}_{25}} \frac{\eta_{25}}{\eta_{23}} = 0.9312 \pm 0.0075$$

from Table IX, a Maxwellian value of 0.097 ± 0.024 is obtained for U^{233} . This derived value is consistent but of low precision.

The measured values for the ratio $\overline{\nu}_{23}/\overline{\nu}_{25}$ are given in Table VII and the weighted mean value of the four accepted values for a Maxwellian spectrum at 20°C is 1.029 ± 0.005. Since the value of 1.043 ± 0.022 by Diven et al.(14) was not measured with a Maxwellian spectrum but was measured with 80-kev neutrons and corrected to thermal energies, it has been used as a consistency check only. If included, the average would be raised to 1.031 ± 0.005.

TABLE VII

 $ar{
u_{23}}/ar{
u_{25}}$ - Maxwellian Spectra Including Delayed Neutrons

	x _i ± ^σ i	x_i/σ_i^2
Sanders '56 McMillan, <u>et al.</u> '55 deSaussure, Silver '59 Kalashnikova, <u>et al.</u> '55 Colvin, Sowerby '58 Anderson May '52	$1.001 \pm 0.016*$ 1.017 ± 0.033 1.020 ± 0.01 1.031 ± 0.010 1.034 ± 0.008 $1.079 \pm 0.02*$	934 10200 10310 16156
Least squares wt av	1.029 ± 0.0053	37600

*Not used in average.

The two measured values for $\overline{\nu}_{\rm M}$ for U²³³ listed in Table VIII are not precise primarily because accurately calibrated sources with appropriate neutron spectra have not been available for calibrating the fast neutron detectors. More precise values can be derived from $\eta_{\rm M}$ and $1 + \alpha_{\rm M}$ for U²³³ and from $\overline{\nu}_{25}$ and $\overline{\nu}_{23}/\nu_{25}$. The weighted mean for these two values is 2.501 ± 0.019.

The values of η , $1 + \alpha$, and $\overline{\nu}$ and their ratios for U^{233} for Maxwellian spectra are summarized in Table IX. The significance of the last figure in most of the values is doubtful, but they were retained to prevent

TABLE VIII

	$x_i \pm \sigma_i$	x_i/σ_i^2
Sanders '56	2.47 ± 0.15*	(110)
$\eta = 2.284 \pm 0.015, \\ \alpha = 0.0934 \pm 0.0028 $	2.497 ± 0.022	5159
$\overline{\nu}_{25} = 2.44 \pm 0.03,$ $\overline{\nu}_{22}/\overline{\nu}_{25} = 1.029 \pm 0.006$	2.511 ± 0.035	2050
Kalashnikova, <u>et al</u> . '55	2.64 ± 0.11*	
Least squares wt av	2.501 ± 0.019	7209

U²³³ Absolute Nu - Maxwellian Spectra

*Not used in average.

the introduction of calculational errors. The values for U^{235} predicted from the U^{233} values, with their errors expanded to allow for the transformation to 2200-m/sec values, are quite close to the June, 1959 recommended values of Safford and Havens listed in Table I.

TABLE IX

Consistent Network of U²³³ and U²³⁵ Parameters for Maxwellian Spectra

Based on η and $1 + \alpha$ values for U^{233} and ratio measurements. The input data are marked with an asterisk *. The values for η_{23} and η_{23}/η_{25} are not completely independent data.

	U ²³³	Ratio	U ²³⁵
η	$2.284 \pm 0.015* -$	- 1.105 ± 0.006* -	► 2.067 ± 0.018
1 + α	1.0934 ± 0.0028*-	- 0.9312 ± 0.0075 -	► 1.174 ± 0.010
$\overline{ u}$	2.497 ± 0.022 —	- 1.029 ± 0.006* -	► 2.427 ± 0.026

The details of obtaining $\pm 0.35\%$ as the uncertainty of transforming $\eta_{\rm M}$ to a 2200-m/sec value of 2.28 \pm 0.02 can be found in IDO-16554. The energy dependence of η is shown in Fig. 4. These direct measurements of η are very difficult measurements. Many fairly large corrections are required. The multilevel parameter fit obtained from MTR $\sigma_{\rm T}$ and $\sigma_{\rm nf}$ measurements agree reasonably well with the direct η measurements.


As mentioned earlier, we have spent an appreciable effort in trying to develop simple methods of correcting α measurements made in pile spectra to α_{M} and α for 2200-m/sec neutrons. The derivations can be found in IDO-16554, but the following formula shows the result:

$$\alpha_{\mathbf{M}} = \frac{\mathbf{r}}{\mathbf{r}-1} \begin{bmatrix} \mathbf{R} \\ \mathbf{R}-1 \\ \mathbf{\alpha}_{\mathbf{pile}} \end{bmatrix} - \frac{1}{\mathbf{R}-1} \\ \alpha_{\mathbf{res}} \end{bmatrix} - \frac{1}{\mathbf{r}-1} \\ \alpha_{\mathbf{5kT-0.4}}$$

where

$$R-l \equiv \frac{\varphi_{0fth}}{\theta \int_{0.4ev}^{\infty} \sigma_{f}(E) dE/E}$$

 and

$$\mathbf{r} - \mathbf{l} \equiv \frac{\theta \mathbf{M}^{\sigma} \mathbf{f} \mathbf{M}}{\theta \int_{\mathbf{5kT}}^{\mathbf{0} \cdot \mathbf{4} \cdot \mathbf{ev}} \sigma_{\mathbf{f}}(\mathbf{E}) \, \mathrm{d} \mathbf{E} / \mathbf{E}}$$

If only a 2-region consideration is adequate, r is large and only the portion of the expression in brackets need be used. The 3-region expression will be most needed for U^{235} , Pu^{239} and Pu^{241} which have resonances in the 0.3-ev region. It is important for these considerations that reactor spectra be specified in terms of Cd ratios for fission in thin fissile samples.

We have also derived an approximate method for obtaining a 2200-m/sec value of α from α_M and the approximate energy dependence of α through the Maxwellian region:

$$\alpha_{2200} = \alpha_{M} - (1-x) E_0 (\Delta \alpha / \Delta E)$$

where E_0 is the most probable neutron energy in the Maxwellian, x is the power designating the energy dependence of $\sigma_{nf} = k/E^x$, and $\Delta \alpha / \Delta E$ is the slope of the α vs. E curve. The method used can easily be extended to allow α to be described in terms of a polynomial in E.

In the thermal region $\overline{\nu}_{23}$ is believed to be effectively constant, even though this conclusion is supported primarily by circumstantial evidence. Measurements show that $\overline{\nu}(E)$ for U^{235} and Pu^{239} varies less than 1% through the thermal region (16) and η for U²³³ is constant to within 1% up to 0.1 ev.(17) Since it has been shown that mass yields of fission fragments change from resonance to resonance (18) and that the number of neutrons per fragment varies with the fragment mass ratio, (19) small changes in $\overline{\nu}$ would also be expected. In recent work by Regier, Tromp and Burgus at MTR the peak to valley ratio of Pu²³⁹ fission changes by a factor of 3 in going from thermal neutrons to 0.3-ev neutrons. In the case of U^{233} the variation in mass yields is quite small from resonance to resonance. In all cases the $\overline{\nu}$ values measured have been constant within the experimental errors, in some cases < 1%. The changes would be expected to take place between resonances, and the variation through regions narrow compared to width of resonances, such as the Maxwellian region, would be expected to be a small fraction of the total variation. Since any such small changes would be small compared to other errors in $\overline{\nu}$ values, $\overline{\nu}$ is considered to be constant throughout the thermal region. Therefore, the recommended 2200-m/sec value of $\overline{\nu}$ is 2.50 ± 0.02.

Conclusions

The recommended 2200-m/sec values for the U²³³ cross sections and fission constants are shown in Table X. The mean values arrived at present a consistent picture with the accuracies of all important constants within 1%. The modifications to values from recent summaries(8,9) is not large and are mainly due to the lower absorption cross section from the new measurements used in this report. The effect of lowering the value of the absorption cross section is somewhat compensated by a lower value of α , so the fission cross section and η values are essentially unchanged. A net result is a lower value of $\overline{\nu}$, which tends to be in agreement with the recent more accurate measurements on $\overline{\nu}$ for U²³⁵.(20)

TABLE X

Recommended U²³³ 2200-m/sec Values

σ _{nT}	586 ± 3 b	Onf	524 ± 4 b
σ _{nn}	13 ± 3 b	σ	49 ± 2 b
σ _n χ	573 ± 4 b	η	2.28 ± 0.02
1 + α	$1.093_5 \pm 0.004$	ν	2.50 ± 0.02

At the time of the 1958 Geneva Conference fairly serious doubt as to the correctness of η for U²³³ was raised by the British. And on a general basis considerable doubt existed as to the validity of reactor theory, since most η results were based upon reactivity, criticality, or reactor types of measurements. When these fears were coupled with the lack of agreement between various fission cross-section measurements, it appeared that the basic reactor constants were much more uncertain than suspected and that U^{233} was much less promising as a possible thermal breeding cycle material than formerly believed.

The results obtained here tend to support the values which have been accepted as correct in the past, and the modifications in this report can certainly be considered statistical. The ORNL experiments of Magnuson and $Gwin^{(21)}$ and of Macklin and deSaussure⁽²²⁾ should provide strong verification for the correctness of the reactor approach. It must be remembered that the use of standard deviations as an expression of errors does not place great confidence in the limits quoted, and further experiments are justified to increase the confidence in the basic reactor constants. More accurate and independent experiments, as well as additional study of existing measurements, are needed.

The error quoted for the U²³³ fission cross section seems small, and the development of absolute fission counting techniques and standards needs to be stressed. The approach here has been to throw out all direct fission measurement results, and the internal agreement observed with the network of constants provides strong support for this approach. A danger exists that systematic errors might be present throughout the field. This again stresses the need for independent measurements.

A proper statistical analysis involving error correlations needs to be attempted, and this approach probably fits into the field of operational analysis. This would assist in assaying for possible systematic errors, and should be applied to all of the fissionable isotopes to be most effective. The difficulty of obtaining experimental details is a major problem to this approach, and a need for careful reporting exists.

When results are obtained which are several standard deviations from the accepted mean error, a reluctance to accept the value should result unless the technique used provides an unquestionable improvement in accuracy. In general, such results should not be included in the best fits until independent experimental verification is available.

Consideration of the use of U^{233} as a standard for reactor measurements is recommended. The nearly 1/v dependence of the cross sections provides important simplifications for reactor measurements. Also, very high-purity U^{233} can be obtained by chemical separation processes by isolating Pa²³³ from the irradiated thorium.

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Discussion of Paper Presented by Mr. Evans

MR. BOLLINGER:

Are there any questions about this paper ?

MR. SAMPSON:

This is not really a question, but a word of praise to this great age of people getting sophisticated about really tying these constants down. I felt this way in conversations with the data people for a long time, particularly on the subject of intelligent analysis of the constants and rapid reporting. I would just like to say in addition that there is need for a reactor physicist or nuclear physicist who could be seriously interested in correlating these and putting out the official publication.

A REPORT ON THE MEASUREMENTS OF ETA OF U²³³ AT ORNL

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Introduction

The value of eta, the number of fission neutrons emitted by a fissile nucleus per neutron absorbed, is probably the most fundamental parameter of nuclear energy. The thermal value of eta for the isotope U^{233} is especially important because on it depends the feasibility of thermal breeding in the U^{233} -thorium cycle.

Because of this importance one would expect the thermal value of eta of U²³³ to have been one of the first nuclear parameters experimentally determined to the best attainable accuracy. And indeed many measurements of this constant have been carried on in the US, in Canada, in England and in the USSR. Until very recently most of the direct measurements of eta were obtained by the "danger coefficient" or the "pile oscillator" techniques. Those techniques consist in measuring the "apparent" absorption cross section of the fissile material; by observing its effect on the criticality of a nuclear reactor, eta can then be obtained from the measurement if the real absorption cross section is known.

Measurements of eta of U^{233} by the pile oscillator method were carried out as early as 1945-46 by Zinn at Argonne and by Littler and Ward at Chalk River. The values quoted by Littler in 1948 for those two measurements were 2.36 \pm 0.06 and 2.38 \pm 0.08, respectively.⁽¹⁾ Those high values and the large errors reflect mainly a poor knowledge of the absorption cross section. Indeed, the Argonne measurement, reanalyzed in 1955 using the better determined absorption cross section quoted in BNL-325, came down to a value of 2.29 \pm 0.03.

By 1957 the world consistent value of 2.28 ± 0.02 , compiled from measurements in the US, the UK and the USSR for the supplement to BNL-325, was generally accepted.

In 1958, however, a series of new measurements, mainly British measurements, were made, which led indirectly to a much lower value of eta for U^{233} . At the second Geneva Conference, Rose, et al.,⁽²⁾ described a measurement of the ratio of eta of U^{233} to eta of U^{235} , obtained by the pile oscillator technique with the reactors Dimple and Gleep. For this ratio, averaged over a thermal spectrum of 20°C, they obtained a value of 1.074 ± 0.015, or some 3 per cent lower than the previously accepted value. Shortly after the Geneva Conference, Sanders et al.,⁽³⁾ also at Harwell, described a very careful measurement of nu for U^{235} . This constant was obtained by comparison with

the spontaneous fission neutron source of Pu^{240} calibrated by Richmond. A value of 2.420 \pm 0.037 was obtained. Combining this value of nu with a value of 0.192 \pm 0.003 obtained by Craig <u>et al.</u>, at NRX, Sanders <u>et al.</u>, arrived at a value of 2.02 \pm 0.03 for eta of U^{235} . Using this value and the ratio measurement of Rose <u>et al.</u>, mentioned above, a value of 2.17 \pm 0.05 is obtained for eta of U^{233} . With such a low value of eta of U^{233} the feasibility of thermal breeding may be questioned.

Because of the importance of the value of eta for U^{233} in determining the feasibility of a thermal breeder and in view of the discrepancy between values obtained by the recent British experiments and the previously accepted value, ORNL has started an intensive program of eta measurements. This program is now nearly completed. It includes a direct measurement of the variation of eta of U^{233} with energy, a determination of the ratio of the value of eta of U^{233} to the value of eta of U^{235} by comparison of reactivity effects in a flux trap critical assembly, another determination of this ratio by comparison of critical experiments with aqueous solutions of U^{233} and U^{235} , respectively, and measurements of the absolute thermal value of eta for U^{233} by criticality experiments and, directly, by the manganese bath technique. Those measurements will now be described in turn, and the results obtained will be discussed.

Direct Measurement of the Variation of Eta of U²³³ with Energy (Measurements by J. A. Harvey, R. C. Block, G. G. Slaughter and N. J. Pattenden)

The variation of the ratio of the fission cross section to the capture cross section with neutron energy, nu being constant, results in a variation of eta. The knowledge of the energy dependence of eta for U²³³ is of considerable importance to the breeder reactor program, first, because of the emphasis on fast breeder reactors, and also because even in thermal reactors about 10 per cent of the fission are caused by epithermal neutrons. Furthermore, the energy variation of eta has a direct bearing on the temperature coefficient of reactivity and on the interpretation of absolute measurements of eta over a sub-cadmium spectrum. The variation of eta may be calculated from the energy dependence of the fission and absorption cross sections. This procedure has, however, been found unreliable, particularly in the neighborhood of resonances, where uncertainties due to resolution effects are not necessarily the same in the measurements of the fission and absorption cross sections. Thus direct methods of measuring the energy dependence of eta were developed, originally at Brookhaven. The principle of the ORNL measurements, similar to the original BNL measurements is illustrated in Fig. 1, which shows the apparatus used by Harvey <u>et al</u>. Under exactly the same experimental conditions, two



Fig. 1. Equipment to Measure $\eta(E)$ for U²³³

measurements are made simultaneously: (1) the emission rate of fission neutrons from a thick foil of fissionable material as a function of the energy of the incident neutrons, and (2) the change with energy of the flux of neutrons incident upon the foil. From the ratio of the two measurements the change of eta with energy is obtained. The fission neutrons are detected by means of a set of two Hornyak buttons disposed around the sample as shown in the figure. A thick U²³³ sample is used, so that nearly all the incident neutrons are absorbed and a maximum counting rate is obtained in the detector. The incident flux is monitored by a thin BF_3 proportional counter which absorbs only a few per cent of the incident beam. The boron cross section in the energy range of the measurements varies as 1/v and, since the detector is thin, the efficiency of the counter varies very nearly as the cross section. The energy of the incident neutrons was determined by a time-of-flight method, using the ORNL high-intensity fast chopper spectrometer. Corrections were made for the transmission of the sample, however, since nearly all the incident neutrons were absorbed; the results depended only in a insensitive way on the absorption cross section. Other small corrections were made for impurities in the sample (less than 1 per cent), departure of the BF₃ counter from 1/v law (about 1 per cent), and internal scattering in the foil (about 1 per cent also). No attempt was made in this experiment to make an absolute determination of eta. The data were normalized to a value of eta of 2.30 at 2200 m/sec. The determination of this normalization constant is the purpose of experiments that will be discussed later.

Direct measurements of the variation of eta of U^{233} with incident neutron energy were made by the technique just discussed in the region from 0.2 ev to 3 ev. Those data are shown in Fig. 2. In the region from



0.2 to 1.0 ev, eta is almost constant. This implies that the effective value of eta for a Maxwellian distribution will equal the value for 2200 -m/sec neutrons and will be rather temperature independent.

Additional measurements will be made with a fast neutron detector of higher efficiency to increase the statistical accuracy of the data. The measurements will also be extended to higher energies.

Determination of the Thermal Value of Eta of U²³³ Relative to the Thermal Value of Eta of U²³⁵ by Comparison of Reactivity Effects in a Flux Trap Critical Assembly (Measurements by D. W. Magnuson and R. Gwin)

The thermal value of eta for U^{233} relative to the thermal value of eta for U^{235} was determined by a method in which reactivity contributions made by small samples of the uranium isotopes and purely absorbing materials to a critical assembly were compared. The flux trap critical assembly is represented in Fig. 3. It consists of a 15-cm dia. water column located axially within a critical cylindrical annulus of uranyl fluoride (UO_2F_2) solution enriched to 93.2 per cent in U^{235} . The outside diameter of the annulus is 38.1 cm. The height is nominally 51 cm but was varied ± 1 cm in order to control the assembly. Samples of aqueous solutions of the fissionable and absorbing materials were inserted axially into the water column after a stable critical system had been achieved, and the resulting



Fig. 3. Experimental Configuration for Annular Critical Experiment.

reactivities were measured by means of period determinations and the Keepin delayed-neutron data. The sensitivity of the system was sufficiently high to use samples which were essentially infinitely dilute (maximum density of 3.2 g/liter for the uranium samples). The samples were contained in a 136-ml polyethylene bottle which, in turn, was placed in a Lucite bottle holder. This assembly was suspended on a phosphor bronze cable and located successively in and out of the center of the critical assembly, and the resulting stable periods were measured. Samples of aqueous solutions of $U^{233}O_2(NO_3)_2$ (enriched to 97.7 per cent), $U^{235}O_2F_2$ (enriched to 93.2 per cent), Pu²³⁹ (NO₃)₄, H₃BO₃, LiOH, In $(NO_3)_3$ and pure water were used.

Radial flux measurements were carried out across the assembly in order to study the shape of the thermal flux and the proportion of epithermal flux. Bare and cadmium-

covered gold and U^{235} metal foils (2 mils thick) were irradiated in the flux of the assembly at a power of approximately 1 w, and the induced gamma-ray activity was subsequently measured. The relative activity of the bare and cadmium-covered gold foils as a function of distance from the centerline of the assembly are shown in Fig. 4. This figure shows



that both the thermal and the epithermal fluxes were constant in the region occupied by the samples. At the center of the water column the cadmium ratio of the gold activity was 12.0. Figure 5 shows the radial distribution of the fraction of U^{235} fissions above the cadmium cutoff as a function of distance from the centerline of the assembly. The cadmium ratio of the fission in U^{235} at the center of the water column was 82.



Fig. 5. Radial Distribution of the Fraction of Fissions Above Cadmium Cutoff.

The flux at the sample position was assumed to be Maxwellian below 0.2 ev and 1/E above 0.2 ev. The temperature of the Maxwellian was assumed to be 300°K, the measured physical temperature being 25°C. The proportionality constant between the resonance and the Maxwellian parts of the spectrum was determined by the cadmium ratios of the gold and U²³⁵ activities discussed before, and by comparing the reactivity contributions of samples of indium with samples of boron and lithium. Using value of the resonance fission integral of U²³⁵ calculated from Oracle cross-section tapes by W. E. Kinney and a resonance integral of In calculated by L. Dresner, values of λ between 0.019 and 0.022 were obtained.

The reactivity measurements yield essentially comparisons of the product of the absorption cross sections and eta for the two uranium isotopes. The epithermal contributions were evaluated assuring a 1/E fast flux, and the experimentally determined value of $\lambda = 0.021$. The ratio obtained and the derived value for the ratio of etas is shown in Table I.

This ratio for the values of eta will be later compared with other values of this ratio obtained at ORNL and at other laboratories.

TABLE I

RESULTS OF FLUX TRAP EXPERIMENT

$\eta\sigma(U^{233})$	$= 0.957 \pm 0.013$	$\frac{\eta^{23}}{\eta^{23}} = 1.102 +$	0 022
$\eta\sigma(U^{235})$		η^{25}	0.022

The ratios are averaged over a Maxwellian neutron spectrum at $20^{\circ}C$,

The cross sections used were: $\sigma_a^{23} = 575 \pm 6b$ f-factor = 1.000 $\sigma_a^{25} = 680 \pm 7b$ f-factor = 0.974

Comparison of Critical Experiments for the Determination of the Thermal Value of Eta of U²³³ Relative to the Thermal Value of Eta of U²³⁵ (Measurements by D. W. Magnuson and R. Gwin)

The ratio of eta for U^{233} and eta for U^{235} has been determined by measuring critical concentrations of U^{233} and U^{235} in aqueous solutions for two spheres of 69.2 cm and 122 cm diameter, respectively. Figure 6 is a photograph of the larger sphere. The diameter of this sphere is 122 cm, its volume is 949 liters, and the aluminum wall is 0.77 cm thick. Concentrated solutions of UO_2 (NO₃)₂ of the desired isotope were diluted in water in the flat storage tank seen in back of the picture and then introduced into the sphere. The critical concentrations for this sphere were 14.11 g/liter and 12.93 g/liter for the U^{235} and U^{233} , respectively. In the 69.2-cm dia sphere, five concentrations of U^{233} and four concentrations of U^{235} were made critical. Boric acid was added as a poison in those experiments in order to vary the critical concentration.

The critical equations used to interpret the results are as follows:

$$k_{eff} = \frac{K(B) \left[\frac{\eta}{1 + L^2 B^2} \int_0^{0.2 \text{ ev}} \phi_M \Sigma_{au} dE + \lambda \nu \int_{0.2}^{10^6} \frac{\Sigma_f}{E} dE \right]}{\int_0^{0.2 \text{ ev}} \phi_M \Sigma_{at} dE + \lambda \int_{0.2}^{10^6} \frac{\Sigma_{at}}{E} dE} \qquad (1)$$

$$\lambda = \frac{\int_0^{0.2 \text{ ev}} \Sigma_{at} \phi_M dE}{\zeta \Sigma_S} \qquad (2)$$



Fig. 6. Apparatus for Critical Experiment

In this equation K(B) represents the nonleakage probability of fission neutrons to thermal energies; it is a function of the buckling B^2 . This factor could not be calculated because of the uncertainty in the age. Rather, it was eliminated between the results for U^{233} and those for U^{235} to obtain a ratio of eta for the two isotopes. In these equations ϕ_M represents a Maxwellian flux. The physical temperature of the solution was 25°C and the neutron temperature was assumed to be 23°C (that is, 300°K). The ratio of the resonance flux to the Maxwellian, λ , was obtained by a neutron balance at the thermal cut off. The calculated values of λ were in agreement with experimental values determined from the fission product activities of bare and cadmium-covered U²³³ and U^{235} foils. In the large sphere, values of λ were about 0.035; in the smaller sphere they were between 0.043 and 0.060, depending on the concentration and the isotope. The resonance contributions (above 0.2 ev) were obtained by W. E. Kinney, assuming a 1/v behavior of the boron cross sections and using Oracle tapes for the uranium isotopes. The nonleakage probability of thermal neutrons is also a function of buckling. An extrapolation length of 2.8 ± 0.3 cm was used, obtained by flux traverses taken in a cylindrical geometry. Table II shows the results that were obtained. The ratios of eta and of eta times the thermal utilization are all averaged over a thermal spectrum. The cross sections used are those obtained recently at ORNL, as indicated. The result for the 69-cm dia sphere is the average over nine experiments of different concentrations. The probable errors include a 10 per cent error in λ and in the resonance integrals, a 1 per cent error in the thermal cross sections, and a 0.5 per cent error in the concentration.

TABLE II

Results with the 122-cli Spher	Results	with	the	122-cm	Sphere
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 $\frac{(\eta f)^{23}}{(\eta f)^{25}} = 0.988 \pm 0.005 \qquad f^{23} = 0.5179 \pm f^{25} = 0.4625$ $\frac{\eta^{23}}{\eta^{25}} = \begin{cases} 1.106 \pm 0.015 & 122 \text{ cm dia sphere} \\ 1.100 \pm 0.015 & 69 \text{ cm dia sphere} \end{cases}$ $\sigma_{a}^{25} = 680 \pm 7b \qquad \sigma_{a}^{23} = 575 \pm 6b$ $f-factor = 0.974 \qquad f-factor = 1.000$

Absolute Determination of the Thermal Value of Eta of U²³³ from Critical Experiments in Spherical and Cylindrical Geometry (Measurements by D. W. Magnuson and R. Gwin)

In a set of critical experiments in a cylinder of 60.92-cm diameter with solutions of U²³³, the nonleakage probability was determined experimentally, by kinetic measurements, for various heights and concentrations.



Fig. 7. The Cylindrical Assembly

The data were analyzed by using the criticality equation and the cross sections shown previously. Figure 7 shows a photograph of this cylindrical assembly. Four concentrations were made critical with heights varying between 19.49 to 54.49 in. Table III shows the results of this set of experiments. An average thermal value of 2.297 \pm 0.010 was obtained for U²³³. The error quoted in this average does not include possible systematic errors in the cross sections or in the model.

TABLE III

IN CYLINDRICAL GEOMETRY Cylinder dia: 60.92 in., constants used: $\sigma_a^{23} = 575 \pm 0.066$ f-factor for U²³³ = 1.000 CYLINDER **EXPERIMENTAL** U²³³ CONCENTRATION HEIGHT NON-LEAKAGE η g/l IN. PROBABILITY $14.12 \pm 0.5\%$ 0.8865 ± 0.0077 19.49 2.302 ± 0.025 23.31 $13.53 \pm 0.5\%$ 0.9093 ± 0.0059 2.295 ± 0.021 12.88 ± 0.5% 30.58 0.9341 ± 0.0040 2.293 ± 0.016 54.59 $12.21 \pm 0.5\%$ 0.9602 ± 0.0022 2.296 ± 0.014

EXPERIMENTAL DATA AND RESULTS OF CRITICAL EXPERIMENTS

WEIGHTED AVERAGE: $\eta = 2.297 \pm 0.011$

From this set of experiments a value of the nonleakage probability will be calculated for the 122-cm dia sphere, and from the critical experiment in that sphere another absolute value of eta will be obtained. This calculation is not yet completed.

Direct Measurement of the Absolute Thermal Value of Eta of U²³³ by the Manganese Bath Technique

(Measurements by R. L. Macklin, G. deSaussure, J. D. Kington and W. S. Lyon)

In this experiment a thermal neutron beam, defined by cadmium differences was introduced into the center of a 1-meter dia sphere filled with a dilute solution of manganous sulphate in water, and the direct activation of the bath by the beam was determined. The beam was then totally absorbed in a sample of U²³³ and the activation of the bath by the fission neutrons from this sample was measured. The ratio of the two activities is equal to eta, except for small corrections.

This experiment has been designed to obtain a high-precision, absolute value of eta. In contrast to the experiments described before, the results do not depend in a sensitive way on the cross section or on the nonleakage probability of fission neutrons.

Figure 8 shows a diagram of the experimental equipment. A D_2O can and an air-filled cylinder are mounted rigidly on the Bulk Shielding Reactor (BSR)



Fig. 8. Experimental Arrangement for Measurement of Eta of U²³³.

grid plate. The D_2O provides a source of thermal neutrons, that form a beam along the air-filled cylinder. A collimator defined by two cadmium openings is mounted on the manganese bath sphere, ashore. Then this assembly is submerged and aligned on the neutron beam for irradiation (about $\frac{1}{2}$ hr with a BSR power of 1 Mw). The neutron beam is monitored by a thin manganese foil (20 mg/cm²) at the entrance of the sphere. The insert shows a detail of the fuel sample: this consists in 4 x 0.025-in. discs of high-purity U²³³, about 1.5 in. in diameter. A 0.020-in. thick cadmium disc covers the sample on one side, and cadmium differences are obtained by simply reversing the orientation of the sample. The uranium foils are spaced as shown in order to reduce the multiplication in the foil (analog to the fast effect in fuel rods). The manganese bath sphere has a 1-meter diameter, so that the leakage of fission neutrons is smaller than 0.5 per cent. Figure 9 is a picture of the equipment being irradiated next to the BSR.



Fig. 9. Equipment for Measuring Eta for U^{233}

For each measurement of eta, four measurements are made with the manganese bath: one with the fuel sample, one with the sample covered with cadmium, one with open beam, and one with a cadmium disc. The value of eta is then obtained from the following equation:

$$\eta^* = \frac{R_U - R_{CdU}}{R - R_{Cd}}$$

where
$$R_i = \frac{Mn^{56} \text{ activity in bath sample}}{Mn^{56} \text{ activity in monitor}}$$



Fig. 10. Counter Arrangement

Here R; represent the ratios of the activity of an aliquot sample of the solution to the activity of the monitor. Since both activities are measured on the same detector, the ratio is independent of small changes in the efficiency of the detector from one day to the next. Also, because differences between two measurements are taken, any constant activation of the manganese solution (due to leakage of the reactor) will be cancelled in the final result. Eta is starred in the equation to indicate that some minor corrections have been applied to this measured value. Those corrections will be discussed later.

After an irradiation the solution is stirred for 20 min. Then a one-liter sample is weighed and its activation is measured on a 3×3 -in. NaI(T1) crystal by detecting the 0.85 Mev gamma ray from the 2.56-hr Mn⁵⁶ The counter arrangement is shown in Fig. 10. The solution is contained in a plastic cylinder; this reduces the sensitivity to the exact position of the detector. For the measurement of the activation of the bath when the U²³³ was in, the solution was diluted by a factor of 2.5 before being

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counted. In this way all count rates were kept around 10^3 cps, avoiding problems of overload.

Figure 11 shows the arrangement of the same detector for the counting of the monitor activation. With this arrangement the ratio of the



Fig. 11. Counter Arrangement for Monitor Activities

activation of a solution to the activation of a monitor could be reproduced to better than 0.3 per cent from one day to the next. All counts were duplicated and repeated over two or three different detectors to guard against any error.

Table IV shows the list of the corrections that were considered and representative values as to their order of magnitude. The largest correction is due to the fast effect in the foil; this was reduced by splitting the foil. There is a correction connected with the sample being not black to all neutrons below the cadmium cut-off, one from scattering from the sample, and one due to the excess absorption of fission neutrons in the 340-ev resonance in manganese. The absorption of thermal neutrons in the aluminum is larger than that of the fission neutrons. Also, some fission neutrons may be scattered back into the sample and cause further fissions. Some fission neutrons leak out of the bath; some escape through the collimator; some thermal neutrons are absorbed in impurities in the sample; finally, some fission neutrons cause threshold re-

actions in the manganese bath. A further correction has been included to convert the thermal eta to an eta at 2200 m/sec.

TABLE IV

1. Fast multiplication in the metal foils $\eta^*/\eta = 1 + \frac{P_c'(\nu - 1 - \alpha)\sigma_f/\sigma}{1 - P_c^n(\nu\sigma_f - \sigma_s)} = 1.031 \pm 0.002$ (U^{233}) 2. Beam transmission through sample (0.992)(0.998)3. Beam scattering from sample 4. Excess resonance absorption in Mn for (1.005)fission neutrons 5. Absorption in structural materials (1.004)(chiefly thermal neutrons) 6. Indirect multiplication (1.008)(0.992)7. Leakage of fission neutrons 8. Duct streaming (0.999)9. Isotopic impurities (0.999)10. Chemical impurities (0.999)11. Variation of η over the sub-Cd spectrum (0.997)12. (n,α) and (n,p) reactions in O and S (0.996)Total Correction (1.020)

The value that was obtained for eta of U^{233} corrected to 2200 m/sec was 2.30 ± 0.02. The error will probably be reduced to 0.015 or better when all the corrections are finally evaluated.

Conclusions

Table V shows a comparison of various values of the ratio of eta for U^{233} to eta for U^{235} . All those ratios are averaged over a thermal spectrum at 20°C. The value of Harvey and Sanders is an average of British and US measurements previous to 1955. It is in excellent agreement with all of the ORNL measurements. The value of Rose <u>et al.</u>, appears slightly lower.

Table VI shows a comparison of values of eta for U^{233} . The value obtained by the British measurements is definitely lower than the ORNL

results. The latter results are slightly higher than the conventional BNL-325 value of 2.28 \pm 0.02, and perhaps a value of 2.30 \pm 0.015 is now more appropriate. The uncertainty of the manganese bath measurement will be reduced when all the corrections are entirely evaluated. This last measurement gives a value somewhat more reliable since it is almost independent of the absorption cross section of U²³³.

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COMPARISON OF EXPERIMENTAL VALUES OF η^{23}/η^{25}	
(AVERAGED OVER MAXWELLIAN SPECTRUM)	

HARVEY AND SANDERS ^(a)	1.10 ± 0.02
ROSE <u>et al</u> . ^(b)	1.074 ± 0.015
ORNL FLUX TRAP	1.102 ± 0.022
CRITICAL SPHERES	1.103 ± 0.011
MANGANESE BATH	1.108 ± 0.015

^(a) PROG. IN NUCL. ENERGY SERIES 1, VOL. 1, p 34. ^(b) A/CONF. 15/P/14.

TABLE VI

COMPARISON OF VALUES OF η OF U-233

BNL-325, 2nd ed. WORLD-CONSISTENT	2.28 ± 0.02
UK ^α	2.17 ±0.03
ORNL CRITICAL CYLINDER	2.297 ± 0.011
MANGANESE BATH	2.30 ± 0.02

^aFROM $\eta^{25} = 2.02 \pm 0.03$ AERE R/R 2212 R and $\eta^{23}/\eta^{25} = 1.074 \pm 0.015$ A/CONF. 15/P/14.

References

- 1. CRP-378 (1958).
- 2. Proc. 2nd Intnl. UN Conference on Peaceful Uses of Atomic Energy, United Nations, Geneva (1958) Vol. 16, p. 34.
- 3. AERE R/R 2212 (Revised).

Discussion of Paper Presented by Mr. deSaussure

MR. BOLLINGER:

Are there any questions concerning this paper?

MR. SAFFORD:

The first question has to do with the first slide you showed on the relative eta measurements. I noticed that the thin portion of the foil was at a right angle to the beam leaving the full length of the foil at right angles to the photomultiplier. This, as I understand it, is opposite to the so-called thick-thin method that is used where your foil is almost tangential (almost parallel) to your beam to avoid large scattering effects.

MR. deSAUSSURE:

I believe the foil was perpendicular to the beam. Yes?

VOICE FROM FLOOR:

Doesn't this give rise to considerable scattering corrections?

MR. deSAUSSURE:

No, I am not sure why; I don't understand why it should.

VOICE:

Because, in putting the foil tangential to the beam, this gives you the highest one minus the transmission value for the foil and yet allows the neutrons emitted to be emitted through a very thin section of the foil to your photomultiplier.

MR. deSAUSSURE:

I see. I don't believe that the two are in competition. I think the transmission of the foil was measured by bringing the foil back, and then the foil was laid back in the thin position.

MR. BOLLINGER:

Are there other questions ?

MR. BARTELS:

I am afraid along toward the end of your report I wasn't listening as closely as I should have been. I gather, if I heard you correctly, that you have revised upwards slightly your estimated value of eta as a result of the manganese bath measures, upwards from the values you reported at the ANS meeting.

MR. deSAUSSURE:

I think we increased the value from 2.29 ± 0.02 to 2.30 ± 0.02 . These two numbers are consistent within the errors and a more definite value will have to wait until these errors are fully evaluated.

RECENT BRITISH MEASUREMENTS ON NEUTRON YIELDS FROM U²³³ AND U²³⁵

J. E. Sanders UKAEA, Winfrith

Introduction

The nuclear data of fissile nuclides having most bearing on the breeding or conversion ratio of a system are values of eta, the neutron yield per neutron absorbed. The value of eta determines the number of spare neutrons available for capture in fertile material.

Recent British measurements in the nuclear data field have included a study of the ratios of eta values of the three most important fissile nuclides for thermal neutrons, and absolute determinations of the value of nu, the neutron yield per fission, which is related to eta by the expression

 $\nu = \eta (1 + \alpha) ,$

where alpha is the ratio of capture to fission. Since in the case of U^{233} and U^{235} alpha is much less than unity and can be measured by mass spectrometry and counting of irradiated material to reasonable accuracy, (1 + alpha) is a precisely known quantity for these two nuclides. An absolute measurement of nu thus gives an absolute value of eta to comparable accuracy.

Absolute measurements of eta by the pile oscillator should strictly be regarded as "integral" data, as explained in the next section, so that for the purpose of obtaining eta values which can be regarded as fundamental nuclear data, the approach via nu and alpha is a more consistent procedure.

While in the slow neutron region nu can be considered a constant, eta is an energy-sensitive quantity, so that eta values quoted must be coupled with a statement of the appropriate neutron spectrum.

Recent Dimple and Gleep Oscillator Measurements on Eta Ratios

The perturbation cross section of a fissile nuclide can be written in terms of its absorption cross section and eta value in the form

$$\sigma_{p} = \sigma_{abs} (1 - W\eta)$$

where W is the relative importance to the reactor of fission neutrons emitted and slow neutrons absorbed by the sample. Transposing,

$$\eta = \frac{1}{W} (1 - \sigma_{\rm p} / \sigma_{\rm abs})$$

While the value of W in a system where there is predominantly thermal absorption is expected to be the same for all fissile samples of the same geometry, the absolute value of W is not easy to determine accurately. Its theoretical derivation involves a lattice calculation for the oscillator reactor with the assumption of some particular model. The value of W, and hence that of eta, is likely to show some variation depending on the type of calculation employed, so that one is entering the realm of "integral data" when deriving absolute eta values from oscillator experiments. The situation is similar to that when eta values are obtained from correlation of subcritical or critical experiments.

The aim of the Harwell oscillator experiments on fissile materials is at present concentrated on the determination of eta ratios, which do not involve a knowledge of W and hence may be regarded as in the field of "pure" nuclear data. Particular attention has been paid in this work to accurate characterization of the neutron-energy spectrum of the oscillator reactors and to the use of samples of different origin and of carefully checked chemical composition.

Results on the eta ratios of U^{233} , U^{235} and Pu^{239} were presented at the 1958 Geneva Conference by Rose, Cooper and Tattersall. Because of an apparent discrepancy between the measured U^{233}/U^{235} ratio and the value predicted from current nuclear data, and because of the importance of the U^{233} data for discussions of the breeding potentialities of the U^{233} . Th cycle, further measurements on these two nuclides have been made and will be discussed here.

The spectral conditions of the sample positions in the Dimple and Gleep reactors used in the oscillator experiments are summarized below, in terms of effective neutron temperature and r-value. In Dimple, the sample is located at the center of a heavy water "Thermal pit" of 50-cm radius. In Gleep, the sample is located in an interstitial position of the uranium-graphite lattice. The eta values and their ratios predicted from Westcott's compilation are given in Table I, together with the measured ratios as of 1958, to indicate the magnitude of the previous discrepency. It is seen that the measured ratios are somewhat lower than the predicted ratios, the discrepancy, in the case of Dimple at least, appearing significant.

V	'a.	lue	S	oİ	E	ta

Reactor	Spectrum		Predicted Eta			Measured Eta
	T (°C)	r	U ²³³	U ²³⁵	Ratio	Ratio, 1958
Dimple Gleep	2 1 66	0 0.042	2.283 2.277	2.077 2.068	1.099 1.102	1.057 ± 0.015 1.091 ± 0.015

In the more recent measurements, previously used samples have been reanalysed and the oscillator swings repeated. Further samples of both nuclides have been prepared, including some from the USA. All the samples are in the form of a solution of the sulphate or fluoride in heavy water. The results of this extended programme are as follows:

> (a) Re-analyses of the solutions used to obtain the eta ratios given at Geneva and quoted above have indicated significantly different fissile contents than were provided by the original analyses. The reason for this discrepancy is not entirely clear. All the more recent analyses have been made by a volumetric technique, which is believed to give uranium contents to a precision of $\pm 0.2\%$ Confirmation of the more recent analyses is obtained from the fact that a solution of U²³³ from ORNL when analysed gave results very close to the ORNL assay.

The repeat of the oscillator measurements on these original samples gave results agreeing, within errors, with those previously obtained.

- (b) Fresh samples have been made up using UK material. They yield results in satisfactory accord with those from the original solutions, with the more recent chemical analysis. Check analyses on these samples will be made at ORNL.
- (c) U²³³ and U²³⁵ samples have been supplied from the USA, including samples of solution used in the ORNL critical sphere experiments. The measurements so far indicate the somewhat surprising result that US U²³³ has about 4% higher reactivity per gram than the UK material.

The results of the more recent measurements are summarized in Table II.

TABLE II

Deseter	Sample 1			
Reactor	UK	US (U ²³³)	Predicted	
Dimple Gleep	1.097 ± 0.013 1.106 ± 0.013	1.129 ± 0.014 1.139 ± 0.015	1.099 1.102	

1959 Measurements on Eta Ratios

While the UK samples now yield eta ratios in good agreement with the predicted values using the best current nuclear data, the situation is confused by the higher ratios obtained with the US U^{233} , this appearing a significant discrepancy.

A search for impurities of high cross sections in the British U²³³ is being made, but so far nothing significant has been detected. The level of impurity required to explain the discrepancy is relatively large, being several thousand ppm of natural samarium.

Only UK samples of U²³⁵ have so far been measured with the Harwell oscillators, and the situation may be clarified when measurements are made with the US samples. Furthermore, samples from Harwell are being sent to NRTS, Idaho for reactivity measurement in the RMF and for detailed chemical analysis.

Recent Measurements of the Absolute Value of Nu

The determination of nu is in principle a simple process of counting fission neutrons in coincidence with fission fragment pulses. However, the accuracy of the earlier absolute measurements was limited by the accuracy with which the neutron detector could be calibrated. Recent British work in this field has aimed at the calibration of suitable neutron detectors to high precision. The two approaches that have been made are as follows:

(a) Pu²⁴⁰ Standard Source

A spontaneous fission neutron source consisting of a 180-gram sphere of plutonium metal containing 8.5% Pu^{240} has been calibrated to an accuracy of about $\pm l \frac{1}{2}\%$ by absolute methods and by comparison with a series of standard neutron sources from various other laboratories in the UK, USA and Europe. The spontaneous fission source has the merits of stability of output and spectral similarity to the fission neutrons to be measured in the determination of nu. Its chief drawbacks are the rather low output (2.036 x 10^4 per second) and possibility of multiplication effects when placed in the vicinity of moderators. This source has been used in three distinct experiments for the determination of nu for U^{235} fission.

(b) Boron Pile

A high-sensitivity (around 60% at 265 kev) neutron-detection system having a rather flat energy response has been constructed at Harwell by means of an array of BF₃ proportional counters in a graphite stack. This "Boron-Pile" has been described by Colvin and Sowerby in Geneva paper 52. An absolute calibration of this system is being made using a deuterium photodisintegration chamber irradiated with gamma rays of various energies from radioactive sources and charged particle reactions. A one-to-one correspondence exists between photoprotons counted in the chamber and photoneutrons injected into the pile, thus enabling the efficiency to be determined in a coincidence experiment.

The Pu^{240} spontaneous source has been used by Kenward, Richmond and Sanders to calibrate a BF₃-wax castle neutron counter employed in a coincidence experiment with a U^{235} fission chamber irradiated by a thermal neutron beam, to give a value of nu. It has also been used to calibrate the Boron Pile in a similar fashion, by Colvin and Sowerby. Thirdly it has been used at Aldermaston by Moat, McTaggart and Mather to obtain nu for 75-kev neutrons on U^{235} , via the intermediate step of obtaining nu for spontaneous fission of Cf^{252} . The Pu^{240} source and a Cf^{252} source in the form of a thin foil were compared in a wax-castle counter, the fission rate of the Cf^{252} being subsequently measured in a fission chamber. The value of nu for Cf^{252} spontaneous fission is thus obtained. The ratio of nu for 75-kev U^{235} fission and Cf^{252} spontaneous was then coincidence-measured in a large liquid scintillator system having an overall efficiency of 65%.

The three experiments give the thermal nu values (including delayed neutrons) in Table III, based on the same value of the Harwell Pu²⁴⁰ source.

TABLE III

Valu	es	of	Nu	for	U^{235}
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Experiment	Nu
Colvin and Sowerby, 1959	2.409 ± 0.039
Kenward <u>et al</u> ., 1958	2.420 ± 0.037
Moat <u>et al</u> ., 1959	2.354 ± 0.053

These data yield a value of 2.40 ± 0.04 . This value, rather than that of any one of the three contributory results in isolation, should be considered the best estimate of nu from the calibrated source approach. Though the Aldermaston value appears low, it is less than 2 standard deviation from the mean (after the common uncertainty of the source calibration has been subtracted), and it is by no means certain that a real discrepancy exists here.

The value of nu for Cf^{252} spontaneous fission has been measured in terms of the Pu^{240} source in the Boron Pile giving a result in good agreement with the Aldermaston determination. However the ratio of nu for Cf^{252} spontaneous and U^{235} thermal fission measured in the Boron Pile yields a result (1.513) lower than that obtained at Aldermaston, which, if coupled with the value of Cf²⁵² nu, gives a value for U²³⁵ in good agreement with the Harwell measurements. There is some doubt on the correction for scintillation detector efficiency between Cf and U²³⁵ in the Aldermaston ratio measurement, though it is felt that this cannot introduce an uncertainty of more than 1% into the measurement. In the Boron Pile experiments with the Pu²⁴⁰ source, there is some uncertainty regarding multiplication effects due to neutrons moderated in the graphite.

An entirely independent calibration of the Boron Pile is provided by the deuterium photodisintegration technique. So far these measurements have been made only with Na²⁴ as the gamma-ray source yielding photoneutrons of 265-kev energy. A calibration at this neutron energy together with a calculated sensitivity vs energy characteristics has given a value of nu for U^{235} thermal fission between the limits of 2.43 and 2.49: the major uncertainty remaining here appears to be in the possible anisotropy of the response of the Boron Pile.

It is clearly not possible to arrive at any sort of "final" data at this stage: however, both approaches give results compatible with a value of 2.43 ± 0.04 , which may be taken as a strictly provisional result of the direct nu measurements. This value is to be compared with that of 2.47 ± 0.03 given in the 2nd Edition of BNL-325. This latter value rests heavily on the absolute value of eta from pile oscillator measurements. The direct approach to eta via the absolute determination of nu yields slightly lower values than those previously accepted, although with the uncertainty at present put on the nu measurements the differences are well covered by the errors. In the present somewhat tentative state of the data, and in view of discrepancies which exist in both the nu and eta experiments, one would not wish to place too much emphasis on these results at the present stage. It is interesting though that there seems an indication of somewhat a lower U²³⁵ eta value which has generally been the trend of the eta derived from subcritical and critical experiments.

Acknowledgements

The work on eta measurements described in this paper has been done by the Harwell Pile Oscillator Group under the general direction of H. Rose, with chemical analysis of samples by M. J. Cabell of Harwell Chemistry Division. The calibration of the Pu²⁴⁰ source was made under the guidance of R. Richmond of Reactor Division, Harwell. The Boron Pile Measurements are being made by M. G. Sowerby and D. W. Colvin of Nuclear Physics Division Harwell. The Aldermaston experiments are the work of A. Moat, M. H. McTaggart and D. S. Mather. I am grateful to several of the aforementioned for interesting discussions while preparing this paper. The following unclassified reports are relevant to the data presented in this paper:

Rose, H. W. A. Cooper and R. B. Tattersall <u>The Use of the Pile Oscillator in Thermal Reactor Problems</u> Proc. of the 2nd U.N. Intnl. Conf. on the Peaceful Uses of Atomic Energy, Geneva (1958) Vol. 16, p 34.

Richmond R. and B. J. Gardner <u>Calibration of Spontaneous Fission Neutron Sources</u>, <u>AERE R/R 2097 (1957)</u>

Kenward, C. J., R. Richmond and J. E. Sanders <u>A Measurement of the Neutron Yield in Thermal Fission of U²³⁵,</u> <u>AERE R/R 2212 (Revised) (1958)</u>

Colvin, D. W. and M. G. Sowerby <u>Precision Measurements of Nu by the Boron Pile</u>, <u>Proc. of the 2nd U.N. Intnl. Conf on the Peaceful Uses of Atomic</u> Energy, Geneva (1958) Vol. 16, p 121

Moat, A., M. H. McTaggart and D. S. Mather $\frac{\text{Discrepancies in Nu Values}}{\text{AWRE NR/P 1/59}}$

Discussion of Paper Presented by Mr. Sanders

MR. BOLLINGER:

Are there any questions concerning these new results from the UK ? MR. GREEBLER:

Just one question on the Dimple experiments. Does that r = 0.04 in the Westcott cross section mean that 4 per cent of the fission are epithermal fission ?

MR. SANDERS:

With a 1/v absorber, 4 per cent of the absorptions occur above 5 kT.

MR. GREEBLER:

Can you from this crank out an epithermal eta for $U^{233}\ ?$

MR. SANDERS:

I don't think the accuracy would allow it.

MR. GREEBLER:

The 0.04 is too small to permit this.

MR. SANDERS:

Yes, in Dimple one can go to an r value of about 0.1 by crowding in the uranium but well, we do this sometimes, I think.

MR. GREEBLER:

Thank you. I see.

MR. BOLLINGER:

Could you explain exactly what the nature of these errors are ?

MR. SANDERS:

They are standard deviations.

MR. BOLLINGER:

In that case the difference is not really outside of experimental error.

MR. SANDERS:

No, well, if you take a value which is midway between these, I think one would probably be not too unhappy. I think if one takes the mean of the results of UK and US samples, one would come out with a value which is quite close to the values which Dr. Evans had on his slides.

MR. DeBOISBLANC:

I think if you would ask Dr. Rose from Harwell if he thought that the error in just comparing the Harwell sample to the American sample, which in effect was done, then the errors quoted there would not prevail; that is, he could make that comparison to somewhat better than 1 per cent, perhaps one-half per cent. I think he has already made this comparison and is quite sure that there is a distinct difference.

MR. SANDERS:

It is a 4 per cent difference in reactivity change and the error is about 1 per cent.

MR. DeBOISBLANC:

That difference could lie in the chemical analysis, that is, the error in the chemical analysis, and therefore occupies the greater part of this spread. If the chemical analysis is really a good half of one per cent, then you should not observe that discrepancy in the measurement. What is being said is that these two errors here are not independent.

MR. SANDERS:

No, they are not independent. If I was comparing the samples of the same material, the error would be much less than it is being quoted there. So there is a real discrepancy between the US and UK samples of U^{233} .

MR. DeBOISBLANC:

A point that relates to the previous paper and to this one is that it is always good to have a watch bird in these measurements, and we have been elected by the Commission to sort of be a watch bird on these two sets of measurements. From each of the samples of the sphere measurements made at Oak Ridge a sample was taken for our use, and we have so far collected 11 of these of which 7 have already been measured in the RMF, which is a flux trap type of measurement similar to that reported by Gwin from Oak Ridge. Now we have recently received a one-gram sample of the U^{235} and U^{233} from the United Kingdom, and this is presently being dissolved totally and will be divided into four parts. One part will go to Harwell for chemical analysis and one to Oak Ridge for chemical analysis; one will be analyzed in our own plant and the other one will be measured in the RMF. We hope to get a consistent set of measurements which might unravel the disagreement between the British and American samples based on 3 powerful sets of chemistry.

MR. WOLFE:

I would like to direct this to anyone who will answer among the speakers. Are there any measurements other than those reported for eta for U^{233} above 10 ev; I think the previous two speakers reported on data within the 10-ev range and it was not clear to me whether that data agreed. I wondered if there was any further data on this?

MR. EVANS:

I think the only data in the higher region have been that of the KAPL people.

MR. CAMPISE:

The only other data I have seen in the literature are those of the Russians, which apparently go to about a kilovolt. They show eta to be fairly constant, something like 2.25. The eta data that the Russians have published in the kilovolt range is about 2.25; they don't show any dips.

DR. COHEN:

There are also some photoneutron source measurements, by Spivak <u>et al</u>. These are reported in that issue of The Journal of Nuclear Energy which combines about 4 issues into one, and it is all the translations from the Russians' atomic energy journal. I think it is the June issue of this year.

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SESSION V

Wednesday Morning, October 21, 1959

Chairman: B. I. Spinrad Secretary: C. N. Kelber

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BREEDING POTENTIAL OF NEAR THERMAL REACTORS

Jack Chernick Brookhaven National Laboratory Upton, New York

Introduction

The US thermal breeder program has suffered a number of setbacks during the past year. The first shock was the low value reported by the British at the 2nd Geneva Conference for the thermal value of η for U²³³. Another setback was the recommendation of the "Ad Hoc Advisory Committee on Reactor Policies and Programs" that "until better knowledge of eta is obtained, work on thermal breeders should be held to a minimum" and in addition, that not more than one fluid fuel reactor project for producing lowcost nuclear power should be supported. Finally, the majority of the members of a "Fluid Fuel Reactors Task Force" questioned the feasibility of maintaining proposed circulating fluid fuel reactors and estimated their probable power costs in the neighborhood of 11 mills/kwh. Their report was much more pessimistic than those of previous evaluations of fluid fuel reactors, although the likelihood of breeding was conceded. As a result, the fluid fuel reactor program at ORNL, BNL and B&W has been drastically cut; in particular, the LMFR project has been cancelled.

In view of these events, we regarded it important to seek answers to the following questions: 1) whether the knowledge of eta has now improved sufficiently to enable one to decide if positive breeding is still possible in other than fast reactors; 2) in the light of the technical difficulties that have been experienced with the AHR, whether thermal breeding is possible with any moderator other than D_2O ; and 3) in view of the questioned feasibility of all of the proposed fluid fuel reactors, whether thermal breeding is possible in solid fuel reactors? Our answers to these questions, as we shall develop them here, are all in the affirmative.

The Value of Eta for U^{233}

Of course, the thermal value of eta of U^{233} is very important to the determination of the breeding ratio of thermal reactors. The value which we have assumed in our studies to date is 2.28 \pm 0.02. Such a value was, in fact, recommended by the Reactor Physics Planning Committee at a time when alarm over the measurements of H. Rose <u>et al</u>. at Harwell was at its height. It was the value used by the "Fluid Fuel Reactors Task Force" in estimating the likely breeding ratio of a full-scale AHR at 1.09 and that of the graphite-moderated LMFR and MSR breeders at 1.05. These estimates still appear to be sound ones at the present moment. The results of experimental work at ORNL are yielding a thermal value of η of about 2.30 for U^{233} . In the mean-time, errors have been discovered in the British measurements and the situation has not changed from that reported by deBoisblanc and Ergen at the

Gatlinburg meeting of the ANS. The preliminary results of experiments still in progress at ORNL and at the MTR to measure eta and U^{233} cross sections more accurately as a function of energy do not appear to affect greatly the results which we reported at the Gatlinburg meeting.

In Fig. 1, two estimates of the dependence of eta for U^{233} on the neutron spectrum of the reactor are shown. The spectrum parameter chosen is $\xi \Sigma_{e}/N_{23}$, the slowing-down power of the moderator per atom of U^{233} . The



Fig. 1 Dependence of Eta of U²³³ on Neutron Spectrum

curves show a slow decrease in eta as the spectrum hardens, with a minimum in intermediate-energy reactors, where both thermal and fast fission are small. The solid curve is based on old, unpublished data on total cross sections by V. Sailor at BNL and MTR data on fission cross sections reported by R. Fluharty <u>et al</u>. at the 2nd Geneva Conference. More recent data, communicated to us by J. Harvey at ORNL and by O. D. Simpson and M. S. Moore at the MTR, have been included in the second curve.

The importance of considering nonthermal absorption is also indicated. Over the range generally considered for "thermal breeders," $10,000 > \xi \Sigma_s / N_{23} > 2000$, the absorptions above 0.5 ev range from about 10 per cent to 35 per cent. Hence the generic term "near-thermal breeder" would be more appropriate. Over the same range, the difference in the estimates of $\overline{\eta}$ varies from 0.005 to 0.02, which is not regarded as serious, since the thermal value of eta which we have assumed is probably low by about 0.02.

Dependence of Breeding Gain on Moderator

The breeding ratio of a reactor is reduced by neutron absorption in the moderator. We have calculated the maximum breeding ratios obtainable with conventional moderators. The results are shown in Table I. These calculations are based on a multigroup analysis with values of eta for each group obtained from the data which we have discussed on U^{233} cross sections. The moderator absorption cross sections assumed are 1.5 mb per deuterium atom, 4 mb for graphite, 10 mb for beryllium, and 330 mb per hydrogen atom. The results show that optimum breeding ratios are obtained in incompletely thermalized reactors. While the largest value, 1.26, is obtained with heavy water as moderator, the values for graphite and beryllium are not much smaller, and even ordinary water turns out to be a possible moderator for a near-thermal breeder.

TABLE I

Dependence of Breeding Ratio on Moderator

Ulu Data						
$\xi \Sigma_{s}/N_{23}$	Thermality	D ₂ O	С	Be*	H ₂ O	
1,000	40.5	1.184	1.179	1.177	1.170	
2,000	63.7	1.223	1.211	1.204	1.183	
5,000	83.5	1.251	1.214	1.194	1.131	
10,000	91.4	1.258	1.180	1.136	1.003	
20,000	95.6	1.255	1.094	1.005	-	

Breeding Ratio vs. Neutron Spectrum

		2			
$\xi \Sigma_{s} / N_{23}$	Thermality	D ₂ O	С	Be*	H₂O
1,000	41.1	1.212	1.208	1.206	1.199
2,000	64.1	1.240	1.228	1.221	1.200
5,000	83.7	1.259	1.222	1.202	1.139
10,000	91.5	1.262	1.184	1.140	1.007
20,000	95.6	1.257	1.096	1.007	-
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New Data

*Does not include fast effect in Be.

The fast effect in beryllium was not considered in the above calculations but can be substantial. Estimates of $\epsilon = 1.061$ and 1.066, respectively, were obtained for pure beryllium based on the (n,2n) data of Fischer and of Marion and Cranberg. The results are shown in Table II. The calculations were exact but were based on isotropic scattering. A rough calculation indicates that forward scattering in beryllium may increase ϵ by about 1.5 per cent. In addition, there are indications that measured (n,2n) cross sections may be low. We have therefore revised our estimate of ϵ upward to 1.075 ± 0.020 . Because of its possible application to thermal breeders, beryllium should receive greater emphasis as a reactor material than it has in the past.

TABLE II

Fast Effect in Beryllium

Energy Interval (Mev)	(n,2n) lst Flight	(n,2n) Total	(n,α) lst Flight	(n,α) Total
6 – ∞	6.2x10 ⁻³	8.5x10 ⁻³	0.3x10 ⁻³	0.5x10-3
5 - 6	8.3	13.2	0.7	1.1
4 - 5	16.1	27.8	1.9	3.2
2.5 - 4	23.9	50.0	5.7	13.3
1 - 2.5			6.2	20.8
Totals	5.4%	10.0%	1.5%	3.9%
	E	= 1.061		

(Fischer's Data, Isotropic Scattering)

Energy Interval (Mev)	(n,2n) lst Flight	(n,2n) Total	(n,α) lst Flight	(n,α) Total
6 -∞	6.2x10 ⁻³	8.5x10 ⁻³	0.3x10 ⁻³	0.5x10-3
5 - 6	7.9	12.3	0.7	1.1
4 - 5	12.5	21.6	1.9	3.2
2.5 - 4	18.3	38.3	5.7	13.6
1.65 - 2.5	7.7	24.3		
1 - 2.5			6.2	20.6
Totals	5.3%	10.5%	1.5%	3.9%
	e	= 1.066		

Potentiality of Near-thermal Breeders

The maximum breeding ratios listed in Table I indicate a satisfactory margin for positive breeding. It is true that the breeding gain will be greatly reduced in a practical system where neutron losses to higher uranium isotopes, to protoactinium, to xenon, samarium and other fission products, to structural materials and leakage, etc., must be permitted. Some of these losses will be discussed in a companion paper by Dr. Levine. It is also true that, as long as fissionable fuel costs are low, breeders cannot be expected to compete on a cost basis against their nonbreeding counterparts, because of necessary restrictions on neutron losses. For example, the breeder will generally be larger in order to reduce neutron leakage losses, and the processing costs of a breeder will generally run higher in order to reduce fission product poisoning. The premium paid for breeding need not be high, however, and the emphasis on near-thermal breeders is due to their apparent economic advantages as a class.

It is clear that liquid fuel reactors have advantages as breeders in terms of reactivity control, xenon poisoning and possibly in the cost of processing fuel, fertile material and fission products. However, positive breeding in solid-fueled, near-thermal U^{233} reactors also appears to be within reach if pains are taken to minimize neutron losses.

Conclusions

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In conclusion, the analysis of recent data indicates that positive breeding on the U^{233} -thorium cycle is possible in near-thermal reactors with D_2O , graphite, beryllium or even ordinary water as the principal moderator. In view of the urgency of achieving this goal in the near future, additional power breeder projects are needed. Breeder proposals should be compared primarily on the basis of technical feasibility and economics. The question of doubling time, which is presently being given major emphasis, is of minor importance in these reactors and should not be considered. In our opinion, a solid-fueled, U^{233} -thorium breeder belongs in the short term US powerdemonstration program.

Discussion of Paper Presented by Mr. Chernick

MR. SPINRAD:

Dr. Greebler.

MR. GREEBLER:

I would like to ask a question pertaining to your beryllium data, specifically with regard to the Be (n,2n) effect.

The 6 per cent reactivity gain that you show in the clean condition is very encouraging. What I thought was a little discouraging in your data is the ratio of the (n,2n) to the (n,α) reaction, which was only something like $2\frac{1}{2}$ to 1. This means in a short time the (n,α) poisoning is essentially doubled as the lithium reaches saturation.

MR. CHERNICK:

I don't agree it is a short time compared to fuel-processing cycles. I think that there may be a point if beryllium is used as the moderator as well as the fuel-bearing material, and that this may be an uncomfortable question. Whether or not we have to allow the lithium to build up to the equilibrium concentration is not a fuel problem; because as the lithium comes in it can be looked at as an additional fission product poison with, at most, a 4 per cent yield and with something like a 550-barn cross section. We know that fission products of that nature eventually become uncomfortable in fuel elements and have to be removed, but they present nothing like the problem of samarium, for example, where you have to concede the loss.

In other words, we don't concede the loss of a 550-barn cross-section material. I think that fuel elements are processed more often than that. Assuming that the loss is to lithium, and assuming there is a process, it then can be removed and there will still be an advantage in using beryllium.

MR. GREEBLER:

Thank you.

MR. HALL:

Would you care to elaborate on your statement that the doubling time is not important?

MR. CHERNICK:

The problem is this. The essential thing is to use up fuel; in this connection the doubling time is irrelevant. The key question is to obtain breeding.

MR. HALL:

In my mind the doubling time concept is important because if at some point in time you wish to build reactors because they are more economical and it is a better way to make power, then you need some material with which to start the new reactor, possibly to expand the economy. Unless you have a rather short doubling time you have no capital fuel to provide this expansion.

MR. CHERNICK:

I think that comes very much later in time. The key problem, then, is first the question of fuel utilization and, secondly, a question of achieving self-sufficiency with the fuel that you are using. Any breeder can use, if it runs long enough, all the fertile material in the world, and I don't understand what we hope to accomplish with pushing the doubling time, which, it seems to me, is going to greatly increase the cost. If you attempt to maximize these reactors for doubling time you are maximizing on a relatively minor parameter, and this is what I am worried about because it is already apparent that people are doing this; they are not just trying to get breeders. We haven't got any. I think that is the first problem, to get a breeder that isn't going to cost so much money that nobody is going to want to build it. To give it a chance, you have got to forget about the question of doubling time.

MR. HALL:

I think the doubling time has the same importance as the fuel utilization.

MR. CHERNICK:

The doubling time should keep pace with our power requirements, assuming that it is all nuclear and we have no more coal and no more U^{235} , and that, it seems to me, is something in the distant future.

MR. SPINRAD:

If I may be permitted to comment on this, I think that what we are talking about is really what Mr. Chernick said, a question of economics now and a question of conservation in the future. Doubling times and considerations of this sort are still very important in systems like the fast reactor breeders where there is an enormous financial investment in the fuel, and by lack of its dilution a relatively limited power per amount of fuel loaded. In the thermal breeders it is less important now, though it will undoubtedly become important in the future, and the only question is, what is the time scale? Can we afford to be economical now rather than preparatory for the future?

I would like to make one further point. In regard to the remarks about breeding or not breeding, in a thermal sister-system (much laughter), many years ago, just at the start of the civilian power program, a number of studies were made on thorium fuel systems, U^{233} fuel, which were potential breeders. In every case it was discovered that the minimum fuel cost system was one which failed to breed by some very small margin - it was still an efficient converter, but the extra work to make it a breeder wasn't worth it dollar-wise.

MR. CHERNICK:

I might say that I agree with everything Mr. Spinrad has said. I would like to add this: for the present I am also concerned about the fact that we are paying very little attention to the conversion in the power reactors that we are building today. They are gradually going down and down. I mean the old Hanford reactors were pretty good converters compared to the present crop of power reactors.

MR. VonHERMANN:

You mentioned a number of 11 mills per kilowatt in your introduction, and you seemed to imply that your talk would show that this number has been lowered. Is that true or not ?

MR. CHERNICK:

I would like to spend a few minutes talking about this. This question of economics of reactors is a very tricky one, because people can come up with all sorts of numbers depending upon the degree of optimism or pessimism as to what the reactors will do and what it will cost to operate and maintain them, - even what the capital cost will be. For example, the Project Dynamo in 1953, in which both Spinrad and I were members, came up with numbers which undoubtedly were low, but of the order of something like 5 mills per kilowatt hour. There were a lot of very fine engineers on this study and this was their considered opinion of what these reactors might do, ten or fifteen years from the date when the study was made.

Since then, the question of maintenance of these radioactive circulating fuel reactors has been the cause of increasing concern. Now there have been developed programs to try to determine how you actually maintain such a system, and here again you can get all graduations of opinions of whether the methods that are being developed will, in fact, be feasible. Additional questions come up; things like contingency funds for these reactors, for example. When you get a reactor, such as the circulating fuel reactors, that has not been built, this particular evaluation group uses very large contingency funds compared to what is normal practice in the power field. Unless one is a good bookkeeper,one cannot check item by item where the difference in cost is, but when they add the whole think up, as I have already mentioned, the cost can double from the estimate at Dynamo of 5 mills per kilowatt hour to estimates of the order of 11 mills per kilowatt hour for all three of the fluid fuel reactors.

MR. BARTELS:

Comparing your maximum beryllium conversion ratio and your maximum D_2O conversion ratio, you have 1 26 for D_2O and 1.22 for beryllium. If we assume that the beryllium can be chemically reprocessed, I infer that we could get a gain of about 0.07.

MR. CHERNICK:

It would be higher than for D_2O . If you multiply these factors, it would be much higher than the maximum you get in D_2O .

MR. BARTELS:

Now, this is the question. You are talking about 1.29 versus 1.26, or maybe 1.30 versus 1.26. Is this difference significant?

MR. CHERNICK:

I think it goes above 1.30.

MR. BARTELS:

Then the chemical processing of the beryllium really is quite a significant question.

MR. KASTEN:

I would like to review something Dr. Hall was talking about. You stated that breeding ratio economics were important, and that doubling time was not important. It appears to me that breeding ratio in itself is less of an economic factor than doubling time and that we are talking about economics.

MR. CHERNICK:

I didn't say breeding ratio was important. We would like to have it greater than 1. Technical feasibility and economics are the important things.

MR. KASTEN:

Well, the information you gave here was primarily based on some nuclear calculations on averaged eta. Now you imply that there is considerably more information on cost, and as I am not aware of it, could you enlighten me on that information?

MR. CHERNICK:

Oh yes, of course. The latest cost estimate was the fluid fuels evaluation. Project Dynamo, the 1953 study, is also available. There were a number of additional studies, particularly on the liquid fuel reactor, carried out. There was one carried out at Brookhaven and at B&W, and a number of other groups looked into the cost problem. There is a huge amount of material, if you want to get it. Of course, there are lots of Oak Ridge studies of the economics of their systems. In fact, you are sitting next to Mr. MacPherson who's responsible for the molten-salt reactor, and I'm sure he can give you some more references on their estimates of the cost of the molten-salt reactor. There aren't any though, that I know of, on solid fuel reactors. So you have to remember that none of these ractors was really pushed as a breeder; they were pushed as low-cost power reactors, which some day might breed. Breeding was never a part of the program of AEC.

MR. CAMPISE:

I have three questions. Number one, you implied that your better near-thermal systems had about 10 to 35 per cent of the absorptions above thermal; what is this energy range?

MR. CHERNICK:

In that calculation, all captures occurring above 0.5 ev are considered above thermal.

MR. CAMPISE:

Do you know approximately where their center was?

MR. CHERNICK:

Yes, we do have a report, a paper by Miss Oleksa and myself, which is going to be published in the Journal of Nuclear Science and Engineering, and this has a detailed breakdown where the absorptions occur in energy.

MR. CAMPISE:

Does this imply that eta is fairly good above the thermal range?

MR. CHERNICK:

We actually found that when we got above 30 ev the data get rather ragged. We used the data from 30 to 1000 ev, which is still of some significance for these reactors, and take one value of eta: 2.20, and quite a bit less than what the Russian measurements indicate, but it's more in line from what appears to be likely based upon the low-energy resonances.

MR. CAMPISE:

What assumptions did you make about the scattering cross sections inside the resonance?

MR. CHERNICK:

We have taken a flat $12\frac{1}{2}$ barns for the scattering cross section, but again in this paper we have three graphs of the scattering cross section versus energy as calculated by a number of different people, and they fluctuate around and do not exceed 20 barns and go down to somewhat below 10 barns. The $12\frac{1}{2}$ barns still looks like a good average for these studies.

THE EFFECT OF HIGHER ISOTOPE PRODUCTION ON BREEDING

M. M. Levine Brookhaven National Laboratory Upton, N.Y.

Introduction

Breeding ratios for clean systems containing only U^{233} and moderator have been presented by Chernick and Oleksa Moore.⁽¹⁾ This is extended here to the case where equilibrium amounts of higher isotopes and fission products are present. These depress the breeding ratio because the absorption of neutrons in the nonfissile isotopes rob neutrons from the fuel, and even the absorptions in U^{235} are less productive than those in U^{233} . Furthermore, the presence of these other isotopes may be expected to raise the spectrum somewhat, causing those absorptions which take place in U^{233} to be at higher neutron energies where η is lower.

The breeding ratio is defined here as

B.R. =
$$\left\{ \left[\text{neutron productions in } U^{233} \text{ and } U^{235} \right] \right.$$

- $\left[\text{neutron absorptions in moderator, } U^{233}, U^{235}, \text{ and } U^{236} \right] \right\} (1)$
 $\left. \div \left\{ \text{neutron absorptions in } U^{233} \text{ and } U^{235} \right\}$.

(Absorptions in U^{234} do not appear here since, like absorptions in Th^{232} , they give rise to fissile nuclei.)

In the absence of higher isotopes and fission products, this definition coincides with that employed by Chernick and Oleksa Moore. When higher isotopes are present, this is equivalent to defining the breeding ratio as fuel atoms produced per fuel atom consumed, but neglecting neutron consumption through absorptions in Pa^{233} , fission products, and other materials (such as structure and control rods) and through leakage. Some of these other losses will be considered subsequently.

Higher Uranium Isotopes

The concentrations of higher isotopes will be functions of time and approach constant, steady-state values only when there is continuous fuel processing. However, even if this is not the case the concentrations will eventually remain near steady-state values based on some sort of continuous'averaged processing, so constant concentrations are assumed. These will be achieved in a reasonably short time, depending on flux levels.

The steady- state conditions will be as follows:

Absorptions in U^{234} = Captures in U^{233} ; Absorptions in U^{235} = Absorptions in U^{234} ; Absorptions in U^{236} = Captures in U^{235} .

No absorptions past U^{236} are considered here. By letting $A_E \equiv$ absorptions in species E and $\mu_E =$ (fissions \div absorptions) in species E, the steady-state equations can be symbolized as follows:

$$A_{24} = (1 - \mu_{23})A_{23}$$

$$A_{25} = A_{24}$$

$$A_{26} = (1 - \mu_{25})A_{25} \qquad . \qquad (2)$$

These allow Eq. (1) to be re-written as

B.R. =
$$\frac{\nu_{23}\mu_{23} + \nu_{25}\mu_{25}(1-\mu_{23}) - [(A_{mod}/A_{23}) + 1 + (1-\mu_{23}) + (1-\mu_{23})(1-\mu_{25})]}{1 + (1-\mu_{23})}$$
 (3)

Hence the problem of obtaining the breeding ratio is reduced to finding and manipulating the quantities μ_{23} and μ_{25} together with the ratio of moderator to U²³³ absorptions.

Multigroup calculations showed that these quantities are only slightly affected by the spectrum changes brought about by the presence of equilibrium concentrations of the higher isotopes. A treatment of the spectrum as though it is unperturbed by the higher isotopes yielded values of μ_{23} , μ_{25} , and $A_{\rm mod}/A_{23}$ within the necessary limits of accuracy, as shown in Table I.

TABLE I

B.R. vs Moderator and Moderating Power

$\xi \Sigma_{s}/N_{23}$	D ₂ O	С	Be*	H ₂ O
1000	1.151	1.147	1.145	1.141
2000	1.198	1.187	1.181	1.162
5000	1.228	1.194	1.175	1.118
10000	1.234	1.162	1.122	1.000

(New Cross Sections)

*Does not include the fast effect in Be.

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Pa²³³ Capture

As is evident from the high value of its resonance integral, Pa^{233} may capture a forbiddingly large number of neutrons when the spectrum is fast enough. This is a double loss, for each time a neutron is absorbed in Pa^{233} both a neutron and a potential fuel atom is destroyed.

The case of uniform mixture of equilibrium Pa^{233} with U^{233} is easy to examine. At equilibrium;

decays of Pa²³³ = absorptions in U²³³
decays of Pa²³³ ÷
$$\lambda_{13}$$
 = absorptions in Pa²³³ ÷ $\phi_{th} \left[\frac{\sigma_{13} + R_{13} \sigma_{th}}{p(\xi \Sigma_s / N_{23})} \right]$.(4)

Here λ_{13} , σ_{13} , and R_{13} are the decay constant, the thermal cross section, and the resonance integral of Pa^{233} , respectively, σ_{th} is the thermal cross section of the core material expressed in barns per U^{233} atom,p is the resonance escape probability and ϕ_{th} is the thermal flux.*

The thermal flux itself can be related to the specific power, P/M, and the cross sections; following Chernick,⁽³⁾

$$\phi_{\rm th} = \frac{1.2 \times 10^{16}}{[\sigma_{23}^{\rm F} + R_{23}^{\rm F} \sigma_{\rm th}/P(\xi \Sigma_{\rm s}/N_{23})]} \frac{P}{M} , \qquad (5)$$

where $\sigma \frac{F}{23}$ and R_{23}^{F} are the thermal fission cross sections and the fission resonance integral of U^{233} , respectively. Here the equivalent fission cross section in a thermal flux of U^{233} appears in the denominator. (Cross sections are in barns, P/M is in megawatts per kilogram of fuel.)

A combination of Eqs. (4) and (5) yields

$$A_{13}/A_{23} = \frac{\sigma_{13} + R_{13} \sigma_{th}/p(\xi \Sigma_{s}/N_{23})}{\sigma_{23}^{F} + R_{23}^{F} \sigma_{th}/p(\xi \Sigma_{s}/N_{23})} \frac{1.2 \times 10^{-8}}{\lambda_{13}} \frac{P}{M}$$
(6)

*The equivalent cross section in a thermal flux $[\sigma_{13} + R_{13}\sigma_{th}/p(\xi \Sigma_s/N_{23})]$ used above can be derived from a two-group model as follows:

Absorptions in Pa^{233} = thermal absorptions + resonance absorptions

$$N_{13}\sigma_{13} (equiv) \phi_{th} = N_{13}\sigma_{13}\phi_{th} + (N_{13}R_{13}/\xi\Sigma_s)q$$
; (a)

the thermal flux and the slowing-down density q are related by

$$pq = N_{23}\sigma_{th}\phi_{th} \qquad (b)$$

Equations (a) and (b) can be combined to give

$$\sigma_{13}(\text{equiv}) = [\sigma_{13} + R_{13}\sigma_{th}/p(\xi \Sigma_s/N_{23})]$$

For σ_{13} and R_{13} Stoughton and Halperin ⁽²⁾ give 70 and 1200 barns, respectively. Usually σ_{th} is not far from 600 or 700 barns.

Table II gives the ratio of Pa^{233} to U^{233} absorptions per unit of specific power and the effect on the breeding ratio, $\Delta B.R.$, per unit of specific power (taking into account the double loss involved in neutron capture by Pa^{233}). This is done for the two values of the resonance integral of Pa^{233} 900 and 1200 barns. Clearly under these assumptions the absorption in protactinium can be very high for large specific powers, P/M.

TABLE II

~~ /NI	$R_{13} = 90$	0 b	$R_{13} \approx 12$	200 Ъ
^{حک} / ^{۱۹} 23 s	$(A_{13}/A_{23})/(P/M)$	$\Delta BR/(P/M)$	$(A_{13}/A_{23}/(P/M))$	Δ B.R./(P/M)
1000 2000 5000 10000	0.124 0.068 0.033 0.023	-0.108 -0.061 -0.030 -0.021	0.163 0.087 0.041 0.027	-0.146 -0.079 -0.033 -0.024

Effect of Pa²³³ Absorptions

However, when the fertile thorium is in a blanket surrounding a fuel-bearing core, the concentration of Pa^{233} will not be so high as the equilibrium value used here (and even for high-flux cores only a small layer of the blanket will have such high epithermal fluxes as to give high absorptions in protactinium).

For specific powers up to 10 Mw/kg the decay rate of the protactinium is still much greater than the absorption rate, so that it still has an effective half-life (including burn-out) of 27 days. This means it takes about a month to come within 50 per cent of equilibrium.

Fast Fission in the Higher Isotopes

There is a bonus from fast fissions in U^{234} and U^{236} . For given $\xi\Sigma_s/N_{23}$ (taken at resonance energies, that is, from a few ev to a few hundred ev), the fast fission effect is larger in H_2O than in the other moderators examined here, since its cross section drops off so strongly at high energies. (This decrease in cross section leads to weaker slowing down and therefore to stronger flux.)

The contribution from U^{234} is considerably larger than from U^{236} because its equilibrium concentration and its fission cross section are larger.

For water moderation with $\xi \Sigma_s / N_{23} = 1000$, the ratio of U^{234} to U^{233} fissions is calculated from a spectrum based on slowing down by hydrogen:

$$\frac{F_{24}}{F_{23}} = \frac{F_{24}}{\mu_{23}A_{23}} = \left\{ \frac{N_{24}}{N_H} \int \frac{[s(u) + S(u)]\sigma_{24}^F(u)}{\sigma_H(u)} du \right\} / \mu_{23}A_{23} = 0.011 \quad . (7)$$
(Definitions: $s(u) = \text{fission spectrum}$
 $s(u) = \text{integrated fission spectrum} = \int_0^u s(u') du'$
 $\sigma_{24}^F = \text{fission cross section of } U^{234}$

 $\sigma_{\rm H}$ = cross section of hydrogen.)

This yields

$$\epsilon_{24} \simeq 1 + \frac{\nu - 1}{\nu} \frac{F_{24}}{F_{23}} = 1 + \frac{1.7}{2.7} 0.011 = 1.007$$
 (8)

The extra contribution to the breeding ratio is

$$\Delta B.R. = \frac{\nu_{24} F_{24}}{A_{23} + A_{25}} = 0.024 \qquad (out of a value of B.R. equal to 1.141) when the fast fission effect is ignored).$$
(9)

The contribution from U^{236} is much less, of the order of a tenth of this.

For carbon moderation at $\xi \Sigma_s / N_{23} = 1000$ there is obtained $\epsilon = 1.0015$, and the contribution to the breeding ratio is $\Delta B.R. = 0.005$.

Since the equilibrium ratio of N_{24}/N_{23} does not change very much over the range of moderating powers considered here, N_{24}/N_H will be about inversely proportional to the moderating power $\xi \Sigma_s/N_{23}$ and so will be the fast fission contribution.

Fission Products

Fission products are divided into two groups according to whether or not they saturate. For simplicity it is assumed that only those having cross sections greater than 10^4 barns saturate and that all the others have concentrations proportional to the fuel burnup.

Saturating Fission Products

These are Xe^{135} , Sm^{149} , and Sm^{151} . There are others with cross sections exceeding 10^4 barns, but their fission yields are too small to need to be considered here. The absorption rate, A_E , in a saturated fission product of species E is

$$A_{E} = (\gamma_{E,23} F_{23} + \gamma_{E,25} F_{25}) \frac{\sigma_{F}^{eq} \phi_{th}}{\lambda_{E} + \sigma_{E}^{eq} \phi_{th}} , \qquad (10)$$

where $\gamma_{E,23}$ is the fission yield of species E by fissions in U²³³, and F₂₃ is the fission rate in U²³³ (similarly for U²³⁵).

The factor outside the parenthesis in Eq. (10) is different from unity only for Xe¹³⁵ (among the saturating fission products being considered here) since the others are stable ($\lambda = 0$). In Eq. (10) $\sigma \stackrel{eq}{}_{E}$ is the equivalent cross section in a thermal flux defined in the section on Pa²³³ absorptions. (For Xe¹³⁵ with a cut-off between thermal and resonance groups of 0.5 ev, the resonance integral is zero and $\sigma \stackrel{eq}{} = \sigma$.) This competition factor is nearly unity at high fluxes, as may be seen from Table III.

TABLE III

65 /NI	¢ /(p/x)	σφ/(σ	φ + λ)
ξ ^Δ s/ N ₂₃	$\Psi_{\rm th}/(P/M)$	for $P/M = 10^{\circ}$	for $P/M = 10^1$
1000	1.3×10^{13}	0.65	0.95
2000	2.0×10^{13}	0.74	0.97
5000	2.3×10^{13}	0.77	0.97
10000	$2.4 \ge 10^{13}$	0.78	0.97

Competition Factor for Xe¹³⁵

The yields of Xe^{135} , Sm^{149} , and Sm^{151} are given by Eastwood <u>et al.</u>, ⁽⁴⁾ and are shown in Table IV.

TABLE IV

Fission Yields

E	$\gamma_{\rm E,23}$	$\gamma_{\rm E,25}$
Xe ¹³⁵	0.060	0.0641
Sm ¹⁴⁹	0.008	0.0113
Sm ¹⁵¹	0.003	0.0045

The fission rates at steady state are, according to Eq. (2) and the definition of μ , given by

 $F_{23} = \mu_{23}A_{23}$

$$\mathbf{F}_{25} = \mu_{25} \mathbf{A}_{25} = \mu_{25} \left(1 - \mu_{23} \right) \mathbf{A}_{23} \quad . \tag{11}$$

Equations (10) and (11) yield the values in Table V for the ratio of absorption in saturating fission products to fuel absorptions and the corresponding effect breeding ratio.

TABLE V

65 /2	T	P/M =	10 ⁰	P/M =	= 10 ¹
ζ _s /n	23	A_{FP}/A_{23}	Δ B.R.	A_{FP}/A_{23}	Δ B.R.
100 1000	0 0	0.051 0.058	-0.045 -0.053	0.067 0.069	-0.060 -0.063

Saturating Fission Product Absorptions

Nonsaturating Fission Products

These are taken to comprise all other species of fission products. The relevant cross sections are

 $\sigma_{\rm FP}$ = 45 barns/fission estimated from curves of Waller,(5)

and

 $R_{FP} = 200 \text{ barns/fission; Srikantiah.}(6)$

Thus, in terms of fractional fuel burnup, B, of fissionable material,

 $(N\sigma)_{FP}/N_{23} = \mu_{23} B \sigma_{FP}$

.

$$(NR)_{FP} / \xi \Sigma_{s} = \mu_{23} B R_{FP} / (\xi \Sigma_{s} / N_{23})$$
 (12)

Then the absorption in these fission products can be estimated from the equation

$$A_{FP} = (NR)_{FP} / \xi \Sigma_{s} + p(N\sigma)_{FP} / N_{23} \sigma_{th}$$
$$= \mu_{23} B \left\{ R_{FP} / (\xi \Sigma_{s} / N_{23}) + p \sigma_{FP} / \sigma_{th} \right\} .$$
(13)

Results are given in Table VI.

TABLE VI

Nonsaturating Fission Product Absorption

$\xi \Sigma_{s}/N_{23}$	$\frac{1}{B}(A_{FP}/A_{23})$	$\frac{1}{B}(\Delta B.R.)$
1000	0.21	-0.19
2000	0.14	-0.12
5000	0.10	-0.09
10000	0.09	-0.08

Additional Notes

A concession was made to the non-l/v behaviour of the U^{235} absorption and fission cross sections by taking a "non-l/v" factor for them of 0.9 in all of the calculations.

 U^{233} cross sections were the "new cross sections" of Chernick and Moore.⁽¹⁾ Resonance integrals for U^{234} and U^{236} as functions of self-shielding were obtained in a private communication from R. A. Vernon.

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Discussion of Paper Presented by Mr. Levine

MR. SPINRAD:

The paper is now open for discussion.

MR. SANDERS:

I would like to ask two questions. Firstly, do I understand correctly that these calculations that Dr. Chernick and you have been giving are for homogeneous mixtures of fuel and moderator and physically infinite systems?

MR. LEVINE:

That is correct.

MR. SANDERS:

Could you comment on the loss of breeding ratio likely to be obtained due to disadvantage factor in lumping fuels; is this likely to be a large effect? Secondly, could you comment on the value of the thorium resonance integral that you would think would be the value to take in these calculations?

MR. LEVINE:

Certainly, there will be important effects from the disadvantage factors. I think it is very difficult to estimate these without talking about a specific system, and I'm afraid I'm not prepared to quote these for any particular system. There is another effect, too, that will come into that, I think, and that is the effect of neutron temperature for the particular form of the arrangement of the materials.

As for the resonance integral of thorium, this has been side-stepped here by assuming that all of the neutrons that are not used up in one of the places that have been mentioned go into the fertile material.

MR. OKRENT:

I didn't catch that; are you including the gases in these fission product losses ?

MR. LEVINE:

Yes.

MR. OKRENT:

I wonder if you could be so kind as to indicate what these losses would do in a reasonable system.

MR. CHERNICK:

I think we can only talk about the systems that we know about and in these systems it appears that xenon will be a very small problem. The reason one has to be fairly specific here is that it does depend on the type of system. For example, the Oak Ridge people do not expect to take any appreciable losses, although they may use numbers of the order of 1 per cent to be on the safe side in the aqueous homogeneous reactor. On the other hand, in the slurry type, PAR, the design was such that they expected to take, essentially, full xenon loss. So it does depend on the choice of design. Again, with the graphite-moderated systems there is still some uncertainty about xenon holdup in the graphite structure. This hasn't been completely resolved, but on the other hand there is not yet any definite indication that the losses will be serious.

MR. REYNOLDS:

I am aware of the 1200-barn resonance integral that Stoughton and Halperin for protactinium and the 670-barn value that Smith of MTR reports, and I wondered where you obtained the 900-barn resonance integral?

MR. LEVINE:

This was mostly a reaction against the horror of the results with 1200 barns. I wanted to take a smaller number to exhibit the effect of possible uncertainties.

MR. SPINRAD:

Again I would like to make one closing comment if I might, and that is that apparently the net result of the various effects of higher isotopes is relatively small. If it were larger, it would again become significant, however, to consider the self-purified nature of the process; that is, when equilibrium is set up, there are probably too many higher isotopes in the fuel if you are constantly purifying it by removing the bred material. This can be a significant effect if the corrections you have just mentioned are themselves significant.

BREEDING RATIOS, DOUBLING TIMES, AND FUEL COSTS IN AQUEOUS HOMOGENEOUS THORIUM BREEDER REACTORS

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In a free economy a nuclear reactor station producing electrical power will normally operate under those conditions associated with minimum costs per electrical kwhr. However, due to lack of data and experience, it is difficult to predict nuclear power costs accurately, and so simpler concepts are often considered as criteria for desirable reactor configurations and operating conditions. Presumably fuel cost can be evaluated with more certainty than total power cost; however, minimum fuel cost is not necessarily associated with minimum power cost. Nevertheless, fuel cost is often used as a criterion for specifying desirable reactor conditions.

Further removed from that of power cost is the concept of doubling time. As considered here, doubling time is that time associated with doubling the fuel inventory associated with the entire fuel cycle under equilibrium conditions. Thus, fuel held up for cooling and decay, processing and storage is included, and the fuel composition leaving and entering the reactor proper is that associated with equilibrium, or steady-state, operating conditions. The results presented here give doubling times on the basis of an 80 per cent load factor for the reactor station. Use of doubling time as a criterion for desirable reactor conditions implies that the economic conditions of the future will be such that doubling time is a predominant economic factor.

Even further removed from power cost is the breeding ratio of the reactor. In this study breeding ratio is defined as the net rate of fissionable-fuel production relative to the fuel-destruction rate, and thus considers the fuel processing losses. The breeding ratio determines the net yield of fuel from a reactor and thus does have some influence upon fuel cost; however, based on present economic conditions, this influence per se is not appreciable for one per cent change in breeding ratio.

The reactors considered here were two-region systems in which the core region contained UO_2SO_4 dissolved in heavy water, while the blanket region contained thoria, urania, and heavy water. The thorium concentration in the blanket was considered to be 1000 g Th/liter of slurry. Four - and five - ft diameter cores were considered along with various blanket thicknesses and blanket-fuel concentrations. The power level was normally 200 Mw thermal. Since the fuel fluid is circulated through an external heat exchanger, there is a relatively large fuel inventory outside the reactor

proper The external inventory was based on an equivalent heat removal capability of 20 kw per liter of core fluid and 14 kw per liter of blanket slurry

Based on economic conditions at this time, operation of aqueous homogeneous reactors at minimum fuel cost does not lead to minimum doubling times This is indicated in Fig 1, which gives power cost as a



Fig 1 Effect of Temperature on Unit Cost (Conditions given in ORNL-1810)

function of reactor temperature for reactor core surrounded by a blanket containing 1000 g Th/liter and 3 g U^{233} /liter The absolute values given on the ordinate are based on what are now believed to be some optimistic assumptions and so the given values are too low, however, the trend shown 1s believed to be significant The factors considered in obtaining these results were those associated with changes in station efficiency and turbo-generator costs with increasing fluid temperature As indicated, the minimum power cost for the assumed economic conditions was associated with an average reactor temperature of about 320°C However,

there are physical limitations on the operating temperature, since for temperatures greater than ~ 320 °C the fuel solution may be unstable Because of this, the average reactor temperature is normally assumed to be 280 °C Neglecting this limitation, the power cost would tend to decrease with increasing temperature up to about 320 °C, but for a given reactor the breeding ratio would decrease and the doubling time would increase In order to reduce doubling time, the reactor temperature should be as low as possible, although there are practical limitations in this direction also

There is considerable leeway with regard to reactor specifications without affecting fuel costs to a marked degree This is shown in Fig 2, which gives the fuel cost as a function of blanket thickness, blanket thorium concentration and core diameter These results indicate that certain design and operating conditions can be changed significantly without affecting fuel costs appreciably The variation would depend on the particular economic conditions which applied The results⁽¹⁾ given here are based on a 12 per cent inventory charge for fuel, moderator, and fertile material, and a fuel



Fig. 2. Effect of Blanket Thickness on Fuel Cost (Conditions given in ORNL-1810)

value of $\frac{20}{g}$ for fissionable fuel; moderator value was $\frac{40}{lb}$, and processing charges were $(\frac{3}{kg} Th) + (\frac{50.50}{g} U) + (\frac{50.35}{l} D_2 O)$ plus a fixed charge for processing of 0.75 mill/kwhr electrical.

Although doubling time is often tactically assumed to be relatively independent of costs, this cannot be so in a general sense, since doubling time can be decreased by increasing the rate of fuel processing, which in turn increases processing costs. It is also clear that there is a minimum doubling time associated with the fuel cycle. If the fuel is processed too rapidly, there is a tendency for the inventory in the processing plant to become predominant. Too slow a processing rate, on the other hand, leads to high fuel concentrations in the reactor and a correspondingly high reactor inventory. The specific processing scheme considered for these reactors is indicated in Fig. 3. Two types of processing were assumed; the blanket was considered to be processed by the Thorex process, while the core fuel was processed by means of hydroclones as well as Thorex. The blanket material was assumed to be held up 55 days before initial processing, after which time an additional 150 days was associated with the remaining Pa^{233} to permit its decay to U^{233} ; at the end of this time the U²³³ was separated from the fission products.

The fission products in the core were divided into three groups. Group 1 considered the high cross-section gaseous fission products; these were assumed to be removed from the system at such a rate that the equivalent poison fraction of this group was 1 per cent (the poison fraction was taken to be the ratio of the poison-to-fission macroscopic cross sections). Group 2 considered the high cross-section nongaseous fission products; these were assumed to contribute a poison fraction of 0.8 per cent.



Fig. 3. Schematic Fuel-processing Flowsheet for a Two-region Homogeneous Thorium Breeder Reactor

Group 3 constituted the remaining fission products. It was assumed that 75 per cent of the Group 3 fission products could be removed from the core as insoluble oxides by means of the hydroclones. These fission products are essentially the rare earth elements and were designated as Group 3A. The remaining 25 per cent of the Group 3 poisons were designated Group 3B, and assumed to be soluble and removed only by Thorex processing (these poisons correspond for the most part to gaseous fission products which decay rapidly to elements that are soluble). It was further assumed that the amount of Group 3A poisons present in the reactor was equivalent to 1 per cent poison fraction (this corresponds to the solubility associated with the solubility product and an allowance for the circulation of a small portion of these solids).

The effective cross ection of the Group 3 poisons will be a function of the age of the fission products in the reactor. Hurst(2) calculated the effective cross section of the Group 3 fission products as a function of neutron exposure, and found that the 2200-m/sec value varied from about 46 b/fission at time zero to about 18 b/fission after 5 fuel burnups. The core cycle time associated with such a burnup will be a function of reactor design and operating conditions; for the reactors considered here the cycle time associated with such a burnup will be a function of reactor design and operating conditions; for the reactors considered here the cycle time associated with 5 fuel burnups would be about 300 days. In most of the studies the cross section for the Group 3 fission products was taken as 20 b (2200-m/sec value) independent of core cycle time. (In practice the cross section will be a function of the core cycle time and this variation should be considered.)

The effect of poison cross section on breeding ratio and fuel cost is shown in Fig 4 as a function of poison level in the core. As shown, increasing the poison cross section increased fuel cost and decreased breeding ratio. The breeding ratio curves decreased at low poison levels



Fig. 4. Effect of Group-3 Poisons on Fuel Cost and Breeding Ratio (Conditions given in ORNL-1810)

because of processing losses. At 0.7 per cent variable poison fraction and processing losses of 0.1 per cent per cycle, the decrease in breeding ratio due to processing losses was about 0.003. The poison fraction at which the minimum fuel cost occurs will be a function of the assumed economic conditions. The results given in Fig. 4 are based on the same cost bases used in obtaining Fig. 2.

During circulation of the fluid there will be some corrosion of the wall surface. Because of the relatively large area of the heat exchanger surface, nearly all

the corrosion products result from corrosion in the heat exchanger itself. Since a relatively small fraction of the total power is generated in the blanket region and the fertile material competes effectively with corrosion products for neutrons (for blanket cycle times of interest and observed corrosion rates in slurry systems), the primary effect of corrosion on reactor performance is associated with the corrosion in the core circuit. In comparison with results based on zero corrosion, corrosion will increase the fuel cost and also the doubling time. The corrosion products consist primarily of the oxides of iron and chromium, and soluble forms of nickel and manganese. With a solution-type core, the insoluble products can be removed by hydroclones; however, nickel and manganese would not be removed, and since these constitute about 15 and 9 per cent, respectively, of the stainless-steel cross section, corrosion can lead to a significant increase in poison fraction for a given cycle time. At present an electrolysis process⁽³⁾ has operated successfully in the laboratory which removes essentially all of the nickel and manganese. In using such processing, a side stream would be taken from the reactor core, sent to the electrolysis process, and returned to the core circuit after the removal of nickel and manganese. There would be an equilibrium amount of nickel and manganese in solution which would be a function of the cycle time, but the Mn and Ni could be readily kept at levels such that the associated poison fraction would be less than 0.005.

The nickel and manganese would also be removed in the Thorex process, and by decreasing the Thorex core cycle time the poison fraction due to the Ni and Mn would be decreased. Decrease in core cycle time will increase the processing cost, but not significantly so long as the core fuel can be added to the blanket processing stream without increasing the effective cost per kg thorium processed; however, there would be an increase in processing losses due to the increased processing rate. With zero corrosion rate and 0.1 per cent fuel loss per core cycle, the variable poison fraction can be decreased to 0.007 before there is any significant decrease in the breeding ratio (associated with the processing loss). The core cycle time for such a variable poison fraction would be from 70 to 100 days depending on the particular reactor. For a corrosion rate of about $\frac{1}{2}$ mil/yr in the core circuit (equivalent to doubling the yield of the fission product poisons), the effect of corrosion would either increase the variable poison fraction to 0.014 with a core cycle time of 70-100 days or would decrease the breeding ratio by 0.005 if the core cycle time were 35-50 days.

Figure 5 gives the relations between core cycle time (T_c) , fuel cost, and breeding ratio (B.R.) as a function of variable poison fraction with zero corrosion and also with a corrosion rate of 1 mil/yr.(4) Such a corrosion rate is equivalent to increasing the fission product yield by a factor of 3, and so for the same variable poison fraction the effect of corrosion would decrease the core cycle time by about a factor of 3. In obtaining these results the processing cost was assumed to be 50 cents/g of U processed,





and so the fuel cost increased as the core cycle time decreased. The particular curves shown in Fig. 5 for breeding ratio correspond to a 0.4 per cent fuel loss per core cycle; a fuel loss of 0.1 per cent/cycle is believed attainable (although this is yet to be demonstrated in a large plant) and for this circumstance the breeding ratio curves have no maximum for the variable poison fractions shown in Fig. 5. Rather, the breeding ratio increases linearly with decreasing poison fraction both with and without corrosion. For the reactor considered in Fig. 5, a vari-

able poison fraction of three per cent and zero corrosion would correspond to a core cycle time of about 600 days; if the average corrosion rate were 1 mil/yr, the associated core cycle time would be about 200 days, which would decrease the breeding ratio by 0.002. It is believed that the average corrosion rate in reactor systems will be $\frac{1}{2}$ mil/yr or less.⁽⁵⁾

The results in Fig. 2 indicated that changes in some reactor conditions have relatively little effect on fuel cost; Fig. 6 illustrates that several reactor designs can be considered which lead to about the same doubling time. For the reactors considered, as the blanket thickness increased the breeding ratio increased, but the doubling time stayed about the same.



Fig. 6. Breeding Ratio and Doubling Time for Various Reactor Sizes

The results shown assume that the blanket contained 3 g of $U^{233}/liter$; the 4-ft diameter core reactor had the higher breeding ratio and the lower doubling time. The minimum doubling time was associated with a blanket thickness of about $2\frac{1}{2}$ ft.

An important parameter affecting doubling time is the blanket fuel concentration. Figure 7 gives the doubling time and fuel cost as a function



Fig. 7. Doubling Time and Fuel Costs vs. Blanket Fuel Concentrations (HRE-3)

of blanket fuel concentration; the reactor with the 4-ft diameter core and 9-ft diameter pressure vessel had lower doubling times and fuel costs. The results shown are based on equilibrium calculations⁽⁶⁾ with 3g U^{233} /liter in the blanket; for lower blanket fuel concentrations the reactor inventory was decreased in accordance with the change in blanket fuel inventory, while fuel costs increased in accordance with the appropriate blanket cycle time and a processing charge of 28/kg Th. Recent results from equilibrium calculations with a blanket concentration of 1 g U^{233} /liter gave doubling times very close to those shown in Fig. 7.

The doubling time will tend to decrease as the poison fraction in the core is decreased; however, this trend cannot go on indefinitely, since eventually the inventory held up outside the core for processing will become predominant, causing the doubling time to pass through a minimum with decreasing core poison fraction. With zero corrosion, the core poison fraction corresponding to minimum doubling time was from 4 to 5 per cent poison fraction; based on present economic conditions, minimum fuel cost appears associated with a core poison fraction of about 7 per cent, and a blanket fuel concentration of 4 g U²³³/kg Th.⁽⁷⁾ The results given above are based on calculations for spherical reactors. For cylindrical geometry the results will be somewhat different. Figure 8 gives results for a reactor having a 4-ft x 12-ft cylindrical core surrounded by a 2-ft-thick blanket. The fuel cost, breeding ratio, and doubling time are given as a function of blanket fuel concentration. As shown, the breeding ratio was rather insensitive to the blanket fuel concentration for the values considered. For the assumed processing cost of 28/kg Th, the doubling time can be decreased from about 20 to 10 years by increasing the fuel cost by about 0.5 mill/kwhr.



Fig. 8. Fuel Cost, Doubling Time and Breeding Ratio vs. Concentration

There are several factors which contribute to neutron losses in aqueous homogeneous reactors. Table I lists those which should be considered in obtaining breeding ratio and doubling time. The neutron losses (given in terms of neutron losses per 100 neutron absorptions in fuel) will be a function of the specific reactor design. For example, a 5-ft dia Zr - 2 core absorbs about 3 neutrons per 100 fuel absorptions; in a 4-ft dia core the loss would be about 2 (wall thicknesses were 0.5 and 0.4 in. for the 5 - and 4-ft dia cores, respectively).

TABLE I

Neutron Lo	ss	e	s
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Moderator Fission Product Poisons	Copper Nickel, Manganese		
Higher Isotopes	Leakage		
Core Wall	End Effects		
Blanket Shroud	Protactinium		

The addition of 0.01 g mol of copper/liter of solution (the copper effectively recombines decomposition gases) increased neutron losses by about 0.6 per 100 fuel absorptions in a 5-ft dia core and by about 0.3 in a 4-ft dia core. The presence of 0.25 per cent H_2O in heavy water absorbed about one neutron per 100 fuel absorptions in a 5-ft core and about 0.5 per cent in a 4-ft core. Table II lists typical neutron losses per 100 neutron absorptions in fuel for a reactor having a 4-ft core diameter, a 9-ft pressure-vessel diameter, 1000 g Th/liter in the blanket, and 3 g U²³³/liter in the blanket.

TABLE II

Material	Neutron loss (neutrons/100 neutrons absorbed 1n fuel)		
Moderator (0 25% H ₂ O)			
Core	2 2		
Blanket	0.2		
Fission Product Poisons			
Core	5 0 ^a		
Blanket	0.2		
Higher Isotopes			
Core (\overline{U}^{235}) effective	1 6 ^b		
U^{236}	0 5		
Blanket U^{234} , U^{235} , U^{236}	0 2°		
Core Wall (0 4 in thick)	2 0		
Blanket Shroud (0.25 in. thick,			
3 in from core wall)	0 6		
Cu (0 025 molal)	0 8		
N1 and Mn $\left(\frac{1}{2}$ -mpy corrosion,			
no electrolytic processing)	2 6R ^d		
Leakage	2 0		
End Effects:			
neck	0 4		
8-in protrusion	0 2		
Pa ²³³ 1n blanket	2 0		
Delayed neutrons	0.7		
Corrosion Product Solids	0 6 ^e		
Total	21 8		

Typical Neutron	Losses in Aqueous	Homogeneous	Reactor	Having 4-ft
Core Diameter	(blanket thickness,	$2\frac{1}{2}$ ft, blanket u	ıranıum,	$3 g U^{233}/\ell$;
solution core, 280°C)				

^a About 2 5 of this is associated with fixed poisons (Xe, Sm, circulating group 3A poisons); the corresponding core cycle time is about 400 days (no corrosion)

^b Corresponds to effective loss associated with the lower value of η^{25} relative to η^{23}

^C Not all of this is a loss

dR is ratio of core cycle time to 400 days

^e Corresponds to $\frac{1}{2}$ -mpy corrosion rate and assumes that $\frac{1}{2}$ % by weight of solids removed by hydroclones is uranium which is not recovered

The listed corrosion products are based on Thorex being utilized to remove nickel and manganese corrosion products. Use of the electrolysis process would keep the nickel and manganese at a concentration of 0.005 g mol nickel/liter, which would correspond to about 0.4 neutron loss/100 fuel absorptions. The effective loss associated with U^{235} takes into consideration that the value of eta for U^{235} is lower than the eta for U^{233} . The end effects were calculated⁽⁸⁾ by means of a two-dimensional calculation (PDQ program); the reactor was considered to be cylindrical with a neck at the top of the core. The pipe bringing fluid into the core was assumed to extend 8 in. into the core region.

Most of the calculations performed assumed the two-group model, although a few multigroup calculations were also performed. The multigroup results usually gave lower leakages than the two-group results, but otherwise the results agreed quite well. The effective thermal cross section of Pa²³³ was taken to be 146 b. The blanket shroud was placed in the blanket region to permit a high fluid velocity near the core wall for cooling purposes. This shroud was assumed to be $\frac{1}{4}$ in. thick and placed 3 in. from the wall.

The corrosion- and fission-product solids (removed by the hydroclones) will adsorb uranium, and some of this uranium will be difficult to recover. The losses given in Table II for Corrosion Product Solids correspond to such uranium losses. Actually, such losses directly decrease the effective breeding ratio, but they can also be converted into effective neutron losses, as was done here. For a corrosion rate of $\frac{1}{2}$ mil/yr and $\frac{1}{2}$ per cent of the hydroclone-removed solids being uranium, the effect of change in breeding ratio would be about 0.006.

The results given previously in Figs. 4-8 did not include all the neutron losses listed in Table II; the net effect of including all of them would increase the doubling time from 2 to 5 years, depending on the specific reactor and the treatment of corrosion products. The results given in Table II indicate a breeding ratio of 1.07 (for $\eta^{23} = 2.29$); if electrolytic processing were employed, the breeding ratio would increase to 1.09.

The relation between doubling time and fuel cost is dependent upon the economic conditions which apply. It is clear that a decrease in fuel value will <u>decrease</u> the minimum fuel cost but will <u>increase</u> the doubling time associated with minimum fuel cost. As the fuel value increases, there will be a tendency to build a reactor which has a fuel inventory charge equal to the credit obtained from the net fuel production. Thus, as the fuel value increases the minimum fuel cost will increase and the optimum doubling time (associated with minimum fuel cost) will decrease. For the conditions assumed in this study, the minimum doubling time under optimistic conditions appears to be about 10 years (80% load factor); taking into account presently known factors may increase this doubling time by 3 to 5 years. To decrease doubling time significantly it will be necessary to increase the heat-removal capability of the external core circuit or to decrease the holdup time associated with fuel processing.

It appears that homogeneous reactors have doubling times which are rather insensitive to reactor power. This results from the high inventory of fuel held up outside the core, which is a function of the reactor power level. Thus a 60-Mw thermal reactor has about the same doubling time as a 500-Mw system. Aqueous homogeneous reactors appear unique as thermal reactors inasmuch that based on today's economic conditions those reactors which have minimum fuel costs are also breeders with doubling times from 20-30 years. By increasing processing costs by about 1 mill/kwhr, these doubling times can be reduced to 10-20 years.

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Discussion of Paper

Presented by Mr. Kasten

MR. GREEBLER:

What did you say was the plant utilization factor you were assuming in these calculations of doubling time?

MR. KASTEN:

We assumed that the doubling time was based on a 80 per cent load factor and the doubling time of the fuel cycle inventory. There was no factor of a Log_{10} in here based on a system inventory doubling time.

MR. FREUND:

Would you comment on single region versus two region in regards to some of the conclusions you drew on fuel cost and so on?

MR. KASTEN:

Based on today's economics, the two-region reactor has lower fuel costs than the single-region reactor. There are several factors here, one being that in the one-region reactor, a slurry would be used and you cannot remove the fission products separately in a hydroclone processing cycle like you can in the two-region reactor. The most economic single-region reactor is about a break-even system. The breeding ratio, in itself, is not a determining criteria if today's economics are used.

LIQUID METAL THORIUM BREEDER REACTORS

C. E. Thomas Babcock and Wilcox Company Lynchburg, Virginia

The Babcock and Wilcox Company, under contract to the Atomic Energy Commission, is currently engaged in a comprehensive study to evaluate the technical and economic feasibility of the Liquid Metal Fuel Reactor Concept as a thorium breeder.

Reactor Design

The Liquid Metal Thorium Breeder Reactor (LMTBR) systems employ graphite as moderator, liquid bismuth as the coolant and fuel carrier, thorium as the fertile material, and U^{233} as the primary fissionable material. The reactor designs may be classified according to the method of heat removal. If the fission heat is generated in the fuel-coolant stream and transported to a heat exchanger external to the core, the reactor is said to be externally cooled. If the fission heat is generated in the fuel solution and transferred to a separate coolant in the core as in fixed fuel reactors, the reactor is internally cooled.

Both types of reactors have been studied, and the externally cooled type is currently favored because of its ease of construction based upon present technology. The internally cooled core has the advantage of a lower fuel volume and higher breeding ratio potential, but it has the limitation on power density imposed by heat transfer from the fuel through the graphite wall to the coolant, and is more complex mechanically to build. While both types of designs are worthy of consideration, only work concerning the externally cooled concept will be discussed further in this paper for the sake of brevity.

While single-region reactors can be "hold-your-own" breeders, the influence of specific power on doubling time requires the use of two-region designs - i.e., a core region surrounded by a blanket region - where doubling time is an important consideration.

The consideration of an externally cooled core is made possible by a major improvement in the primary system design. The reactor core and blanket, the primary heat exchanger, and primary pumps are all located in one vessel. This compact arrangement has reduced the external volume of fuel required to transport heat by a factor of three to four over previous LMFR designs.⁽¹⁾ The fuel enters the reactor at 1000°F and exists at 1300°F. The only materials exposed to the 1300°F fuel are tantalum and graphite; all other primary system materials are exposed isothermally to 1000°F fuel.
Breeding Ratio

For an extremely large homogeneous medium of U^{233} and graphite, Chernick and Moore⁽²⁾ have calculated a maximum breeding ratio of 1.22, corresponding to a value of $\xi \Sigma_{\rm s}/N_{23} = 2,000-4,000$. This value of breeding ratio takes into account the reduction in eta due to resonance absorption and the neutron losses due to captures in the graphite. Utilizing the tworegion design, the maximum practical breeding ratio is approximately 1.22, including the reduction in η from resonance absorption and the neutron losses from absorption in the bismuth and graphite.

Material Cross Sections

The high potential breeding ratio of the LMTBR is due to the excellent moderating ratio $(\xi \sigma_s / \sigma_a)$ of the graphite and the low neutron cross section of bismuth. Bismuth has a thermal absorption cross section of 0.032 barn and is primarily a 1/v absorber. The contribution of resonance absorption in bismuth has been calculated to be in the range from 0.7 to 1.6 per cent of the thermal cross section.⁽³⁾ At a temperature of 1150°F liquid bismuth has a density of 9.64 g/cc. This results in a macroscopic thermal absorption cross section at 2200 m/s of 0.000898 cm⁻¹.

Commercial grades of nuclear graphite are currently available with a microscopic thermal absorption cross section at 2200 m/s of 0.0037 barn. At a density of 1.80 g/cc, then, Σ_a (2200 m/s) is 0.0003346 cm⁻¹.

Reactor Design

The LMTBR reactor designs do not require cladding material, metal core tank, or other structural material in the core and other high-flux regions. The liquid bismuth is compatible with high density and unclad graphite, and the core and blanket fluids are separated by proper arrangement and fabrication of graphite blocks to eliminate the need for a core tank.

The principal problem of designing reactors with high breeding ratios is the development of a blanket region which contains a high concentration of thorium and completely surrounds the core region containing a low concentration of thorium. Feasible core and blanket designs have been developed which achieve this objective.

The variation of breeding ratio with core diameter and thorium content in the core is shown in Figs. 1 and 2. Although thorium is not contained directly in the fuel solution, the amount of thorium in the core region can be easily varied by the reactor designer. The amount of thorium in the core region determines the relative breeding between the core and blanket. For



Fig. 1. Breeding Ratio vs. Core Diameter $(N_{23}/N_{Bi} in$ Blanket = 90 x 10⁻⁶)

Fig. 2. Breeding Ratio vs. Th Concentration in Core Bi $(N_{23}/N_{Bi} \text{ in Blanket} = 90 \times 10^{-6})$

a fixed thorium concentration in the core, neutron leakage to the more efficient blanket region decreases as diameter increases; therefore breeding ratio decreases. For a fixed core diameter, an increase in the thorium content of the core region improves the breeding efficiency of the core and increases the overall breeding ratio (see Fig. 2). All results shown in the graphs were obtained with a 3-ft thick blanket having a slurry-to-graphite volume ratio of 1.0 with 10 w/o thorium in the ThO₂-Bi slurry.

Fission Product Poisons

Research and development have shown that xenon can be removed continuously from the liquid bismuth fuel solution; therefore, it appears that the neutron loss from absorption in volatile fission products can be held to approximately 0.01. Other fission products build up more slowly, but will constantly decrease breeding ratio until they are removed from the fuel by chemical processing. The LMTBR has the advantage common to all fluid fuel reactors, that is, fission products can be removed continuously by chemical processing on a rapid cycle. It appears practical to maintain concentration levels of the nonvolatile fission products by chemical processing such that the reduction in breeding ratio can be held to approximately 0.03 under equilibrium conditions. Thus with proper chemical processing rates, the total neutron loss to all fission products may be kept to approximately 0.04.

Higher Uranium Isotopes

By recycling the fuel, the higher uranium isotopes: U^{234} , U^{235} , and U^{236} will gradually build up to equilibrium values. The U^{234} isotope may be considered a fertile material, since a neutron absorption in this isotope results in the production of U^{235} , a fissionable element. Since the average value of eta for U^{235} is lower than that for U^{233} , the average value of eta for the mixture of U^{235} and U^{233} will be reduced approximately 0.02 under equilibrium operating conditions. The radiative captures in U^{235} result in the production of U^{236} , which acts as a neutron poison. Under equilibrium conditions this poison will decrease the breeding ratio by approximately 0.01. Thus, the total net loss in breeding ratio from higher uranium isotopes will be approximately 0.03. These values assume that the net fuel produced is removed from the core and contains equilibrium concentrations of the higher isotopes.

Protactinium Losses

Neutron absorption in the fertile material, thorium, results in the production of Pa^{233} , which decays to U^{233} . Since the half-life of Pa^{233} is 27.4 days, substantial concentrations of this isotope will be present in the reactor. Neutron absorption by Pa^{233} results in a poison effect as well as a reduction in the amount of U^{233} resulting from the absorption in thorium. This loss in breeding ratio due to Pa^{233} is dependent upon the neutron flux to which the Pa^{233} is subjected and the chemical processing rate for the removal of this isotope from the reactor. The average flux in the blanket is lower by an order of magnitude than the average flux in the core; therefore, it is mandatory that the residence time of thorium and Pa^{233} in the core region be small compared to the residence time in the blanket region and external loop.

For the flux values associated with the LMTBR designs, the Pa^{233} losses may vary from less than 0.01 for rapid chemical processing rates of the blanket to 0.028 for very slow processing rates. The effective resonance integral for Pa^{233} contains a large uncertainty; therefore this loss in breeding ratio cannot be predicted accurately. With chemical processing economics and reactor parameters as variables, however, the loss in breeding ratio due to Pa^{233} can probably be kept in the range of 0.01-0.03 for many types of designs.

Neutron Leakage

The neutron losses due to leakage may be held very low by proper blanket design. In the LMTBR a blanket containing 50 per cent graphite by volume and 50 per cent thorium oxide-bismuth slurry (10 w/o thorium) with a thickness of 3 ft will keep the neutron leakage loss to less than 0.01 for almost all core designs considered in this study.

A neutron balance is shown in Table I for the reactor core having a diameter of 6.5 feet and a thorium content of 1.5 w/o of the bismuth. The ratio of bismuth to graphite is 1.25 by volume. The neutron losses are listed for equilibrium operating conditions for two concentrations of fuel in the blanket fluid. The low concentration $(N_F/N_{Bi} = 90 \times 10^{-6})$ represents rapid processing of the blanket fluid, and the fuel-to-bismuth atom ratio of 500 x 10^{-6} corresponds to a moderate processing cycle time of approximately 150 full-power days. The difference in breeding ratio between the two cases is due primarily to the difference in protactinium losses and leakage. The values of doubling times reflect these differences in breeding ratio as well as the difference in inventory due to the fuel in the blanket system.

TABLE I

Neutron Balance

	N_{F}/N_{Bi} in Blanket		
	90×10^{-6}	500×10^{-6}	
Absorptions in U ²³³	0.920	0.927	
Absorptions in U ²³⁵	0.080	0.073	
Absorptions in Th ²³²	1.036	1.028	
Absorptions in U ²³⁴	0.089	0.087	
Absorptions in Graphite	0.011	0.010	
Absorptions in Bismuth	0.034	0.031	
Absorption in Fission Products	0.024	0.022	
Absorptions in U ²³⁶	0.008	0.007	
Absorptions in Pa ²³³	0.009	0.022	
Leakage	0.009	0.015	
Chemical Processing and	0.010	0.010	
Miscellaneous Losses			
Average Value of Eta	2.230	2.232	
Breeding Ratio	1.116	1.093	
Specific Power, Mwh/kg	2.9	2.4	
Doubling Time, yr	6.8	10.6	

(Based upon eta thermal for U^{233} of 2.28)

Specific Power

Specific power is related directly to the total fuel inventory in the reactor system and the power capability of the reactor. Figures 3 and 4 show the total fuel inventory as functions of thorium concentrations in the core, core diameter, and fuel concentrations in the blanket.



Blanket = 90×10^{-6})

Fig. 4. Fuel Inventory vs. N_{23}/N_{Bi} in Blanket (Core Diameter = 6.42 ft.)

The external system volume of the core solution is dependent upon the power level of the reactor, which in turn is dependent upon the core diameter. The complex relationship among the parameters - critical fuel concentration, core diameter, power level, and thorium concentration in the core - are shown in Fig. 3. Fuel inventory decreases as core diameter increases, but the decrease is significant only for the core that has 1.5 w/othorium in the core. For any diameter the fuel inventory increases substantially with the increase of thorium content in the core because of the increase in critical fuel concentration.

The effect of fuel concentration in the blanket fluid on fuel inventory is shown in Fig. 4. Since the total volume of the blanket fluid is comparable to the total volume of core fluid, an increase in equilibrium fuel concentration in the blanket fluid substantially increases the total fuel inventory.



Fig. 5. Specific Power vs. Core Diameter

The results of these variations upon specific power are shown in Fig. 5. In this graph specific power is plotted against core diameter for three values of thorium concentration in the core region. The abscissas at the top of the graph gives the power capability of the reactor corresponding to the various core diameters. These values of power capability, together with the inventories shown in Figs. 3 and 4, were used to compute the values of specific power.

The specific power shown in this graph is in the units of Mwh/kg of fissionable material as normally defined. This is a measure of merit in the production of fissionable material in breeder reactors. Specific power expressed in units of Mwe/kg of fuel is a measure of merit in the production of electrical power. In a power reactor economy the latter definition has more significance, not

only in economics, but also in the efficiency with which raw materials are used in the production of electricity. This definition emphasizes the advantage of high thermal efficiency (43 to 50 per cent) obtainable in the Liquid Metal Thorium Breeder Reactors.

Doubling Time

Doubling time in full-power years is defined by the equation

D.T. = $1/S.P. \times B \times G \times 365$ where S.P. is specific power (Mwh/kg), B is fuel burned (kg/MWD), and G is breeding gain (B.R. - 1).

Doubling time has been computed for a series of LMTBR designs, and the results are shown in Figs. 6, 7, and 8. For low fuel concentrations in the blanket fuel $(N_{23}/N_{Bi} = 90 \times 10^{-6})$, the minimum doubling times for each thorium concentration in the core are approximately equal (see Fig. 6). If the ratio of fuel to bismuth in the blanket is allowed to build up to higher concentrations, 500-1,000 $\times 10^{-6}$, the most attractive doubling times are obtained with 1.0 to 1.5 w/o thorium in the core bismuth (see Figs. 7 and 8).



Fig. 6. Doubling Time vs. Core Diameter $(N_{23}/N_{Bi} \text{ in} B \text{ lanket} = 90 \times 10^{-6})$



Fig. 8. Doubling Time vs. Core Diameter $(N_{23}/N_{Bi} in Blanket = 1000 \times 10^{-6})$





While the increase in fuel in the blanket has little effect upon the overall breeding ratio of the reactor, the increased inventory causes the doubling time to be higher for the cases of higher concentrations in the blanket. The parametric studies represented by these graphs indicate that LMTBR designs will provide doubling times of 10 to 15 years with a wide latitude in many design and economic parameters, such as core diameter, thorium concentration in the core, and fuel concentration in the blanket - which corresponds to chemical processing rate.

Reactor specifications for a typical design are listed in Table II.

Economics

Several previous LMFR studies⁽⁴⁾ conducted by the B and W Company have illustrated the economical potential of nonbreeding LMFR designs. These studies were based upon one reactor per station and a station capacity of 315 Mwe. The range of energy costs may be broken down as follows:

Fixed charges on capital investment	-	4.1-4.6 mills/kwh
Operation and Maintenance	-	1.1-1.6 mills/kwh
Fuel Costs	-	1.1-1.6 mills/kwh
TOTAL	_	6.3-7.8 mills/kwh

TABLE II

Reactor Power, Mwh 1130 Reactor Power, Mwe 500 Number of Reactors per Station 2 Core: 6.5 Height, ft 6.5 Diameter, ft 0.45 Volume fraction of graphite Volume fraction of U-Bi fuel solution 0.47 Volume fraction of Th-Bi slurry 0.08 1540×10^{-6} N_{23}/N_{Bi} in U-Bi fuel solution Thorium to Bismuth ratio, w/o 1.5 Mass of fuel in core system, kg 330 Blanket: Thickness, ft 3.0 Volume fraction of graphite 0.50 0.50 Volume fraction of slurry Thorium content of slurry, w/o 10 $90-500 \times 10^{-6}$ N_{23}/N_{B_i} in blanket fluid Mass of fuel in blanket system, kg 23-130 Specific Power, Mwe/kg 1.0-1.25

Reactor Design Specifications

A preliminary evaluation of breeder economics indicates that energy costs for a LMTBR will depend somewhat upon the incentive for a minimum doubling time, but good "doublers" can be designed with potential costs equal to or better than those previously reported.

Capital Investment, Operation and Maintenance

The more important factors which contribute to lower costs for the LMTBR designs over previous LMFR designs are summarized below:

Improved engineering designs have led to more compact primary systems contained within the reactor vessel, elimination of primary piping,

reductions in shield requirements, and lower building and primary system costs.

The steam cycle has been modified and improved to provide increased thermal efficiency - thus, lower reactor thermal power and reduced costs for the heat transport system.

Maintenance has been simplified and maintenance costs have been reduced by the improved primary system design.

If breeder economics is developed for a station of 1000-Mwe capacity, the larger sized units will result in reduced costs. Also land, buildings, and maintenance equipment will serve two or more reactors on the same site with little or no additional costs.

Preliminary cost estimates based upon previous studies indicate the capital costs listed below for a 1000-Mwe station.

Number of	Capital	Annual	Fixed Charges
Reactors per	Investment	Amortization	on Capital
Station	(dollars)	Rate (%)	Investment, Mills/kwh
2	178,000,000	14	3.6
3	216,000,000	14	4.3
4	232,000,000	14	4.6

To these costs, maintenance and plant operation cost must be added. These costs are estimated to be 0.6 mill/kwh and 0.4 mill/kwh, respectively. Plant costs, excluding fuel costs, should range between 4.6 and 5.6 mills/kwh.

Fuel Economy in a LMTBR

The fuel costs of a LMTBR are composed of the following items: (1) bismuth inventory, (2) thorium inventory, (3) fuel inventory, (4) fuel sold, and (5) chemical processing.

The net fuel produced in a breeder with a positive breeding gain will be a profit instead of a loss. The chemical processing charges should include fixed charges on chemical plant investment, operating and maintenance of the chemical plant, uranium losses during processing, and waste disposal.

Bismuth Inventory

Assuming a 20-year life of the bismuth, the annual fixed charges will be 15 per cent of the investment. At a density of 600 lb/ft^3 and a unit cost of \$2.25/lb bismuth, investment will be \$1,350 per cu ft, and the annual

fixed charges are $202.50/\text{ft}^3$ -yr. Bismuth inventory charges are inversely proportional to power density, and the power density ranges from 0.211 Mwe/ft³ for a 4.18-ft diameter core producing 452 Mwh, to 0.314 Mwe/ft³ for a 6.42-ft dia core producing 1100 Mwh. The corresponding inventory charges are 0.137 and 0.092 mill/kwh, respectively.

Thorium Inventory

Assuming a 20-year life for thorium, a unit cost of 42/kg and a plant factor of 80 per cent, thorium inventory charges are equal to $9 \times 10^{-4} M_{02/P}$. The quantity $M_{02/P}$ (kg of thorium per Mwe) ranges in value from 49.8 kg/Mwe for a core diameter of 4.18 ft to 40.7 kg/Mwe for a core diameter of 6.42 ft, and the thorium inventory charges range from 0.045 to 0.037 mill/kwh.

Fuel Inventory and Fuel Production

Assuming a plant factor of 80 per cent, the fuel inventory charge is given by

$$I = \frac{M_{f} \times V_{f} \times R}{7,000 P_{e}} = \frac{R}{7,000 S.P_{e}} \times V_{f},$$

and the value of net fuel produced is

B.U. =
$$\frac{P_h \times \beta \times G \times V_f}{24 P_e}$$
 = $\frac{\beta G}{24\epsilon} \times V_f$

here

B.U. = Value of net fuel produced, mills/kwh

- M_f = Total inventory of fuel, kg
- $V_f = Value of fuel,$ \$/kg
- R = Annual lease charge or fixed charge on capital investment in fuel
- $P_e = Reactor Power, Mwe$

$$S.P_e$$
 = Specific Power, Mwe/kg = P_e/M_f

 P_h = Reactor Power, Mwh

- β = Fuel burned, kg/MWD
- G = Breeding Gain, B.R. 1
- ϵ = Net station efficiency, P_e/P_h

Since both I and B.U. are direct functions of the value of the fuel, V_f , the combined fuel costs are independent of the value placed on the fuel when

$$\frac{R}{7,000 \text{ S.P}_{e}} = \frac{\beta G}{24\epsilon}$$

If $\frac{\beta G}{24\epsilon} > \frac{R}{7,000 \text{ S.P}_{e}}$, the net contribution will be a credit. Using a net station

efficiency of 43 per cent for the LMTBR and a value of $1.18 \times 10^{-3} \text{ kg/MWD}$ for β , the combined costs are zero and fuel costs are independent of the value of the fuel for the following sets of values:

R	$s.p_e$	G
·	<u> </u>	
0.04	0.5	0.10
0.08	1.0	0.10
0.12	1.5	0.10
0.04	2.15	0.0186
0.08	2.15	0.0372
0.12	2.15	0.0558

The value of 2.15 corresponds to a value of $S.P_h$ of 5 Mwh/kg, a value which can be obtained in several LMTBR designs. Simultaneous values of $S.P_e$ and G well above those required to obtain the above conditions can be obtained in many LMTBR designs. It follows that the combined fuel inventory and burnup costs can be a substantial credit even if lease charges on the fuel are as high as 12 per cent per year.

An illustration of the potential credit is given below for a LMTBR having the following characteristics:

S.P_e = 1.0 Mwe/kg
R = 0.04
G = 0.10

$$\epsilon$$
 = 43 per cent
V_f = \$15,000/kg

Net Credit = 0.086 mill/kwh (Fuel sales less fuel inventory charges).

Chemical Processing

The chemical processing costs are dependent upon the processes selected and the processing rates, which are in turn a function of reactor system parameters, such as system volumes, inventories, and power split between the core and blanket. Several chemical processing methods are compatible with the fuel streams. Pyroprocessing methods, at an advanced stage of development, may be used thereby eliminating the need for inventory holdup for radioactive decay. The comparative evaluation of the possible chemical processing methods has not yet been completed; therefore, their contribution to fuel costs have not been clearly defined.

Summary

The combined bismuth and thorium inventory charges for the LMTBR should be in the range from 0.13 to 0.18.

The combined fuel inventory charges and the credit from sales of the net fuel produced should always result in a credit to the fuel costs for any value of the bred uranium.

In a LMTBR producing 500 Mwe with a fuel value of 15,000/kg and 4 per cent annual lease charges, the credit from the net fuel produced will approximately balance all other fuel costs except chemical processing.

Total fuel costs are approximately the chemical processing charges.

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Discussion of Paper Presented by Mr. Thomas

MR. SPINRAD:

The paper is now open for discussion.

MR. AVERY:

First, has anything occurred in the last few months that makes you feel far more optimistic than you did at the time of the fluid fuel task force evaluation group (Jan. 1959), and I ask this fully aware that at the time of the evaluation there had not been the full-scale evaluation of the breeding potential?

Secondly, do you really believe that you have realistically taken into consideration all of the engineering aspects that might tend to lower the breeding potentials?

Finally, since I knew at the time of the evaluation that you had been doing some thinking in terms of comparison with other fuel systems, do you really believe the LMFR is a better system than the aqueous homogeneous in terms of doubling time?

MR. THOMAS:

The principal change in our opinion of the physics of the LMFR core is this: Previous to the task force days, we had looked at the two-region designs with only a uranium-bismuth solution in the core and all of the thorium located externally - in the blanket. The problem was how to get enough power out of it to make it truly attractive from an economic point of view, and secondly, how to get a realistic blanket around the core. We could not see our way clear and therefore we chose at that time to stick with the simpler one-region design because of its ability to be built with present technology. Here the studies have been extended to incorporate the use of internal breeding in the core. We find that we can keep the breeding ratio fairly constant as a function of core diameter by simply varying the thorium in the core.

Secondly, we have put in a lot of effort and we feel that this core and reactor arrangement now is well engineered at the conceptual stage. We feel that the technical feasibility, based on research and development which has been carried on in the past 2 to 3 years in the LMFR program, shows that this reactor vessel primary system can be built. We see no obstacle of a technical nature which would limit the breeding ratio as we have presented here. Does that answer your question fully?

MR. KASTEN:

I would like to comment on this comparison. I think it is very difficult to compare systems done by two different groups in which the degree of optimism taken by one group is different from another. It is very difficult to assess first hand just what assumptions went into the definition of doubling time.

MR. THOMAS:

The doubling time as defined by me, in full-power years, is proportional to the reciprocal of the specific power (with specific power computed on the total inventory throughout the core blanket and external systems).

MR. KASTEN:

Another thing is, I don't think one can compare our doubling times with your figure of $6\frac{1}{2}$ years because your figure was based on no fuel in the blanket, and I don't really see how one can get $6\frac{1}{2}$ years unless you consider a large fraction of fuel in the external system to keep the blanket concentration at that level.

MR. THOMAS:

That is certainly true. I put this comparison here to establish what is essentially an ultimate that one could even consider. I believe the 500 ppm will turn out to be fairly economical to achieve. I would prefer to say that where economics is essential, the 10 to 12-year doubling times are better to talk about.

MR. deBOISBLANC:

It isn't quite clear to me from looking at your slide in which you showed that for a given core size an increase in the internal breeding in the overall breeding gain was achieved by increasing the thorium content in the core. Is there an optimum thorium content? If there is not, why would you always be driven in the direction of maximizing the thorium content?

MR. THOMAS:

As one adds thorium, eventually one can achieve a maximum breeding ratio. The thing that will keep you from going that far in an externally cooled core is the huge buildup in inventory which comes with an increase in fuel concentration.

MR. BARTELS:

One of the areas requiring development in the LMFR all along has been the use of graphite in this particular application, in which it serves as a structural material. You have complicated the core design from the original design. Still you are talking about a 1000-Mw heat reactor with high fluxes, high power densities around the graphite region, and, I infer from the diagram, you are talking about something like a 3 or 4-in. graphite web between the various low channels. Is it more?

MR. THOMAS:

No, it is much less. There is one thing that I would like to point out in our core: the thermal stresses in the graphite are not a limitation in any way. The volume fraction of bismuth to the volume fraction of carbon is equal to 1.25 in the core and to 1.0 in the blanket. The ligaments are fairly small, so that thermal stresses are not a limitation in any way.

In answer to your other question, I would like to say that in general we are taking large blocks of high-density graphite, and this is only a small extrapolation over what is currently available. We are simply drilling channels through these blocks. Now in the core, since the block is different from the end blanket, we drill a differently sized channel, with a transition region from one diameter to the other. The only additional machinings which were not incorporated in our other simple one-region designs are the plenums arranged there. For example, there is a ring which goes around the entire top, accomplished simply by machining or selecting the size of this graphite block properly. The core region is made up of quadrants. The external blanket is made up of 8 sections clamped together by temperature-compensated circumferential clamps in order to hold the graphite tightly together throughout life with thermal expansion. I feel, really, that this is still a simple structure.

MR. SPINRAD:

I am afraid I will have to break off this discussion at this time in order to get on with the next paper. It has been a very stimulating discussion, and I would recommend members of the audience who want to question the speaker further to see him privately.

OPTIMIZING THE MOLTEN-SALT REACTOR FOR MINIMUM DOUBLING TIME

H. G. MacPherson Oak Ridge National Laboratory Oak Ridge, Tennessee

The molten-salt reactor is a graphite-moderated reactor that utilizes a molten homogeneous mixture of $\text{Li}^7 F$ and $\text{Be} F_2$ as a carrier for UF_4 and ThF_4 . At usable temperatures, up to 15 mole per cent ThF_4 (1500 g/liter of thorium) can be incorporated in solution in the salt. Uranium can replace any desired amount of the thorium, or uranium can be dissolved in the base salt with no thorium present. Breeder reactors will have a blanket, since a very large size, one-region reactor, capable of generating more power than could be used in a single station, would be required to reduce the leakage to a negligible value. All the blanket can be molten salt, since the salt has more than half the slowing-down power of graphite, and all nuclear calculations to date have been made with such a blanket. However, in practice, one would replace part of the blanket volume with graphite, since it is far less expensive than the salt. Two general constructions are proposed; both are circulating-fuel, externally cooled reactors. In Fig. 1 is shown the simplest of these, at least in concept. It is made of several large blocks of impervious graphite, each block the full diameter of the reactor core. I call this structure the graphite core-shell reactor.

The second type of construction is shown in Fig. 2. It is of the unit fuel-tube construction. It will be noted that some of the blanket is used to cool the moderator graphite. It has three advantages over the core-shell type. The fuel salt contacts only the tube wall and so cannot soak into the bulk of the core moderator graphite. Since it is of a unit tube construction, a reactor of any physical size or power level can be constructed by increasing the number of tubes. Furthermore, it is capable of practical demonstration of feasibility at a minimum of cost. Its principal disadvantage is that it involves the two salt carriers in the core, and so there is more carrier salt absorption for a given uranium concentration.

The molten-salt system has several features that distinguish it from other breeder reactors. One of these is, of course, the solubility of thorium fluoride, which enables easy use of thorium in the core, either mixed with the fuel or in a separate stream. This adds a degree of freedom to the design of reactors.

Another feature is the existence of an easy method of removing uranium from the salt. By bubbling fluorine through the salt, the uranium is removed as UF_6 . The UF_6 can be converted back to UF_4 and reconstituted



in fresh salt. Presumably this operation can be carried out without a cooling period, so that there is no need for a large inventory of uranium outside the reactor core.

We believe that the process will be inexpensive enough so that the amount of uranium in the blanket circuit can be kept to no more than 10 per cent of that in the core circuit. This has three benefits: it reduces blanket leakage by reducing the number of fissions that occur in the blanket; it keeps the rate of buildup of fission products in the blanket down to a very low figure, making it unnecessary to give the blanket salt a complete purification for long periods of time; and it of course allows a shorter doubling time as a result of the reduced blanket inventory.

For fuel salts that contain thorium, the volatility of UF₆ provides a way of reducing the protactinium losses in a reactor of high specific power. This is accomplished by removing the uranium from the core salt in a cycle that is short compared with the half-life of protactinium and storing the salt with its thorium and protactinium outside the reactor. In the meantime, the uranium can be reconstituted into some of the previously stored core salt.

These freedoms of the molten salt system allow one to vary the core diameter, the volume fraction of fuel in the core, and the thorium concentration in the core. The purpose of this paper is to show how these may be adjusted to design a reactor with a minimum doubling time.

A number of multigroup calculations have been performed on molten salt reactors by L. G. Alexander, J. W. Miller, L. A. Mann and M. E. Lackey at ORNL, and many of these have been published, at least in ORNL reports. Two of the pieces of information available from the calculations are plotted in Fig. 3. The uranium concentration in kilograms per cubic foot of the



total core volume is plotted against the carbon absorptions per fissionable uranium absorption in the core. This curve, obtained empirically, is just what one should expect to find, but we have found it quite useful. The carbon absorptions, or carbon poisoning, in a core composed largely of carbon is a convenient index of the thermality of the neutron spectrum, and is therefore an index of the strength of other thermal absorbers and of the effective value of eta of the uranium. The carbon absorption figure is equivalent to Chernick's slowing-down power per uranium atom and to A. M. Perry's number of carbon atoms per uranium atom.

A wide diversity of calculations is represented on this plot. It includes calculations of reactors with core diameters of 3, 4, 5 and 14 ft. It includes fuels with thorium concentrations of 1, 4, 7 and 13 mole per cent ThF₄, and fuel salt volume fractions of 10, 12.5, 15, 18.3 and 20 per cent. It includes calculations of both the initial state of these reactors with pure U^{233} and the state achieved after 20 years of operation with a mixture of U^{233} and U^{235} .

By noting the relative positions of the points for different per cents of ThF_4 and for different core diameters, an empirical estimation can be made of the effects of these changes on the carbon absorptions or the thermality of the reactor. This is useful in estimating the combinations of thorium in the core and core diameter required to design a reactor that can cover any desired position on the curve.

If the ordinate of this curve is divided by the volume fraction of fuel in the core, it becomes the concentration of uranium in the fuel salt itself. Of course, this introduces a vertical spread of the data, and there will be a separate line for each volume fraction of salt in the core. Figure 4 shows



Fig. 4. Specific Powers at various Concentrations of Fuel

how these curves appear idealized and extended to higher volume fractions than we have calculated points for. Since the power that can be removed in the external cooling system is proportional to the volume of the fuel salt, the ordinates can be converted into the reciprocal of specific power, or kilograms of fissionable uranium in the core salt per megawatt. This has been done on this curve.

From Chernick's curves of maximum breeding ratio vs. moderating power, we know that the region of greatest interest for breeders will be in the range of carbon absorption from 0.03 to 0.05. It is apparent also



Fig. 5. Absorptions in Salt Carrier as a Reaction of Absorptions in Carbon from this curve that for high specific power, we prefer to have a high value of the volume fraction of the fuel in the core. However, as is shown in Fig. 5, the absorption in the carrier salt increases with volume fraction of fuel. On this figure are plotted the available calculations and some idealized extrapolations.

The parasitic absorptions, other than for carbon and carrier salt, are assumed constant under equilibrium conditions. Table I results from three sets of assumptions that have been made. The sets marked I and II are for a small core reactor in which most of the thorium absorptions are in the blanket and represent different degrees of optimism and different fuel reprocessing cycles.

The set III is for a large core reactor with most of the breeding occurring in the core and with the same order of optimism as the set I.

TABLE I

Neutron Losses other than Carbon and Salt

	Ι	II	III
Fission Products U ²³⁶	.022 .01	.032 .01	.022 .01
$2 \ge 2 $ Pa Absorption	.01	.01	.01
Leakage, Blanket Salt Delayed Neutrons FP Absorbed in	.035	.035	.015
Graphite, etc.	-	.02	-
Total	.077	.107	.057

CASE

To get the breeding ratio, we subtract one plus the parasitic losses from the value of eta. Figure 6 shows a set of values of eta for an equilibrium mixture of U^{233} and U^{235} as a function of carbon absorptions. The curve is not intended to be definitive and should not be used as a reference, but it does represent a reasonable approximation taken from recent data.



Fig. 6. Eta of Uranium as a Function of Absorptions in Carbon

The breeding ratios calculated as above have been combined with specific power in the core salt to obtain regions of minimum doubling time (see Fig. 7). The three circles represent the approximate position on the plot of these minimum doubling times for the three sets of assumptions as to parasitic absorptions other than carbon and salt carrier.

The absolute value of the doubling time will depend on the amount of uranium inventory outside the core circuit. As explained above, the ease of transferring uranium from one salt to another leads to the belief that the ratio of total inventory to core salt inventory will be low. A value of 1.2 has been assumed in

giving the number of years doubling time on the circles I and II. In the case of assumption III on parasitic absorptions, a further correction was made to take into account the fact that not all of the salt volume in the core contains uranium, since this case can only correspond to a large reactor using the unit fuel-tube construction.

Any reactor of a type designed to operate within any of these roughly circular regions should have a doubling time less than the value marked on that curve. It can be seen that if a reactor can be designed to operate with a volume fraction of 25 per cent and with a carbon poison fraction of 0.04, it should have near optimum performance in doubling time. It appears that if there is no ThF₄ in the core, a right cylindrical core about 4 ft in diameter and 4 ft high will meet this criterion. With ThF_4 in the core region, larger equivalent diameter will be required.



Fig. 7. Regions of Minimum Doubling Time

Discussion of Paper Presented by Mr. MacPherson

MR. SPINRAD:

Thank you, Mr. MacPherson. This paper is now open for discussion.

MR. WOLFE:

Please discuss further the reasons for and against cladding the graphite.

MR. MacPHERSON:

The graphite is not clad in the designs presented here. It is my personal belief that it will be easier to get graphite into which the salt will not penetrate in fine-grain extruded graphite material. During this year we hope we will be able to prove out graphite, at least in the tubular size, as far as the amount of salt that it will pick up and its ability to hold the salt. This is one reason we like the tube construction.

MR. SPINRAD:

I would like to thank the speakers for keeping exactly to the morning schedule.

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SESSION VI

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Wednesday Afternoon, October 21, 1959

Chairman: J. R. Dietrich Secretary: D. H. Shaftman

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GAS-COOLED BREEDER REACTORS

Alfred M. Perry Oak Ridge National Laboratory Oak Ridge, Tennessee

The subject of gas-cooled breeder reactors is potentially a very broad one. Since only the coolant is specified - and that in very general terms - the choices of moderator and fuel are left open. In fact, several different moderators are not only possible, but likely to be interesting. Graphite, BeO, and D_2O all deserve consideration, and even H_2O cannot be excluded, as was pointed out in a previous paper by Chernick. A variety of fuel forms is possible also, even including gaseous suspensions. There is thus presented a great diversity of possible reactor configurations in the large family of gas-cooled reactors. None of these has been studied in very great detail with specific emphasis on breeding, and many have hardly been considered beyond the recognition of their possibility. Therefore, rather than present detailed performance estimates for any specific reference reactor, it would seem more instructive at this time to draw attention to certain general characteristics which have the greatest bearing on the suitability of various gas-cooled reactors for breeding. Mention will be made of the neutron economy, power density-specific power relationships, and fuel forms. Since it is doubtful if the power density required in a fast reactor could be achieved with a gas coolant, the discussion may be restricted at the outset to reactors with a thermal or intermediate neutron spectrum and hence, at least for the present, to the $Th-U^{233}$ cycle.

A significant feature of gas-cooled reactors, which may make them unique among solid-fuel reactors, is the possibility of eliminating structural materials from the chain-reacting region, leaving only fuel, moderator, and fertile material. As pointed out by Chernick this morning, the moderating ratio, $\xi \Sigma_s / \Sigma_a$, is a useful index of the breeding performance of thermal and intermediate reactors, because of a balance that exists between parasitic moderator absorptions and the influence of the neutron spectrum on the average value of eta. While a homogeneous reactor model was employed in developing these considerations, essentially the same results apply to heterogeneous systems when corrected for flux variations. Both graphite and BeO are capable of providing all the structure required, and reactors using these may be designed to be essentially homogeneous.

It is possible also to divert neutrons from the fertile blanket to the core as fission poisons accumulate, by means of a movable curtain of moderating material, thus avoiding a neutron loss in control poisons.

The application of these considerations to a graphite-moderated helium-cooled breeder was studied at ORNL in the summer of 1958. The results were discussed at the Detroit meeting of the American Nuclear Society last December, (1,2) and will be reviewed here only very briefly.

The study was conditioned at the outset by the objective of achieving a doubling time as short as possible. Simple considerations suffice to show that with a net breeding gain of 0.05, a specific power of 3400 kw/kg is required to achieve a doubling time of ten years. Since the in-core inventory can hardly exceed one-third to one-half the total inventory of fissionable material chargeable to the reactor, the specific power in the core must approach $10,000 \, \text{kw/kg}$. At this specific-powerlevel, 10 per cent of the Pa²³³ would be destroyed by neutron capture if the fertile material were in the core. This line of reasoning led to the choice of a two-region reactor with all of the thorium in a graphite blanket surrounding the core. Between the core and blanket was placed a six-inch thick buffer region of graphite, whose effective density could in practice be varied from 0.3 to 1.65 g/cm³. To have a conversion ratio of order unity, the neutron leakage from the core would have to be about 50 per cent. Thus, at least one dimension of the core should be small. In a gascooled reactor, there tends to be a loss of neutrons by streaming out of the cooling channels; for this reason, coupled with the greater difficulty of providing a blanket in the axial direction, the configuration chosen was a long slender cylinder. Fluid flow considerations led to a length of about 20 ft, and by introducing the coolant at the midplane, as in the Brookhaven reactor, the core may be made 40 ft long, resulting in nearly negligible axial neutron leakage. Radial dimensions, selected on the basis of multigroup diffusiontheory calculations, were: 39-in. core diameter, 6-in. buffer thickness, and 3-ft blanket thickness. In this study, blanket thickness and thorium concentration were not optimized in terms of economy of thorium inventory, but were simply adequate to minimize neutron leakage. The multigroup results indicated that a 1.5 to 2-ft blanket would have been nearly as effective. The overall diameter is thus less than 10 ft, and a gas pressure in excess of 1000 psi is therefore possible.

The concentration of U^{233} in the core was determined by a series of multigroup calculations for a large (33-ft diameter) graphite- U^{233} core. These calculations were undertaken to explore the effect of U^{233} concentration on the neutron spectrum, parasitic capture, and breeding ratio. Results of these calculations indicated, as also observed by Chernick, that the breeding gain in a graphite- U^{233} system has a maximum at a carbon- U^{233} ratio of about 7000. At lower ratios, resonance captures in U^{233} degrade the average value of eta, while at higher ratios parasitic capture in the graphite becomes serious. The breeding ratio is shown in Fig. 1 as a function of N_c/N_{23} , the atom ratio of carbon to U^{233} . (Quantities in Fig. 1 represent averages over the entire neutron flux spectrum.) These results were based on intermediate-energy values of η of U^{233} that are quite possibly too low. A comparison with Chernick's results indicates that a maximum breeding gain 0.03 to 0.04 higher than indicated here may be inferred from available data on U^{233} .



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Fig. 1 $~\eta$ and $\eta_{\rm f}$ vs. $\rm U^{233}$ Concentration

For the 39-in. diameter core described above, Fig. 2 shows the trend in η and in the percentage of fissions caused by the thermal neutron group during the core burnup, designated "% thermal."



Fig. 2 $~\eta$, $\eta_{f}~$ and Per Cent Thermal vs. Time

Figure 3 shows the trend in buffer density and breeding ratio; $\rho_0 = 1.65 \text{ gm/cm}^3$. For the first core, a burnup of about 40 per cent of the U²³³ is achieved in 32 days, at which time the maximum amount of buffer control has been used. In Table I are listed the neutron absorptions at various stages in the core and blanket cycles.





TABLE I

Core Number	1	1	3	6
Exposure Time (days)	0	30	15	30
U ²³³	1.0000	0.9946	0.9986	0.9900
U ²³⁵		0.0054	0.0014	0.0100
Th^{232}	1.0660	0.9822	1.0317	0.9831
U ²³⁴		0.0220	0.0118	0.0271
η_{av}	2.1630	2.2020	2.1990	2.2160
B.R.	1.0660	1.0040	1.0435	1.0100
Other:	(0.1070)	(0.1973)	(0.1595)	(0.2061)
Core:	(0.0751)	(0.1511)	(0.1058)	(0.1356)
С	0.0225	0.0359	0.0262	0.0322
Xe	0.0526	0.0586	0.0534	0.0434
Sm		0.0126	0.0065	0.0093
Fission Products		0.0440	0.0197	0.0507
Buffer	0.0043	0.0191	0.0079	0.0178
Blanket	(0.0174)	(0.0190)	(0.0362)	(0.0422)
С	0.0072	0.0080	0.0068	0.0057
Xe		0.0012	0.0048	0.0144
Sm		0.0002	0.0011	0.0037
Fission Products		0.0007	0.0008	0.0077
Pa ²³³		0.0089	0.0131	0.0107
Leakage	0.0102	0.0081	0.0096	0.0105

Neutron Absorptions

Table II gives the material balance over six core cycles, at the end of which the blanket was discharged. With chemical processing losses taken into account, it appears that the system just about breaks even. This result is strongly dependent on the values of η employed in the calculations. The larger breeding gain suggested by Chernick's calculations would bring the doubling time into the range of 10 to 15 years.

TABLE II

Summary of Core and Blanket Inventories
through Six Core Cycles

Duration days	249
Duration, days	
Th ²³² burnup, kg	210.0
Pa ²³³ loss, kg	2.5
U ²³³ burnup in blanket, kg	30.1
Final blanket inventory, kg	177.4
U ²³³ burnup 1n core, kg	172.6
Average conversion ratio	1.036
Blanket processing loss (0.8%), kg	1.4
Core processing loss (1.0%), kg	2.1
Net amount of bred U^{233} , kg	1.3
Average breeding gain (dimensionless)	0.0064

There are several possibilities, not explored in the 1958 study, which are now under investigation at ORNL. These include the effect of continuous fuel loading and the possibility of incorporation of thorium in the core by feeding it rapidly through the core, with intervening storage periods in the blanket to allow Pa^{233} to decay. The effect of using an equilibrium feed material containing U^{234} , U^{235} , and U^{236} , as well as U^{233} , is also being examined.

The 1958 study does show, however, that the average neutron losses are about 0.15 (see Table I), of which more than half comes from xenon, samarium, and other fission products. The fission product cross sections used in this study implied a resonance integral of 120 barns. The fission product absorption (other than of Xe^{135} and Sm^{149}) was taken to be linear with fuel exposure, i.e., nonsaturating. Recent studies by Gordeev and Pupko,⁽³⁾ by Pattenden, (4) and by Nephew at ORNL indicate that the resonance integral of the fission products of U²³⁵ is closer to 190 or 200 barns, but of course the higher cross-section poisons saturate with increasing fuel exposure. Nephew's study, complete as yet only for the case of U²³⁵, indicates a resonance integral of about 125 barns/fission for a burnup of 0.5 fission /initial U^{235} atom in a spectrum in which 60 per cent of the U^{235} fissions are caused by thermal neutrons. For a given percentage of thermal fissions, however, the fission product will saturate less rapidly with fuel burnup in a U²³³ reactor than in a U²³⁵ reactor, because of the much larger resonance integral of U²³³. These considerations imply that the fission product absorptions shown in Table I are low, by an amount estimated to be about 10 per cent.

The high specific power assumed in this study implies of course a correspondingly high power density, and it needs to be shown that a gascooled reactor is capable of the required power density.

For an average carbon- U^{233} atom ratio of 5000, and a specific power of 10,000 kw/kg in the fissionable material, the necessary power density in the graphite is readily calculated to be 60 w/cm³. Assuming a coolant void



Fig. 4 Power Density in Heliumcooled Graphite-U²³³ Reactor.

fraction of 30 per cent in the core, the average core power density will be more than 40 kw/liter; the maximum of course will be considerably greater. If only a portion of the graphite in the core contains fuel, the power density in the fueled portion must be correspondingly greater. That power densities of this magnitude can be achieved in a helium-cooled graphite-U²³³ reactor is shown in Fig. 4.(5) The core is assumed to consist of parallel plates of fuel-bearing graphite. Heat generation within a graphite plate is limited by a thermal stress of 800 psi. Heat removal from the plates is limited by an assumed pumping-power allowance of 2 per cent of the heat transferred. If one restricts attention to plates no thinner than 0.25 in., and to a coolant temperature rise of roughly 50°F/ft (to give an overall rise of about 1000°F in a 20-ft channel), it may be seen that a power density of 300 kw/liter is possible with a maximum film drop of 500 to 600°F, which is not unacceptable for a ceramic fuel element.

Figure $5^{(5)}$ shows similar results for BeO plates cooled by CO₂ at 500 psi. While the allowable stress in BeO is ten times greater than in graphite, the Young's modulus is also very

much greater, and the conductivity somewhat lower; the result is that a BeO plate can generate only about 10 per cent as much heat as a graphite plate of the same thickness.



Fig. 5 Power Densities with BeO Plates Cooled with CO₂

thin as 0.1 in. prove to be feasible then, as Fig. 5 shows, maximum power densities of 200 to 300 kw/ liter can be achieved. With BeO, the optimum Be/U^{233} atom ratio is about 3000, and the density is about 2.8 g/cm³, as compared with 1.6 to 1.7 g/cm³ for graphite. Thus, the target power density is around 90 w/cm³ in the BeO, or 45 kw/liter for 0.1-in. plates with 0.1-in. spacing. In the case of BeO, however, unlike the case of graphite, it will be economically unattractive to have a large fraction of the BeO containing fuel. If the fabricated cost of BeO fuel elements is optimistically placed at \$100/kg of BeO, exclusive of U²³³ cost, then 50 per cent burnup of the contained U^{233} implies a contribution to the power cost of about 3 mills/kwh. If only 20 per cent of the BeO contains fuel, i.e., if the Be/U^{233} ratio in the fueled BeO is 600 instead of 3000, the contribution to the power cost is only 0.6 mill / kwh. Now, however, the required power density in fueled BeO is 450 w/cm³, instead of 90, even before taking into account the powerdensity distribution throughout the core. It thus appears that a BeO reactor probably can not simultaneously satisfy the requirements of optimum Be/U^{233} ratio, high specific power, and low BeO supply cost. Some compromises will be

If BeO plates (or tubes) as

necessary, most likely in the direction of reduced specific power. It is interesting to note that the limitation on performance is imposed by thermal stresses in the BeO, rather than by heat transfer to the gas. As a coolant, the gas is well matched to the fuel. (Recent experimental results indicate that at high irradiation temperatures the thermal stresses in BeO may be relieved by plastic deformation, it is thus possible that higher power densities in BeO may prove to be feasible for power reactor applications. This possibility has not been factored into the calculations reported here.) A potential advantage of BeO is the fast effect, due to the (n,2n) reaction, which has already been discussed at this meeting by W. Haefele. If the full reactivity bonus of 0.03 to 0.05 computed for BeO is to be realized, it is desirable for the Li⁶ produced to be able to diffuse out of the BeO in a time small compared to $(\sigma \phi)^{-1}$, which is of the order of one year. In preliminary experiments at ORNL, BeO of density about 2.3 g/cm³ was heated in air at 1450°C for less than an hour, and an approximately tenfold reduction of lithium concentration was observed. How rapidly the lithium will diffuse out of BeO at lower temperatures remains to be determined, but it is possible that the Li⁶ poisoning effect may prove to be small compared to the Be (n,α) effect.

The suggestion is sometimes made that Xe^{135} poisoning may be reduced in ceramic fuel-element reactors by diffusion of xenon out of the fuel. While this possibility cannot be altogether excluded at this time, it does not appear to be very likely. At a flux level of 10^{14} neutrons/cm² sec, the effective half-life of Xe^{135} , including neutron absorption, is less than an hour. In general, the fission gases do not diffuse out of fuel materials this quickly. Estimates of the fraction of fission products released from UO_2 at 1400°C, as a function of their decay constant, have been made by Culver at ORNL⁽⁶⁾ and are shown in Fig. 6. It is evident that under these conditions very little of the Xe¹³⁵ can escape from the fuel before absorbing a neutron.



The case of a D_2O -moderated gas-cooled reactor is unlike that of the graphite or BeO-moderated reactors in that a structural barrier is required to separate the coolant and the moderator. While detailed characteristics of such a system designed for breeding are not available, it can be shown on the basis of rather general arguments that the well-known advantages of a D_2O system can be realized even in a heterogeneous reactor. For purposes of discussion, we assume a pressure-tube concept, with the D_2O and the pressure tube kept below 100°C. The pressure tube is assumed to be fabricated of Zircaloy. For an allowable stress of σ psi, a gas pressure of p psi, the tube thickness-to-diameter ratio is $p/2\sigma$; and the volume of zirconium to the enclosed volume in the tube is $2p/\sigma$. If one assumes parallel-plate fuel elements of fueled graphite, occupying a fraction v of the tube volume, the Zr/fuel volume ratio is $(2p/\sigma v)$. It is then easily shown that the ratio of neutron absorptions in zirconium to absorptions in U^{233} is approximately given by

$$\Sigma_{\rm Zr}/\Sigma_{\rm U} = 0.01 \ (ps/\sigma vQ)$$

where s is the specific power in kw/kg U^{233} and Q is the power density in the fuel plate, limited by fuel plate thickness, t. Table III shows values of Σ_{Zr}/Σ_U for assumed values of the other variables.

TABLE III

t (in.)	Q (w/cm ³)	v	s (kw/kg)	p (psi)	σ (psi)	Σ_{Zr}/Σ_{U}
0.3	550 1200	0.3	10,000	500	7000	0.043
0.2	1200	0.3	10,000	500	7000	0.012

Relative Absorptions in Zirconium

These examples illustrate the possibility of reducing the zirconium absorptions to acceptable levels. If a pressure-vessel concept were employed, the coolant tubes could be considerably thinner, and the zirconium absorptions correspondingly lower. The important point is that the breeding performance is limited not by the D/U ratio but by the Zr/U ratio, and this ratio in turn is largely governed by the thermal performance of the fuel element.

In this rather cursory review of gas-cooled breeder reactors, the following points have been developed: (1) The performance of the reactors, as breeders, is not limited by the choice of a gas as coolant, and, in fact, the possibility of removing all structural materials from the core is an advantage of the gas-cooled reactor. (2) The inability to remove fission poisons from the core is a disadvantage that remains a limiting feature of a solid-fuel reactor. (3) The fast effect in BeO may well prove to be a permanent, rather than a transient, asset in the life of the reactor. (4) In a D_2O reactor the absorptions in moderator-plus-structure can probably be kept significantly lower than in graphite or BeO; the fission poisons are present, but may be somewhat less serious because the neutron spectrum is more nearly thermal.

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Discussion of Paper Presented by Mr. Perry

MR. DIETRICH:

Is there a discussion of Mr. Perry's paper?

MR. WATT:

I would like to take issue with you on the diffusion lifetime of fission products out of the system for the specific case of the graphite plates that were used. The lifetime of diffusion out of those plates at 1900 is about 100 seconds.

MR. PERRY:

Nineteen hundred centigrade?

MR. WATT:

Yes.

MR. PERRY:

I see. Well, I confess not to have been thinking in terms of such a high temperature.

MR. WATT:

It is not clear what the lifetime versus temperature will be.

MR. PERRY:

Yes.

MR. CAMPISE:

In reference to this question about diffusion of fission fragments out of a graphite plate containing uranium, have you looked into what happens to the delayed neutron fraction in reference to control?

MR. PERRY:

No, not at all.

I should think that the same considerations apply. There you are talking about matters of tens of seconds to 100 seconds or so, and I should think it would take a very high temperature, quite a lot higher than in these rather thick elements, which is in the neighborhood of 1000 to 1400°C, to get diffusion rates that are at all comparable to the lifetime of the delayed neutron emitters. Is this different from your view?

You see, the internal temperatures in such elements are not high. We are talking about gas temperatures, let's say, up to 1500°F, and if the film drops are on the order of 500°F, which is quite a healthy drop, the central temperatures of a fuel element need not get above 1200-1400°C.

MR. OKRENT:

Do you expect the beryllium oxide to be able to stand up for a long time in the reactor if you have it separated from the fuel?

MR. PERRY:

Well, I know that experiments which indicate crumbling of beryllium oxide at very high irradiation have been reported. The particular experiments I had in mind were done at rather low temperatures, and the failure of the oxide was attributed to accumulation of helium in the body. What the situation would be at somewhat higher temperatures is an interesting question, but I don't think we know the answer to it. Experiments will certainly be required
to find out what the characteristics are at high irradiations. One point that I would like to make is that graphite also suffers deleterious effects upon high irradiation, and there has been, so far as I can see, no mention of this whatsoever. I just don't know what happens in some of these other graphite systems after quite a long time.

MR. SANDERS:

What is the reactivity needed to compensate for the rise to operating temperature? How does this compare with the reactivity invested in this graphite buffer zone ?

MR. PERRY:

The reactivity change due to temperature, I believe, is rather small in this graphite- U^{233} system. I confess to not knowing what the temperature coefficient is in a graphite- U^{233} system. I know that it is usually thought that a homogeneous ceramic fuel reactor would have to operate on a thorium cycle rather than on a plutonium cycle just because of the temperature coefficient, and yet I am not one hundred per cent satisfied myself at this stage of the game that this is the case, because of the fact that the U^{233} cross section varies less rapidly than 1/v. But there may not be a small positive coefficient with U^{233} . I feel that this depends on really very nice calculations of the thermal spectrum and probably even more precise knowledge of eta than exists in the cross sections we have. I don't really think it is a question that can be resolved properly today.

The reactivity control capacity of the buffer zone is about 18 per cent in the particular example shown. You see, this comes about because the reactor is small and has very high leakage.

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MODERATOR CONTROL REACTOR

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Introduction

The Moderator Control Reactor, (MCR) utilizes a varying mixture of D_2O and H_2O to control reactivity. The addition of D_2O to the moderator lowers $\xi \Sigma_s$ and hence increases the resonance capture in fertile material. At the beginning of lifetime the Moderator Control Reactor has about 80 per cent D_2O as the moderator and coolant. By varying the amount of D_2O as the core life progresses, the multiplication factor is maintained at unity. Control through varying D_2O concentration rather than inserting parasitic material increases the conversion ratio and allows a much greater degree of power flattening.

Plant Performance

A list of the major performance characteristics is given in Table I.

TABLE I

Performance Characteristics of Moderator Control Reactor Rated Power 1064 Mw (heat) Gross Electrical Output 325 Mwe 300 Mwe Net Electrical Output Design Pressure 1900 psig 1650 psia Normal Operating Pressure 60×10^{6} lb/hr Coolant Flow Rate 520°F Average Coolant Temperature 4,064,000 lb/hr Total Steam Flow Steam Conditions (steam drum outlet) 500 psig, dry, saturated 340°F Feed Water Temperature

Figure 1 shows the core layout. The reactor has 16 control rods, used for power doppler and safety shutdown. Xenon, samarium, fuel burnup and fission product buildup are controlled by changing the concentration of D_2O .

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Fig. 1. Core Arrangement - Moderator Control Reactor.

The reactor is designed such that at full power all control rods will be out of the core. With no control rods in the core, it is possible to flatten the power distribution to a much greater extent than in a conventional pressurized water reactor. For example, zone loading in the axial direction is not feasible in a conventional pressurized water reactor, but in a moderator control reactor, with no control rods inserted at full power, axial loading can be used. In addition, the radial flattening in the moderator control reactor is much easier since only the case with no control rods inserted must be solved.

The moderator control reactor core is zone-loaded with three zones in both the radial and axial directions. Figure 2 is a two-dimensional power plot of the MCR. For the moderator control reactor the peak-to-average power varies from 1.8 to 2.0. For a conventional pressurized water reactor a minimum obtainable peak-to-average power in a well-designed core varies from 3.0 to 3.6.

In Fig. 2 the contour lines show the peak-to-average power in RZ calculation. The peak-to-average power given in Fig. 2 does not include the local factor of 1.2.



Fig. 2. Contour Map. Peak-to-Average Power Ratio. Moderator Control Reactor.

Increased Conversion Ratio

Since the whole concept of moderator control is based on the absorption of excess neutrons in fertile material rather than in parasitic absorbers, the Moderator Control Reactor offers an increase of about 0.2 in conversion ratio. With U^{235} as a fuel, the Moderator Control Reactor has an integrated conversion ratio of 0.82, while conventional water reactors have conversion ratios in the neighborhood of 0.6. From a breeding aspect the interesting fuel is U^{233} . However, little if any U^{233} is available; therefore the reactor designer is obliged to consider the case of starting with U^{235} and recycling to attain an equilibrium fuel.

For the Moderator Control Reactor, calculations have been performed for fuel recycling and compared with the fuel recycling case of a conventional water reactor. Figure 3 shows the results. It can be seen that the maximum conversion ratio for a conventional water reactor is approximately 0.72, while a Moderator Control Reactor can attain a conversion ratio slightly in excess of 1 for the equilibrium fuel. These calculations were performed for the core with the minimum power-generating costs.





Encouraged by the results, design changes were made to the fuel arrangement within the core and a shorter core life was assumed. With these changes, an average breeding ratio of 1.07 was attained together with a doubling time in the neighborhood of 35 years. An increase in the breeding ratio above 1.07 can be obtained by a further decrease in the core lifetime. With on-site processing and remote fabrication, an appreciably smaller doubling time can be attained. Table II shows a neutron balance for the core with the average breeding ratio of 1.07.

TABLE II

	End of Life Average: 10,000 Mwd/Ton	Beginning of Life
Th ²³²	0.956	1.034
U ²³⁴	0.084	0.110
U ²³³	0.908	0.913
U ²³⁵	0.092	0.087
U^{236} and Pa^{233}	0.04	0.038
Xe + Sm	0.054	
F. P.	0.056	
Zr	0.018	0.017
H ₂ O	0.041	0.007
Leakage	0.02	0.033
	2.269	2.239
Fast Fission in Thorium	0.029	0.029
η_{av}	2.24	2.21
-	2.269	2.24

Neutron Balance for the Equilibrium Reactor

At the beginning of life the neutron loss to U^{236} is 0.038. However, by the time Pa^{233} has built up, loss to U^{236} has decreased to 0.02. Hence, the total losses to U^{236} and Pa^{233} are essentially constant. It should be noted further that the average value of η increases with core life. The Moderator Control Reactor, due to the fact that its spectrum is changing, has built in some compensating increases in the conversion ratio during the core lifetime.

One of the uncertainties in the breeding ratio quoted above is in the resonance integral of thorium. Figure 4 shows a comparison of the resonance integral curve used and that obtained from the Nordheim formula⁽¹⁾ as a function of D_2O concentration in the core moderator.



Fig. 4. Effective Resonance Integral of Th^{232} vs. D_2O Concentration Zero Power 500°F

Not shown in the figure is an additional point recently measured in the TUPE experiments. (The TUPE experiments consist of a series of criticals using thorium and fully enriched U^{235} , clad with aluminum, being performed jointly by the Argonne National Laboratory and the Babcock and Wilcox Company.) This datum point lies closer to the Nordheim curve than to the curve which was used in the calculations. In the formulas shown in Fig. 4, the quantity γ is the Dancoff-Ginsburg factor with the D₂O at room temperature, and f(T) is a correction factor accounting for the effects of the rise to D₂O temperature T. Of course, a higher resonance integral would lead to a higher conversion ratio. Experiments are currently under way by Dr. W. G. Pettus at our Critical Experiment Laboratory to measure the resonance integral as a function of lattice spacing and D₂O concentration.

Fuel Recycling

In order for a U^{233} -thorium fuel system to be practical, a satisfactory fuel-recycling scheme must be developed. Probably, the ultimate goal is cheap, remote hot fabrication. However, this goal seems to be in the far future. At the B and W Company a fuel-recycling scheme is in the conceptual design stage; we hope to fabricate recycled fuel for less than 30 per cent extra cost than fresh fuel. In this process the irradiated fuel is cooled, reprocessed in the normal manner, and then shipped to the fuel-recycling plant. The first stage consists of removing Th²²⁸ and its daughter products by using solvent extraction. The fuel is then fabricated into rods by swaging. The swaging process does not have to be remote, although it would probably be a glove-box operation. The assembly of the fuel rods into bundles would be performed remotely. This whole concept for fabricating recycled fuel depends mainly on assembly-line operation. The fuel must be fabricated into rods quickly to avoid a buildup of Th²²⁸ and its daughters.

Conclusions

The Moderator Control Reactor may not have as short a doubling time as some of the reactors which are not yet designed in detail. However, the Moderator Control Reactor is almost completely designed, so that all neutron losses resulting from practical considerations in reactor design are accounted for. Further, the neutron balance has been worked out for an equilibrium fuel. I might mention here that the effect of including the higher uranium isotopes is two-fold in its effect on doubling time. First, the presence of higher isotopes decreases the conversion ratio; secondly, these higher isotopes greatly increase the inventory.

References

- F. T. Adler, G. W. Hinman, and L. W. Nordheim, <u>The Quantitative</u> <u>Evaluation of Resonance Integrals</u>, Proc. of the 2nd UN Int. Conf. on Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 16, p. 155.
- 2. Private Communication from W. G. Pettus; new data, as yet unpublished.

Discussion of Paper Presented by Mr. Webb

MR. DIETRICH:

Is there a discussion of this paper?

MR. CAMPISE:

In reference to moderator control, what kind of kinetic problems would you have in a system of this type?

MR. WEBB:

I think you are referring to the power overshoot. We use control rods to control the reactor, so far as maneuvering is concerned. We program these four rods in such a way that the hot spot factor will always stay less than two. As a result, we put on a limitation of the maneuvering rate. In the case of accidents, the rods shown here are sufficient to scram the reactor in a hot condition, allowing 3 per cent margin for stuck rods.

MR. CAMPISE:

I was under the impression that the control rods were out during power operation.

MR. WEBB:

They are out during full-power operation. They can be in during operation at reduced power. Along with this, I might point out that at reduced power we can allow a larger hot-spot factor. For example, at 50 per cent power one could permit a hot-spot factor as high as 4.

MR. WOLFE:

Aside from the low hot-spot factor of your reactor, are there other advantages over a plain D_2O reactor?

MR. WEBB:

This is only one advantage. The other advantage is increased neutron economy, with its tie-in to the picture of economics. As you know, if you overload a reactor for long life, the conversion ratio goes way down. This is because you overload it and put in the parasitic poisons in the form of control rods. But in this reactor we control it by changing the ratio of deuterium to hydrogen. By increasing this ratio for added control, we increase the capture in the fertile material, and this provides an advantage over a D_2O reactor controlled by parasitic absorbers.

MR. WOLFE:

If you ignore the very first few years of operation and consider the equilibrium core, then the reactivity variation due to the burnup is quite small; it is only a couple of per cent once you reach equilibrium xenon and some fission products.

MR. WEBB:

If you get through the first 50 or 60 days, you are right. You do not have a problem. It is that first 60 days, though, that could make a difference.

MR. WOLFE:

It seems like a considerable complication to get you over the hump.

MR. WEBB:

There is another problem, which perhaps I didn't mention, that we considered. That is the fact there is now very little U^{233} . In order to get to a decent, so-called equilibrium cycle, we must have a reactor with a high conversion ratio in the beginning. To do this you can not use the over-loading concept. For example, as you know, if you take a normal reactor and start with U^{235} you would not have the same shaped curve that you have at equilibrium, so you would have to load it heavily with fuel and you would end up with a rather low conversion ratio. However, in this design we can achieve an initial conversion ratio of approximately 0.8. This conversion ratio increases from cycle to cycle. Unless we did this, we could not reach a very good equilibrium fuel. I will grant you that perhaps the advantage is somewhat reduced once you reach an equilibrium cycle. The problem is how do you get there.

MR. GREEBLER:

If this advantage is limited only to the initial core, then I will withdraw the comment I am about to make.

So far as the equilibrium core is concerned, you show a very rapid reactivity decrease initially. This would suggest to me that you are using what you might call single-batch loading, that is, your normal reloading pattern is to reload the entire core each time. Could you not improve the situation somewhat by batch loading?

MR. WEBB:

Batch loading always improves the situation because it enables you to remove fission products.

MR. GREEBLER:

Therefore, you don't have the control problem of having to have a lot of poison in the core to hold down excess operating reactivity.

MR. WEBB:

Except in the first sixty days.

MR. GREEBLER:

Again you are talking about the first core, not the -

MR. WEBB:

In any core, whether it is equilibrium or the first core. You have a problem through the xenon, samarium and Pa^{233} holdup.

MR. GREEBLER:

Well, not with batch loading, not if you are putting in only a small fraction -

MR. WEBB:

If your concept is such that you control reactors by adding fuel even during the initial period of time, you are quite correct.

MR. GREEBLER:

If you have an equilibrium core and you are always adding a small amount every two months, or every four, five or six months, whatever the case may be, at any one time you replace perhaps 10 per cent of your core with fresh fuel, so that you get only very small fluctuations in reactivity.

MR. WEBB:

This would be an advantage, but this does not answer the question for the first 50 or 60 days. However, if you wanted to do it more often it would answer that question.

I don't think I quite understand your question.

MR. GREEBLER:

I don't think that there is anything unusual about the first 50 or 60 days if you use this batch loading. This period is unusual only in the case

that you have a single-batch core. If you reload the entire core at once, and allow it all to run down, then you have the 60-day problem. Otherwise you do not.

MR. WEBB:

Once you get beyond the point where you started out with clean fuel, you are quite correct, I understand. We have not gone into any other thing besides single-batch loading.

MR. HERRMANN:

What is the temperature coefficient in that core?

MR. WEBB:

The temperature coefficient in this core is variant and -

MR. HERRMANN:

Well, at the beginning of the cycle.

MR. WEBB:

- at the end of life there is essentially no D_2O present, and the coefficient is about the same as for an H_2O -moderated core, namely, around 10^{-4} , even for this size. At the other end it is about three to four times higher.

MR. HERRMANN:

But you do have a curve of coefficient versus life?

MR. WEBB:

Yes, we do.

MR. HERRMANN:

The other question is connected with your peaking factor due to the fact that you operate with the rods out. Do you mean by this that you achieve a power flattening and thereby decrease your hot spot -

MR. WEBB:

The problem of power flattening is quite difficult and you have to solve it for several rod configurations. This is one of the most difficult

problems that a reactor designer has. If you remove all rods, burnup tends to flatten the power further.

MR. HERRMANN:

Isn't there the additional problem that when you have a control rod channel such as you have in here that you get peaking at the channel?

MR. WEBB:

In the one slide that was on the board, I believe, I did not include the local factor. The local factor we found to be in the neighborhood of 1.2.

MR. HERRMANN:

I have one last question. You optimized to cost; what is the cost?

MR. WEBB:

The only figure that I am allowed to give out is that it is nearly competitive on the West Coast and in the New England states.

MR. OKRENT:

Since you are working with D_2O at least part of the time, why can't you start up with D_2O and get rid of the need for the reactivity in control rods by, well, reflector control or the equivalent of reflector control, let's say displacement of D_2O by gas over part of the cycle? What is the relative advantage of mixing the two types of water over working with just one?

MR. WEBB:

What you mean is the void method control which has been reported in several other places. Well, the void method control has some mechanical difficulties on which I am not in a position to comment.

VOICE FROM FLOOR:

If I understand this concept correctly, you gain a measure of advantage by increasing the leakage into the fertile blanket as you increase the concentration of D_2O in the system. Now, you therefore net an advantage by capturing neutrons that would otherwise be captured in the necessary control elements as poisons. In this way you raise the conversion ratio. What if you made your control rods out of thorium?

MR. WEBB:

This has been considered by several people. I think you will find that it will take too many for the amount of control that you need.

MR. DIETRICH:

Is it true that the main effect is a leakage into this blanket or -

MR. WEBB:

No, this is not true. The main effect is the k_∞ change by changing the resonance capture in the thorium.

MR. DIETRICH:

Are you increasing the moderation as time goes on and decreasing the resonance absorption?

MR. WEBB:

That is correct.

MR. SHAFTMAN:

With regard to this void- D_2O concept, in a conceptual design carried out at Argonne, tanks of moderator were used, and the level of moderator in the tank was adjusted to adjust the resonance escape probability. Is this what you meant by the void- D_2O case? This was not a displacement of coolant by void, it was a displacement of moderator.

MR. WEBB:

Let me say that we have done no work on a void case. There is a reason why we probably will not. The reason is that this reactor is already designed with the largest pressure vessel we can ship. If we take up any more core volume, we would have to lower our power level.

VOICE FROM FLOOR:

Have you evaluated a temperature or a void coefficient for this reactor when it is cold and has its maximum light water content?

MR. WEBB:

When it has its maximum light water content it is essentially the same as for a normal pressurized light-water reactor. I have not calculated the temperature coefficient for that case, but it might even be positive when the reactor is cold. MR. DIETRICH:

Do you have a number for the average neutron loss to hydrogen over the life of the core?

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MR. WEBB:

The neutron loss to hydrogen is variant over core life, and it varies from 0.01 to 0.04.

MR. MOREWITZ:

With this concept - where all control rods are out - how do you take care of xenon transients across such a large water-moderated core- the xenon oscillation problem?

MR. WEBB:

We are now looking into the xenon oscillation problem. According to preliminary information, we do not expect a problem.

A MIXED REACTOR FOR HIGH CONVERSION RATIO

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R. C. Ross, K. Kniel, P. Marnell Nuclear Development Corporation of America White Plains, New York (Presented by Mr. Ross)

Introduction

The difficulties with both fully fast and fully thermal reactors for breeding have led to efforts to develop mixed spectrum reactor concepts which may offer, in particular instances, more useful characteristics than either. One can mention in particular the coupled fast-thermal reactors investigated here at Argonne and previously described at this meeting. (1) In the course of our Sodium-Deuterium Reactor (SDR) program, we were led to a somewhat different concept for a mixed reactor, which appears to have some potential for breeding gain and may also have the low-fuel requirement characteristic of thermal reactors. (2)

In arriving at the concept, one can approach the mixed reactor either from the fast or from the thermal direction. One considers adding moderator to the fast reactor in an effort to lower fuel enrichment and/or improve control characteristics without destroying the breeding capability; or one investigates the thermal reactor to see how to improve the conversion ratio. Our original effort was aimed at improving the conversion ratio of a nearthermal reactor. We were not looking for a breeder with a low doubling time, but rather hoped to make the conversion ratio at least high enough that the reactor could burn essentially all of the natural uranium or depleted uranium feed. It was not until the concept had evolved significantly that we noted that the reactor we were considering could be thought of as a coupled reactor. I would like to describe our approach to this objective, describe briefly the reactor concept that evolved and some of its features and problems, and then consider useful avenues for future exploration.

Approach

I would like to write the neutron balance for a mixed reactor in the following form:

I.C.R. = $\overline{\eta}\epsilon - 1 - L - A$

(these simple relationships are developed here because it has been found convenient to use slightly different definitions than are customary in thermal reactors).

Consider a single fuel material and a single fertile material. Let one neutron be absorbed by the fuel, producing an average number of fission neutrons $\overline{\eta}$, representing the appropriate average over neutron energy. These fission neutrons, in leaving the fuel elements, cause fissions in the fertile material, so that a total of $\eta \epsilon$ neutrons are available for the various processes which may take place. By this definition,

$$\epsilon = 1 + \frac{(\nu_{\text{fertile material}} - 1)}{\nu_{\text{fuel}}} \frac{(\text{fertile material fissions})}{\text{fuel fissions}}$$

without the usual corrections for fast absorptions in fuel and fertile material. For criticality, one neutron must be absorbed in the fuel isotope, leaving $\overline{\eta}\epsilon$ - 1 neutrons potentially available for breeding. After subtracting off neutrons leaking and absorbed in moderator, structure, fission products, etc., one obtains the initial conversion ratio (I.C.R.) or breeding ratio for the reactor. Note that these definitions distinguish between events in the fuel and events in the fertile material, and do not distinguish between events at thermal vs other energies. These definitions are simpler for describing breeding or conversion ratio than the customary definitions which are intended to be useful in computing reactivity.

In a typical advanced natural uranium SDR configuration, consisting of fuel clusters about 5 in. in diameter on about 14-in. centers, $\overline{\eta}$ is assumed to be 2.07 (all U²³⁵ absorptions taking place at thermal) and ϵ is calculated to be 1.036, giving $\overline{\eta}\epsilon$ of 2.14, or a potential initial conversion ratio of 1.14. After correcting for parasitic absorption, leakage, etc., the calculated initial conversion ratio is 0.79.

Initial 3-group calculations indicated that the conversion ratio of the SDR could be raised to unity or greater by (1) increasing the size of the fuel clusters to the order of 1 foot or so in diameter, thereby greatly increasing the fast fission factor, (2) maintaining reactor criticality by an increase in the average enrichment of a fuel cluster to a few percent, thereby reducing the effects of parasitic absorption, and (3) using a breeder blanket to collect most of the leakage neutrons.

Since conversion ratios greater than unity were in sight, it became of interest to consider a breeding cycle using plutonium as fuel and U^{238} as fertile material. To test this concept, several multigroup calculations were made on typical single fuel cells of interest, and the conceptual design of a high-conversion power reactor based on this principle was considered.

Calculation Procedure

Calculations were made using the STAR 10-group diffusion theory code in a one-dimensional cylindrical geometry. These calculations gave the neutron balance for a single lattice cell (consisting of a fuel cluster with its surrounding D_2O) with a boundary condition of no neutron leakage. The neutron balance could then be corrected for leakage in a finite geometry. The group distribution was chosen to cover as adequately as possible fast

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fission effects, U^{238} resonance absorptions, and Pu resonance absorptions, as well as thermal effects. The group distribution and constants are given in an Appendix to this paper.

These calculations considered only Pu^{239} as fuel and U^{238} as fertile material. Only the initial breeding ratios were calculated in detail, and no fission products except xenon and samarium were considered. The objectives of this investigation were to determine whether any breeding potential existed in this type of reactor, and to indicate possible problem areas and directions of approach. Although no detailed consideration was given to overall fuel cycles, preliminary calculations indicated that neither the buildup of higher isotopes of plutonium nor the presence of small amounts of U^{235} from the depleted uranium feed would significantly affect the conversion ratios calculated.

Conceptual Design

A typical fuel cluster is shown in Fig. 1. The fuel rods of mixed uranium and plutonium carbide, each 0.75 in. in diameter, are clustered in a 14.5-in. diameter niobium tube. Around the fuel tube are a Zircaloy barrier tube and an aluminum calandria tube, with $\frac{1}{4}$ -in. gaps between each pair of tubes. A number of such fuel tubes are combined in a tank of D_2O to form the reactor assembly.



Fig. 1 Breeder (High Conversion) Design - Schematic of Cross Section of Fuel Cluster

The composition of the fuel cluster, within the fuel tube, was approximately 30 per cent coolant, 5 per cent cladding, and 65 per cent fuel. As is well known, the value of η for plutonium is extremely favorable at high neutron energies, becomes quite unfavorable at intermediate resonance energies, and finally becomes almost tolerable again at thermal. The object of this configuration is to make the fuel cluster large enough to give a sizeable increase in the number of fast fissions occurring in the plutonium (and also in the U²³⁸), and to surround the cluster with enough D₂O moderator to minimize resonance plutonium absorptions and encourage thermal absorptions.

In the fuel cluster, the central region will have predominantly fast fissions and the outer region predominantly thermal fissions. In order to avoid an excessive power peak at the outer surface, it was found desirable to lower the enrichment in that region and to increase the enrichment in the center. In the case shown, three different fuel enrichments were used: 10 per cent in the inner region, 1.5 per cent in the intermediate region, and 0.25 per cent in the outer region, for an average fuel enrichment of not quite 5 per cent. The power pattern for this fuel distribution is shown in Fig. 2. The fuel enrichments and the region boundaries have been adjusted to give the flattest power distribution possible with three fuel enrichments. Considering the outer region to be one row of fuel rods, the maximum power of a fuel rod is about 1.35 times the average power of a fuel rod. The shape of the power pattern confirms that the outer two regions are predominantly thermal and the inner region is predominantly fast. About half of all fissions occur at thermal.



Fig. 2 Power Density Distribution for a Nonuniformly Enriched Cell

A simplified balance for this calculation is given in Table I. (A more detailed neutron balance is given in an Appendix to this paper.)

TABLE I

Calculated Neutron Balance

$\overline{\overline{\eta}}$		2.117
E		1.109
$\overline{\eta}\epsilon$		2.348
Neutrons Available ($\overline{\eta}\epsilon$ - 1)		1.348
Parasitic Captures:		
Niobium Clad	0.0196	
Sodium Coolant	0.0201	
Niobium Fuel Tube	0.0257	
Zircaloy Barrier Tube	0.0033	
Aluminum Calandria Tube	0.0124	
Moderator	0.0060	
Xenon-135	0.0303	
Samarium-149	0.0253	
Total Parasitic Captures		0.142
Leakage Through Blankets		0.027
U ²³⁸ Captures		
In Core	0.911	
In Blanket	0.268	
Total Initial Conversion Ratio		1.179
Total Neutron Consumption		1.348

The fast plutonium fissions bring the average value of η for plutonium to 2.12. The fast fission factor, ϵ , is 1.11, giving 2.35 neutrons available. After subtracting various parasitic absorptions plus an allowance for leakage through a breeder blanket, one is left with a modest initial breeding ratio of about 1.15.

What limits the conversion ratio in the design? The desire to operate with a high specific power requires diluting the fuel element with considerable quantities of coolant, which lowers the spectrum in the fuel rod and limits the number of fast fissions that can be obtained. The aim of high specific power also dictates a low enrichment, which further limits the fast fissions and pushes the design toward a thermal spectrum.

Let us look briefly at the specific power of this reactor. In line with the advanced SDR program, a reactor to produce 200 Mw_e (540 Mw_t) was considered, and the sodium was assumed to enter at 750°F and leave at 1100°F,

with a maximum velocity of 35 ft/sec. A preliminary analysis led to a 12-tube design, as indicated in Fig. 3. Large amounts of moderator are provided around each tube (the tubes are placed on 23.5-in. centers), leading to an effective core diameter of about $7\frac{1}{2}$ ft. The core height was taken as 6 ft. These dimensions are compatible with reactor criticality and with the desired power, and lead to a fuel inventory of about 880 kg of plutonium. This gives the rather low specific power of just over 600 kw_{th}/kg of plutonium.



Fig. 3 Breeder (High Conversion) Design -Schematic Plan View of Reactor Vessel - 200 Mw_e

Why is the specific power only about the same as that of the Fermi reactor, although the fuel enrichment is lower by a factor of five? This design is limited by heat transport, and with the specifications given, this is all the heat that can be removed by the sodium. The principal reasons for the low volumetric specific power in this case are the greater core height (the sodium undergoes its temperature rise over a 6-ft height instead of over a $2\frac{1}{2}$ -ft height), and a fairly poor maximum-to-average power. In addition to the power variation within a fuel cluster, the central tubes generate considerably more power than the outer tubes.

How can we improve the reactor performance? There may be some room for modest improvement by increasing the maximum sodium velocity and the sodium Δt , but both of these values are fairly large already. One can increase the sodium volume fraction at the expense of the conversion ratio. The greatest improvements can be obtained by shortening the core height, improving the power distribution. and/or lowering the average fuel enrichment. We have looked briefly at a 6-tube design (and also a little at an annular design which is roughly equivalent), and find that we can more than double the volumetric specific power by the resultant reduction in core height and improvement in power pattern. We also note that the central region of the fuel clusters contain more than 90 per cent of the fuel but produce less than 40 per cent of the power in the fuel tube. Considerable improvement in specific power is possible by lowering the enrichment in that region. Each of these changes requires some increase in enrichment of the outer layer, and will have some adverse effect on the breeding ratio.

An additional problem with this design appears to be fission product poisoning in the low fuel enrichment section. More will be said about this later.

This design evolved from a thermal reactor. Let us review briefly the principal features of the design, and see if they are justified. The D_2O moderator appears to be the best possible material for getting neutrons past the plutonium resonance without excessive moderator absorptions. A brief comparison of D_2O , graphite, and H_2O indicated somewhat less favorable results with these other moderators, and also indicated, incidentally, that more D_2O is provided in the design shown than is really desirable.

The sodium coolant is reasonably nonmoderating and has fairly good heat transport capability. The use of light or heavy water could improve heat transport (particularly if a phase change were allowed), but the additional moderation would probably lead to excessive resonance absorption in plutonium. The use of steam or other gas coolant would not improve heat transport unless a greater temperature rise were permitted than with sodium. The recent AEC announcement of the availability of lithium-7 of extremely high purity at reasonable cost suggests that at least some consideration should be given to its use. Lithium has twice the volumetric heat capacity of sodium, and also about twice the moderating power.

The use of carbide fuel is of some interest. Although the carbon content both dilutes the fuel and provides undesired moderation, these effects are not as severe as they would be with oxide fuel. Metallic fuel elements tend to be unattractive because we need high performance but cannot tolerate absorbing alloy materials in the thermal region. Note that the thermal properties of the carbide are so favorable that in this design fuel elements of 0.75-in. diameter are permitted, not only reducing fabrication cost, but perhaps making possible a smaller total amount of cladding.

Along this line, I should mention that NDA, with the Carborundum Company as subcontractor, is now in the first phase of a program to study the technology of the UC-PuC fuel cycle for breeder reactor applications.⁽³⁾ Plutonium is not a very satisfactory fuel for the thermal region. The use of plutonium puts severe requirements on the moderator to avoid resonance captures, and even then the resultant thermal captures are not particularly favorable. One might consider the use of U^{233} for the fuel in the outer region. A rough estimate of the effect of U^{233} can be made by assuming that the same number of captures occur as before, but using η of U^{233} instead of η of plutonium in the two outer regions. This indicates a possible increase of about 0.15 in $\overline{\eta}\epsilon$. The use of U^{233} would allow considerable reduction in the amount of moderation required, since the resonance problem is much less severe, and might therefore permit a higher enrichment in the outer region, easing the fission product poisoning difficulty.

With this system, the outer blanket would be thorium, and the core fuel elements would contain plutonium and U^{238} in the central region, and U^{233} and U^{238} in the outer region. It appears to be possible to balance the design so that sufficient breeding takes place in the blanket to replace the U^{233} burned in the thermal part of the core.

This makes an interesting concept - a mixed fast-thermal breeder, using plutonium in the fast region where it is advantageous, and U^{233} in the thermal region where it is most useful. As yet it is only a concept, and much more investigation is required to see if it can lead to a useful reactor design.

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Appendix 1

Fission		Lowest	U ²³⁸			Pu ²³⁹			
Group	Fraction	Energy	σ _c	σ _f	ν	ס c	¯, of	ν	
1	0.3417	2.2 Mev	0.0193	0.588	2.65	0.037	1.94	3.217	
2	0.2312	1.32 Mev	0.0623	0.407	2.65	0.045	1.95	3.049	
3	0.3938	180 kev	0.140	0.00596	2.65	0.108	1.70	2.929	
4	0.0255	67 kev	0.259			0.411	1.75	2.92	
5	0.0078	9.1 kev	0.472			0.825	2.19	2.91	
6		101 ev	0.444			5.37	7.11	2.91	
7		5.0 ev	2.333			24.5	35	2.91	
8		0.102 ev	0.621			13.1*	34.6*	2.91	
9		0.0286 ev	1.92			242	527	2.91	
10		Thermal	2.26			279	672	2.91	

Ten-group Constants Used for U^{238} and Pu^{239}

*To obtain the group 8 cross sections, two curves of absorption cross sections were prepared, corresponding to "optimistic" and "pessimistic" assumptions of the energy dependence of α These curves and the fission cross-section curve were then integrated to obtain group-averaged cross sections, both by integrating the cross-section curve vs lethargy directly (corresponding to an infinitely dilute fuel) and by integrating the cross-section curve weighted with the reciprocal of the total plutonium cross section (corresponding to pure Pu²³⁹ fuel). The values given above are for the optimistic α case and pure Pu²³⁹ fuel. For infinite dilution, the group 8 $\overline{\sigma}_{\rm f}$ was calculated to be 477 b, and $\overline{\sigma}_{\rm C}$ was 248 b (optimistic α) or 324 b (pessimistic α), respectively.

NOTE: All values of the microscopic cross sections listed are in "barns" (10^{-24} cm^2) .

Appendix 2

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Neutron Balance for Reference Case

(Basıs 8 45951 Total Neutrons)

A Composition

Region	Nature	Region Outer Radius, cm	Composition, Volume Fractions of Fall Density Material				
			Pu ²³⁹	U ²³⁸	Na	Fe	D ₂ O
1	High Enrichment (10%)	11 25	0 0463	0 4167	0 491	0 0460	
2	Medium Enrichment (1 5%)	14 07	0 00695	0 4561	0 491	0 0460	
3	Low Enrichment (0 25%)	16 89	0 00115	0 46185	0 491	0 0460	
4	Structure	18 77				0 020	1
5	Moderator	31 83					1 000

B Neutron Balance

	Central Region		Intermediate Region		Outer Region		Total for All Regions		
Group	Pu ²³⁹ Fissions	U ²³⁸ Fissions	Pu ²³⁹ Fissions	U ²³⁸ Fissions	Pu ²³⁹ Fissions	U ²³⁸ Fissions	Pu ²³⁹ Fissions	U ²³⁸ Fissions	Total Fissions
1	0 05037	0 13548	0 00396	0 07848	0 00022	0 07259	0 0 5 4 5 5	0 28655	0 34110
2	0 04559	0 08466	0 00 3 4 7	0 04750	0 00054	0 04410	0 04960	0 17626	0 22586
3	0 29092	0 00900	0 02022	0 00465	0 00304	0 00422	0 31418	0 01787	0 33205
4	0 10102		0 00701		0 00114		0 10917		0 10917
5	0 11199		0 00897		0 00174		0 12270		0 12270
6	0 13380		0 02199	1	0 00630		0 16209		0 16209
7	0 04893		0 02400		0 01105	2	0 08398		0 08398
8	0 15355		0 04435		0 01428		0 21218		0 21218
9	0 02251		0 06687		0 04880		0 1 3 8 1 8		0 13818
10	0 19336		0 85172		0 70461		1 74969		1 74969
	1 15204	0 22914	1 05256	0 13063	0 79172	0 12091	2 99632	0 48068	3 47700

Group	U ²³⁸ Captures	Iron Captures	Sodium Captures	Heavy Water Captures
1	0 01009			0 00736
2	0 02746			0 00000
3	0 42280			0 00000
4	0 27064			0 00000
5	0 49556			0 00000
6	0 32534			0 00007
7	0 42570	0 00245	0 00150	0 00018
8	0 17750	0 02248	0 01413	0 00037
9	0 07493	0 01073	0 00685	0 00056
10	1 1 3 0 7 4	0 19014	0 11956	0 01607
	3 36076	0 22580	0 14204	0 02461



MR. DIE TRICH:

Is there discussion of this paper?

MR. CHERNICK:

You discussed so many reactors that it is hard to decide how to direct this question, but I believe I am thinking of your plutonium- U^{238} system. You mentioned the fact that there were 50 per cent thermal absorptions.

MR. ROSS:

Thermal fissions.

MR. CHERNICK:

Thermal fissions. Now I think it is important for us to know where you cut off the thermal from the epithermal group in this case. What do you call a thermal fission? Below - ?

MR. ROSS:

In our 10-group calculation, we had one group that embraced the plutonium resonance. That was the eighth group, and I added a ninth and tenth together and called those thermal.

MR. CHERNICK:

Does it include the 0.3-ev resonance ?

MR. ROSS:

No, the 0.3-ev resonance region was a group all by itself.

MR. SHAFTMAN:

I have two questions I would like to ask. The first question is, what kind of a cell calculation would be appropriate under these circumstances; the second question is, how reliable do you feel that your spectrum calculation would be in, say, the thermal group in the outer two annuli of a cluster?

MR. ROSS:

Let me tell you what we did. I don't think it represents the best possible calculation that could be made. We did single-cell calculations in which the fuel cluster was taken with the appropriate amount of moderator that corresponds to a single lattice cell and a zero-leakage condition was used at the cell boundary. That gave us an infinite-reactor calculation. Then we made fairly crude estimates of the leakage in the overall cell by essentially just an L^2B^2 -type correction. We were not too concerned with the leakage; the idea was that there would be a blanket. One can change the k ∞ of an individual cell very markedly by a relatively small change in the enrichment of the outer layer, which we don't think would make a very large difference in $\eta \epsilon$.

MR. SHAFTMAN:

Was k_{∞} very sensitive to the amount of D_2O that you used?

MR. ROSS:

No, we had plenty of D_2O . We didn't really optimize on that, but in the reactor shown we had about 3 volumes of D_2O around the cluster per volume of fuel cluster. This meant that we had a D_2O thickness of something like a foot around the fuel cluster and then a zero-current condition at the cell boundary. Going to somewhat smaller or somewhat larger D_2O thickness didn't make much difference.

MR. SHAFTMAN:

Then, in your computations you put a one-foot thickness of D_2O around the outer clusters as well and cut down the D_2O thickness of your reflector. Is that correct?

MR. ROSS:

Yes.

MR. DeBOISBLANC:

It seems that you have made a great effort to skip the resonance absorption in plutonium, and I would like to ask what fraction of the absorptions in plutonium occurred above and what fraction occurred below this resonance as a result of this elegant scheme? I asked this question because in shifting the spectrum to the thermal region you introduce a competition with the captures of neutrons by the sodium. I presume you still have a sodium coolant in this.

MR. ROSS:

Right.

MR. DeBOISBLANC:

Then my question has the other part. Do you not lose as many neutrons to sodium as you would have lost by having a lower value of eta in the plutonium resonance region?

MR. ROSS:

I don't have these numbers at my fingertips for you. I think that I can find the table in the report. It very well may be. I think that use of U^{233} in that region would solve our questions, of course, because then we would not have to worry about a resonance there. I think something like 10 per cent, or a little more of the fissions in the outer regions still occurred in the resonance group, in spite of all of the D_2O we had. I am not sure whether we are above or below optimum with regard to the balance between the plutoniumresonance captures and the sodium and niobium. We use niobium for the outer fuel tube because of its lower thermal absorption cross section, in spite of its probably higher fast absorption cross section.

MR. CHERNICK:

Just one more question on the thermal value of eta. Did you calculate this as a function of neutron spectrum? Did you calculate the thermal group value of eta as a function of neutron spectrum?

MR. ROSS:

We used 2.05 for eta. The moderator is assumed to be a cool moderator. I am not sure just what that corresponds to. I think it was something like 145°F for the D₂O temperature. That was one of the difficulties. In considering a graphite-moderated version, the disadvantage of the graphite was that we thought that the graphite temperature would be so high that the value of η in the thermal region would be quite low. With the cool D₂O, calculation yielded an $\eta = 2.05$ in the thermal region.

MR. CHERNICK:

Then you did not look at the effect of fuel, which is quite dense here, on the neutron spectrum?

MR. ROSS:

Well, the spectrum is probably fairly well controlled by the moderator in this lumped-fuel arrangement.

MR. CHERNICK:

What was your volume ratio of fuel to D_2O ?

MR. ROSS:

It was about 3 to 1, moderator to total fuel cluster, something like 7 to 1 moderator to carbide, which would be maybe 12 to 1 moderator to equivalent full-density fuel, or even higher^{*} than that.

MR. MacPHERSON:

I presume that these calculations were made with Pu²³⁹. Have you looked into what might happen if you get a fraction of higher plutonium isotopes?

MR. ROSS:

We have not looked at this, but we assumed that it would not be a major effect, because, as people have said today, Pu^{240} is roughly equivalent to U^{238} and the Pu^{241} is roughly equivalent to Pu^{239} . We did not think it was a big effect and we have not looked at it.

MR. SHAFTMAN:

I should like to raise again an earlier question, which was partially raised by Jack Chernick. The major purpose of my question about how you decided how much D_2O to assign was in connection with the spectrum. I think that the spectrum computation is very difficult, especially in the case of Pu^{239} . I think that the amount of D_2O used can have very important effects on reactivity and on conversion ratio in those two annuli,

MR. ROSS:

Probably my best answer would be that we were not very careful in calculating this thermal effect. We were looking for the general ball park here just to find out where we were, and I think it would be worth while to do this much more carefully.

MR. KIEHN:

In the paper I gave on Monday, which was about the involute of this system, about 10 per cent of the fissions were in the resonance region. It did not seem to improve the production ratios to increase enrichments in the fast region to beyond 5 per cent plutonium.

REMARKS MADE IN SUMMARY OF THE CONFERENCE ON THE PHYSICS OF BREEDING

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I have been charged with the task of summarizing the meeting.

Actually, I would call this my impression of the conference, rather than a summary. A conference of this scope and importance should be summarized more thoughtfully than time has permitted. Further, I don't think it is reasonable to summarize what I think constitutes some of the most valuable information presented: the numerical results of crosssection measurements and cross-section evaluations.(4-6,16-21)

The conference started off appropriately with a paper by Mr. MacPherson on the relationship of breeding to our long-term energy supply.(2) I think the fact brought out in this paper that was new to many of us was that the energy resources of uranium and/or thorium in the earth's crust appear to be comparable with the energy resources represented by the deuterium in the seas, on the assumption that we could use the deuterium in thermonuclear plants.

Of course, this does not mean that the uranium is as easy to get at. On the other hand, it may be a good deal easier to use. At any rate, I think this observation was very satisfying to us who are in the fission power business, for two reasons: it assures us that we are not promoting an industry that will squander the energy resources of our descendants, and it assures us that there is a reasonable chance that the work we do will have some permanent significance.

Now in the discussion of breeders, it has been said here that two characteristics may be considered as figures of merit; these have been discussed at some length, but I will add a few words. These two figures of merit are the breeding ratio and the doubling time. It has also been suggested here that really the two figures of merit should be the breeding ratio - or, possibly, simply the statement that the reactor will breed or will not breed - and the power cost. Of these latter quantities, the first would determine, of course, whether technically we have it in our power to make available the very large store of energy that is in the earth's crust; the latter would presumably determine how much of this energy, in the form of coal, oil, and gas, or in the form of uranium or thorium, we will waste before we get around to doing the thing that may be feasible technically. Consequently, I think that this question of the cost of power from a breeder is certainly an important one, whatever one's attitude toward the doubling time. Certainly the doubling time itself is a property related to the economic factors, but the relationship is not a simple one and in many cases it is not an obvious one, as has been pointed out. The most obvious case in which the doubling time is important occurs when the fissionable isotope inventory of the reactor represents a large cost. In that case, as Mr. Kasten⁽²⁴⁾ pointed out, if new fissionable isotopes can be made at an annual fractional rate equal to the annual rate of charge against the money value of the inventory, then the two effectively cancel, and the inventory charge is not a concern. Of course, the chances are that if the doubling time is short the specific power is high, and perhaps the inventory charge will not be excessive anyway.

From the point of view of conservation, there are two obvious places where the doubling time enters importantly. First, if there is not enough fuel that can be mined to give the fissionable isotope inventory necessary to start new reactors, then bred fuel must be used to start them, and the doubling time determines how rapidly the total number of reactors may be expanded. Secondly, the doubling time determines the fraction of the total fissionable isotope on hand at any given time that can be diverted to nonbreeder uses without using up the basic supply.

It does seem to me that the question of getting a substantial breeding gain is a much more fundamental consideration for the long-term energy prospect than that of achieving a short doubling time. If we do not breed fissionable isotopes we may, with respect to energy supplies, impoverish our descendants. If we breed with a long doubling time, we may, perhaps, simply present them with a practical problem. While it seems to me that we should not impoverish our descendants, I don't believe we have to solve all their problems for them.

With respect to fast reactors, I think we can say that we know how to build a breeder that is feasible technically. Although no great effort has gone into the engineering on thorium as a fertile material for such reactors, I think it is reasonable to say that by the fast breeder we can extract the energy from either U^{238} or thorium in a technically feasible manner. I don't think we know how to do this in economic competition with fossil fuels, of course. It is apparent that the economic problems are mainly engineering ones. Those which involve reactor physics most closely, even though they are to a large extent engineering problems, are the problems that are connected with the fuel: the fuel elements, the reprocessing, and so on. We have not yet recycled plutonium on a scale which can give us confidence in the economics of that process. I gather from some remarks that came out in the discussion here that we do not really have a plutonium fuel element which comes close to being economically attractive. I still say that this is economics, however, and that if we had no other way of getting energy, we would run plutonium fuel elements at temperatures as low as 300°C if we had to.

Now the solution of these fuel problems and many of the other problems connected with the fast breeder will no doubt require some changes in design philosophy which must be factored into the reactor physics. It seems to me that the main activities of reactor physics in the further development of the fast breeder will be directed to the ends of helping to find more attractive engineering solutions, rather than to further demonstrations of the fundamental breeding potentialities. Consequently, such work as the work on the fast cross sections reported by Moldauer,(6) Cranberg,(5) and Terrell(4) at this conference continues to be highly important, and is beginning to have, one might say, engineering significance, as well as significance from the point of view of long-range energy resources. It will have still further significance in developing an accurate and practical reactor physics.

I think, in this connection, that it is gratifying to see the developments that have come about in fast reactor physics in the last several years. Although they were not specifically emphasized here, I think the results were quite evident. It is good to see that the multigroup techniques that we have adopted have been so successful, not only because they allow us to calculate many practical situations, but also because they allow us to fit incremental experimental information into the body of the theory as it becomes available. This may be a trivial observation, but I can remember not many years ago, when one wondered whether it was worth while to use as many as four or five groups to make a fast reactor calculation. It is also good to see, as pointed out by Dr. Okrent,(8) that the important nuclear performance characteristics of the fast reactor are not extremely sensitive to changes in some of the less well-known microscopic properties of reactor materials.

It seems to me the conclusion that the fast reactor will develop into a practical breeder is bolstered by the relatively high breeding gains that were shown by several sets of reactor calculations, in which the nature of the fuel, and in some cases the nature of the reactor, were varied over quite a wide range. The fuels considered ranged from solid(8) and molten(9)metals to uranium and plutonium oxides,(10) and to carbides and mixtures of carbides and metals.(12)

When we consider breeders other than the fast breeders, our motivation is presumably mainly economic, although I think that there is also a motivation to see where we are headed in some of the reactor types that have achieved a degree of success as power producers without regard to the question of breeding. The obvious example, of course. is the H_2O reactor: the pressurized water or the boiling water reactor. In this regard I think that Dr. Chernick's paper(22) was quite interesting, aside from its more direct function of discussing what can be done with the best thermal breeders. I don't know that we have come to the conclusion here that any of the presently most highly developed types of power reactors - the solid-fueled thermal reactors - are potential breeders, but it does appear that at

least they are capable of rather high neutron economy and high conversion ratio. After all, one doesn't go in one day, or in one year, from a situation in which the cost of nuclear power is the over-riding consideration to one in which breeding is all-important. There is an intermediate stage, and to me it is a very interesting one, for I expect to be around at least for this stage.

I think this objective, of seeing where we can get with the technology that we have already developed, is also the motivation for the study on the epithermal-sodium-graphite reactor, (15) and I think we might remark, in passing, that this is one place where further experimental cross-section data are very much needed.

In the field of thermal or near-thermal breeders, of course, the main interest has been centered about the question of η for U²³³. It appears that the previously accepted value of 2.28, or perhaps a slightly higher value, is being vindicated in the United States, and, as Dr. Sanders described, (21) the British value is moving up, although there seem to be still some loose ends that haven't been tied up. I found the measurements that were described by Dr. deSaussure (20) to have great interest, not only from this point of view, but also as examples of important new techniques and improvements of techniques.

Since the breeding gain in thermal reactors is not high, the neutron contributions and losses, due to such things as the(n, 2n)reactions, fission product absorptions, absorptions in protactinium, nonproductive absorptions in higher isotopes, and so on, are particularly important, and I think the careful studies of these effects reported by Mr. Haefele, (13) by Mr. Pattenden, (16) and by Mr. Levine (23) have been very much worth while. Apparently, without such careful considerations of these effects which add to or subtract from the breeding gain, we really can't make much of an assessment of the thermal breeder. I think I am correct in stating that these studies have not shown any results that are particularly discouraging to the thermal breeder - that is, the potential neutron economy does not appear to be less good by any large margin than we had previously believed. Of course, the analysis of the (n, 2n) reaction in beryllium has been rather encouraging.

In summing up the whole thermal breeder situation, Dr. Chernik's paper (22) has been quite encouraging. It goes without saying that this study, which was concerned with the fundamentals of the problem, leaves plenty of engineering questions. In some ways it is too bad that we couldn't put more of our time into looking at some of the engineering questions, for they seem to be crucial ones in this as well as in the fast breeder field.

We have looked at some conceptual designs for several thermal breeder reactors and, in particular, representatives of the main fluid fuel reactors.(24,25,26) All of these studies show a rather attractive performance in terms of breeding ratio and doubling time, and I think we will have to say that even if we accept the doubling time as the criterion of excellence for a breeder, we can't make any decision among these types on that basis - at least not without considering engineering questions which are out of place at a conference on the physics of breeding.

The last few papers covering the solid-fueled thermal breeders or perhaps I should say the solid-fueled non-fast breeders - have been extremely interesting. They have, I think, pointed up the fact that it is difficult to breed with such a reactor, for most of them employ specialized and novel designs to improve the neutron economy. It does appear that perhaps reactors of these types can "hold their own." It is not obvious whether they will have a doubling time short enough to be significant.

I believe this is a summary of what we have heard, more from the point of view of a reactor designer than from that of a physicist. But it does cover things that I found most interesting. Would anyone care to ask a question or make a comment ?

References: PROGRAM OF THE CONFERENCE

1.	WELCOME	N. Hilberry, ANL
2.	BURNING THE ROCKS	H. G. MacPherson, ORNI
3.	ON THE DEFINITION OF BREEDING	B. I. Spinrad, ANL
4.	THE STATUS OF MEASUREMENTS OF ν AND $\sigma_{capture}$ FOR FAST NEUTRONS	J. Terrell, LASL
5.	FAST NEUTRON SCATTERING BY U ²³⁵ , Pu ²³⁹ , and U ²³⁸	L. Cranberg, LASL
6.	ON THE ESTIMATION OF FAST NEUTRON CROSS SECTIONS	P. Moldauer, ANL
7.	A COMPUTATIONAL SURVEY OF IDEALIZED FAST BREEDER REACTIONS	W. H. Roach, LASL
8.	THE SENSITIVITY OF BREEDING RATIO IN FAST REACTORS TO	D. Okrent, ANL

UNCERTAINTIES IN CROSS SECTIONS

9. BREEDING: INTERNAL OR EXTERNAL

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- PARAMETRIC ANALYSIS OF A PuO₂-UO₂ FUELED FAST BREEDER
- 11. BREEDING IN FAST PLUTONIUM METAL SYSTEMS WITH EBR-II TYPE RECYCLE
- 12. BREEDING CHARACTERISTICS OF FAST REACTOR SYSTEMS USING PLUTONIUM AND U²³³ CERAMIC DISPERSIONS IN URANIUM AND THORIUM MATRIX MATERIALS
- 13. THE FAST MULTIPLICATION EFFECT DUE TO THE (n, 2n) REACTION IN BERYLLIUM AND BERYLLIUM OXIDE
- 14. BREEDING IN COUPLED FAST-THERMAL SYSTEMS
- 15. ADVANCED EPITHERMAL THORIUM BREEDER CONCEPT
- 16. FISSION PRODUCT POISONING DATA
- 17. REVIEW OF FISSION PARAMETERS OF U²³⁵
- 18. STATUS OF LOW ENERGY CROSS SECTIONS OF PLUTONIUM
- EVALUATION OF LOW ENERGY CROSS SECTION DATA FOR U²³³
- 20. MEASUREMENTS OF η OF U²³³ AT ORNL
- 21. RECENT BRITISH MEASUREMENTS
- 22. BREEDING POTENTIAL OF THERMAL REACTORS

- R. M. Kiehn, LASL
- Paul Greebler* and Peter Aline,G.E., San Jose
- H. H. Hummel, ANL
- J. B. Nims, APDA
- W. Haefele, ORNL (and Kernreaktor, Karlsruhe, West Germany)
- R. Avery, ANL
- A. Z. Campise,* E. R. Cohen, and D. T. Eggen, Atomics International
- N. J. Pattenden, ORNL (and Harwell)
- G. Safford, Columbia University
- B. Leonard, G. E., Hanford

J. E. Evans* and R. G. Fluharty, Phillips Petroleum

G. deSaussure, ORNL

- J. E. Sanders, UKAEA, Winfrith
- J. Chernick, BNL

- 23. THE EFFECT OF HIGHER ISOTOPE M. M. Levine, BNL PRODUCTION IN BREEDING
- 24. BREEDING RATIOS, DOUBLING TIME, AND FUEL COSTS IN AQUEOUS HOMOGENEOUS THORIUM-BREEDER REACTORS
- 25. THE LIQUID METAL FUEL THERMAL BREEDER REACTOR
- 26. OPTIMIZING THE MOLTEN-SALT REACTOR FOR MINIMUM DOUBLING TIME
- 27. GAS COOLED BREEDER REACTORS
- 28. A MODERATOR-CONTROL, HETEROGENEOUS BREEDER
- 29. A MIXED REACTOR FOR HIGH CONVERSION RATIO
- 30. REMARKS MADE IN SUMMARY OF THE CONFERENCE ON THE PHYSICS OF BREEDING
 - (* denotes person presenting the paper)

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- J. R. Dietrich, General Nuclear Engineering Corporation

Discussion of Paper Presented by Mr. Dietrich

MR. MacPHERSON:

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I thought it might be worth while to attempt a very brief defense of doubling time as a measure of the breeding performance of a reactor. This is entirely aside from whether one believes that the doubling time figure of itself has significance or not.

To have a short doubling time one must have a breeding ratio well in excess of unity. In calculating the performance of a reactor one is apt to be, perhaps, optimistic in overlooking a few small neutron losses. So, if one has a short doubling time, the potential breeding ratio is probably high enough that the final reactor when built will actually be a breeder of some kind. Furthermore, the other factor that goes into the doubling time is the specific power. I believe that nearly every study of the future of power reactors shows the importance of high specific power during the period of very fast growth of the nuclear power industry, if we are to provide the initial fissionable material for reactor inventory during the period of rapid growth. So I believe that specific power in itself has a very high significance.

MR. GREEBLER:

I think you stated that you feel the justification for the thermal breeder is an economic one. The implication was that perhaps it isn't important to have short doubling times and therefore people are seeking out ways of attaining breeders at lower cost. I think the story is quite the reverse. I think the proponents of thermal breeding claim that they can get shorter doubling times because of the higher specific powers in the thermal reactors. Now I think everyone here will agree that the cost for any breeder type is uncertain at best, but I believe that with the fast breeder we can really run circles around the thermal breeders on a cost basis - economic feasibility and so on.

MR. DIETRICH:

I think that what you have said shows the need for a clarification of my own remarks. I believe we can say with some certainty that the problem of the long-range energy supply can be solved by the fast reactor. Then we must answer the question of why we look at the other breeder types - obviously because some think they can do the job, or part of the job, better. It was in this broad sense, of possibly doing the job better - whatever better may mean in the future - that I used the term economic. I did not intend my remarks on doubling time to have any particular significance with respect to reactor types.
MR. CAMPISE:

One general question concerns plans that might be made for a meeting, say a year or so from now, to look at the progress that has been made on all breeder concepts since the present meeting.

The second comment I wanted to make is that I believe what we are trying to prove is, first, that breeding is possible and, secondly, that a useful power unit can be built. If we answer these two questions, then the question of whether or not we are producing enough fuel to fuel all the reactors might be considered. If all the reactors breed, to some extent, I think this is a step forward.

MR. DIETRICH:

In answer to your first remark, I think it would be an excellent idea to have further meetings of this kind. I wish I could offer to be your host, but I will turn that over to the Argonne or AEC people. I would have no further comment on your second comment; I believe it is substantially the way I feel.

MR. SPINRAD:

Just in general affirmation of the previous remarks, I wanted to say that we had originally started off by thinking of holding a meeting on the physics of breeding and of high-conversion reactors. It now appears that there is a real difference between these two types of systems. A breeder system, when all is said and done, has as its primary incentive the production of new fissionable material. I think that is a background summary which perhaps no one else but I is so foolish as to infer from this conference, but if we didn't really feel that there was something good about breeding with most of the systems we described, we would possibly go for power just a little bit cheaper.

On the other hand, there is something to be said, particularly in the thermal breeding systems, for the idea of trying, at least, for high conversion as a means of getting efficient use of the fuel in a power economy, rather than efficient production of fuel for the power economy.

What I would like to suggest is that most of the breeding systems, whose breeding gains - however they may be defined - are less than perhaps 15 per cent, can more accurately be described as super-efficient burners rather than as true breeders whose motivation is production.

MR. OKRENT:

It seems to me that no one except the proponents of fast reactors and those of the coupled reactor, about which Dr. Avery spoke, (14) presented a breeder that is satisfactory for the U²³⁸-plutonium system. I will not be willing to accept the last paper (29) on this basis, yet, if you will forgive me.

It seems to me that a solution has to be found, and will be found, for some fast reactor with the uranium-plutonium system. It may not compete on the West Coast or New England, but at least in India, let's say, it will be slightly overpriced at the worst. If we do not, we are impoverishing the world by something like half of its energy - I don't know what the exact figure is. If this is the case, it seems to me that the thermal reactor people have a real challenge. If we solve the problem in fast reactors for one system it will almost be solved for the other - the uranium-thorium system. If you have the technology of the one system it carries over rather simply, I believe, in almost all aspects. The fuel cycles differ, but all the hardware and so forth looks the same, and many of the problems in the fuel cycle itself - the techniques - are the same. So I think that the thermal reactor people have real competition here to overcome if they wish to see their reactors successful in the long run. While I admit to some prejudice, I think it is not completely without reason.

MR. DIETRICH:

Do you wish to answer that, Dr. Chernick?

MR. CHERNICK:

I really think there is a need for fast breeders and there is a need for thermal breeders. I don't want to start any recrimination between the fast and the thermal reactor people.

I do think that if we depended only on fast reactors they would have quite a job supplying the Navy and the Merchant Marine, for example, with the large amounts of fuel that they need to run their reactors with conversion ratios of the order of 0.3. I think this is a serious concern and I don't think the immediate fast reactor program is going to solve it. I think that as far as the thermal breeders are concerned, we are certainly not as far advanced as the fast reactor. We don't have a demonstrated experiment. The fuel needed is practically nonexistent and we need some reactor which has a chance of breeding; it may miss by a few per cent, but I don't think that is terribly important if perhaps the second reactor makes it. I think that is the problem of the thermal breeder. Everybody recognizes the need for complete utilization of fuel. We are already 15 years into the atomic energy era and we are practically starting all over again now with the problem of developing thermal breeders.

MR. DIETRICH:

I might say that I think what you say, Dr. Chernick, is most appropriate. This is one reason I said that I felt much interested in the question of very high conversion ratios in the thermal solid-fueled reactors. After all, we are talking about developing a nuclear power industry in this country under a system of free enterprise, and questions of immediate economics will often take precedence over long-range goals. It does seem to me that, once nuclear power becomes really competitive, the economic forces will become so great that it will be very hard for any agency to exercise control over the direction of development. So perhaps the main importance of what we do today is that it sets the stage, one might say, for tomorrow.

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MR. HAEFELE:

In comparing these two breeder types, I was wondering that the safety aspect was not considered as an important one in the competition, at least in heavily populated areas, as perhaps in Europe. I understand that some years ago that aspect was considered to be more of a problem than now, but I think the safety aspect in fast breeders is still an important one.

MR. DIETRICH:

Would anyone like to reply to this question?

MR. OKRENT:

I would only like to say, if it turns out that all fast reactors do have a safety problem in the sense that there is a feeling that they are hazardous for largely populated areas, I really do believe one can satisfy this problem by going to the coupled reactor. It is sufficiently thermal so that one can rid himself of this stigma. From what we know now, I don't see that there is any reason to think there will be a safety problem for fast reactors that we cannot solve. There are imaginary problems now, for which we don't yet know enough to specify the answers completely, and we need to learn more, really, to help answer them specifically and satisfactorily, rather than to say whether they exist as insoluble problems.

MR. CAMPISE:

I appreciate what Jack Chernick mentioned in reference to the facts that it took us 15 years to get where we are and it will probably take us 15 more before we have a decent breeder system. This leaves a little room for epithermal intermediate systems to make a start. We know that alpha is fairly small in the fast intermediate region. The question, as far as we are concerned, is whether it still is small in the epithermal intermediate range. In reference to what Dave Okrent just mentioned, epithermal systems would have an easier control problem relative to fast systems. This is one of the reasons we are looking at them.

MR. GREEBLER:

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With regard to the safety problem, I don't think it is really a community safety problem under any circumstances. I think people have tried to make rather serious experiments with reactors and have not been too terribly successful with it. I think that the overall energy potential in, let's say a sodium-cooled fast breeder reactor, is probably a good deal less than that available from the metal-water reaction in a water-cooled thermal reactor. I think there is a problem in safety from the standpoint of damage to the capital equipment but not a community safety problem.

MR. DIETRICH:

I believe that it will be a real disappointment to the fast reactor people if it turns out that safety considerations are really limitations on the application of fast reactors as electrical generators. I don't believe they are expecting to find themselves in this situation.

I would like to close the session with a word of thanks to Argonne for a very interesting and worthwhile symposium on a very important subject.

WHEREUPON, the Conference on The Physics of Breeding was concluded.

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