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THE STATUS AND DEVELOPMENT POTENTIAL OF PLATE-TYPE FUELS FOR RESEARCH AND TEST REACTORS

by

D. Stahl

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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Materials Science Division

March 1979

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ABSTRACT

Recent U.S. Department of State action to restrict the shipment and use of highly enriched uranium for research and test reactors has renewed fuel development activity. The objective of these development activities is to increase the total uranium loading in the fuel meat so that enrichment reduction can be accomplished without significant performance penalties. This report characterizes the status and the potential for development of the currently utilized plate-type fuels for research and test reactors. The report also characterizes the newer high-density fuels which could be utilized in these reactors and indicates the impact of the utilization of both the new and current fuels on enrichment reduction.

I. INTRODUCTION

In the 1950s and 1960s, low-power research reactors were built worldwide which utilized Materials Test Reactor (MTR)-type fuel elements containing 20% enriched uranium. [MTR elements consist of 17-19 curved plates within an 81 x 76 mm (3.2 x 3 in.) box. Fuel length is 603 mm (23.75 in.).] However, the demand for higher specific power to provide higher neutron flux levels created a need for greater uranium-235 concentrations. Early difficulties in increasing uranium content led to the substitution of highly (90-93%) enriched uranium in place of the 20% enriched fuel previously utilized. The highly enriched material also yielded other benefits including longer core residence time, higher specific reactivity and somewhat lower fuel-cycle cost. Highly enriched material then became readily available and was used for high-power reactors as well as low-power reactors where 20% enriched material would have sufficed. The trend toward higher and higher specific power also led to the development of the dispersion-type fuels which utilized highly enriched uranium at a concentration of about 40 wt. %.

In the 1970s, however, concerns were raised about the proliferation resistance of fuels and fuel cycles. As a consequence, the U.S. Department of State has recently prohibited the foreign shipment of highly enriched material, except where prior contractual obligation or specific merit exists. This has impacted on the availability and utilization of highly enriched uranium for research and test reactor fuel. It has also stimulated development programs on fuels with higher uranium content which would allow the use of uranium of lower enrichment. Fuel development programs are under way in the U.S., Canada, France, the Federal Republic of Germany (FRG), and Argentina.

The major fuel development activity in the U.S. is part of the Reduced Enrichment Research & Test Reactors (RERTR) program which is managed by Argonne National Laboratory (ANL) for the Department of Energy. An objective of this program is to develop existing and new plate-type and, to some extent, rod-type research and test reactor fuels to their maximum uranium loading, which would make enrichment reduction possible. The enrichment-reduction potentials of the current and new fuels are shown in Table I. It is evident from the table that extensions of currently utilized fuels will permit enrichment reductions to 20% enriched fuel in low- and high-power research and test reactors, but that only the new fuels with higher uranium contents will permit such reductions for very high-power reactors. It is also evident from the table that enrichment reduction to 20% for low-power reactor fuels could be accomplished with existing technology.

The purpose of this report is to characterize the presently utilized fuels and the newer fuels as to uranium content and performance and to estimate the upper limits of uranium loading of these fuels for plate-type reactors.

II. URANIUM-ALUMINUM ALLOY FUEL

A. Introduction

Alloys of up to about 30 wt. % uranium in aluminum can be easily fabricated by melting and casting techniques to yield uniform uranium distribution, in the form of UAl_3 and UAl_4 precipitates in an essentially uranium-free aluminum matrix. By proper casting and heat treating, the amount of the more brittle UAl_4 phase can be reduced. However, with increasing uranium content, it becomes increasingly difficult to achieve uranium uniformity and the ductility of the alloy decreases as well.[1,2]

The ductility may be increased by suppressing the formation of the brittle UAl_4 phase in favor of the more ductile UAl_3 phase through the use of certain ternary additions. Silicon has been successfully used as a suppressant. Additions of 0.8 and 3.0 wt. % Si were found to completely suppress UAl_4 formation in 20 and 48 wt. % uranium-aluminum alloys, respectively.[3] Other ternary additions which suppress UAl_4 formation are tin, germanium, zirconium, and titanium.[2] The ternary alloy additions, however, complicate the standard reprocessing schemes.[4] This difficulty, coupled with the introduction of the aluminum dispersion fuels, has curtailed further exploration of these systems.

B. Irradiation Performance

Fuel-plate assemblies with a meat (the uranium-containing portion of the fuel) composed of a U-Al alloy with 18-22 wt. % U have been extensively tested in the MTR and the Experimental Test Reactor (ETR).[1] The average burnup of the fuel has exceeded 25% with peaks of 1.8 x 10^{21} fissions/cm^3. Experimental assemblies have been run to 75% burnup with no deleterious effect.[5] Swelling appears to be linearly related to irradiation exposure, with a value of 6.38% $\Delta V/V$ per 10^{21} fissions/cm^3. Unmodified U-Al fuels have performed well at concentrations of 24 and 30 wt. % uranium. For example, the 50-MW General Electric Test Reactor has been utilizing U-Al fuel meat containing 30.5 wt. % uranium.[6] The core average fuel-burnup limit is 50%, with a maximum of 2.0 x 10^{21} fissions/cm^3 (Ref. 7).

Experimental irradiations were also performed with U-Al alloys containing ternary additions. Small plates containing 48 wt. % U and 3 wt. % Si were successfully irradiated to burnups of 83% (7.50 x 10^{20} fissions/cm^3).[8] Standard plates containing up to 50 wt. % uranium with additions of silicon and tin were irradiated to large burnups (85% or 1.0 x 10^{21} fissions/cm^3) under MTR conditions without dimensional distortion.[9]

C. Development Potential

As noted above, U-Al alloy plates have been produced which contained up to 50 wt. % uranium plus small ternary additions to suppress the formation of the brittle UAl_4 phase. Recent development activity at NUKEM in the FRG and at Savannah River Laboratory (SRL) has been directed toward increasing the uranium concentration to 40 and to perhaps 45 wt. % without ternary additions by proper casting and hot-working techniques.[10,11] Because of its limited potential as indicated in Table I, U-Al alloy fuel was not included in the RERTR program.

III. ALUMINUM DISPERSION FUEL

A. Introduction

As noted in Section II, the difficulty in achieving uniform fuel distribution in U-Al alloy fuels led to development of configurations in which fuel particles were dispersed in a metallic matrix. The dispersion approach provides a distinct separation of the fissile fuel particle and the metal matrix so that their desirable properties are retained. The metal matrix material must possess the following properties: high formability, high thermal conductivity, high corrosion and radiation resistance, and low thermal-neutron cross section. Among others, alloys of Al, Be, Mg, Nb and Zr, and stainless steel have been employed as matrix materials.[12]

The fuel particles must possess high radiation and corrosion resistance and high uranium density and must be compatible with the matrix material. The particles should also possess some hot formability when present in high concentrations. The particle size should be large enough (>100 μm) so that fission-product damage is concentrated in the particle. Particle spacing should be large enough so that damage zones, which surround the particles, do not link up. Porosity, which is usually associated with the particles, also provides some swelling accommodation. A number of fissile compounds are given in Table II. The theoretical uranium content per unit volume of dispersed phase, its relation to the density of uranium metal, and the melting points of the compounds, which provide a relative indication of stability, have also been included in the table.

Considerable work was performed with UO_2 dispersed in stainless steel or aluminum because of its stability and high uranium concentration. The 1955 Geneva Conference Reactor (GCR) used an UO_2-Al fuel. Great difficulty was experienced during manufacture of the GCR fuel elements because of abnormal dimensional growth during elevated-temperature fabrication procedures.[2] The growth of the fuel plates was traced to volume changes accompanying the reaction of UO_2 and aluminum. The reaction reached 90 to 100% completion in 10 h at 600°C in 52 wt. % UO_2 fuel plates.[14] Fortunately, experimenters at ANL discovered that U_3O_8 was more stable than UO_2 with aluminum and recomended its use.[15] Later experiments at Oak Ridge National Laboratory (ORNL) confirmed that the reaction of U_3O_8 with aluminum was much slower at the temperatures needed for fabrication.[16] It was shown that 3000 h were required for complete reaction at 600°C.[17] Fuel plates could, therefore, be fabricated from U_3O_8 and aluminum with relatively little difficulty arising from dimensional changes due to a reaction between the components. This process, however, leads to a fuel element which retains the potential for an exothermic (thermite*) reaction between the components.

The reactions of U_3Si, U_3Si_2, UC, and UC_2 with aluminum were also evaluated, in 620°C tests lasting up to 48 h. No reaction occurred with UC_2; however, U_3Si, U_3Si_2, and UC were found to react with aluminum, but at a slower rate than UO_2.[13] Studies on aluminides[18] showed that UAl_2 reacts rapidly, and UAl_3 reacts slowly, with aluminum to form the stable compound UAl_4, which undergoes no further reaction with Al. A mixture of aluminides, designated UAl_x but compositionally close to UAl_3, will therefore react with aluminum in the reactor, but at a slow rate. The stable UAl_4 is not used directly, since it is very brittle and not easily formable with present plate-fuel technology.

B. UAl_x-Al Dispersions

1. Irradiation Performance. Dispersions of UAl_x in aluminum have performed satisfactorily in the ETR and Advanced Test Reactor at 22 and 41 wt. % uranium contents, respectively.[1,19] In these tests, prototypic elements are exposed to average burnup levels of 30% (0.8×10^{21} fissions/cm^3) and peak levels of ~90% (~2.3×10^{21} fissions/cm^3).[19,20] Experimental plates have performed successfully after irradiation in the MTR, ETR, High Flux Isotope Reactor (HFIR), and FR2 (Karlsruhe) under a wide variety of conditions with the maximum burnup closely approaching total depletion of the uranium-235 (~2.8×10^{21} fissions/cm^3).[1,9,19,21] Swelling rates were generally about one half that of U-Al alloy at about 3% $\Delta V/V$ per 10^{21} fissions/cm^3 (Ref. 1). However, for the MTR experiments, which utilized slightly lower temperatures and water pressures, swelling rates were found to approach

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*A thermite reaction is one that involves the ignition of aluminum powder with an oxide to rapidly produce a great deal of heat.

that of the U-Al alloy. It is postulated that under these conditions, the process of in-pile densification, which reduces swelling, is impeded. For most irradiations, swelling was roughly linear with burnup to the maximum value tested. The exception was the FR2 (Karlsruhe) data, which showed an increase in swelling rate to about 12% $\Delta V/V$ per 10^{21} fissions/cm^3 after a burnup of 40% ($\sim$1.5 x 10^{21} fissions/cm^3).[21]

2. Development Potential. Potential exists for increasing the uranium loading of UAl_x-Al dispersion fuels. In the past, experimental loadings of only 45 wt. % uranium were fabricated.[20] However, it is anticipated that 50 wt. % uranium loadings, equivalent to about 50 vol % UAl_x, can be easily fabricated as part of a development program. This limit may possibly be extended to 55-60 wt. % uranium; however, careful control of fabrication parameters will be necessary to prevent enlargement of the ends of the fuel-meat cores (dogboning) and to maintain uniform meat thickness, width, and uranium distribution.

C. U_3O_8-Al Dispersions

1. Irradiation Performance. The HFIR at ORNL utilizes aluminum-clad fuel plates with fuel meats consisting of a dispersion of uranium oxide (U_3O_8) in aluminum. The maximum fuel loading in the meat of these fuel plates is $\sim$40 wt. % uranium ($\sim$47 wt. % U_3O_8); the meat thickness varies from about 250 to 760 μm (about 0.010 to 0.030 in.). The average burnup of the fissile atoms is 31% (0.9 x 10^{21} fissions/cm^3), with peaks of $\sim$65% ($\sim$1.9 x 10^{21} fissions/cm^3).[22] Experimental plates have experienced burnups of $\sim$75% ($\sim$2.1 x 10^{21} fissions/cm^3) without failure or gross dimensional change. As part of the U_3O_8 dispersion-fuel development program for HFIR and the Puerto Rico Nuclear Center Reactor, test samples were made, irradiated, and evaluated at ORNL.[8,23] The fuel loading was 55 wt. % uranium (65 wt. % U_3O_8) in the meat, which was $\sim$610 μm ($\sim$0.024 in.) in thickness. Short plate-type specimens were found to be dimensionally stable after a burnup of 7.7 x 10^{20} fissions/cm^3 (>90% U^{235} depletion). A full-size MTR-type element was found to be dimensionally stable after a burnup of $\sim$4 x 10^{20} fissions/cm^3 ($\sim$50% U^{235} depletion).[8] Thus, 55 wt. % uranium represents the present maximum fuel loading of U_3O_8 dispersion-type aluminum plate fuel.

Related recent development activity at SRL on U_3O_8-Al dispersion-fueled tubes has indicated that 48 wt. % uranium (58 wt. % U_3O_8) loadings have performed well after 141 irradiation days.[11]

2. Development Potential. The potential for development of higher uranium loadings for U_3O_8 dispersion-type aluminum plate fuel does exist. As part of the U_3O_8 dispersion-fuel development program at ORNL for HFIR and at SRL, test samples were made that contained up to 100 wt. % U_3O_8 in the meat.[11,22] As expected, for loadings greater than about 60 wt. % uranium (about 70 wt. % U_3O_8), depending on void content, the continuous phase of the dispersions changed from aluminum to U_3O_8. However, all cold-pressed U_3O_8-Al dispersions exhibited sufficient green strength after compacting to permit some handling. All of the plate fuel appeared to roll-clad satisfactorily, as was evident from the absence of internal cracks. However, for the tubular fuel elements, high extrusion ratios led to overheating which

produced a thermite reaction between U_3O_8 and aluminum. The meat thickness and extrusion ratio are being reduced to eliminate this problem.[11]

In addition to the potential for a thermite reaction, technical issues which must be addressed to increase the uranium loading beyond 55 wt. % include: adequate control of cladding thickness, dogboning, a continuous aluminum phase in the meat, uranium homogeneity, and fuel behavior under long-term irradiation (i.e., unacceptable swelling and shrinking). Of perhaps critical importance is the maintenance of a continuous aluminum phase that will facilitate heat flow out of the meat. Based on simple geometric considerations, a uranium loading of about 60 vol %, which is about 70 wt. % (about 80 wt. % U_3O_8), appears to be the theoretical limit for maintenance of the continuous aluminum phase in a U_3O_8-Al dispersion with U_3O_8 spheres (see Fig. 1). Slightly higher loadings may be possible with control of particle shape and size distribution.

3. Thermite-reaction Concerns. As noted above, the potential for a thermite reaction must be evaluated as part of the safety analysis. Both off-normal and transient effects must be considered. Fortunately, some data are available under both of these conditions.

Fleming and Johnson heated cold-pressed pellets of mixtures of U_3O_8 and aluminum at 33°C/min.[24] At a composition range of 65-75 wt. % U_3O_8, violent exothermic reactions occurred between 900 and 1100°C with a peak energy release of 1.2 MW-s/kg fuel. This reaction was also studied by workers at ANL using various compositions, pretreatments, environments, and particle size distributions.[25,26] However, a slow heat-up rate (25°C/min) was used in these tests, and the violent reactions reported by Fleming and Johnson were not duplicated; only temperature excursions to a maximum of 1300°C were observed. Similar results were obtained for specimens inserted into a furnace at 1250°C. In contrast, for specimens which were sintered at 600°C after pressing, violent reactions were observed with compositions of 75 and 85 wt. % U_3O_8. Thus, the reaction rate can be strongly influenced by the preconditioning of the compacts.

Large-scale tests with full-size elements were performed by SRL for $\leq$57 wt. % U_3O_8-Al cores.[11] It was found that the maximum fuel temperature was 1370°C, only 370°C above the furnace temperature. Thus the heat of reaction was not an important energy source for these fuel elements.

Transient-reactor (TREAT) experiments on U_3O_8-Al HFIR fuel plates were performed by ANL.[27] The tests simulated nuclear excursion accidents in a water-cooled reactor. Specimens of U-Al fuel were included for comparison. The major conclusions were that the U_3O_8-Al specimens retained their shape up to 1400°C (a significantly higher temperature than that observed for U-Al fuel) and that the exothermic reaction was not an important energy source.

IV. POTENTIAL OF NEW FUEL TYPES

A. Introduction

Although the development of increased uranium loadings in current research-reactor fuel types, if successful, will allow many research reactors to utilize lower-enrichment uranium in the fuel, there will still be several research and test reactors which cannot be converted to the use of low-enrichment uranium fuel. These are the reactors that are already operating with highly enriched fuel at the highest uranium loadings currently qualified. Also, these are in general the more advanced (higher power-density) and more highly utilized research reactors which are designed for extra-long reactivity-core life.

Thus, for new research reactors, and for replacement fuels in the higher power-density research reactors, one option is to develop a different research-reactor fuel specifically designed for high uranium loadings. This option could allow the research reactor to convert to low-enrichment uranium fuel with a minimum impact on the core design or operation as compared with other options such as complete core modification or decommissioning. Development of this fuel option also provides a backup if the planned developments based on current fuel do not work out.

Several advanced fuel types have been and are being studied. The properties of some of these fuels are shown in Table III (taken from Ref. 28). Not all of them are compatible with a water-cooled research reactor environment. The candidate fuel materials possess much higher uranium densities than the currently utilized dispersion fuels, which possess a uranium loading of only 1.7 g/cm^3. The corrosion resistance, irradiation performance, and development potential of some of these candidates fuels is given in the following sections. These materials could be utilized alone or even as a dispersant since their uranium densities, as listed in Table II, are much greater than that of UAl_x or U_3O_8.

B. Corrosion Resistance

Metallic uranium alloys and compounds were extensively investigated in the U.S., principally at ANL, Battelle, Knolls Atomic Power Laboratory, and Westinghouse, in the late 1940s and the 1950s as fuels for light-water reactors. Several thorough reviews of the subject have been published.[29,30] These investigations focused on alloying to improve the poor corrosion resistance of uranium and to ameliorate the anisotropic growth problem inherent in the unalloyed material. In these early efforts, almost every metallic element was alloyed with uranium in an attempt to produce a fuel which would be resistant to high-temperature water. The alloys with good to moderate corrosion resistance fall structurally into two groups: metastable gamma alloys and distorted alpha alloys. The former are produced by quenching from 800-1000°C. Molybdenum and niobium compositions in the range of 7 to 15 wt. % and 6 to 20 wt. %, respectively, have been studied. Corrosion resistance was dependent on heat-treatment conditions, with molybdenum superior to niobium. The alpha-alloy group consisted mainly of alloys with zirconium (up to 10 wt. %) or niobium (up to 6 wt. %) and ternary alloys containing both these elements. However, when these alloys, in wrought form, were heat treated or aged for maximum corrosion resistance, they exhibited poor dimensional stability under irradiation.

It is also possible to provide corrosion resistance by the formation of intermetallic compounds. U_3Si has been studied because it possesses reasonable ductility and a moderately high decomposition point. With proper heat treatment, this compound is nearly as corrosion resistant as the best uranium-molybdenum alloy.[31] More recent work on this alloy system by the Atomic Energy of Canada Limited (AECL) has shown that the addition of 1-1/2% aluminum substantially increases the corrosion resistance of the fuel.[32]

Another approach to the problem of providing corrosion resistance was developed at SRL.[33] Corrosion resistance can be provided by a diffusion bond between an intermediate layer and both the fuel and cladding. The fuel was bonded to the aluminum cladding by utilizing an Al-Si dip-canning process on early fuel elements and an electroplated nickel layer on later fuel elements. Both methods were followed by a hot die-sizing process to form the diffusion bond. This technique substantially reduces the exposure of the fuel to hot water in the event the cladding is penetrated. A diffusion bond between a uranium-zirconium alloy and the Zircaloy-2 cladding, created by coextrusion, was also found to provide excellent corrosion resistance.[30]

Of the ceramic compounds listed in Table III, only UO_2 possesses chemical inertness and resistance to high-temperature water.

C. Irradiation Performance

Uranium-molybdenum alloys generally exhibit satisfactory irradiation behavior. In early work, the U-10 wt. % Mo alloy was shown to be stable to at least 5 at. % burnup except for abnormal swelling of some samples operated at 335-390°C, possibly due to plastic flow effects near the alpha-beta transition temperature.[30] The corrosion resistance of these materials was relatively good, since the gamma phase was stabilized by the irradiation. Similar behavior was found in more recent work at SRL on alloys containing lower Mo concentrations and 0.1 wt. % silicon.[34] Swelling rates of 2-4%/at. % burnup were found after irradiations of 9000 and 12000 MWd/t up to 600°C maximum fuel temperature, again with the exception of abnormal swelling at $\sim$400°C. The irradiation behavior of uranium-niobium alloys was found to be similar to that of uranium-molybdenum.[30] However, additions of 4-6 wt. % of zirconium to U-10 wt. % Nb raised the abnormal swelling temperature to above 450°C.

The irradiation behavior of uranium-zirconium and uranium-zirconium-niobium alloys generally has been less favorable.[30,34] However, additions of carbon were found to greatly modify swelling behavior in low-burnup tests.[30] Good irradiation performance has been obtained with a U-5 wt. % Fs alloy (where the fissium is made up of the mixture of fission products expected in recycled fuel) to burnups exceeding 10 at. % in the EBR-II.[35] However, these cast and heat-treated rods are bonded to the cladding with sodium; this provides volume for $\sim$30% swelling up to about 4 at. % burnup, after which the cladding can easily restrain the spongy fuel.

Ongoing experiments at SRL with very small additions of Cr, Al, Si, Fe, and Mo (either separately or in combination) to uranium have produced interesting results.[34] Most of these alloys were quenched from 725°C, then irradiated to 9000 and 13000 MWd/t at various temperatures ranging from 200 to 600°C. Swelling rates of 3-4%/at. % burnup were found for alloys containing 800-1150 ppm Al + Si at temperatures below the swelling threshold of 350°C. Alloys containing the above Al + Si concentrations were further improved by the addition of 250-350 ppm Fe, 200 ppm Cr, and 1000 ppm Mo. An alloy containing 800 ppm Al, 350 ppm Si, 350 ppm Fe, and 1000 ppm Mo was found to be stable to approximately 425°C.

The early irradiations of U_3Si fuel material produced mixed results.[30] Extruded samples irradiated to 0.17 at. % burnup by Westinghouse-Atomic Power Division were found to have increased in both length and diameter. Several samples were bowed and all exhibited cracks, blisters and bands. However, in tests performed by ANL, both cast and extruded samples exhibited good irradiation stability. In recent experiments at AECL, U_3Si and aluminum-modified U_3Si showed excellent irradiation performance.[36,37] Diameter increases for Zircaloy-clad U_3Si in the form of annular fuel rods were less than 1% after 10,000 MWd/t and about 1.2% after 19,000 MWd/t at a maximum temperature of about 500-600°C. With the addition of 1.5 and 2.4 wt. % aluminum, diameter increases were reduced slightly and, as noted earlier, the corrosion resistance of the alloy fuel was improved.

Despite its poor thermal conductivity, UO_2 performs well since it can be operated at steady-state central temperatures approaching its melting point.[38] UO_2 was used as the fuel material for this country's first prototype power reactors - the Shippingport pressurized water reactor (PWR) and the Vallecitos boiling water reactor (BWR) - which were built in the late 1950s. However, metal fuels were utilized in the early experimental reactors (the Ship Thermal Reactor developed by ANL, Westinghouse, and the Naval Reactors Division - USAEC; and the Experimental Boiling Water Reactor developed by ANL).[39] The shift to oxide fuel occurred because the behavior of metal alloy fuels was found to be inadequate.[38]

The reference fuel for both the PWR and the BWR is still UO_2 in the form of pellets of ∿93% theoretical density. Satisfactory performance has been obtained at burnups of over 40,000 MWd/t.[40,41] This led to the consideration of oxide fuel for liquid metal-cooled fast breeder reactors and to the development of an information data base on oxide fuels for this application.[42] A similar document is available for light water reactor oxide-fuel behavior.[43] UO_2 fuel rods which contain ∿5% enriched uranium have also been used in the PULSTAR reactors.

More recently, the French have utilized UO_2 wafers (caramels) in a compartmentalized Zircaloy cladding arrangement for plate-type reactors.[44] The plates have performed well to a maximum burnup of 30,000 MWd/t. The French design is similar in concept to that employed in the second core of Shippingport, which utilized UO_2-ZrO_2 as fuel and UO_2 in the blanket. As part of the development of the UO_2 plates for the blanket, Westinghouse performed high-temperature irradiations to burnups as high as 140,000 MWd/t

(4×10^{21} fission/cm^3).[45] However, plate failures occurred at a burnup as low as 20,000 MWd/t ($\sim 0.6 \times 10^{21}$ fission/cm^3) as a result of fission-gas release.

D. Development Potential

The data presented above suggest that U_3Si, probably modified with aluminum, has the greatest potential for providing the necessary uranium density and irradiation performance levels. Uranium-molybdenum alloys appear to be the second choice, with UO_2 third. UO_2 would have to be fabricated in the form of very thin members because of its poor thermal conductivity. Interestingly, the French have chosen U-Mo alloys as a backup for their UO_2 caramel fuel.[45]

It is likely that U_3Si can be utilized as a dispersant in aluminum, provided that the positive volume change upon reaction to form UAl_3 can be reduced or slowed by alloy addition. As shown in Fig. 1, U_3Si provides a much higher uranium weight fraction than does an equal volume of U_3O_8 or UAl_x. For example, an expected volume-fraction range of 0.5-0.7 would yield uranium weight fractions of about 0.8-0.9, as indicated in Table I. Thus an alloy addition would not greatly reduce the uranium loading. The use of an aluminum dispersion would mean that present dispersion fuel-meat technology could be utilized, rather than some advanced technology which might require cladding with Zircaloy or some other material which is stiffer than aluminum. However, the greater density difference between U_3Si and aluminum would necessitate care in the blending of powders to maintain uniform fuel-particle distribution.

Another potential problem with U_3Si concerns reprocessing. Silicon in some weight-fraction ranges has been shown to interfere with phase separation in the extraction column.[4] A study is under way at ANL to determine whether the aqueous process can be successfully modified or whether a suitable nonaqueous method can be utilized to accomplish the reprocessing of plate-type fuels containing U_3Si.

V. SUMMARY AND CONCLUSIONS

The current emphasis on the conversion of plate-type research and test reactor fuel from fully enriched uranium to less than 20% enrichment or to some intermediate enrichment, perhaps 45%, has stimulated the development of more highly uranium-loaded fuels. The development effort has been logically divided into two areas: the extension of currently utilized fuels to their maximum uranium loading, and the development of new high-density fuels.

The uranium-density and enrichment-reduction potential of current fuels was summarized in Table I. It is anticipated that U-Al alloy fuel which contains 40-45 wt. % uranium can be developed; this loading will permit reductions to less than 20% enrichment for low-power reactors, but will permit reductions to only intermediate enrichment for other reactors.

Aluminide and U_3O_8 dispersion fuels could reach uranium densities as high as 60 and 70 wt. %, respectively, which would make possible the use of less than 20%-enriched uranium in high-power reactors. This assumes that dispersions containing 60 vol % of the dispersed fuel phase are possible. For both of these materials, fuel fabricators and developers have stated[47] that ∿55 vol % will be the maximum loading possible as a result of development efforts. If only 50 vol % dispersions are possible, which is a more likely outcome, then the aluminide dispersion might not permit the conversion of high-power reactors without some modification of the fuel-meat thickness and element geometry. This may be true for U_3O_8 dispersions as well. Little or no high-burnup experience exists for these extensions of currently utilized fuels; in contrast, considerable positive experience exists for the lower uranium loadings. This suggests that if uniform dispersions of highly loaded fuel can be successfully fabricated, there is a strong likelihood that they will exhibit satisfactory irradiation behavior.

For the new high-density fuels, a nearly opposite situation exists. Very little low-temperature, high-burnup irradiation experience has been obtained. For U_3Si and U-Mo alloys, the maximum burnup was less than 20,000 MWd/t, while for UO_2 plates, the maximum burnup was less than 30,000 MWd/t. As far as uranium loading is concerned, these materials are more than adequate, and dispersions may be utilized to take advantage of existing dispersion-fuel technology. However, the compatibility of the fuel compound with the matrix must be assessed. U_3Si, for example, must be stabilized so that its slow in-pile reaction with aluminum does not yield large volume increases. Such dispersions with 50 or more volume percent of dispersed fuel would allow conversion of even the very high-power reactors.

Thus it is likely that the fuel development effort now under way will provide the technical means of reducing enrichment in research and test reactors. Extensions of currently utilized fuels should allow the conversion of low- and high-power reactors and the new high-density fuels should allow the conversion of very high-power reactors.

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TABLE I. Uranium Density and Enrichment Reduction Potential of Candidate Fuels for Research and Test Reactors.

Fuel Type	Near-term Uranium Loading, wt. % (g/cm^3)	Long-term Uranium Loading, wt. % (g/cm^3)	Near-Term/Long-Term Enrichment-reduction Potential, %		
			Low-power Reactors	High-power Reactors	Very High-power Reactors
U-Al Alloy	30 (1.1)	40-45 (1.6-1.9)	20/20	70/45	93/45
UAl_x-Al	42 (1.7)	50-60 (2.2-3.0)	20/20	45/20	93/45
U_3O_8-Al	42 (1.7)	60-70 (3.1-4.2)	20/20	45/20	93/45
UO_2 Caramel Fuel		85 (9.6)	20	20	20[a]
U_3Si, U_3Si-Al		80-90 (7.0-13.4)	20	20	20
U-10 Mo		90 (14.8)	20	20	20

[a] For very high-power reactors, UO_2 would have to be fabricated in very thin sections to provide proper heat removal.

TABLE II. Fissile Dispersants[a]

Compound	Melting Point, °C	Density, g/cm^3	Uranium Loading, g/cm^3	Relative Uranium Loading
U	1133	19.1	19.1	1.00
UAl_2	1590	8.1	6.6	0.35
UAl_3	1350	6.7	5.0	0.26
UAl_4	730	6.0	4.1	0.21
UBe_{13}	2000	4.37	2.9	0.15
UC	2500	13.6	13.0	0.68
UC_2	∿2500	11.7	10.6	0.56
UO_2	2875	10.96	9.7	0.51
U_3O_8	–[b]	8.40	7.1	0.37
UN	2630	14.3	13.5	0.71
U_6Fe	815	17.7	17.0	0.89
UFe_2	1235	13.2	9.0	0.47
U_3Si	930[b]	15.6	15.0	0.79
U_3Si_2	1665	12.2	11.3	0.59

[a] From Ref. 13.

[b] Decomposes.

TABLE III. Properties of Fuels

	U	U-10% Mo	U_3Si	U-Fs[c]	UO_2	UC	UC_2
Melting point, °K	1405	1423	1203	1275	3138	2780 ± 25	2773
Density, g/cm^3	19.12	17.12	15.58	18	10.96	13.61	12.86
Heavy metal density, g/cm^3	19.12	16.38	14.91	17.5	9.65	12.97	11.68
Crystal structure	[a]	[b]	bct	bcc (>1000 K)	fcc (CaF_2)	fcc (NaCl)	fcc (CaF_2)
Thermal conductivity. W/cm-°K	0.35 (670 K)	0.29 (870 K)	0.2 (to 1170 K)	0.33 (820 K)	0.03 (1270 K)	0.216 (to 1270 K)	0.35 (to 1270 K)
Thermal expansion. 10^{-6}/°K	19 (to 920 K)	12.3 (to 670 K)	16 (to 1070 K)	17 (to 820 K)	10.1 (to 1270 K)	11.6 (to 1470 K)	18.1 (1970 K)
Electrical resistivity, ohm-cm	35×10^{-6} (298°K)		75×10^{-6} (to 1070 K)		1×10^3	40.3×10^{-6} (298 K)	
Specific heat, cal/g-°K	0.026 (to 773 K)	0.035 (to 773 K)	0.043 (to 773 K)		0.065 (700 K)	0.048 (298 K)	0.12 (298 K)
Heat of fusion, cal/mole	4760				16,000	11,700	
Vapor pressure, atm	5×10^{-6} (2300 K)	5×10^{-6} (2300 K)			8.5×10^{-8} (2000 K)	1.7×10^{-10} (2300 K)	2.5×10^{-11} (2300 K)
Debye temperature, °K	200 K				<600 K, 870 K.	--	--
Free energy of formation, kcal/mole	-				-218 (1000 K)	-23.4 (298 K)	
Heat of formation, kcal/mole	-				-260 (to 1500 K)	-23.63 (298 K)	-23 (298 K)
Entropy, cal/mole-°K	-				18.6 (298 K)	14.15 (298 K)	16.2 (298 K)
Poisson ratio	0.21	0.35			0.3	0.284	
Modulus of rupture, MPa					80		
Modulus of elasticity, MPa	1.7×10^5	10^5		6×10^4	1.8×10^5	2×10^5	
Shear modulus, MPa	0.85×10^5	3×10^4			0.75×10^5	0.873×10^5	
Tensile strength, MPa	400	300	600	270	35		
Compressive strength, MPa			2000		1000	350	
Thermal neutron fission cross section, barns	4.18 (natural)	4.18 (natural)	0.159 (natural)		0.102 (natural)	0.137 (natural)	0.112 (natural)
Thermal neutron absorption cross section, barns	7.68 (natural)	6.68 (natural)	0.293 (natural)		0.107 (natural)	0.252 (natural)	0.207 (natural)
Eta (η)[d]	1.34	1.34	1.34	1.34	1.34	1.34	1.34

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[a] Orthorhombic (<936°K), tetragonal (936-1043°K), body-centered cubic (>1043°K).

[b] Orthorhombic plus tetragonal (<838°K), body-centered cubic (>838°K).

[c] U containing 5% fissium (0.22% Zr + 2.5% Mo + 1.5% Ru + 0.3% Rh + 0.5% Pd). U-5% fissium is bcc above 1000°K, bcc + monoclinic U_2Ru between 825°K and 1000°K, and bcc + U_2Ru + tetragonal below 825°K.

[d] Number of fission neutrons released per neutron absorbed.

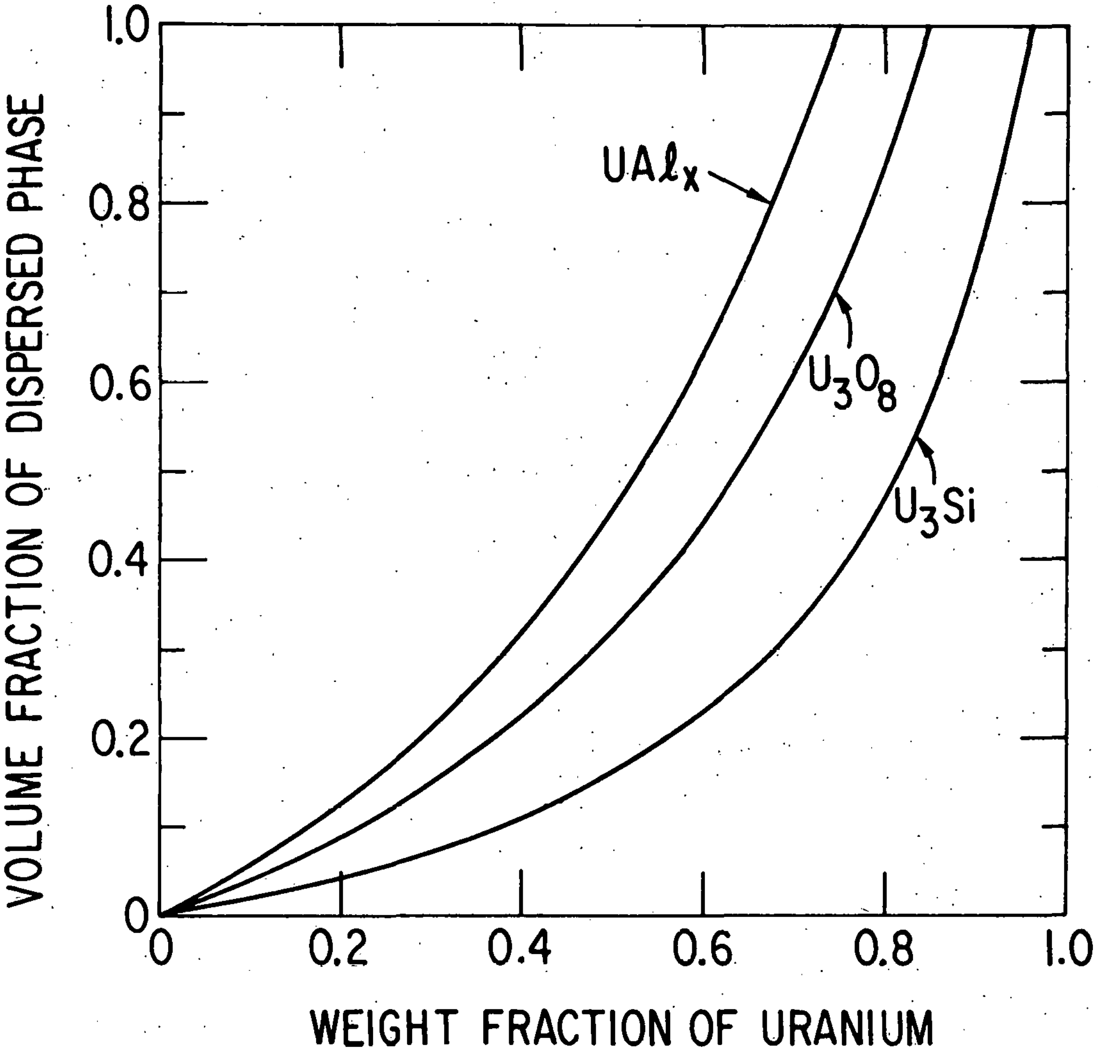

Fig. 1. Volume-Weight Relationships for Uranium Dispersion Fuels.

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