

RM E52B28

NACA RM E52B28



RESEARCH MEMORANDUM

CALCULATION OF INTERNAL PRESSURES IN THE
FUEL TUBE OF A NUCLEAR REACTOR

By B. M. Rosenbaum and G. Allen

Lewis Flight Propulsion Laboratory
Cleveland, Ohio

FOR REFERENCE

NOT TO BE TAKEN FROM THIS ROOM

LIBRARY COPY

MAY 23 1958

LANGLEY AERONAUTICAL LABORATORY
LIBRARY, NACA
LANGLEY FIELD, VIRGINIA

NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

WASHINGTON

July 1952

Declassified April 1958

NACA RM E52B28

NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

RESEARCH MEMORANDUM

CALCULATION OF INTERNAL PRESSURES IN THE FUEL TUBE
OF A NUCLEAR REACTOR

By B. M. Rosenbaum and G. Allen

SUMMARY

General procedures for computing internal pressures in fuel tubes of nuclear reactors are described and the effects on the pressure of varying neutron flux, fissioning material, and operating temperature are discussed. A general proof is given that during pile operation each fission product is monotonically increasing and therefore a maximum amount of all elements is present at the time of shutdown. The post-shutdown build-up of elements that are held in check during pile operation because of their inordinately high capture cross sections is calculated quantitatively. An account of chemical interactions between the many fission-product elements and the resulting effect on the total pressure completes the discussion.

The general methods are illustrated by calculations applied to a system consisting of 90 percent enriched U^{235} in the form of UO_2 packed into a hollow metal cylinder or "pin", operating at a flux of 8×10^{14} at $2000^\circ F$. Calculations of the pressure inside a pin are made with and without a sodium metal heat-transfer additive. The bulk of the pressure is shown to depend on the four elements, xenon, krypton, rubidium, and cesium; the amount of free oxygen, however, was also significant. For a shutdown time of 10^6 seconds, the pressure was about 100 atmospheres.

INTRODUCTION

In some reactors of interest for power production, the U^{235} is in the form of a powder contained within a sealed chamber and it is anticipated that a large part of the gases generated during fission will collect in the chamber and exert considerable pressure on the chamber walls. The pressure may be sufficient under some conditions to break the chamber walls. In order to provide some background for fuel-tube design, an analysis of the factors contributing to internal pressure in the fuel tubes of nuclear reactors has therefore been made at the NACA Lewis laboratory and is presented herein.

2420

1-40

The problem is very complicated and it is convenient to break up the calculations in the following manner in this report:

I. Calculation of Number of Atoms of Fission-Product Elements

- (a) Pile Operation Period
- (b) Period Following Pile Shutdown

II. Calculation of Pressure

- (a) Calculations of Chemical Composition
- (b) Calculation of Pressure Contributions of Compounds

The considerations in part I are entirely physical and result in a determination of the quantity of each and every material in the system at every instant of time. In addition to the fission products, all other substances present in the system such as heat-transfer additives are included.

Part II involves chemical as well as physical computations, one of the main aspects being a consideration of the chemical reactions occurring among the elements in part I at the operating temperature (or, after pile shutdown, at temperatures below operating temperature). This consideration is of the utmost importance because it is the equilibrium composition of the mixture, not the free element, which determines the pressure.

Following an analysis based on the aforementioned general considerations, an example illustrating the method is presented. In the example, actual calculations are made of the pressure on the inside of a small hollow cylinder (or pin as it will be called) packed with 90 percent enriched U^{235} in the form of UO_2 . The operating temperature is taken as $2000^\circ F$ and calculations are made both with and without liquid sodium metal in the fuel pin as a heat-transfer material.

SYMBOLS

The following symbols are used in this report:

a_n	$\lambda_n + (nv)\sigma_{cn}$
ΔF	free energy of formation (kcal/mol)
ΔH	change in enthalpy (kcal/mol)
$M_{f,g}$	moles of gaseous fission product

N_f	number of fissions per second
N_n	number of atoms of n^{th} daughter
$N_{n,r}$	number of atoms of n^{th} daughter, n^{th} daughter radioactive
$N_{n,s}$	number of atoms of n^{th} daughter, n^{th} daughter stable
(nv)	thermal neutron flux $\left(\frac{\text{number of thermal neutrons}}{\text{cm}^2\text{sec}}\right)$
$P_{f,g}$	pressure of gaseous fission product
R	universal gas constant (1.986 cal/mol ($^{\circ}\text{K}$))
ΔS	change in entropy (cal/mol ($^{\circ}\text{K}$))
T	operating temperature ($^{\circ}\text{K}$)
T_B	boiling point ($^{\circ}\text{K}$)
t	time (sec)
t'	variable of integration
Y_n	fission yields, number of atoms of n^{th} daughter formed per fission
λ_n	decay constant of n^{th} daughter (sec^{-1})
$\sigma_{c,n}$	neutron capture cross section of n^{th} daughter

ANALYSIS

I. Calculation of Number of Atoms of Fission-Product Elements

(a) Pile Operation Period

The equations and the calculations employed in this section are well known and the number of atoms of each fission product at any time t during pile operation may be easily determined provided that sufficient data on the quantitative chemical analysis of the fission products are available. Such data exist for U^{235} fission (references 1 and 2) and for five other nuclear fuels (reference 3).

The number of atoms of a fission product isotope is given by

$$N_n = \sum_{i=1}^m N_{n,i} \quad (1)$$

where

- N number of atoms of isotope
 n designates generation of daughters
 i chain considered
 m total number of chains which give rise to N

The defining equation for $N_{n,i}$ is:

$$\frac{dN_{n,i}}{dt} = N_f Y_{n,i} + \lambda_{n-1,i} N_{n-1,i} - [\lambda_{n,i} + \sigma_{cn,i}(nv)] N_{n,i} \quad (2)$$

where

- t time
 N_f number of fissions per second
 Y yield, number of atoms formed per fission
 λ decay constant
 σ_c thermal neutron capture cross section $\left(\frac{\text{cm}^2}{\text{neutron}}\right)$

The solution to this equation may be written

$$N_{n,i} = e^{-a_{n,i}t} \int_0^t F(t') e^{a_{n,i}t'} dt' \quad (3)$$

where

$$a_{n,i} = \lambda_{n,i} + \sigma_{cn,i}(nv)$$

$$F(t') = N_f Y_{n,i} + \lambda_{n-1,i} N_{n-1,i}$$

Wherever multiple parents or daughters exist, the chain may be split up into subchains, each of which are completely described by equations (2) and (3).

The n^{th} daughter of the chain may be either radioactive, whereupon it decays to the $(n+1)^{\text{st}}$ daughter of the chain; or stable

($\lambda_{n,i} = 0$), whereupon it is the last daughter of the chain. Also, $\sigma_{cn,i}^{(nv)}$ may be neglected in almost all cases. With this assumption, equation (3) may be written as

$$N_{n,i,r} = e^{-\lambda_{n,i}t} \int_0^t F(t') e^{\lambda_{n,i}t'} dt' \quad (3.1)$$

or

$$N_{n,i,s} = \int_0^t F(t') dt' \quad (3.2)$$

where the subscripts r and s designate radioactive and stable isotopes, respectively.

At this point, it is worthwhile to note:

(1) The expressions for every $N_{n,i}$, whether stable or radioactive, all contain N_F as a multiplicative factor. As a consequence, the number of atoms of each fission product is directly proportional to N_F with one exception. This exception occurs when the $(nv)\sigma_{cn}$ term may not be neglected, inasmuch as (nv) effects changes in the equation for $N_{n,i}$ other than as a simple multiplication factor.

(2) It is shown in appendix A that, while the reactor is operating, the number of atoms of every fission product increases monotonically, becoming a maximum at shutdown. Therefore, the maximum number within the pin will be developed either at the moment of shutdown or at some time during the shutdown period. Since the assumption will be made that the pressure is proportional to the number of atoms, calculation of the numbers at shutdown time will suffice for obtaining a maximum during pile operation.

In order to summarize the previous results, the method of calculating the number of atoms of an isotope in the case where $\sigma_{cn,i}^{(nv)}$ is negligible follows:

First, an explicit expression for an isotope formed only by direct fission is obtained. Here the $\lambda_{n-1} N_{n-1}$ term in the integrand is missing. If this isotope is stable, equation (3.2) yields the value of the number of atoms of this isotope $N_{0,s}$ at any time t . If this isotope is radioactive, it will be the parent of a radioactive series and the number of atoms $N_{0,r}$ of this isotope is given by use of

equation (3.1). To find the number of atoms of daughter N_1 , either equation (3.1) or (3.2) is used, depending on whether the first daughter is radioactive or stable. The expression found for $N_{0,r}$ corresponds to $N_{n-1,r}$ in arriving at a relation for N_1 . This process is repeated until the last isotope of the radioactive chain is reached, which is necessarily stable.

(b) Pile Shutdown Period

During the pile shutdown period there is zero (nv) and therefore no more fission takes place and the number of atoms is governed by the ordinary laws of radioactive decay. An equation of sufficient generality to cover this situation is

$$\frac{dN_n}{dt} = \lambda_{n-1} N_{n-1} - \lambda_n N_n \quad (2a)$$

which is merely a modification of equation (2). If shutdown time is taken as zero, the solution to this equation is given by

$$N_n = e^{-\lambda_n t} \left[N_n(t_s) + \int_0^t e^{\lambda_n t'} \lambda_{n-1} N_{n-1} dt' \right] \quad (3a)$$

where

N_n number of atoms of n^{th} daughter of chain present in pin at time t after shutdown

t time after shutdown

$N_n(t_s)$ number of atoms of n^{th} daughter of chain present in pin at time t_s of shutdown

Proceeding as was done in the pile operation period, equation (3a) takes on two forms and notation similar to that employed in equations (3.1) and (3.2) is employed to differentiate the two forms:

$$N_{n,r} = e^{-\lambda_n t} \left[N_{n,r}(t_s) + \int_0^t e^{\lambda_n t'} \lambda_{n-1} N_{n-1,r} dt' \right] \quad (3.1a)$$

and

$$N_{n,s} = N_{n,s}(t_s) + \int_0^t \lambda_{n-1} N_{n-1,r} dt' \quad (3.2a)$$

Equations (3.1a) and (3.2a) are used like equations (3.1) and (3.2) to obtain values of N_n for any element of a given radioactive chain at any time t after pile shutdown.

II. Calculation of Pressure

(a) Calculations of Chemical Composition

The problem of determining the chemical composition of the mixture of fission products at high temperatures is very complicated and perhaps impossible at this time. In order to obtain reasonable results, simplifying assumptions must be made. The assumptions which will be used herein are described in the following two cases:

(1) As a zeroth approximation, the case of no chemical interactions of any kind may be assumed. The calculations for this case merely involve adding up all the numbers of atoms of each isotope found in part I to get the number of atoms of element concerned. Each element is then assumed to remain in uncombined form. This includes any oxygen released from UO_2 fission or any possible alkali metal used as a heat-transfer medium. It is obvious that this case leads to a high maximum for the pressure.

(2) The following assumptions lead to a more realistic approximation:

(a) The chemical reactions that take place are those with most favorable free energies leading to the formation of simple two-element compounds that are chemically stable at the operating temperature.

(b) All reactions take place until the supply of one of the reacting elements is exhausted.

When explicit data are available, the free energy of formation will be approximated by

$$\Delta F = \Delta H_{298} - T \Delta S_{298}$$

where

ΔF free energy of formation $\left(\frac{\text{kcal}}{\text{mole}}\right)$

ΔH_{298} heat of formation at 298°K $\left(\frac{\text{kcal}}{\text{mole}}\right)$

T operating temperature ($^\circ\text{K}$)

ΔS_{298} entropy of formation at 298°K ($\text{cal}/^\circ\text{K}$)

All reactions for which $|\Delta F|$ is less than 10 will be neglected.

The calculations of chemical composition are the same for pile operation as for period following shutdown except that the drop in temperature in the shutdown period must be considered. In practice, this modification is relatively unimportant and the composition at the higher temperature may be assumed throughout.

(b) Calculation of Pressure Contributions of Compounds

The assumption is made that the pressure, like the number of atoms of fission-product isotope, increases monotonically during the operating period. Actually, because of the different rates of formation of various elements, the possibility of a pressure peak exists. However, this seems unlikely.

In calculating the pressure exerted by those substances that can be considered gases, the assumption is made that the gases act in accordance with the ideal gas law

$$pV = RT$$

where

p pressure

V free volume

R universal gas constant

T operating temperature ($^{\circ}$ K)

The pressure contributions of liquids or solids will be computed from Clapeyron's equation, Trouton's rule being used to estimate ΔH when necessary.

The calculations will be the same for the pile-operating period as for period following pile shutdown except that the drop in temperature during the shutdown period is very important and effects a considerable pressure reduction.

Division of the problem in the predescribed manner is useful pedagogically because the lines of thought in the two sections of Section I, are identical, whereas the considerations involved in Section II, are of an entirely different nature. However, the analysis of part II will almost always show that a relatively small number of elements are responsible for the pressure. For this reason, the computations will be shortened if the calculations of pile-operation period are carried out completely and then the pile shutdown calculations performed. Thus, the order of computation is: (1) Part I (a);

(2) Part II(a) and (b) applied to I(a); (3) Part I(b); 4 Part II(a) and (b) applied to I(b).

The example which follows illustrates this point well.

EXAMPLE OF CALCULATION OF PRESSURE

2420 Consider a pin containing 90 percent enriched U^{235} as fuel in the form of UO_2 . The internal operating temperature is $2000^\circ F$, $(nv) = 8 \times 10^{14}$, $N_f = 7.37 \times 10^{13}$, and there remains 0.0638-cubic-centimeter free volume in each pin after loading it with 0.447 gram UO_2 and 0.538 gram of a heat-transfer material (liquid sodium metal). Shutdown time is 10^6 seconds. What is the maximum internal pressure developed?

2-410 In calculating the amount of each fission product, references 1, 2, and 3 were consulted. The theoretical number of atoms produced per fission should be very close to 2.00 assuming two large fragments per fission. On page 2437 of reference 1, there is a graph of percentage of fission yield against mass number. A smooth curve has been drawn through the experimental points and the area under this curve is 197 percent, indicating good agreement with the theoretical value of 200 percent. However, addition of all the experimental points over 0.1 percent yields 116.88 percent, which is much too low. Many mass numbers above 0.1 percent on the curve had not been reported as found. Several of these missing points were subsequently reported in references 2 and 3. Addition of all the points listed in reference 2 yields the value 166 percent but many of the points are theoretical. The latest available data (reference 3) have only 129 percent listed for experimentally determined fission products.

All the additional reported points lie very close to the curve. Thus, the idea of assuming that the mass numbers not yet reported also lie on the curve and contribute to the number of atoms in the proportion indicated is a temptation. If the missing points are added (using the revised curve on p. 2007 of reference 3) the total is 191.3 percent which is very close to the theoretical value. At least, adding these points is more reasonable than assuming that all the mass numbers were reported too low in the ratio 1.66/2.00 (or 1.29/2.00) and correcting the results by multiplying every value by 2.00/1.66 (or 2.00/1.29).

The known isotopes of the missing mass numbers were investigated with the aid of reference 4 and the following conclusions regarding them were drawn:

There is a possibility of about 0.3×10^{18} atoms of Xe^{128} and 1.55×10^{18} atoms of Xe^{130} . These numbers assume that all the missing mass members 128 and 130 became xenon gas and are thus maxima.

In view of the lack of any more data, the proper assumption to be made here is that the estimates of fission yield are all too low and are to be multiplied by the factor 2.00/1.91 or 1.045.

This procedure of filling in the missing points on the curve to determine the total number of all the atoms is the accepted one at present (reference 5).

Unfortunately, reference 3 was published after all the computations had been completed. Therefore, the values given in tables I, II, and IV are those based on the data in reference 2, and have not been corrected by the factor 1.045. Comparison with the data in reference 3 indicates that it would not be profitable to recalculate the values in these tables. (See the last page of appendix B).

The one instance for which $(nv)\sigma_c$ becomes appreciable occurs for Xe^{135} whose thermal cross section for capture at this temperature is about 1×10^6 barns. (reference 6). Calculations based on a value of 3×10^6 barns were made since that was the only value available at the time. As is shown in table I, Xe^{135} actually forms a negligible portion of the total amount of xenon gas; therefore the consideration regarding the $(nv)\sigma_c$ term may be totally neglected in this example.

Details of calculations of typical values of the number of atoms of fission-product isotopes present after 10^6 seconds of operation are given in appendix B. The number of atoms of each fission-product isotope in each chain having a yield of 0.1 percent or greater (reference 6) is listed in table I.

Summation of all the values in a given column of table I yields the number of atoms of fission-product elements present after 10^6 seconds of operation. In addition to the actual fission-product elements, oxygen is released upon the fissioning of U^{235} , which was in the form of UO_2 , and sodium is present as the heat-transfer medium.

The number of oxygen atoms released in the pin at the end of 10^6 seconds is given by multiplying the number of fissions occurring in 10^6 seconds by 2 inasmuch as two oxygen atoms are released per fission. Hence, at shutdown time, the number of oxygen atoms present in each pin is equal to $(7.37 \times 10^{13} \times 10^6)2 = 1.474 \times 10^{20}$. The number of atoms of sodium present is obtained by use of the given weight of sodium. All the results described in the preceding two paragraphs are found in table II.

CALCULATION OF PRESSURES

Calculation of Chemical Composition

The free energies at 1366°K of the probable two-element compounds that may be formed from the fission products and the sodium and oxygen

are given in table III (references 7, 8, and 9). For the halides, ΔF is given in kilocalories per gram-atom of oxygen. In the case of the oxides of technetium, ruthenium, rhodium, and palladium, general estimates were employed and are indicated by the parentheses surrounding these estimated values.

On the basis of table III, a list of the most stable oxides of each of the elements present in the pin with $-\Delta F_{1366}$ greater than 10 kilocalories per gram-atom of oxygen may be established. This list is presented in order of decreasing ΔF :

La_2O_3	$(\text{Gd})_2\text{O}_3$	ZrO_2	In_2O_3
Y_2O_3	Eu_2O_3	NbO_2	TcO_2
Ce_2O_3	Sm_2O_3	Na_2O	Rb_2O
Pr_2O_3	SrO	Sb_2O_3	Cs_2O
Nd_2O_3	BaO	MoO_2	CdO

In considering the oxides that might form at 1366°K , the assumption has been made that those oxides having the more negative free energy of formation at 1366°K will be present in the equilibrium composition and the remaining oxides will not be present, if forming the former oxides requires that the complete supply of free oxygen in the pin be exhausted.

From table III, it can be noted that the free energy of the resulting equilibrium composition can be lowered most by assuming that the alkali metals combine with the halogens, bromine and iodine, rather than with the oxygen. Cesium is the element first to combine with the halogens. Inasmuch as the quantity of cesium present in the pin at shutdown is more than enough to exhaust completely the quantities of both bromine and iodine, the only halogens assumed present in the pin at shutdown are cesium bromide and cesium iodide.

Table IV shows the composition calculated on the basis of the preceding paragraph for case 1 where sodium is present as the heat-transfer medium and for case 2 where a chemically inert material exerting no vapor pressure is used as the heat-transfer medium.

Calculations of Pressure Contributions

For those substances that can be considered gases at 1366°K , the ideal gas law is assumed. Upon substitution of the given values for volume and temperature, it is found that

$$p_g = 2.91 \times 10^{-18} M_{fg} \text{ atmospheres}$$

where

p_g pressure due to gas

M_{fg} molecules of gas

For the remaining substances, the vapor pressures were estimated by means of the Clapeyron equation

$$\ln p = \frac{\Delta H}{R} \left(\frac{1}{T_B} - \frac{1}{1366} \right)$$

where T_B the boiling point in $^{\circ}\text{K}$. Where only T_B is known, ΔH was estimated by Trouton's rule $\Delta H \approx 21T_B$. When neither T_B nor ΔH were known, estimates were made based on similar compounds.

The vapor pressure exerted by a substance at 1366°K was considered negligible if that pressure were less than or equal to 0.05 atmosphere. Where the oxides are concerned, the vapor pressures are negligible except for CdO and the possible exceptions of Na_2O , Rb_2O , and Cs_2O . The oxides of these alkali metals do not have a true boiling point but rather vaporize by dissociation to the metal and oxygen gas (references 8 and 9). Calculations of their equilibrium pressure at 1366°K based on free-energy data seem to indicate that these oxides do possess vapor pressures less than 0.05 atmosphere. Hence, the vapor pressures of the alkali metal oxides at 1366°K are considered negligible.

A list of substances with non-negligible vapor pressures as defined herein follows:

Substance	Vapor pressure at 1366°K , atm
Na	3.62
Rb	18.3
Cs	13.7
Sr	0.11
Cd	15.9
Te_2	2.1
CsBr	0.14
CsI	0.20
CdO	0.10

The substances which are in the form of gases are cesium, rubidium, cadmium, tellurium, krypton, xenon. The sum of the number of molecules of these substances present according to table IV is 24.7×10^{18} . To this should be added the total of 1.35×10^{18} possible additional atoms of Xe^{128} and Xe^{130} from unreported mass numbers 128 and 130 which gives a total of 26.05×10^{18} molecules of gas or 77.3 atmospheres.

2420 There remains sodium, cesium, bromine, and cesium iodide, the vapor pressures of which contribute an additional 3.96 atmospheres, making a total of 81.36 atmospheres. If this value is corrected by the factor 1.045, the final pressure becomes 85 atmospheres.

For purposes of comparison, calculations are presented for case 2, wherein the sodium is replaced by an inert substance exerting no vapor pressure. Here Te_2 , O_2 , Kr, and Xe will be present as gases and their pressure contribution is 82.8 atmospheres. In addition, CsBr, CsI, and CdO contribute 0.44 atmosphere for a total of 83.24 atmospheres. This value is now augmented by adding the 5.5 atmosphere contribution of Xe^{128} and Xe^{130} and then correcting the sum of the factor 1.045, so that the final pressure is 92.7 atmospheres.

In order to set a maximum on the attainable pressure, case 3 where no chemical interactions occur is presented. Now the gases present are rubidium, cadmium, oxygen, tellurium, bromine, iodine, krypton, and xenon, which contribute a total of 287.5 atmospheres including the Xe^{128} and Xe^{130} . The vapor pressures of sodium, cesium, and strontium add another 17.4 atmospheres for a corrected grand total of 319 atmospheres.

Pile Shutdown Period

As can be seen from the summation for the first two cases at the end of the preceding section, the gases always contribute the major part of the total pressure and the important gases are cesium, rubidium, krypton, and xenon. Hence, the calculations to determine whether the pressure at any time after pile shutdown exceeds that existing at t_s may be simplified by considering only those chains which contain an appreciable percentage of these four elements. Calculations of the number of atoms of the four gases, cesium, rubidium, krypton, and xenon, show that at the end of 2 hours of shutdown there is only a very slight increase above that number existing at shutdown and even after 50 hours, the percentage increase is less than 5. The decrease in temperature is more than enough to offset this increase. Hence, the possibility that the maximum pressure within the pin occurs at some time during the cooling-off period is rejected, and the maximum calculated pressure remains 85 atmospheres.

Precision and Limits of Results

The case of no chemical interactions sets a maximum for the pressure which is obviously too high. This case is unrealistic and will not be considered further.

Although the thermodynamic computations were of a simple and crude nature implied by the assumptions that only two-element compounds were formed and that all reactions proceed until one of the reactants is completely exhausted, it can be shown that the pressures as computed are accurate to within about 30 percent. This accuracy arises from the fact that the pressure is due mainly to the gases present in the pin. In the least favorable case, the gases contribute 93 percent of the total pressure. Inasmuch as the halides of the metals possess very small vapor pressures and all the oxides can be considered to exert a negligible pressure, it is only necessary to consider those elements which at 1366° K are gases or possess large vapor pressures. If it can be assumed that very little free oxygen is present in the pin, the elements to be considered are xenon, krypton, rubidium, and cesium. Therefore, a very good estimate of a maximum may be arrived at by adding the pressure due to considering only these four elements in the uncombined state at 1366° K and the vapor pressure of sodium if sodium is to be present. The point that must be emphasized is that free oxygen can only be present in very small amounts if this maximum is to hold. For case 1, this last condition, of course, holds true. For case 2, it is obvious that this condition is not fulfilled.

An estimate may also be made of the minimum pressure that occurs within the pin by just considering the inert gases xenon and krypton plus the vapor pressure of sodium if sodium is present. In every instance, these two gases, contributed at least 70 percent of the total pressure provided again that the assumption can be made that very little free oxygen is present in the pin at the equilibrium composition.

This method of calculating the pressure in the pin at 1366° K yields a very good approximation to the pressure as calculated and hence offers a simple method for estimating the pressure existing in the pin at shutdown if the assumption can be made that only a small amount of free oxygen is present in the pin. At times less than shutdown, this approximate method wherein only the quantity of xenon, krypton, rubidium, and cesium need be determined can also be used to estimate the pressure in the pin.

At temperatures below 1366° K, this simplified method may be applied without change, the proportionate effect of rubidium and cesium on the total pressure decreasing as the temperature drops because of their rapidly decreasing vapor pressures. However, at higher temperatures, the calculations become more complex as the oxides, the halides, and elements other than the four considered begin to exert significant vapor pressures.

It should be noted that, xenon by itself exerts more than 60 percent of the pressure as calculated for each case so that a fairly good estimation of the pressure in the pin may be obtained just by a consideration of xenon alone. In a pile of lower thermal flux, the proportion of xenon is slightly reduced, cesium gaining at the expense of the xenon.

DISCUSSION

Factors Affecting Pressure

As the general analysis implied, the variables that are important in the calculations considered are: (1) the thermal neutron flux, (2) the free volume of the pin, (3) the operating temperature, (4) the shutdown time, and (5) the fuel. These factors are discussed in the order indicated.

Thermal neutron flux. - The example has shown that for U^{235} , the $(nv)\sigma_c$ term may be neglected so that the number of molecules (and the pressure) vary linearly with the flux. Thus, the pressure for any other flux (everything else remaining constant) can be found directly from the values in these tables. If a different fuel is used, it becomes necessary to redo the calculation in order to determine the importance of Xe^{135} .

Fuel volume. - It seems reasonable to assume that the pressure will be due mainly to gases. If this is the case and the ideal gas law is used, free volume also affects the pressure linearly except that the relation is one of inverse proportionality. Note that a compromise must be reached inasmuch as an increase in free volume lowers the over-all heat-transfer coefficient, resulting in higher temperatures within the pin for the same temperature rise of the coolant.

Operating temperature. - The dependence of the pressure on the temperature is quite complicated. Of course, for the gases, the relation is again linear. Nevertheless, the important effect is qualitative, that is, the temperature determines not only the states of the substances but also, to some extent, their chemical composition.

In this connection, it may be mentioned that at temperatures below $1366^\circ K$, the simplified method of computing the pressure, which was described in the preceding section, may be applied without change because the proportionate effect of rubidium and cesium decreases because of lowered vapor pressures. At higher temperatures, however, the calculations become more complex as the oxides, the halides, and elements other than the four considered herein begin to exert significant vapor pressures.

In general, temperature will strongly affect the computation in Section I but will leave all results in Section II unchanged.

Shutdown time. - The expressions in appendix B show clearly that the number of atoms of each isotope is a complicated function of time. Thus, the number of atoms of a given element are even more complicated functions of time. Any change in the shutdown time, therefore necessitates a complete recalculation of results from start to finish. It should be remembered, however, that the number of atoms of each element (and hence the pressure) increases monotonically with the time.

Fuel. - Although U^{235} seems to be the preferred fuel in the majority of cases at this time, as more data on other fuels become available, they will undoubtedly be used in some designs. Because the fission-yield curve of each fuel is unique, calculations for each would have to be made anew. Reference 3 seems to indicate that the same elements would be present in all the known fuels as are present from U^{235} fission, but the amounts of each vary from fuel to fuel.

OTHER FACTORS

A discussion of the specific design for which calculations were made in the example will illustrate some of the other factors which may affect the pressure.

In preparing the list of oxides that form in the pin at 1366° K, the most stable oxide of the metal was chosen and no oxide was considered unless its free energy of formation at 1366° K was more negative than 10 kilocalories per gram-atom of oxygen. Inasmuch as these most stable oxides generally have a lower ratio of oxygen to metal than other possible oxides that might exist at the operating temperature of 1366° K (see table III) and oxides such as TeO_2 and RuO_2 probably will form in the high free-oxygen concentration found for case 2, it is very probable that a very much smaller amount of free oxygen would exist than that listed in the pressure tabulation for case 2 in which event the pressure for case 2 might decrease from that calculated by as much as the pressure due to free oxygen, 27.6 atmospheres. Consequently, if no appreciable amount of free oxygen were present in the pin in case 2, the total pressure would be 64.0 atmospheres instead of the value 92.7 calculated. Because oxygen is so important in the calculations for pressure, it would seem that the presence of an oxide former of comparatively low vapor pressure is desirable.

In regard to sodium, Na_2O decomposes into its primary constituents at some temperature above the melting point of 1000° K. Thus, it is impossible to say with certainty that the presence of sodium serves to reduce the pressure.

0230

SUMMARY OF RESULTS

An analysis of the general problem of calculating the internal pressure in the fuel tube of a nuclear reactor was made. The calculation may, in general, be broken down in the following manner:

I. Calculation of Number of Atoms of Fission-Product Elements

- (a) Pile Operating Period
- (b) Period Following Pile Shutdown

II. Calculation of Pressure

- (a) Calculation of Chemical Composition
- (b) Calculations of Pressure Contributions of Compounds

In practice, the number of calculations may be reduced if performed in the following order:

- (1) I(a); (2) All of II; (3) I(b); (4) All of II.

A simplification of the calculations of maximum pressure developed during operation has been presented in the form of a proof that each fission product is monotonically increasing. This means that during operation the maximum amount of each fission product is present at time of shutdown. The importance of this simplification lies in the fact that in order to determine the maximum amounts of fission products during operation, it suffices to make calculations for the single time value t_s , shutdown time.

The general analysis is applied to a closed reactor system consisting of 90 percent enriched U^{235} in the form of UO_2 packed in a small pin operating at a flux of 8×10^{14} at 2000° F. These calculations show that if the reactor is run for 10^6 seconds a design value of 100 atmospheres internal pressure would suffice since the most probable pressure is about 85 atmospheres and 85 atmospheres is conservative.

For this case the bulk of the pressure has been shown to depend on the amounts of the four elements xenon, krypton, rubidium, and cesium present in the pin. The amount of free oxygen present is also important.

Lewis Flight Propulsion Laboratory,
National Advisory Committee for Aeronautics,
Cleveland, Ohio, January 17, 1952.

APPENDIX A

It is obvious that during operation, a stable isotope will continue to increase and also probably during shutdown as well, if it is formed from a series of radioactive isotopes. Therefore, only the radioactive isotopes formed from fission need be examined.

The equation for the number of atoms of a parent isotope is given by

$$N_{n-1,r} = \frac{N_{f,p} Y_{n-1}}{a_{n-1}} (1 - e^{-a_{n-1} t})$$

where

$$a_{n-1} = \lambda_{n-1} + (nv) \sigma_{c_{n-1}}$$

and

$$t \leq t_s$$

where t_s is the time of operation of the pile up to shutdown. Thus, $N_{n-1,r}$ increases monotonically because

$$\frac{dN_{n-1,r}}{dt} = N_{f,p} Y_{n-1} e^{-a_{n-1} t} > 0$$

for all $t \leq t_s$

Now

$$\frac{dN_{n,r}}{dt} = \lambda_{n-1} N_{n-1,r} + N_{f,p} Y_n - a_n N_{n,r}$$

and

$$\frac{d^2 N_{n,r}}{dt^2} = \lambda_{n-1} \frac{dN_{n-1,r}}{dt} - a_n \frac{dN_{n,r}}{dt}$$

If $N_{n,r}$ reaches a maximum at $t_0 < t_s$ then at t_0 ,

$$\frac{dN_{n,r}}{dt} = 0$$

and

$$\frac{d^2 N_{n,r}}{dt^2} < 0$$

However, $\frac{dN_{n-1,r}}{dt} > 0$ for all t . Therefore, no maxima or minima of $N_{n,r}$ occur during operation.

Furthermore, $\frac{dN_{n,r}}{dt} > 0$ for all $t \leq t_s$. For at $t = 0$, $N_{n,r} = 0$, whereas at a time close to $t = 0$, $N_{n,r} > 0$ so that $\frac{dN_{n,r}}{dt} > 0$ for some t such that $t \leq t_s$. In order for $\frac{dN_{n,r}}{dt}$ to be negative, it would have to pass through the value 0 because $N_{n,r}$ is a continuous function of t with continuous n^{th} order derivatives at all points.

Successive application of this argument shows that $\frac{dN_{n,r}}{dt} > 0$ for all t such that $0 \leq t \leq t_s$ and all n . Therefore, the number of atoms of each fission product is a maximum at the time of shutdown.

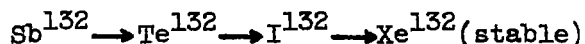
The preceding analysis has shown that the number of atoms of any isotope increases with time while the reactor is in operation and that no maxima exist.

APPENDIX B

An attempt will be made here to include at least one calculation of every type which is involved in obtaining values for table I. Included are (1) plural fission yields with the same mass number, (2) "branching" or plural daughters, (3) plural parents, (4) case where the $(nv)\sigma_c$ term plays a significant role.

Xenon is the most important element for pressure considerations, therefore calculation of a few of the most important chains involving it should prove interesting.

The first step in the calculation of Xe^{132} is to write down the "pedigree" of the isotope. From reference (1) (or (3)), the chain is found to be:



Now recourse to equations (1) to (3) may be had. There is only one chain to consider for mass number 132 and there are no plural parents or daughters in it. For this chain Sb is the parent, Te is the first daughter, I is the second daughter, and Xe is the stable third daughter, so that the number of atoms of each will be designated, respectively, by N_0, N_1, N_2, N_3 with N_3 being stable.

From equation (3.1), it follows that:

$$N_0 = e^{-a_0 t} \int_0^t N_f Y_0 e^{a_0 t'} dt'$$

where

$$a_0 = \lambda_0$$

$$N_0 = \frac{N_f Y_0}{\lambda_0} (1 - e^{-\lambda_0 t}) \quad (B1)$$

Also

$$N_1 = e^{-\lambda_1 t} \int_0^t (N_f Y_1 + \lambda_0 N_0) e^{\lambda_1 t'} dt'$$

which becomes upon substituting equation (B1) for N_0 ,

$$N_1 = N_f \left[\frac{Y_0 + Y_1}{\lambda_1} - \frac{Y_0}{\lambda_1 - \lambda_0} e^{-\lambda_0 t} - \frac{1}{\lambda_1} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_1 - \lambda_0} \right) e^{-\lambda_1 t} \right] \quad (B2)$$

Similarly, the expression for N_2 is

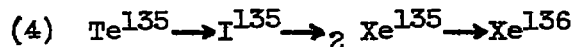
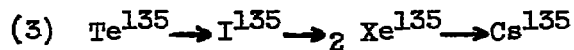
$$N_2 = N_f \left\{ \frac{Y_0 + Y_1 + Y_2}{\lambda_2} - \frac{\lambda_1 Y_0}{(\lambda_2 - \lambda_0)(\lambda_1 - \lambda_0)} e^{-\lambda_0 t} - \frac{1}{\lambda_2 - \lambda_1} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_1 - \lambda_0} \right) e^{-\lambda_1 t} - \frac{1}{\lambda_2} \left[Y_2 - \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_2 - \lambda_0} \right) \right] e^{-\lambda_2 t} \right\} \quad (B3)$$

Finally, equation (3.2) yields

$$N_3 = \int_0^t \lambda_2 N_2 dt$$

The $Y_3 N_f$ term is missing because Y_3 is zero according to available data. Upon integration

$$N_3 = N_f \left\{ - \left[\frac{\lambda_2 \lambda_1 Y_0}{\lambda_0 (\lambda_2 - \lambda_0) (\lambda_1 - \lambda_0)} + \frac{\lambda_2}{\lambda_1 (\lambda_2 - \lambda_1)} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_1 - \lambda_0} \right) + \frac{1}{\lambda_2} \left(Y_2 - \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_2 - \lambda_0} \right) \right) \right] + (Y_0 + Y_1 + Y_2) t + \frac{\lambda_2 \lambda_1 \lambda_0}{\lambda_0 (\lambda_2 - \lambda_0) (\lambda_1 - \lambda_0)} e^{-\lambda_0 t} + \frac{\lambda_2}{\lambda_1 (\lambda_2 - \lambda_1)} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_1 - \lambda_0} \right) e^{-\lambda_1 t} + \frac{1}{\lambda_2} \left[Y_2 - \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(Y_1 - \frac{\lambda_0 Y_0}{\lambda_2 - \lambda_0} \right) \right] e^{-\lambda_2 t} \right\} \quad (B4)$$



For Xe^{135} , equation (B3) may be used with the following simple modifications:

The N_f factor is multiplied by the branch yield, 0.1, and the $Y_{1\text{Xe}^{135}}$ is divided by it, also $\lambda_{1\text{Xe}^{135}}$ is replaced by $a_{1\text{Xe}^{135}}$. The pertinent data are

$$Y_{\text{Te}^{135}} = 3.84 \times 10^{-2}$$

$$\lambda_{\text{Te}^{135}} = 1.93 \times 10^{-2} \text{ sec}^{-1}$$

$$Y_{\text{I}^{135}} = 1.83 \times 10^{-2}$$

$$\lambda_{\text{I}^{135} \rightarrow 1\text{Xe}^{135}} = 2.88 \times 10^{-5} \text{ sec}^{-1}$$

$$Y_{1\text{Xe}^{135}} = 2.36 \times 10^{-4}$$

$$\lambda_{1\text{Xe}^{135}} = 8.9 \times 10^{-4} \text{ sec}^{-1}$$

The $(nv)\sigma_c$ term = $2.4 \times 10^{-3} \text{ sec}^{-1}$ so that $a_{1\text{Xe}^{135}} = (2.4 + 0.89) \times 10^{-3} = 3.29 \times 10^{-3}$. If these values be substituted into the modified equation (B3), a value of 1.32×10^{14} atom will be obtained as given in table I.

The number of atoms of 2Xe^{135} in chains (1) and (2) is given by a formula for N_{3_r} which is not among equations (B1) and (B4). The equation which describes $N_{2\text{Xe}^{135}}$ is:

$$\frac{d N_{2\text{Xe}^{135}}}{dt} = \frac{N_f Y_{2\text{Xe}^{135}}}{10} + \lambda_{1\text{Xe}^{135}} N_{1\text{Xe}^{135}} - (\lambda_{2\text{Xe}^{135}} + (nv)\sigma_c) N_{2\text{Xe}^{135}}$$

The integrated form is more complicated than equation (B4). However, the exponential terms are negligible so only the constant term enters into the calculations. This term is

$$\frac{N_f}{10 a_{2\text{Xe}^{135}}} \left[Y_{2\text{Xe}^{135}} + \frac{\lambda_{1\text{Xe}^{135}}}{a_{1\text{Xe}^{135}}} \left(Y_{\text{Te}^{135}} + Y_{\text{I}^{135}} + 10 Y_{1\text{Xe}^{135}} \right) \right]$$

With the additional data, $\lambda_{2\text{Xe}^{135}} = 2.08 \times 10^{-5} \text{ sec}^{-1}$ and $Y_{2\text{Xe}^{135}} = 2.13 \times 10^{-3}$, the preceding expression becomes 5.51×10^{13} , which is the value given.

The number of atoms of 2Xe^{135} formed from chains (3) and (4) is found by using modified equation (B3), with the branch yield now being 0.9 instead of 0.1.

The final significant series of calculations is the one leading to the number of atoms of Xe^{136} . This isotope is stable but it has several sources of formation: (a) The 1Xe^{135} ; (b) 2Xe^{135} in chains (2) and (4); (c) the simple chain $\text{I}^{136} \rightarrow \text{Xe}^{136}$; (d) the chain $\text{I}^{137} \rightarrow 1\text{Xe}^{136}$; (e) and, according to reference 3, there is a 3 percent direct fission yield. Calculations of the (c) and (d) type have been done already and will not be repeated. For (a) and (b), equation (3.2) is modified so that

$$N_{\text{Xe}^{136}} \text{ (from } 1\text{Xe}^{135}) = \int_0^t (nv)\sigma_c N_{1\text{Xe}^{135}} dt'$$

Otherwise, there is no change. Calculations of type (e) are very simple, the applicable equation being

$$N_{\text{Xe}^{136}} \text{ (from direct fission)} = N_f Y_{\text{Xe}^{136}} t$$

If the difference in fission yield of I^{136} and Xe^{136} in reference 3 is taken as $Y_{\text{Xe}^{136}}$, there is an additional 2×10^{18} atoms of Xe^{136} or about 6 more atmospheres than have been calculated in the body of this report.

Finally, if the corrected σ_c of 1×10^6 barns is used instead of 3×10^6 barns, $N_{1\text{Xe}^{135}}$ will be twice, the $N_{2\text{Xe}^{135}}$ from chains (1) and (2) would be 5.4 times, and that from chains (3) and (4) about three times the values in table I. But what is more important is that $N_{\text{Xe}^{136}}$ is not changed significantly, except that the direct fission yield is only 2 atmospheres instead of 6.

REFERENCES

1. Nuclei Formed in Fission: Decay Characteristics, Fission Yields, and Chain Relationships - Issued by the Plutonium Project. Jour. Am. Chem. Soc., vol. 68, no. 11, Nov. 1946, pp. 2411-2442.
2. Hunter, H. F. and Ballov, N. E.: Simultaneous Slow Neutron Fission of U^{235} Atoms. I. Individual and Total Rates of Decay of the Fission Product. ADC-65, Apr. 11, 1949.
3. Coryell, Charles D., and Sugarman, Nathan: Radiochemical Studies. The Fission Products. Book 3, McGraw-Hill Book Co., Inc., 1951.
4. General Electric Chart of the Nuclides.
5. Private Communication with Nathan Sugarman. Nov. 1951.
6. Xe Effect in an Epi-thermal Reactor. USAEC, Y-F10-17, Oct. 10, 1950.
7. Quill, Lawrence L.: The Chemistry and Metallurgy of Miscellaneous Materials: Thermodynamics. McGraw-Hill Book Co., Inc., 1950, Tables 6.3 and 7.1, pp. 104-115 and 196-207, respectively.
8. Brewer, Leo: The Thermodynamic Properties of the Oxides, UCRL-104.
9. Smithells, Colin J.: Metals Reference Book. Interscience Publishers Inc., 1949.

2420

CE-4

TABLE I - NUMBER OF ATOMS OF EACH FISSION-PRODUCT ISOTOPE AT SHUTDOWN

Mass	Chain	Number of Atoms														
		As	Se	Br	Kr	Rb	Sr	Y	Zr	Nb	Mo					
81	1	5.67x10 ¹¹	3.56x10 ¹³	9.72x10 ¹²	6.56x10 ¹⁵											
81	2	8.85x10 ¹²		1.52x10 ¹⁴	1.05x10 ¹⁷											
83	1		2.04x10 ¹³		2.63x10 ¹⁵	2.06x10 ¹⁵	2.07x10 ¹⁷									
83	2			5.56x10 ¹⁴	2.06x10 ¹⁵	1.61x10 ¹⁵	1.61x10 ¹⁷									
84			9.78x10 ¹³		1.53x10 ¹⁵		8.88x10 ¹⁷									
85	1				2.04x10 ¹⁴	1.84x10 ¹⁸		7.66x10 ¹⁷								
85	2				4.48x10 ¹³		1.72x10 ¹⁷	2.01x10 ¹²								
87	1				5.53x10 ¹²		4.57x10 ¹⁶									
87	2				1.50x10 ¹⁴		1.31x10 ¹⁶	2.01x10 ¹⁸								
88					4.44x10 ¹³		3.68x10 ¹⁶	3.97x10 ¹⁵	8.54x10 ¹⁸							
89							7.2x10 ¹⁴	4.58x10 ¹⁵	3.18x10 ¹⁸	2.5x10 ¹⁷						
90							1.45x10 ¹⁴	5.42x10 ¹²	3.76x10 ¹⁸	7.63x10 ¹⁴	8.92x10 ¹⁴					
91	1						1.37x10 ¹³	1.31x10 ¹⁵	8.04x10 ¹⁶	7.05x10 ¹⁵	1.4x10 ¹⁸	1.0x10 ¹⁷				
91	2						2.06x10 ¹³	1.87x10 ¹⁵	1.21x10 ¹⁷		2.11x10 ¹⁸	1.8x10 ¹⁷				
92							6.84x10 ¹²	4.99x10 ¹²	5.98x10 ¹⁶		7.74x10 ¹⁶	4.13x10 ¹⁸				
93							2.17x10 ¹²	2.53x10 ¹⁴	2.54x10 ¹⁵		2.29x10 ¹⁷		4.19x10 ¹⁸			
94							3.69x10 ¹¹	2.30x10 ¹²	8.42x10 ¹⁴		7.92x10 ¹⁵	4.66x10 ¹⁸				
95	1									2.38x10 ¹⁴	8.78x10 ¹⁶	3.01x10 ¹⁵	2.36x10 ¹⁵	1.43x10 ¹⁴		
95	2									1.13x10 ¹⁶	4.29x10 ¹⁸		2.51x10 ¹⁷	2.02x10 ¹⁶		
97										2.05x10 ¹⁵	3.92x10 ¹⁷		2.98x10 ¹⁶	4.15x10 ¹⁸		

TABLE I - NUMBER OF ATOMS OF EACH FISSION-PRODUCT ISOTOPE AT SHUTDOWN - Continued.

Mass	Chain	Number of Atoms						
		Mo	Tc	Ru	Rh	Pd	Cd	In
99	1	1.43×10^{17}	1.28×10^{18}	2.80×10^{17}				
99	2	1.28×10^{18}	2.64×10^{18}					
101		4.56×10^{15}	4.39×10^{15}	3.81×10^{18}				
102		3.08×10^{15}	1.34×10^{14}	3.10×10^{18}				
103	1			2.22×10^{18}	2.04×10^{15}	2.1×10^{17}		
103	2			6.90×10^{18}	6.0×10^{15}			
105		1.94×10^{13}	1.82×10^{15}	1.73×10^{18}	1.38×10^{17}	5.79×10^{17}		
106				3.65×10^{17}	3.47×10^{11}	4.16×10^{15}		
107			4.90×10^{12}	5.06×10^{13}	3.06×10^{14}	1.47×10^{17}		
115	1						2.11×10^{16}	1.71×10^{15} 5.25×10^{16}
115	2						5.2×10^{15}	5.0×10^{14}

TABLE I - NUMBER OF ATOMS OF EACH FISSION-PRODUCT ISOTOPE AT SHUTDOWN - Continued.

Mass	Chain	Number of Atoms						
		Sb	Te		I	Xe	Cs	Ba
127	1	1.03×10^{16}	1.36×10^{16}	5.83×10^{15}	4.21×10^{14}			
127	2	5.21×10^{16}		5.13×10^{15}	6.6×10^{16}			
129	1	3.02×10^{15}	1.20×10^{17}	1.89×10^{14}	1.6×10^{16}			
129	2	1.29×10^{16}		3.61×10^{15}	5.81×10^{17}			
131			5.73×10^{16}	5.27×10^{15}	1.51×10^{18}	8.50×10^{17}		
132		9.06×10^{14}		1.02×10^{18}	3.20×10^{16}	1.74×10^{18}		
133		1.72×10^{14}		1.72×10^{18}	4.13×10^{17}	1.75×10^{18}	1.44×10^{18}	
134		5.72×10^{13}		1.17×10^{16}	1.83×10^{16}	3.89×10^{18}		
135	1			1.46×10^{13}	1.45×10^{16}	1.322×10^{14}	5.51×10^{13}	1.11×10^{15}
135	2			1.32×10^{14}	1.31×10^{17}		1.61×10^{15}	3.25×10^{16}
136					5.79×10^{14}		4.42×10^{18}	
							$^a 4.347 \times 10^{17}$	
							$^b 3.75 \times 10^{18}$	
							$^c 2.21 \times 10^{17}$	
137	1				6.13×10^{12}			
137	2				8.16×10^{13}		1.24×10^{15}	4.35×10^{18}
								5.61×10^{11}
								7.07×10^{14}

(a) From 135 chain 1.

(b) From 135 chain 2.

(c) From 137.

TABLE I - NUMBER OF ATOMS OF EACH FISSION-PRODUCT ISOTOPE AT SHUTDOWN - Concluded.

Mass	Chain	Number of Atoms															
		I	Xe	Cs	Ba	La	Ce	Pr	Nd	Sm	Su	Fu					
138		2.01×10^{13}	6.24×10^{15}	1.30×10^{18}	4.70×10^{18}												
139		4.71×10^{12}	2.07×10^{14}	2.78×10^{15}	3.42×10^{16}	4.58×10^{18}											
140			5.19×10^{13}	2.33×10^{14}	3.35×10^{18}	3.65×10^{17}	7.8×10^{17}										
141			4.87×10^{12}	5.63×10^{14}	6.72×10^{15}	8.36×10^{16}	3.69×10^{18}	5.7×10^{17}									
142				2.73×10^{14}	1.96×10^{15}	2.69×10^{16}	4.17×10^{18}										
143						6.48×10^{15}	6.82×10^{17}	2.59×10^{18}	7.13×10^{17}								
144						1.79×10^{15}	3.71×10^{18}	1.64×10^{14}	5.41×10^{18}								
145						2.36×10^{14}	3.28×10^{16}	8.28×10^{16}	3.41×10^{18}								
146							3.54×10^{15}	6.30×10^{15}	2.94×10^{18}								
147									1.36×10^{18}	5.59×10^{17}	1.13×10^{15}						
149									9.12×10^{15}	2.48×10^{17}	7.70×10^{17}						
151									5.86×10^{12}	3.83×10^{14}	3.68×10^{17}	2.025×10^{14}					
153										9.30×10^{12}	2.87×10^{16}	8.42×10^{16}					

TABLE II - NUMBER OF ATOMS OF FISSION-PRODUCT ELEMENTS AT SHUTDOWN

Element metals	Number of atoms
Rb	2.79×10^{18}
Cs	5.84
Sr	9.724
Ba	8.09
Y	4.08
La	5.08
Ce	13.07
Pr	3.25
Nd	8.49
(61)	.81
Sm	1.17
Eu	.08
Zr	9.51
Nb	8.67
Mc	5.60
Tc	2.94
Ru	9.38
Rh	.36
Pd	.83
Cd	.03
In	.05
Sb	.08
Te	1.254
Active Nonmetals	
Br	0.12
I	2.78
O ^a	147.4
Inert Gases	
Kr	1.25
Xe	17.07
Na	13,570

(a) Oxygen is released from UO_2 when the uranium fissions.

TABLE III - THERMODYNAMIC PROPERTIES OF THE POSSIBLE TWO-ELEMENT COMPOUNDS
THAT MAY BE FORMED WITHIN A PIN

Compound	ΔF_{1366} , kcal	Compound	ΔF_{1366} , kcal
NaBr	-59.0	Na ₂ O	-52.5
Na I	-47.3	Rb ₂ O	-31.8
Rb Br	-64.0	Cs ₂ O	-31.6
Rb I	-53.0	Sr O	-106.9
Cs Br	-66.6	Ba O	-101.8
Cs I	-56.3	1/3 Y ₂ O ₃	-119
Sr Br ₂	-132.2	1/3 La ₂ O ₃	-122
Sr I ₂	-107.7	1/3 Ce ₂ O ₃	-119
Ba Br ₂	-136.5	1/2 CeO ₂	-93
Ba I ₂	-110.6	1/3 Pr ₂ O ₃	-118
Y Br ₃	-140	1/11 Pr ₆ O ₁₁	-97
Y I ₃	-92.5	1/3 Nd ₂ O ₃	-117
La Br ₃	-163.3	1/3(61) ₂ O ₃	(-115)
La I ₃	-115.5	1/3 Sm ₂ O ₃	-113
Ce Br ₃	-158.3	1/3 Eu ₂ O ₃	(-115)
Ce I ₃	-112.9	1/2 Zr O ₂	-97.7
Pr Br ₃	-157.7	1/2 Nb O ₂	-66
Br Pr I ₃	-111.9	1/5 Cb ₂ O ₅	-62.5
Nd Br ₃	-157	1/2 MoO ₂	-41
Nd I ₃	-106.5	1/3 MoO ₃	-31
(61) Br ₃	-153	1/2 TcO ₂	(-36)
(61) I ₃	-104.5	1/2 RuO ₂	(-45)
Sm Br ₃	-150	1/4 RuO ₄	(-45)
Sm I ₃	-100.5	RhO	(-45)
Sm Br ₂	-137	1/3 Rh ₂ O ₃	(-45)
Sm I ₂	-144	PdO	(-45)
Eu Br ₃	-136	1/2 PdO ₂	(-45)
Eu I ₃	-84.1	1/3 PdO ₃	(-45)
Eu Br ₂	-143	CdO	-22
Eu I ₂	-119.3	1/3 In ₂ O ₃	-40
		1/3 Sb ₂ O ₃	-45.3

TABLE IV - EQUILIBRIUM COMPOSITION IN PIN AT SHUTDOWN

Substance	Number of Molecules	
	Case 1	Case 2
CsBr	0.12x10 ¹⁸	0.12x10 ¹⁸
CsI	2.78x10 ¹⁸	2.78x10 ¹⁸
La ₂ O ₃	2.54x10 ¹⁸	2.54x10 ¹⁸
Y ₂ O ₃	2.04x10 ¹⁸	2.04x10 ¹⁸
Ce ₂ O ₃	6.53x10 ¹⁸	6.53x10 ¹⁸
Pr ₂ O ₃	1.63x10 ¹⁸	1.63x10 ¹⁸
Nd ₂ O ₃	4.24x10 ¹⁸	4.24x10 ¹⁸
(Gd) ₂ O ₃	.40x10 ¹⁸	.40x10 ¹⁸
Eu ₂ O ₃	.04x10 ¹⁸	.04x10 ¹⁸
Sm ₂ O ₃	.58x10 ¹⁸	.58x10 ¹⁸
Sr O	9.72x10 ¹⁸	9.72x10 ¹⁸
BaO	8.09x10 ¹⁸	8.09x10 ¹⁸
ZrO ₂	9.51x10 ¹⁸	9.51x10 ¹⁸
NbO ₂	8.67x10 ¹⁸	8.67x10 ¹⁸
Na ₂ O	39.19x10 ¹⁸	-----
Sb ₂ O ₃	-----	.04x10 ¹⁸
MoO ₂	-----	5.60x10 ¹⁸
In ₂ O ₃	-----	0.03x10 ¹⁸
TcO ₂	-----	2.94x10 ¹⁸
Rb ₂ O	-----	1.40x10 ¹⁸
Cs ₂ O	-----	1.47x10 ¹⁸
CdO	-----	.03x10 ¹⁸
Na	1.355x10 ²²	-----
Rb	2.79x10 ¹⁸	-----
Cs	2.94x10 ¹⁸	-----
Mo	5.60x10 ¹⁸	-----
Tc	2.94x10 ¹⁸	-----
Ru	9.38x10 ¹⁸	9.38x10 ¹⁸
Rh	.36x10 ¹⁸	.36x10 ¹⁸
Pd	.73x10 ¹⁸	.73x10 ¹⁸
Cd	.03x10 ¹⁸	-----
In	.05x10 ¹⁸	-----
Sb	.08x10 ¹⁸	-----
Te ₂	1.254x10 ¹⁸	1.254x10 ¹⁸
O ₂	-----	19.01x10 ¹⁸
Kr	1.25x10 ¹⁸	1.25x10 ¹⁸
Xe	17.07x10 ¹⁸	17.07x10 ¹⁸

NASA Technical Library



3 1176 01436 0037