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FUELS FOR RESEARCH AND TEST REACTORS,
STATUS REVIEW: July 1982

by

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ABSTRACT

A thorough review is provided on nuclear fuels for steady-state thermal research and test reactors. The review was conducted to provide a documented data base in support of recent advances in research and test reactor fuel development, manufacture, and demonstration in response to current U.S. policy on availability of enriched uranium. The review covers current fabrication practice, fabrication development efforts, irradiation performance, and properties affecting fuel utilization, including thermal conductivity, specific heat, density, thermal expansion, corrosion, phase stability, mechanical properties, and fission-product release. The emphasis is on U.S. activities, but major work in Europe and elsewhere is included. The standard fuel types discussed are the U-Al alloy and the UAl_x -Al and U_3O_8 -Al dispersion plate fuels, and the U-Al alloy, $UZrH_x$, and UO_2 rod fuels. Among new fuels, those given major emphasis include U_3Si -Al dispersion and UO_2 caramel plate fuels.

I. INTRODUCTION

Research and test reactors are operated worldwide to provide one or more of the following capabilities: training of reactor operators, materials testing, neutronics studies, radioisotope production, activation analysis, and neutron radiography. Little research on fuels for research and test reactors had been conducted in the ten years prior to 1977, when renewed concerns about the proliferation resistance of fuels and fuel cycles were expressed by the U.S. Government.

Highly (90-93%)-enriched uranium (HEU) fuel is currently used in a majority of research and test reactors in the world. Although a number of relatively low-power research reactors (many of which were supplied under the U.S. Atoms for Peace program) initially used low (<20%)-enriched uranium (LEU) fuel, the more advanced and higher power research and test reactors utilized HEU fuel in order to avoid the need to increase the uranium concentration in the fuel in response to the demand for higher specific powers and neutron fluxes. HEU also yielded other benefits, including longer core residence time, higher specific reactivity, and somewhat lower fuel cycle cost. Since HEU was readily obtainable, even reactors which could have used LEU fuel gradually converted to the use of HEU fuel.

In 1977, President Carter raised the issue of proliferation resistance of nuclear reactor fuels and fuel cycles. As a consequence of this concern, the U.S. has established the Reduced Enrichment Research and Test Reactor (RERTR) Program to provide the technical means for the effective use of reduced enrichment fuels in research and test reactors. The U.S. has indicated that it plans to severely restrict the export of HEU as these technical means become available. These proliferation concerns and the U.S. tightening of HEU export controls have stimulated a number of development programs for fuels with higher uranium content which would allow the use of uranium of lower enrichment.

Reactor operators have begun examining the effect of LEU fuels on reactor performance, and fuel developers and manufacturers have begun evaluating the fabricability and performance of fuels of higher uranium density containing LEU. Fuel development programs are now under way in the U.S., Canada, France, the Federal Republic of Germany (FRG), the United Kingdom (UK), and Argentina. The major fuel development activity in the U.S. is within the RERTR Program, which is managed by Argonne National Laboratory (ANL) for the Department of Energy. An important 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 without significant penalties in safety, performance or fuel cycle cost.

The purposes of this review are (1) to describe the status of the fabrication development work in progress in the U.S. as part of the RERTR Program; (2) to summarize the data base from which these programs were initiated; (3) to show the relationships between the U.S. effort and work going on in other countries; and (4) to discuss the pertinent available data on the properties and irradiation behavior of these fuels and fuel types.

II. FABRICABILITY OF COMMERCIAL FUELS

2.1. Plate fuels

A plate-type fuel element consists of 12-23 flat or curved plates within a total cross-sectional area of 76 x 76 mm (3 x 3 in.). A typical curved-plate element is shown schematically in Fig. 1. Each plate contains a fuel core (an alloy or aluminum dispersion), clad with an aluminum alloy with a thickness of 0.25-0.76 mm (10-30 mils). The active core length is usually 610 mm (24 in.). The enrichment-reduction potentials of both current and plate-type fuels are given in Table 1. As this section will show, enrichment of fuels for low-power reactors can be reduced to 20% or less with existing technology; in the case of high-power reactors, this enrichment reduction will be achievable with extensions of currently utilized fuels. For very high-power reactors, such enrichment reductions will be possible only with new fuels, which have higher uranium contents; these fuels are discussed in Sec. 4.

2.1.1. Uranium-aluminum alloys

Alloys of up to about 30 wt% uranium in aluminum can easily be 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, such as silicon. 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 that suppress UAl_4 formation are tin, germanium, zirconium, and titanium.[2] The ternary

Table 1. Uranium Density and Potential Enrichment Level of Candidate Plate-Type Fuels for Research and Test Reactors

Fuel Type	Uranium Loading, g/cm ³	RERTR Goal	Current/RERTR Goal Potential Enrichment		
			Low-Power Reactors	High-Power Reactors	Very High-Power Reactors
U-Al alloy	1.1	~1.6	<20	70/45	93
UAl _x -Al	1.7	2.6-2.8	<20	45/20	93/45
U ₃ O ₈ -Al	1.7	3.3-3.8	<20	45/20	93/45
UO ₂ caramel	9.1 ^a	-	<20	<20	<20 ^b
U ₃ Si-Al	-	7.0-8.0	<20	93/20	93/20
U ₃ Si (bulk)	-	~11	<20	93/20	93/20

^a8.7 if the Zircaloy spacers are smeared within the fuel core. The density of the UO₂ is 10.3 g/cm³.

^bFor very high-power reactors, UO₂ would have to be fabricated in very thin sections to provide proper heat removal.

alloy additions complicate the standard reprocessing schemes.[4] However, early alloy fuel elements for the Greek Research Reactor-1, which contained 3.0 wt% Si, were successfully reprocessed by UK workers using their standard acid-deficient reprocessing scheme.[5]

Some development work is under way in Denmark on medium (~45%)-enrichment uranium (MEU) U-Al fuels containing 30-40 wt% uranium,[6] for the 10-MW DIDO type reactor. Again, additives have been used to suppress UAl_4 formation and increase ductility. The additives also produce a smaller grain size and a more uniform grain size distribution. It is claimed that these additions do not interfere with standard reprocessing schemes, but this will only be confirmed when the planned irradiation testing is completed. Some development work was performed at NUKEM (FRG) to increase the uranium concentration to 40 wt% and beyond.[7] However, this work was discontinued because it was felt that the dispersion fuels had greater potential and could be produced more economically.

2.1.2. Uranium aluminide-aluminum dispersions

Uranium aluminide-aluminum dispersion fuels are being used in many moderate- and high-power reactors around the world. This fuel was initially developed for the Advanced Test Reactor (ATR), starting in 1962, and was subsequently used in the Materials Test Reactor (MTR) and Experimental Test Reactor (ETR) in the U.S. and in many European reactors including the Belgian Reactor-2 (BR-2) and the High Flux Reactor (HFR)-Grenoble. Aluminide fuel is also used in many U.S. university reactors. The highest loadings are used in the BR-2 (~1.3 g U/cm³) and the ATR (~1.6 g U/cm³). The aluminide is fabricated from a melting and casting operation and as a result contains mostly UAl_3 , some UAl_4 , and a trace of UAl_2 . A typical composition for the ATR contains 63% UAl_3 , 31% UAl_4 and 6% UAl_2 . [8] Such a composition is referred to as UAl_x . The UAl_2 is fairly reactive and will react with excess aluminum to form UAl_3 or UAl_4 . Thus the finished ATR fuel plate usually contains only UAl_3 and UAl_4 as the fuel components.

2.1.2.1. UAl_x -aluminum development at EG&G

EG&G-Idaho, Inc. was chosen as a principal subcontractor by the RERTR program, owing to its expertise in the development of UAl_x -aluminum dispersion fuels for the ATR. EG&G was assisted by Atomics International, which fabricated fuel for the ATR and some U.S. university reactors.

The mini-plates fabricated by EG&G consist of UAl_x -Al cores hot roll-bonded to 6061 aluminum alloy picture frames and covers.[9] The bond surfaces are clad with 1100 aluminum to ensure satisfactory bonding. The plate dimensions, shown in Fig. 2, conform to the size specifications for irradiation testing as determined by mutual agreement among the users of the Oak Ridge Reactor (ORR) irradiation test assembly. Two thicknesses, 1.27 and 1.52 mm, were fabricated. Core thicknesses varied from 0.51 to 0.89 mm. Target fuel loadings in the range of 40 to 56 vol % UAl_x (1.9 to 2.7 g U/cm³) were studied. Most of the plates used UAl_x particles in the size range 44 to 149 μ m. Some of the plates were fabricated with <44 μ m UAl_x particles to study the effects of particle size on fabricability and, eventually, performance in the reactor.

Plate inspection and evaluation consisted of blister testing for clad/core bond integrity; visual examination for surface defects and cleanliness; X-radiography for core dimensions, fuel flaking, and core homogeneity; leak testing; metallographic examination of representative plates for core and clad thickness and grain growth across bonding surfaces; and gamma scanning for uranium content (irradiation plates).

Initial fabrication studies revealed problems with excessive core thickness, particularly at the ends (dogboning), and excessive variation in core thickness. These problems were aggravated both by higher UAl_x loadings and by the use of fine ($<44\text{ }\mu\text{m}$) UAl_x particles. Excessive core thickness precluded the use of plates with UAl_x loadings of 56 vol % U (2.7 g U/cm^3). These variations were observed in radiographs as light and dark bands traversing the width of the core. Excessive thickness was particularly serious at the 52 and 56 vol % UAl_x (2.5 and 2.7 g U/cm^3) loadings and was found to be associated with higher void volume in the more highly loaded plates. The early plates were fabricated on the assumption that the core void volume would be 8%, based on ATR plate fabrication experience. However, immersion density results indicate show that this assumption was correct only for the 40 vol % UAl_x loading and a particle size of 44-149 μm , and that the void volume percentages were greater (10-16%) in the more highly loaded plates. Assuming higher voidages in estimating the core material loads and adjusting the matrix aluminum content resolved the excessive core thickness problem. The effect of this increased voidage on performance has yet to be determined.

Dogboning was the most serious problem encountered in more highly loaded plates. In some cases, dogboning of highly loaded plates was so severe that fuel particles broke through the cladding. This condition was somewhat less pronounced, but still excessive, in plates containing 44-149 μm UAl_x particles. Approaches taken in attempts to alleviate the dogboning problem included modification of the rolling schedule (per-pass reductions were varied from 10 to 30%), slight lowering of the hot-rolling temperature (773 to 753 K) to reduce the hardness differential between the aluminum cladding and the UAl_x fuel, and tapering the leading and trailing edges of the compact. Only tapered compacts yielded any measurable improvement. Various taper configurations were produced in the compacts of some developmental plates by hand filing. A taper of 0.4 rad (22°) appeared to be optimum for controlling dogboning in plates containing up to 52 vol% UAl_x ; no dogboning was seen in plates fabricated from these compacts.

2.1.2.2. UAl_x -aluminum development at CERCA

CERCA (France) has been successfully fabricating aluminide fuel elements since the early 1960s. Elements containing uranium loadings as high as 1.6 g U/cm^3 have been produced. In 1977 a program was initiated to increase the uranium loading in 5 wt% stages.[10] At each stage, over 100 depleted uranium plates were fabricated by procedures [11] that adhered fairly closely to standard commercial practice. Acceptable plates with very good fuel meat uniformity and no dogbones were easily achieved at 45, 50, and 52 wt% U (1.9 , 2.2 , and 2.3 g U/cm^3 , respectively). However, plates fabricated at 55 wt% U (2.5 g U/cm^3) exhibited some dogboning. Hence, above 2.3 g U/cm^3 , the oxide or silicide fuels would be advantageous.

2.1.2.3. UAl_x -aluminum development at NUKEM

NUKEM (Federal Republic of Germany) has also been successfully fabricating aluminide fuel elements since the early 1960s.[12] Elements containing uranium loadings as high as 1.6 g U/cm^3 have been produced. A development program on higher-density UAl_x fuels was initiated in 1977 and a loading of 1.85 g U/cm^3 was easily achieved. This development effort was set aside to concentrate on U_3O_8 powder processing, but was picked up again in 1979.[13] The further development work raised the uranium loading to 2.2 g U/cm^3 . Very good fuel meat uniformity was obtained with little or no dogboning. NUKEM did not push this development work further because U_3O_8 was considered to be advantageous at densities above 2.2 g U/cm^3 (40 wt% U_3O_8).

2.1.3. Uranium oxide (U_3O_8)-aluminum dispersions

U_3O_8 -aluminum dispersion fuels are being used in an increasing number of reactors, since the high density of these fuels relative to other types allows either higher uranium loadings or easier fabrication at the same uranium loadings. The ORR, High Flux Beam Reactor (HFBR), and National Bureau of Standards Reactor (NBSR) are examples of reactors that were recently converted to oxide fuel. The High Flux Isotope Reactor (HFIR) has used U_3O_8 dispersion fuel since 1966. The maximum loading in HFIR is about 1.25 g U/cm^3 (~41 wt% U_3O_8).

The effort on U_3O_8 -aluminum dispersions began in the late 1950s when problems were encountered with the UO_2 -aluminum dispersion fuel used in the 1955 Geneva Conference Reactor (GCR). 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 recommended 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, as discussed later.

The loadings of U_3O_8 were gradually increased to a then maximum level of 2.3 g U/cm^3 (65 wt% U_3O_8). These fuels were developed for possible use in the HFIR and for use in the 1-MW Puerto Rico Nuclear Center (PRNC) reactor. The PRNC reactor operated successfully with a full core loading of U_3O_8 fuel with a fuel meat thickness of 0.61 mm and a 20% enrichment. This 1-MW steady-state reactor was later converted to a pulsing 2-MW Training, Research and Isotope - General Atomic (TRIGA) reactor. Thus the 2.3 g U/cm^3 (65 wt% U_3O_8) loading served as a base for further increases in U_3O_8 content. Some experimental test plates containing up to 100 wt% U_3O_8 were fabricated, but these were not intended for fuel element application.[18,19]

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

2.1.3.1. U_3O_8 -aluminum development at ORNL

ORNL was chosen as a principal subcontractor by the RERT program because of its expertise in the development of U_3O_8 -aluminum dispersion fuels for the HFIR and PRNC reactors. ORNL was assisted by Texas Instruments (TI), which until recently fabricated fuel for the HFIR, ORR, and other reactors. The fabrication techniques used in this investigation were essentially those used for years in both developmental work and commercial fabrication of aluminum-base dispersion fuel plates.[20]

For fuel loadings in use up to now, dogboning has not been a problem for U_3O_8 -Al dispersions. However, dogboning increases with U_3O_8 content and decreases with increasing core thickness; a maximum dogbone of somewhat below 40% is expected for U_3O_8 contents up to 75 wt% and core thicknesses of 0.51 mm (20 mils). As discussed in Sec. 2.1.2.1, dogboning of UAl_x -Al dispersion fuel plates can be effectively reduced by tapering the ends of the fuel compact during pressing.[21] To determine the effectiveness of similarly tapered cores for U_3O_8 -Al dispersions, 75 wt% U_3O_8 -Al plates were fabricated from compacts with (a) a double taper of 0.2 and 1.0 rad (11° and 55°) over 7.6 mm (0.30 in.) as used in the manufacture of the ATR plates,[21] and (b) a single taper of 0.3 rad (18°) over 4.6 mm (0.18 in.). Both types of tapered compacts essentially eliminated dogboning for the 75 wt% U_3O_8 -Al plates.

The effect of increasing U_3O_8 content on the mechanical integrity of clad cores was judged qualitatively by the appearance of the microstructure. Figure 3 [20] shows two polished cross sections of a clad dispersion of 75 wt% U_3O_8 -Al. The aluminum matrix is on the verge of becoming the discontinuous phase. At higher volume fractions of voids plus U_3O_8 , large planar areas with little or no tensile strength in the thickness direction were observed. These areas could accumulate fission gas during irradiation and develop a blister. The cause of the planar areas of voids and U_3O_8 is the particle breakup and stringer formation that occurs during rolling. Previous work [22] showed qualitatively that increasing the particle strength or changing the morphology could delay the onset of particle breakup and thus minimize stringer formation.

Voids formed during the rolling of dispersion fuel plates play an important part in the irradiation performance of the plates by allowing room for solid swelling and fission-gas accumulation. Workers at ORNL [18,23] have investigated the influence of several parameters on void volume and have shown that oxide concentration has the strongest effect. The void volumes obtained in those studies, which are shown in Fig. 4,[20] agree well with those reported earlier. The plates with thicker cores have slightly lower void volumes.

2.1.3.2. U_3O_8 -aluminum development at NUKEM

NUKEM began the development of U_3O_8 -aluminum dispersion fuels late in 1978, recognizing the inherent advantage of this high-density fuel system over aluminide. Rather than rely on obtaining U_3O_8 powder from the U.S., NUKEM developed several processes for the production of U_3O_8 from UF_6 , which is available from several sources and at various enrichment levels.[13] One early method involved the reaction of UF_4 , produced from UF_6 by reaction with tetrachlorethylene, with air and steam at $600^\circ C$. However, the powder produced was

not amenable to easy handling. Two other methods, ammonium uranyl carbonate and peroxide, were explored.[12] These two processes produce particles with densities of about 8.3 g/cm^3 , or about 99% of theoretical density (TD) [24]; in contrast, the density of the ORNL particles was about 8.08 g/cm^3 . Surface area for all particles is about $0.1 \text{ m}^2/\text{g}$, as determined by the (BET) method. An additional sintering step increases the density to 8.38 g/cm^3 , and produces particles that are hard as well as tough.[24]

To investigate the irradiation behavior of fuel elements with low enrichment (19.75 wt% ^{235}U) and to meet the requirements of FRG licensing authorities, six prototype fuel elements have been fabricated with U_3O_8 fuel. These elements are undergoing irradiation as follows: Two (meat thickness 1.5 mm, uranium density 2.4 g/cm^3) in the ORR; two (meat thickness 1.32 mm, uranium density 2.1 g/cm^3) in the HFR, Petten; and two (meat thickness 0.51 mm, uranium density 2.0 g/cm^3) in the Berlin Reactor (BER)-II, FRG.

In addition, high-uranium-density fuel miniplates (dimensions $114 \text{ mm} \times 51 \text{ mm}$) with reduced ^{235}U enrichment are being irradiated in the ORR reactor. Two different meat thicknesses, 0.51 and 0.76 mm, will be tested. The uranium densities are 2.4 and 3.1 g U/cm^3 , respectively. The ^{235}U content and, thus, the ^{235}U areal loading is constant for all plates. The different total uranium densities are adjusted by the ^{235}U enrichment. AlMg 1 is used as the cladding material. The nominal clad thickness is 0.38 mm (minimum 0.2 mm).

2.1.3.3. U_3O_8 -aluminum development at CERCA

CERCA restarted the development of U_3O_8 -aluminum dispersion fuels in mid-1979 after completing its development effort on UAl_x -aluminum dispersion fuels.[11] The previous maximum loading studied was 1.7 g U/cm^3 . The U_3O_8 powder is fabricated from UO_2 PWR-type powder by calcining in air.[11] After crushing and screening, the U_3O_8 powder is compacted and sintered to achieve densification.

A full-size test plate was fabricated with a 0.52-mm-thick core containing 2.7 g U/cm^3 (60 wt% uranium). The fuel core exhibited very good homogeneity and no dogboning.[10] The loadings were then extended to 3.0 g U/cm^3 (65 wt%) uranium. Test plates containing CERCA and ORNL powders were fabricated. Fine ($<40\text{-}\mu\text{m}$)-particle contents of 25 and 40% were evaluated. All plates were fabricated by the standard CERCA picture-frame process. The cladding was AG3 N.E. aluminum alloy, which contains 2.5 to 3.0 wt% magnesium. After cold rolling, the plates were blister annealed at 425°C for one hour. No blisters were found. Radiographically, all plates appeared uniform with no dogboning. A homogeneity check was also performed by x-ray absorption; the fuel region was scanned axially with a 3-mm-diameter spot. These scans showed excellent uniformity, with minimum and maximum peaks within $\pm 5\%$ of the average loading value. Again, no significant differences were detected between the different types of plates. Based on the plate loadings investigated, 3.2 g U/cm^3 loading appears to be a maximum.[25]

Metallographic analysis was performed on at least one of each type of plate. The absence of dogboning was confirmed for all plates. Some stringer formation was observed for the plates containing 40% fines. The particles themselves appeared cracked but not broken up as a result of the rolling. All plates appeared to be acceptable for irradiation.

2.1.3.4. U₃O₈-aluminum tube development at SRL

At the Savannah River Laboratory (SRL), development is directed toward tubular elements made by coextrusion of U₃O₈-Al billets clad with 8001 aluminum alloy.[26,27] Fuel assemblies consist of three concentric tubes with 3.8-m (12.5-ft)-long cores. The diameters of the tubes range from ~4.4 to ~9.4 cm (1.75 to 3.70 in.).

Aluminum-clad billets with up to 80 wt% U₃O₈ in the cores have been coextruded. Above 70 wt%, the yields are low because core cracking occurs in about 40% of the fuel tubes during coextrusion. Variation of particle size, extrusion ratios, and die angles are being investigated to increase yields.

2.2. Rod fuels

2.2.1. Uranium-aluminum alloys

U-Al alloy rods utilized in the NRU and NRX reactors at the Chalk River Nuclear Laboratories (CRNL), Ontario, Canada, contain 21 and 28 wt% uranium (0.7 and 1.0 g U/cm³, respectively). The alloy cores are extrusion clad with aluminum following a procedure which was a modification of that utilized to clad the uranium metal rods for the NRU.[28,29] The cast core is first extruded using an evacuable billet chamber in a semi-continuous manner. For cladding, twin streams of aluminum extruded through the die are forced to merge. The core is enveloped and acts as a moving floating mandrel. The extruded cores are then cold drawn for straightening and final sizing prior to machining, welding, and fit-up.

Development work is currently underway at CRNL on alloys containing 34-40 wt% uranium (1.4-1.5 g U/cm³), using basically the same process described briefly above.[30] This uranium level, if acceptable, could be used with MEU to fuel the NRU reactor. Even at these high uranium levels, the aluminide particles are kept small and good-quality extruded rods have been obtained. Rods with 120-mm-long cores were given intentional defects and subjected to 150 and 180°C autoclave testing for several days. A compatibility test at 400°C for 29 days was also performed. In both tests, only minor changes of the rod were observed.

The extrusion process has also been used to produce annular fuel tubes for the CP-5 and SRL reactors.

2.2.2. Uranium-zirconium hydride

The UZrH fuel rods for TRIGA reactors are produced by casting a uranium-zirconium alloy into graphite molds, machining to size, hydriding at elevated temperature, and grinding to final size. The ~38-mm (~1.5-in)-diameter fuel rods contain a 6.4-mm (0.25-in) drilled central hole to allow faster and more controlled hydriding; the center region is later filled with a zirconium rod. All rods are slip fit into their cladding with a cold gap of 25 to 50 μ m (1 to 2 mils).[31]

A H/Zr ratio of 1.6 is used; this falls within the face-centered cubic delta portion of the phase diagram.[31] As a result of the hydriding step, the uranium is rejected from the alloy as a fine metallic dispersion in the zirconium hydride matrix. Uranium concentrations of 8.5 or 12 wt% have been employed in the past; this corresponds to a maximum uranium volume fraction of 4% (for the 12 wt% uranium fuel). UZrH fuels were further developed during the Space Nuclear Auxiliary Power (SNAP) reactor program to 20 wt% uranium levels.[32] For low-power TRIGA reactors, a uranium enrichment of 19.9% was used. However, for the higher-power TRIGA reactors, 70% enriched fuel was employed with 3% erbium as a burnable poison to increase the core lifetime.[31] In addition, 93% enriched uranium fuel has been used for a 14-MW steady-state TRIGA reactor.

In early 1976, General Atomic undertook the development of fuels containing up to 45 wt% uranium in order to allow the use of LEU fuel to replace the highly enriched fuels while maintaining long core life.[33] These fuels were fabricated successfully, with the required hydrogen content and erbium loading as needed. The structural features of the hydrided LEU fuel were similar to those of the well-proved 8.5 and 12 wt% fuels, as shown by metallography, electron microprobe analysis, and x-ray diffraction. The uniform distribution of the uranium on a macroscale with a volume fraction of 20% for the 45 wt% fuel and the distribution of the various phases were as expected from the experience with the standard fuel. The high-U LEU fuels were subjected to thermal cycling tests, pulsing tests to 725°C, and water quench tests from 1200°C, which they survived successfully. The physical and thermal stability properties of the LEU fuels are acceptable.

2.2.3. Uranium oxide (UO₂)

Sintered UO₂ pellet fuel, clad in Zircaloy-2, is utilized in PULSTAR-type research reactors.[34] This fuel is essentially the same as that employed in light-water power reactors, except that the enrichment has been increased to 4 to 6% in order to give the desired reactivity conditions. Fuel fabrication procedures are identical with those employed for LWR fuel (see Ref. 35, for example). The present cluster design of 25 fuel pins in a 5 × 5 array can be utilized at up to 5 MW. The use of this fuel for research reactors at higher powers and fuel enrichments between 6 and 20% has not been seriously investigated.

III. IRRADIATION PERFORMANCE OF COMMERCIAL FUELS

3.1. Plate fuels

3.1.1. Uranium-aluminum alloys

Fuel-plate assemblies with a U-Al alloy core containing 18-22 wt% uranium (0.6-0.7 g U/cm³) have been extensively tested in the MTR and ETR.[1] The average burnup of the fuel has exceeded 25% with peaks of 40% (8.0×10^{20} fissions/cm³). Experimental assemblies have been run to 75% burnup with no deleterious effect.[36] Swelling appears to be linearly related to irradiation exposure, with a value of 6.38% ΔV/V per 10²¹ fissions/cm³. 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 using U-Al fuel meat containing 30.5 wt% uranium.[37] The core average fuel-burnup limit is 50%, with a maximum of 80% (2.0×10^{21} fissions/cm³).[38]

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% (7.50×10^{20} fissions/cm³). [39] Standard plates containing up to 50 wt% uranium with additions of silicon and tin were irradiated to large burnups (85% or 1.0×10^{21} fissions/cm³) under MTR conditions without dimensional distortion. [40]

3.1.2. Uranium aluminide-aluminum dispersions

Dispersions of UAl_x in aluminum have performed satisfactorily in the ETR and ATR at 22 and 41 wt% uranium (0.7 and 1.6 g U/cm³), respectively. [1,41] In these tests, prototypic elements are exposed to average burnup levels of 30% (0.8×10^{21} fissions/cm³) and peak levels of ~90% ($\sim 2.3 \times 10^{21}$ fissions/cm³). [41,42] Experimental plates have performed successfully after irradiation in the MTR, ETR, HFIR, and FR2 (Karlsruhe) under a wide variety of conditions with the maximum burnup closely approaching total depletion of the ²³⁵U ($\sim 2.8 \times 10^{21}$ fissions/cm³). [1,40,41,43] Swelling rates were generally about one half that of the U-Al alloy at about 3% $\Delta V/V$ per 10^{21} fissions/cm³ (Ref. 1). However, for the MTR experiments, which used slightly lower temperatures and water pressures, swelling rates were found to approach that of the U-Al alloy. It is postulated that under these conditions, the process of in-reactor 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/cm³ after a burnup of 40% ($\sim 1.5 \times 10^{21}$ fissions/cm³). [43]

Aluminide fuel mini-plates containing 44 to 50 wt% uranium (1.9 to 2.3 g U/cm³) have been successfully irradiated in the ORR to over 70% average burnup as part of the U.S. RERTR program. During the irradiation, channel gap spacing measurements were made on roughly a monthly basis to evaluate plate swelling and buckling. No detectable changes were found. These plates are currently cooling in the ORR awaiting post-irradiation examination. Some plates (~60% burnup) were chosen for early blister threshold temperature testing. The blister temperature was found to be ~560°C.

Full-scale prototypic or test elements containing aluminide fuel will be, or are presently being, irradiated in the ORR, the HFR-Petten, and SILOE. One element fabricated by CERCA at ~ 2.2 g U/cm³ and 45% enrichment was irradiated to 50% burnup in the SILOE reactor between February and November 1981. Four elements at 1.7 g U/cm³ and 45% enrichment, two fabricated by NUKEM and two by CERCA, have been under irradiation in the ORR since July 1981. Three of these elements have surpassed 55% average burnup. A third element fabricated by CERCA with a slightly thinner than nominal cladding has been under irradiation in the ORR since April 1982. Two elements fabricated by CERCA with thick (1.32-mm) fuel meats have been under irradiation in the HFR-Petten since January 1982, and two similar elements (1.5-mm meat) will be irradiated in the ORR beginning in late-1982.

In addition, a full-core demonstration of LEU aluminide fuel at ~ 1.7 g U/cm³ was initiated in the Ford Nuclear Reactor (FNR) at the University of Michigan in December 1981. Fuel elements, fabricated by both CERCA and NUKEM, have operated successfully in the FNR.

3.1.3. Uranium oxide (U_3O_8)-aluminum dispersions

The HFIR at ORNL uses 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 ~41 wt% U_3O_8 (~1.25 g U/cm^3); the meat thickness varies from about 0.25 to 0.76 mm (about 0.010 to 0.030 in.). The average burnup of the fissile atoms is 31% (0.9×10^{21} fissions/ cm^3), with peaks of ~65% ($\sim 1.9 \times 10^{21}$ fissions/ cm^3). [44] Experimental plates have experienced burnups of ~75% ($\sim 2.1 \times 10^{21}$ fissions/ cm^3) without failure or gross dimensional change. As part of the U_3O_8 dispersion-fuel development program for HFIR and the PRNC reactor, test samples were made, irradiated, and evaluated at ORNL. [39, 45] The fuel loading was 55 wt% uranium (2.6 g U/cm^3) in the meat, which was ~0.61 mm (~0.024 in.) thick. Short plate-type specimens were found to be dimensionally stable after a burnup of 7.7×10^{20} fissions/ cm^3 (>90% ^{235}U depletion). A full-size MTR-type element was found to be dimensionally stable after a burnup of $\sim 4 \times 10^{20}$ fissions/ cm^3 (~50% ^{235}U depletion). [39] U_3O_8 -Al dispersion fuel tubular elements containing 48 wt% uranium (2.1 g U/cm^3) have performed well for at least 141 irradiation days. [26]

Oxide fuel mini-plates fabricated at ORNL and containing up to 60 wt% uranium (3.1 g U/cm^3) have been irradiated in the ORR to over 70% average burnup as part of the U.S. RERT program. During the irradiation, channel gap spacing measurements were made approximately monthly to detect plate swelling and buckling. No changes were noted for those mini-plates containing ~19.5% enriched uranium. However, three mini-plates containing 2.8-3.1 g U/cm^3 at 45% enrichment blistered during irradiation after experiencing 2.1 - 2.4×10^{21} fissions/ cm^3 . A single, uniform blister covered from ~50 to 100% of the fuel meat area of each plate. The form of blistering is indicative of a fuel meat separation rather than a fuel-meat/cladding separation. The two other 45% enriched mini-plates had swelled substantially (10-11% increase of fuel meat thickness) after experiencing fission densities of the order of 1.9×10^{21} fissions/ cm^3 , but had not failed. On the basis of nondestructive post-irradiation examinations, all of the 19.5% enriched oxide mini-plates successfully withstood the irradiation. It is important to note that 100% burnup of 19.5% enriched, 3.1 g U/cm^3 fuel would correspond to a fission density of only 1.5×10^{21} fissions/ cm^3 (accounting for all U and Pu fissions), well below the apparent failure threshold. Destructive post-irradiation examinations of the oxide mini-plates began in May 1982.

Oxide mini-plates fabricated by NUKEM and containing 2.3-3.1 g U/cm^3 have been irradiated in the ORR to over 70% average burnup. Channel gap spacing measurements on these mini-plates, several of which had accumulated at least 1.6×10^{21} fissions/ cm^3 , gave no indication of excessive swelling. These plates are currently cooling in the ORR pool awaiting post-irradiation examination.

Full-scale prototypic or test elements containing oxide fuel are presently being irradiated in the ORR and the HFR Petten. Four elements fabricated by TI at 1.7 g/ cm^3 and 45% enrichment are being irradiated in the ORR since May 1981. One of the elements has achieved an average burnup of over 70%. Two elements fabricated by NUKEM with 1.32-mm-thick fuel meat, 2.1 g U/cm^3 , and 19.6% enrichment have been under irradiation in the HFR-Petten since late October 1981. Two similar elements with 1.5-mm-thick meat and 2.3 g U/cm^3 will be irradiated in the ORR beginning in late-1982. CERCA is currently fabricating two elements with 0.76-mm-thick meat, 3.2 g U/cm^3 , and 19.6% enrichment for irradiation in the ORR.

3.2. Rod fuels

3.2.1. Uranium-zirconium hydride

Burnups of up to ~0.52 total metal at.% (~75% burnup of the ^{235}U) have been successfully attained with UZrH fuel elements.[31] Fuel swelling has been found to be a function of temperature, hydrogen content, and burnup. Empirical relationships have been developed from the extensive tests in the SNAP reactor program on fuels with H/Zr ratios of 1.4 to 1.8 and uranium contents of 10-19 wt%.[46] Swelling was found to occur in two burnup regimes. Rapid early growth, called off-set swelling, first occurs up to ~0.1 metal at.% (~14% burnup of the ^{235}U) as a result of a vacancy-condensation phenomenon. Swelling in this regime was found to be temperature dependent through an exponential relationship. For an average temperature of 700°C (1300°F), swelling of about 2.5% $\Delta V/V$ was observed at ~0.1 metal at.% burnup. Above this burnup, swelling decreases dramatically to 2.76% $\Delta V/V$ per metal at.% burnup. This second burnup rate, which is due to solid fission products, is relatively temperature insensitive. Hence, for normal steady-state and transient operation, fuel swelling is well within acceptable limits.

TRIGA-LEU fuel is currently undergoing high-power tests in the 30-MW ORR. These tests began in mid-December 1979 on a 16-rod shrouded cluster. The fuel rods, 13.0 mm (0.51 in.) OD and 5.6 cm (22 in.) long, are clad with 0.41-mm (0.016-in.)-thick Incoloy. Fuel samples, with 45, 30, and 20 wt% U (nominally 20% enriched) are undergoing irradiation and will achieve burnup values of up to 50% of the contained ^{235}U . Owing to an interruption, it will take about one more year of irradiation to produce the desired burnup in the 45 wt% U fuel, although the design burnups have already been reached for the other two fuel loadings. Many of the fuel rods have been removed from the cluster for inspection and rearrangement and no growth, swelling, or bending problems have been detected.

3.2.2. Uranium oxide (UO_2)

The UO_2 fuel for PULSTAR-type research reactors is essentially the same as that used for commercial LWRs which has exhibited satisfactory performance to burnups of over 40,000 MWd/t.[35,47,48] The latter references give details of the irradiation behavior of UO_2 pellet fuel.

IV. DEVELOPMENT OF NEW HIGH-DENSITY FUELS

4.1. Requirements

4.1.1. High fuel density

In the RERT program the intention is to use, if at all possible, the present aluminum dispersion fuel-meat technology rather than some advanced technology employing stiffer fuel meats or claddings (e.g., Zircaloy). The most straightforward method of increasing the uranium density of aluminum dispersion fuels is to utilize a fissile dispersant of higher density than those currently employed. Data on a number of current and potential fissile

compounds are given in Table 2.[49] 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. As noted previously, the current maximum qualified fuel loading is about 1.7 g U/cm³. For the oxide and aluminide fuels, this requires fuel loadings of 20 and 30 vol% of the fissile compound, respectively, as seen graphically in Fig. 5. If U₃Si can be used, for example, these volume percentages will provide uranium loadings of 3.0 and 4.6 g/cm³. If dispersions containing 50 vol% of fissile compounds are fabricable, then, from Fig. 5, a uranium loading of about 7.3 g/cm³ can be achieved for U₃Si fuel dispersed in aluminum. This loading will enable the conversion of most research and test reactors to LEU fuels.

4.1.2. Corrosion resistance

Metallic uranium alloys and compounds were extensively investigated in the U.S. in the late 1940s and the 1950s, principally at ANL, Battelle, Knolls Atomic Power Laboratory, and Westinghouse as fuels for light-water reactors. Several thorough reviews of the subject have been published.[50,51] 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. U₃Si 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.[51] 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.[52]

SRL [54] has taken another approach to the problem of providing corrosion resistance, i.e., the diffusion bonding of both the fuel and cladding to an intermediate layer. On early fuel elements, the intermediate layer was formed by an Al-Si dip-canning process; later, an electroplated nickel layer was used. 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.[51]

Table 2. 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
U-10% Mo Alloy	1150	17.1	16.4	0.86
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 ₃ O ₈	- ^b	8.40	7.1	0.37
UN	2630	14.3	13.5	0.71
U ₆ Fe	815	17.7	17.0	0.89
UFe ₂	1235	13.2	9.0	0.47
U ₃ Si	930 ^b	15.6	15.0	0.79
U ₃ Si ₂	1665	12.2	11.3	0.59

^aFrom Ref. 49.^bDecomposes.

4.1.3. 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.[50] 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.[55] Swelling rates of 2-4%/at.% burnup were found after irradiation of 9,000 and 12,000 MWd/t up to 600°C maximum fuel temperature, again with the exception of abnormal swelling at ~400°C. The irradiation behavior of uranium-niobium alloys was found to be similar to that of uranium-molybdenum.[51] 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.[51,55] However, additions of carbon were found to greatly modify swelling behavior in low-burnup tests.[51] Good irradiation performance has been obtained with a U-5 wt% Fs alloy (where the fission is made up of the mixture of fission products expected in recycled fuel) to burnups exceeding 10 at.% in the Experimental Breeder Reactor (EBR)-II.[56] However, these cast and heat-treated rods are bonded to the cladding with sodium; this provides volume for ~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.[55] Most of these alloys were quenched from 725°C, then irradiated to 9,000 and 13,000 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 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 U₃Si fuel material produced mixed results.[51] 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.[57] In recent experiments at AECL, U₃Si and aluminum-modified U₃Si showed excellent irradiation performance.[58,59] Diameter increases for Zircaloy-clad U₃Si 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, UO₂ performs well since it can be operated at steady-state central temperatures approaching its melting point.[60] UO₂ 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).[61] The shift to oxide fuel occurred because the behavior of metal alloy fuels was found to be inadequate.[60]

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.[62,63] 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.[48] A similar document is available for light water reactor oxide-fuel behavior.[49] 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.[64] 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 $\text{UO}_2\text{-ZrO}_2$ 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} fissions/cm³).[65] However, plate failures occurred at a burnup as low as 20,000 MWd/t ($\sim 0.6 \times 10^{21}$ fissions/cm³) as a result of fission-gas release.

4.1.4. Reprocessing potential

Research reactor fuels have been routinely reprocessed at SRL since the 1950s. These include alloy and U_3O_8 and UAl_x dispersion fuels. U-ZrH_x fuels are being stored until suitable quantities to justify a campaign are accumulated. Flow sheets have been developed and tested for this fuel, but the hardware required has not yet been fabricated. For UO_2 fuels, techniques such as the standard PUREX method are available.

No problems are foreseen in the reprocessing of the more highly loaded standard fuels containing LEU. However, confirmatory dissolution studies are being performed. For U_3Si fuels, the reprocessing potential is less clear. Silicon in some weight-fraction ranges has been shown to interfere with phase separation in the extraction column.[4] However, an internal study at ANL by the Chemical Engineering Division has concluded that the standard aqueous process can be used, perhaps with a slight modification. In addition, as noted earlier, U-Al fuel elements from the Greek Research Reactor-1 which contained 3.0 wt% silicon were successfully reprocessed by the UK using their standard acid-deficient reprocessing scheme.[5] Thus, an experimental program has been initiated at SRL to evaluate the reprocessibility of U_3Si -containing fuels and the suitability or adaptability of such reprocessing schemes to the standard SRL flow sheets. Early results are very encouraging.

4.2. Fabrication development

4.2.1. Uranium silicide-aluminum dispersions

4.2.1.1. Introduction

The data presented above suggest that U_3Si , possibly modified with aluminum, has the greatest potential for providing the necessary uranium density and irradiation performance levels. 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. 5, 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. 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 that 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.

4.2.1.2. Phase relationships

The U-Si phase diagram is shown in Fig. 6. The work at ANL under the RERT program is primarily concerned with the U-rich portion of the system up to and slightly beyond U_3Si_2 . [66] The most important feature of the system in this region is the peritectoid formation of U_3Si . As demonstrated by others, and confirmed in Ref. 66, a 72-h anneal at 800°C is sufficient to complete the reaction in an as-cast alloy. In order to be sure that no excess uranium solid solution exists in alloys so heat treated, the composition chosen for the U_3Si alloy was U + 4 wt% Si - slightly to the Si-rich side of the stoichiometric compound. Other alloy compositions selected as candidate fuel alloys include U_3Si_2 (with 7.4 wt% Si, again slightly to the Si-rich side of stoichiometry) and U + 3.5 wt% Si + 1.5 wt% Al. This ternary alloy was developed by AECL researchers during the course of their work with silicides and is referred to as " U_3SiAl " in the present paper. This alloy exhibited improved corrosion resistance and irradiation performance over the binary U_3Si .

Microstructures of U_3Si , U_3Si_2 and U_3SiAl are shown in Figs. 7-9, respectively, both before and after heat treatment. Briefly, the as-cast structure of U_3Si (Fig. 7a) contains primary U_3Si_2 with a matrix of uranium solid solution and U_3Si_2 eutectic. After heat treatment (Fig. 7b), the microstructure consists of U_3Si_2 particles dispersed in U_3Si . The structure of U_3Si_2 in either the as-cast (Fig. 8a) or heat treated (Fig. 8b) condition consists of U_3Si_2 plus a small amount of USi . In the as-cast condition (Fig. 9a), U_3SiAl consists of U_3Si_2 plus UAl_2 in an eutectic matrix of U_3Si_2 plus uranium solid solution. After heat treatment (Fig. 9b), the dominant phase is U_3Si with small amounts of uniformly dispersed U_3Si_2 and UAl_2 .

4.2.1.3. Processing procedures

4.2.1.3.1. Plate fuels

The fabrication procedure is a variation of the standard picture-frame method that finally evolved at ANL for the three candidate fuel alloys (U_3Si , U_3SiAl , and U_3Si_2). Two steps were anticipated to be difficult and are still of concern:

- (a) Problems were expected with the generation of uranium alloy powder in the quantities necessary for fuel plates. Whereas U_3Si_2 is brittle and comminutes easily, the other two alloys are tough and the generation of particulates is difficult. Further, all three alloys tend to be pyrophoric. While all three powders have been successfully produced in an air environment, ignition of fine powder has occasionally occurred during comminution procedures. For the U_3Si and the U_3SiAl , a Spex "Shatterbox" has worked effectively. U_3Si_2 has been crushed with a steel mortar and pestle.
- (b) The production of homogeneous mixtures of fuel alloy particles with commercially available Al powder was expected to be a problem. However, compacts and mini-plates comprising up to about 50 vol% fuel alloy, and falling within the allowable limits of heterogeneity, have been produced by more or less conventional mixing techniques.

The powder of Fig. 10a is typical of the fuel alloy powders employed in this study. The MD-101 Al powder (Fig. 10b), produced by Alcan, is a much finer material, with 100% at -100 mesh and ~85% at -325 mesh.

Hardware before assembly and after rolling is shown in Fig. 11, along with a completed mini-plate. Representative mini-plate cross-sections are shown in Fig. 12.

The uranium loading of the mini-plates has been increased as a result of recent development efforts. The first phase concentrated on U_3Si and U_3SiAl at a fuel volume of 34% (~4.8 g U/cm^3) and U_3Si_2 at a fuel volume of 34% (~3.8 g U/cm^3). The second phase concentrated on volumes of 43-50% or (5.8-6.5 g U/cm^3), principally with U_3SiAl fuel. For the phase-one plates, fuel meat uniformity was excellent and no dogboning was evident. For the phase-two plates, fuel uniformity was still good; however, some dogboning was evident. Recently, U_3SiAl mini-plates containing 7.0 g U/cm^3 were produced. In order to reduce dogboning to an acceptable level, the U_3SiAl -Al compacts were fabricated with tapered edges on both the leading and trailing ends. After satisfactory mini-plates were produced at 7.0 g U/cm^3 , the effort shifted to the development of full-sized highly loaded silicide plates.

The study of silicide dispersion fuels for plate-type elements is also being pursued by CNEA (Argentina), NUKEM (FRG), and CERCA (France). The CNEA effort has been initiated with the fabrication of mini-plates to the same specifications as the ANL plates.[67] Mini-plates have successfully been fabricated with U_3Si containing uranium loadings of 5.2 and 6.1 g U/cm^3 . Fuel uniformity was good; however, some dogboning was evident. NUKEM has been developing U_3Si_2 fuel while CERCA has been developing U_3SiAl fuel, both

as full-sized plates. NUKEM has fabricated U_3Si_2 plates at a uranium loading of 4.75 g/cm^3 (43 vol% U_3Si_2). [68] This fuel was chosen because of its ease of comminution. Earlier work had been performed with U_3Si . The fuel was fairly uniform with no dogboning; however, some slight axial striations were evident. Subsequently, NUKEM has fabricated two full-size fuel elements with a uranium loading of 4.75 g U/cm^3 and an enrichment of 19.7% for irradiation testing in the ORR beginning in May 1982. Homogeneity in these plates was very good. The development of U_3SiAl fuel at CERCA has thus far produced acceptable plates at 5.2 and 6.6 g U/cm^3 . [69] Both loadings produced uniform plates; however, the plates fabricated at 6.6 gU/cm^3 exhibited some dogboning. CERCA is also evaluating the use of higher silicon content as a means of facilitating powder production. CERCA will be fabricating four full-sized fuel plates, two each with uranium loadings of 5.5 and 6.0 g U/cm^3 , for irradiation testing in the SILOE reactor. If irradiation proves successful, a full-sized element will then be fabricated and tested. CERCA will be fabricating two full-sized fuel elements at 4.75 g U/cm^3 for irradiation in the ORR, utilizing the U_3Si_2 compound, which is more easily comminuted.

4.2.1.3.2. Rod fuels

The use of U_3SiAl (also denoted as $USiAl$) fuel in an aluminum dispersion is being evaluated for the NRX and NRU reactors at the CRNL. [70] The NRX element design currently consists of seven similar pins fueled with Al-28 wt% U, while the NRU element design consists of twelve 2.9-m-long pins fueled with Al-21 wt% U; both use HEU. The development program includes production and characterization of $USiAl$ powders and Al- $USiAl$ billets, production of fuel elements by extrusion, out-of-reactor corrosion tests on fuel pins, compatibility studies, defining fuel material properties, and irradiation testing.

The $USiAl$ alloy is very difficult to crush into powder directly from cast rods or buttons. However, suitable powders can be fabricated by machining rods into chips, which are then granulated. At CRNL, this technique was used to produce about 8 kg of $USiAl$ powder for fabrication development and irradiation tests. All size fractions are used to produce the dispersion fuel billets; however, powder in the coarse size range (88 to $149 \mu\text{m}$) is preferred. Figure 13 [70] shows photographs of two polished sections of $USiAl$ powder. The U_3Si matrix exhibits a typical etched structure; U_3Si_2 and UAl_2 particles are visible within it. The powder particles tend to be boulder-shaped rather than acicular. This shape is thought to assist in achieving good homogeneity in the billets and extruded rods. Spectrographic analysis of the powder shows that the impurity level is not significantly higher than the specification required for the as-cast rods.

Since about 500 kg of 20%-enriched $USiAl$ powder would be required each year to refuel the NRX and NRU reactors, faster and better ways of producing such powder are being reviewed. For the longer term, a facility to examine the possibility of directly casting $USiAl$ shot ($\approx 150 \mu\text{m}$) is being set up. About 45 kg of U_3Si shot were produced in 1968 for another program; however, most of the shot was quite coarse ($\approx 2 \text{ mm}$) and would require further attrition. This coarse shot would probably cause considerable wear of the teeth in the granulator. Another possibility for making powder is numerically controlled machining of $USiAl$ rods, with one operator supervising several lathes or mills.

The Al-U alloy for the present NRX and NRU reactor fuels is induction melted and cast into molds. Billets about 37 mm diameter by 150 mm long are produced. The billets are extruded into rods which are drawn to size, stretch straightened, and extrusion clad in aluminum after the end plugs are installed. The proposed fabrication route for Al-USiAl fuels is similar except that billets are produced by powder metallurgical techniques. Billets with fuel compositions ranging from Al-50 wt% USiAl to Al-75 wt% USiAl have been fabricated successfully for extrusion tests, using the following procedure: The -325 mesh aluminum powder and -100 mesh USiAl powder were blended for 30 minutes, vacuum dried overnight at 100°C, and pressed into small billets using a lubricant on the die and plunger. (If a lubricant was not used, the billets deformed badly as they were pushed out of the die.) The powders were pressed at room temperature, at pressures ranging from 100 to 350 MPa (15,000 to 50,000 psi) to produce billets ranging from 75 to 90% TD. The lower-density billets were examined as a possible means of including porosity in extruded fuel pins. The billets were well formed, with shiny aluminum-colored surfaces. Surface finishes on billets pressed at higher pressures were generally smoother than those at lower pressures.

A series of Al-USiAl billets with the composition and densities shown above were heated for one hour at temperatures required for extrusion (300 to 400°C) to determine if oxidation of the fuel is a concern. Results show that heating of dispersion fuel billets in air is not possible because they oxidize badly. Billets heated in an argon-purged furnace also oxidized; those heated in vacuum or pure argon to 400°C showed no change. In comparison, Al-U alloy billets containing 21, 28, or 40 wt% U showed no signs of oxidation after heating at 500°C in air for one hour.

Small billets containing either Al-75 wt% USiAl or Al-55 wt% USiAl at two density levels (about 80 and 94% TD) were vacuum degassed overnight at room temperature before extrusion to remove residual die lubrication. They were then heated in vacuum at temperatures between 300 and 350°C and extruded into air or water to form 5.8-mm-diameter rods. The results of these tests show that:

- (a) The billets required pressures of about 1.1 MN (125 tons).
- (b) Samples extruded into air showed some surface oxidation (~0.2 mm thick) and roughening. A short section of rod produced from low-density billets oxidized completely. Samples extruded into water showed a slight darkening of the surface but no roughening.
- (c) Porosity in billets collapsed during extrusion, resulting in high-density (>97% TD) rods.
- (d) At an extrusion temperature of 300°C, rods produced from Al-75 wt% USiAl showed tear marks over the first half of the rod, indicating that the extrusion temperature was too low. This material extruded well at 350°C. The Al-55 wt% USiAl billets extruded well at 300°C.

Rods of both compositions were drawn 0.3 mm narrower to produce good-quality 5.5-mm-diameter rods. These rods were extrusion clad in aluminum.

Stretch straightening of bare Al-USiAl rods at temperature has not yet been demonstrated.

Metallographic examination of billets and extruded rods show that the homogeneity of the fuel is good. Photographs of polished longitudinal sections of the Al-55 wt% USiAl and Al-75 wt% USiAl rods are shown in Fig. 14 [30]. All sections show a uniform distribution of USiAl particles (dark) in a white aluminum matrix.

4.2.1.4. Properties

4.2.1.4.1. Corrosion tests

Corrosion tests were carried out on two 1.27-mm (0.050-in.)-thick mini-plates in the 0-temper condition.[65] The plates contained 30 vol.% depleted uranium fuel (one U₃Si and one U₃SiAl). A 3.25-mm-diameter hole was drilled through the fuel zone of each plate, and the plates were submerged in separate baths of boiling (100°C) distilled water. The test continued for one week (168 h), with periodic withdrawals for weighing and examination. The U₃Si plate showed no measurable weight change; the weight of the U₃SiAl plate decreased from 32.232 to 32.220 g. The plates darkened during the exposure, but no other changes were noted; no radioactivity was detectable in samples of the water.

Two short extruded mini-elements, containing 55 and 75 wt% U₃SiAl, respectively, were tested in a static autoclave.[30] A 1-mm-diameter hole was drilled through the cladding and into the fuel for these tests. After two weeks in 150°C water or one week in 180°C water, only the Al-75 wt% U₃SiAl element showed changes. It had a slight bow, a 1.5% increase in diameter, and a 1% increase in length. After a total of 3.2 weeks in 150°C water or two weeks in 180°C water, both types of elements showed small defect holes and bubbles at various points along their length. The reaction may be due to the absence of a bond at the fuel-cladding interface.

The results of these corrosion tests are encouraging, since the behavior of the new fuels was not a great deal worse than that expected for currently used fuel under similar conditions. However, the corrosion behavior of advanced fuels in a flowing water environment remains to be determined.

4.2.1.4.2. Compatibility tests

Reactions between the silicide particles and the aluminum matrix or between the fuel and cladding could occur in-reactor as a result of temperature- and fission-induced events. In the reactor the maximum fuel temperature is likely to be around 200°C while the temperature at the fuel-cladding interface will probably be no higher than 150°C.

Unirradiated plate samples of U₃Si, U₃Si₂, and U₃SiAl, approximately 22 mm × 50 mm × 1.27 mm in size and containing 30 vol.% depleted fuel alloy, have been heated for 1000 h at 300°C plus 1000 h at 450°C.[65] Rod samples (≈25 mm long) of Al-75 wt% USiAl and Al-55 wt% USiAl, clad in aluminum, were heated for 700 h in vacuum at 250°C and 400°C.[30] The temperatures were kept higher than those expected in-reactor in an attempt to simulate the enhanced reaction expected as a result of fission-induced events.

X-ray data for fuel meat removed from the thermally treated fuel showed the presence of UAl_3 with dissolved Si, as predicted by the ongoing phase-equilibrium studies. However, whereas all three alloys with 30 vol% fuel should have shown diffraction lines for Al, only two samples displayed such evidence.

After 700 h at 400°C, the rod specimens showed significant increase in diameter and length: 1.5 and 3.7%, respectively, for the 55 wt% U_3SiAl fuel; and 2.5 and 7.9%, respectively, for the 75 wt% U_3SiAl fuel. Preliminary metallographic examination of the dispersion fuel samples heated at 400°C indicated that a severe reaction had occurred and the composition and structure of the fuel particles had changed considerably. The samples are being examined in more detail to identify the phases present. These preliminary results show that the compatibility of the $USiAl$ particles with the Al matrix is not nearly as good as for the UAl_4 particles at 400°C. This temperature may be too severe to simulate in-reactor effects. However, these samples and those heated at 250°C, which have not yet been examined, will provide an opportunity to identify phases and reactions that might also occur in-reactor as a result of temperature- and fission-induced events.

4.2.1.4.3. Thermal conductivity

The thermal conductivity of five samples (19 mm diameter by 22 mm long) with a wide range of silicide-aluminum compositions is presently being determined. Researchers at CRNL have estimated the conductivity of U_3SiAl -Al compacts by employing Maxwell's [71] and Eucken's [72] equations for multi-component systems and the known values of the conductivity of U_3SiAl and aluminum.[30] Maxwell's equation is thought to be valid up to 30 vol% dispersed phase. The conductivities calculated with Maxwell's equation, assuming that either Al or $USiAl$ is the continuous phase, are plotted vs fuel composition in Fig. 15.[30] It is likely that the real curve will be a composite of the left side of curve (A) and the right side of curve (B) with an interconnecting curve (C) between the two. Such S-shaped behavior has been noted by Kingery in two-phase systems.[72] Three data points obtained recently at CRNL [73] are also shown in Fig. 15. Additional data are required.

4.2.2. Uranium oxide (UO_2) caramel wafers

Caramel plate fuel is composed of small square wafers (hence the name "caramel") of UO_2 , separated by Zircaloy wire partitions and frames, between two Zircaloy cladding plates; each such assembly forms one of the plates in a fuel element, as shown in Fig. 16 [74]. Uranium dioxide was chosen for its inertness, its good irradiation performance in standard light-water-cooled power reactors, and its high uranium density (9.1 g U/cm^3), which reduces the required enrichment to as little as 3 to 10%.

The Commissariat à l'Energie Atomique (CEA) of France began the development of caramel fuel for merchant marine propulsion reactors and small-to-medium size power stations in 1972.[74] Investigations began in 1976 to convert the core of Osiris, CEA's most powerful (70 MW) research reactor.[75] The development and testing of the fuel was performed at the Centre d'Etudes Nucléaires (CEN) at Saclay, France.

The wafers are produced by standard powder metallurgy techniques which include cold compacting, sintering at $\sim 1600^{\circ}\text{C}$ under hydrogen, and grinding to size. Wafer thickness can range from 1.5 to 4.9 mm. The thinnest wafers are used in the Osiris reactor in order to increase volumetric power in the fuel to $3000\text{--}4000\text{ W/cm}^3$, while maintaining fuel temperatures at reasonable levels. The wafers are covered by a layer of chromium deposited by cathodic sputtering. The chromium layer provides a barrier to the diffusion of oxygen into the Zircaloy, which could otherwise occur during the diffusion bonding operation. The wafers are placed within a very carefully produced Zircaloy wire grid located within a Zircaloy frame and lower Zircaloy cladding plate. The Zircaloy cover is spot welded on, and the edges are secured by resistance seam welding. The ends of the assembly are then electron-beam welded under vacuum, followed by diffusion bonding of the Zircaloy components at elevated temperature ($>900^{\circ}\text{C}$) and pressure ($\sim 100\text{ MPa}$). After machining, the plates are electron-beam welded into the side plates to form the fuel assemblies.

4.3. Irradiation testing and demonstration

4.3.1. Uranium silicide-aluminum dispersions

One module containing several kinds of silicide-aluminum dispersion fuels has been successfully irradiated in the ORR with an average fuel burnup of $>80\%$ as part of the U.S. RERTR Program. U_3Si and U_3SiAl fuels were loaded to $\sim 4.8\text{ g U/cm}^3$, while the U_3Si_2 fuels were loaded to $\sim 3.8\text{ g U/cm}^3$. In addition, two U_3SiAl mini-plates loaded to 5.6 and 5.9 g U/cm^3 have been irradiated to $\sim 75\%$ average burnup (2.0 and 2.1×10^{21} fissions/ cm^3 , respectively). During the irradiation, channel gap spacing measurements were made on roughly a monthly basis to evaluate plate swelling and buckling. No changes were detected. Post-irradiation examination of these plates has begun.

One module containing U_3Si and U_3SiAl fuels at $\sim 4.8\text{ g U/cm}^3$ was removed from the ORR during October 1980 after a ^{235}U burnup of 34% . During the cooling period in the pool of the ORR, the module was examined visually. A 4X underwater periscope was used to examine the fuel plates without removing them from the modules. No changes were detected. The module was allowed to cool on the pool-side station for three months, and was then shipped to ANL for post-irradiation examination (PIE). The PIE of the 12 silicide mini-plates is now complete. The appearance of these plates was excellent, with no visible distortion or blistering. However, the surface was covered with a thin oxidized aluminum coating. One alloy plate was chosen at random for early burnup analysis, which yielded a value of 29.5% of the original ^{235}U atoms, close to the predicted value.

Immersion densities, micrometer measurements, and gamma scans have been made on all irradiated mini-plates from the module. The density of the plates decreased by 0.2 to 0.6% , with the 1.27-mm -thick plates showing the greatest density change. The results of the micrometer thickness measurements show a negligible change after irradiation, which qualitatively confirms the low level of density change. Gamma scanning for gross, Zr-Nb^{95} , and Cs^{137} counts were performed along the width and length of each plate. The profile for a given isotope was the same for all plates, indicating little or no migration of fission products. The width scans show a depression in counts near the center of the module, caused by flux perturbations.

One U_3SiAl and one U_3Si mini-plate were sectioned for metallographic analysis. The results indicate that all of the fuel particles are surrounded by an interaction layer between the particle surface and the aluminum matrix. The interaction layer is quite uniform and varies from 3 to 6 μm in thickness. Some of the smaller particles have been completely consumed by the interaction layer. Within statistical error, the average fuel particle size and the thickness of the interaction layers around the fuel particles do not vary with position in the plate. Small fission-gas bubbles have been observed within the particles, but they do not appear in the interaction layer. No larger fission-gas bubbles have been observed, nor are there any indications of gross swelling of fuel particles.

A specimen from the U_3SiAl mini-plate has been examined with an electron microprobe. The fission products Zr, Mo, Xe, and Cs (and possibly Ru and Sr) have been identified, and they appear to be uniformly distributed within the fuel particles. Silicon appears in high concentrations in isolated areas of the fuel particles. Initial examination of the interaction layer indicates that it is rich in aluminum. A specimen from the same U_3SiAl mini-plate has been examined in a scanning electron microscope (SEM). The specimen was fractured after immersion in liquid nitrogen, and the examination showed that the fuel particles fractured in the cleavage mode rather than in a ductile mode characteristic of the unirradiated fuel. The spherically shaped and uniformly distributed fission-gas bubbles ranged in size from 0.5 μm in diameter down to the resolution of the SEM ($\sim 0.1 \mu m$). No fission-gas bubbles were observed in the interaction layer.

Conventional blister threshold temperature measurements have been performed for both U_3Si and U_3SiAl mini-plates. The blister threshold temperature was measured to be $515 \pm 10^\circ C$ for both types of fuel. In each case the blistering consisted of a single large blister covering most of the meat width near the trailing edge in the initial roll pass of the plate. Severe bowing of the plates occurred in the temperature range 490 to $500^\circ C$. Sectioning of a U_3SiAl plate revealed that the blister was a result of a separation within the fuel meat. This separation occurred even in areas removed from the blister area. Fuel particle pullout occurred during polishing for metallographic examination, indicating that the particles were weakly bonded to the aluminum matrix. The fuel particles grew considerably during the blister test at the expense of the aluminum matrix. The particles appeared to be in two phases, a central phase surrounded by a 20- to 40- μm -thick interaction layer. An electron microprobe analysis of the sample showed that the interaction area was rich in aluminum and that the aluminum concentration decreased continuously from the edge of a particle towards the surface of its central U_3SiAl phase, while the uranium and silicon concentrations increased. A fifth mini-plate was blister tested with a procedure that was different from the standard procedure. Instead of incrementally increasing the temperature, holding for $\sim 1/2$ h, cooling and examining for blisters, and then repeating the sequence for the next higher temperature, the plate temperature was slowly raised from room temperature to $515^\circ C$ over a period of 6 h. The temperature was held for $\sim 1/2$ h and the plate was removed from the furnace. The plate again showed severe bowing, but no blistering was observed. The plate thickness had increased by 10 to 15% during the test and its length had increased by 2.8%.

The PIE of the high-burnup plates has just begun. One blister threshold temperature test has been run. The blister temperature was about 510°C, which is consistent with data from the low-burnup plates; this indicates that there is essentially no burnup dependence.

Mini-elements containing 5.5-mm-diameter U_3SiAl fuel at a uranium density of 3.5 g/cm³ are undergoing irradiation by the CRNL in the NRU reactor. Interim examinations were performed at 20 and 40% burnup. All elements appeared to be in excellent condition. Volume changes were <1.0% and 0.4%, respectively. The irradiation of higher-density (4.9 g U/cm³) U_3SiAl is planned.

4.3.2. Uranium oxide caramel wafers

Prior to the conversion of the Osiris reactor to caramel fuel, the fuel was extensively tested by CEN at a broad range of specific powers and burnups. [76] An exploratory evaluation program was conducted between 1965 and 1970 in the EL3 reactor at Saclay to define the technological limits of the fuel. The specific power ranged from 1000 to 3000 W/cm³ and the cladding temperature was between 280 and 340°C. Burnups of 30,000 MWd/T were reached. Additional tests were performed during the period from 1973 to 1978 in the SILOE and Osiris reactors; each involved several small samples of caramel fuel. The Osiris experiment was in a pressurized (14 MPa) loop at 300°C while the SILOE experiment was at pool water temperature and pressure. The maximum exposures were 25,000 and 40,000 MWd/T. The fuel behaved well in these experiments, with no detectable release of fission gas. Power cycling tests were also performed on 4-mm-thick caramels after a burnup of 30,000 MWd/T in the Osiris reactor. No problems were encountered after the 3634-cycle test.

To appraise the operational safety of caramel fuel, a test of cladding with artificial defects was run within a loop in the EL3 reactor. A delayed neutron detection system was used to follow fuel behavior. A low level of fuel release was recorded, which corresponded to insignificant changes in the fuel under the previously drilled hole. This confirmed the excellent behavior of the fuel and the failure detection capability of the delayed-neutron system.

Prior to conversion of the Osiris core, three prototypic caramel fuel assemblies were irradiated under a range of operating conditions with no difficulties. The Osiris reactor was completely loaded with caramel fuel in October 1979, with start-up tests performed in January/February 1980. Since that time, the fuel has performed well and burnup levels have been increased to 30,000 MWd/T. Maximum specific power in the fuel is 4300 W/cm³. A systematic test by strain gauge of the water channel spacing in all elements revealed no changes from pre-irradiation values. A destructive evaluation of an element which has reached an average burnup of 30,000 MWd/T (peak of ~40,000 MWd/T) is currently under way.

4.4. Other fuel systems

Other fuel systems have been or are being evaluated as backups to the aluminide, oxide, and silicide fuels now in use. Uranium-molybdenum fuels were chosen by both ANL and CEA as backups for silicide and caramel fuel, respectively. However, development was never initiated, owing to the success of the silicide and caramel fuels. NUKEM has a small development program to evaluate other fuel systems.[67] Efforts to develop a form of UO_2 that was less prone to thermite reaction with aluminum were terminated when diluents used to reduce UO_2 activity, such as alumina, were found to substantially reduce the fuel density. However, an effort on U_6Fe is under way at KfK even though preliminary tests revealed that this fuel was more reactive with aluminum than U_3Si_2 .

Another fuel system currently under evaluation (for the University of Missouri) is the UAl_2 -Al system. Fabrication development was carried out by Atomics International (AI).[77] Mini-plates approximately 25 mm wide, 250 mm long, and 1.27 mm thick were successfully produced at a uranium loading of up to 55 vol% (~ 3.4 g U/cm^3). These plates are being irradiated in the ATR.

V. PROPERTIES OF FUEL SYSTEMS

5.1. Thermal conductivity

5.1.1. Dispersion fuels

No data is available on the thermal conductivity of UAl_x -aluminum dispersions. Some thermal diffusivity experiments were performed on fuel-plate wafers by the pulsed-laser technique.[78] However, the specimens were not characterized, nor was the system calibrated well enough to yield meaningful results. Excellent data on the U-Al alloys have been obtained from heat-flow measurements [79], as shown in Fig. 17. Since the alloy is a dispersion of UAl_3 and UAl_4 particles in an essentially uranium-free aluminum matrix, these data should adequately represent the conductivity of UAl_x -aluminum dispersion fuels. The data have been fitted by a straight line which represents the decreasing thermal conductivity with increasing weight fraction, x , of uranium by the equation

$$k = 216.55 - 276.25x \text{ W/mK.}$$

Data are shown in Fig. 18 for various uranium weight percentages in U-Al alloys.[79] As seen in the figure, the temperature effect on thermal conductivity for these alloys is a small one, with conductivity decreasing very slightly with increasing uranium content.

The thermal conductivity of U_3O_8 -aluminum dispersions at 60°C was derived by the author from unpublished information contained in two ORNL letter reports.[80,81] Disk samples 6.35 mm (0.25 in.) in diameter were cut from areas of the fuel plates that had been selected for uniformity by eddy-current measurements and radiography. The thermal conductivity measurements were made in a comparative heat flow apparatus. The data were joined as shown in Fig. 19 to obtain a smooth curve. For the lightly loaded dispersions, the conductivity decreases linearly with increasing fuel particle volume

fraction. The decrease is due to the substitution of low-conductivity U_3O_8 particles ($k \sim 0.3-0.5$ W/mK, from Ref. 79), which ostensibly act as voids, for aluminum. As more U_3O_8 is added, however, the conductivity drops more dramatically. In the range of interest to the RERTR program ($2.5-3.0$ gU/cm³) the conductivity ranges from 30 down to 12 W/mK. The experimenters at ORNL also evaluated the highly loaded samples at 30 and 90°C.[81] No appreciable temperature dependence was found in the temperature range 30-90°C.

The thermal conductivity of $U_3SiAl-Al$ dispersions was shown in Fig. 15.

5.1.2. UO_2

Much work has been performed in determining the conductivity of UO_2 . The oxygen-to-metal ratio greatly affects the conduction process, since it determines the defect structure (i.e., excess electrons and holes). At elevated temperatures, atmospheric control must be provided to maintain UO_2 stoichiometry. The thermal conductivity is also affected by porosity. Figure 20 [48] shows a compilation of available data on the conductivity of UO_2 as a function of temperature.

5.1.3. $U-ZrH_x$

The thermal conductivity of $U-ZrH_x$ has been measured over a range of temperatures, [82,83] and for uranium contents ranging between 8.5 and 45 wt%.[84] The data are shown in Fig. 21.[84] Curves A through D infer that conductivity increases with hydrogen content. From these curves, researchers at General Atomic determined that the straight line UL through curve D represented a reasonable upper limit for conductivity of $U-ZrH_{1.6}$ material.[84] From instrumented fuel elements, they deduced that the straight line labelled LL was a reasonable lower limit, which produced a most-probable-value line designated by MP.

Diffusivity experiments were performed on samples of $U-ZrH_{1.6}$ with varying uranium contents as a function of temperature, [84] using the pulse-heating technique. When reasonable values for density and specific heat were used, the conductivity was found to be roughly independent of temperature and uranium content, with a value of $18 \text{ W cm}^{-1} \text{ } ^\circ\text{C}^{-1}$ (dashed line A in Fig. 21). The cause of this difference is not understood, but the earlier data, determined by a heat-flow method, may be inaccurate owing to the diffusion of hydrogen during the test.

5.2. Specific heat

5.2.1. Dispersion fuels

The specific heat C_p of the fuel meat is obtained by summing the specific heats of the dispersed fuel phase and the aluminum matrix, each weighted by the respective weight fractions. Thus,

$$C_p = xC_{p\text{fuel}} + (1 - x)C_{pAl} ,$$

where x is the weight fraction of the dispersed fuel phase and $(1 - x)$ is the weight fraction of the aluminum matrix. The specific heats of aluminum and the dispersion fuels for the temperature range of 0-300°C are given by the following approximately linear equations:

$$C_{PA1} = 0.892 + 0.00046T \quad \text{J/gK,} \quad (\text{Ref. 85})$$

$$C_{PU} = 0.109 + 0.00013T \quad \text{J/gK,} \quad (\text{Ref. 85})$$

$$C_{PUAl_x} \approx 0.36 + 0.0003T \quad \text{J/gK,} \quad (\text{Calc. from Ref. 85})$$

$$C_{PU_3O_8} = 0.27 + 0.00030T \quad \text{J/gK,} \quad (\text{Ref. 86})$$

$$C_{PU_3Si} = 0.172 + 0.000013T \quad \text{J/gK,} \quad (\text{Ref. 86})$$

where T is temperature in °C.

5.2.2. UO_2

The specific heat of UO_2 up to temperatures of interest to research reactors (~1000°C) can be given by the following parabolic relationship:

$$C_{PUO_2} = 0.235 + 0.0088(T - 300)^{1/3} \quad \text{J/gK.} \quad (\text{Ref. 48})$$

5.2.3. $U-ZrH_x$

The specific heat of $U-ZrH_x$ can be calculated by summing the specific heats of the ZrH_x matrix and the dispersed uranium, as shown above for dispersion fuels.

$$C_{PZrH_{1.6}} = 0.363 + 0.00075T \quad \text{J/gK.} \quad (\text{Ref. 87})$$

5.3. Density

5.3.1. Dispersion fuels

The density, ρ , of the dispersion fuels can be directly calculated from the measured density of the fuel particle (f.p.) and the density of aluminum, 2.70 g/cm³, provided that the porosity, P , of the dispersion is known.

$$\rho = \frac{(1 - P)}{x_{f.p.}/\rho_{f.p.} + x_{al}/\rho_{al}} = \frac{(1 - P)}{x/\rho_{f.p.} + (1 - x)/2.70}$$

The uranium density can then be determined by multiplying the density of the dispersion fuel by the uranium weight fraction in the fuel core. The uranium density can also be determined directly from the fuel particle density by multiplying the latter by the volume fraction of fuel particles in the dispersion. In either case, the measured densities of the fuel particles are required. These are as follows:

<u>Dispersed Fuel Particle</u>	<u>Measured Uranium Density, g/cm³</u>
UAl _x	6.37
U ₃ O ₈	8.10
U ₃ Si ₂	11.9
U ₃ SiAl	14.2
U ₃ Si	15.2

5.3.2. UO₂

The theoretical density (TD) of UO₂ is given as 10.96 g/cm³. The measured density is given by TD(1 - P), where P is usually about 5%.

5.3.3. U-ZrH_x

The density of U-ZrH_x fuel is shown as a function of H/Zr ratio for five uranium contents in Fig. 22.[84] For the normal delta-phase material, the density can also be calculated from the dispersion fuel formula by substituting the weight fraction and density of ZrH_x for that of aluminum.

5.4. Thermal expansion

5.4.1. Dispersion fuels

The thermal expansion of aluminum dispersion fuels is essentially controlled by the thermal expansion of aluminum, as shown in Table 3.

5.4.2. UO₂

The thermal expansion of UO₂ is given in Table 3.

5.4.3. U-ZrH_x

The thermal expansion of U-ZrH_x fuel is controlled by that of the ZrH_x matrix. The expansion coefficient increases with increasing temperature. [87] The data for the delta and epsilon phases have been fitted to an equation of the form

$$\frac{\Delta l}{l} = -0.288 \times 10^3 + 6.24T + 7.34 \times 10^{-3}T^2,$$

where T is in °C and $\Delta l/l$ is the fractional change in length. The average expansion coefficient for the delta phase over the range of 200° to 850°C is $14.2 \times 10^{-6}/^\circ\text{C}$. Experimental data for ZrH_{1.7} over the range of 93° to 650°C are given below.

<u>Total Expansion of ZrH_{1.7} from Room Temperature, 10⁻⁶</u>				
<u>93°C</u>	<u>205°C</u>	<u>310°C</u>	<u>540°C</u>	<u>650°C</u>
3.3	11.7	35.9	51.1	68.8

Table 3. Thermal Expansion Coefficients of Various Solids

Temperature Range, °C	Coefficient of Thermal Expansion, $10^{-6}/^{\circ}\text{C}$			
	Pure Aluminum[88]	U ₃ Si[89]	UAl ₃ [90]	UO ₂ [90]
20-100	23.86	-	15.6	8.3
20-200	24.58	13.0	15.7	8.4
20-300	25.45	13.4	15.7	8.6
20-400	26.49	14.2	15.7	8.8
20-500	27.68	14.9	15.8	9.1

5.5. Corrosion

5.5.1. Water corrosion of aluminum alloy claddings

5.5.1.1. Introduction

Aluminum alloys have been used in water-cooled production, research, and test reactors since the beginning of the nuclear enterprise. Thus, there is a voluminous literature dealing with the aqueous corrosion of aluminum and its alloys under a variety of conditions. The purpose of this section is not to present a comprehensive review of this literature. Rather, a portion of the literature relevant to research reactors, i.e., that applicable to corrosion in recirculatory systems operating below 200°C and with provision for pH and impurity control, will be considered for the purposes of specifying operating parameters and estimating reasonable service lifetimes.

5.5.1.2 Corrosion behavior

The corrosion behavior of aluminum alloys is sensitive to temperature, pH, and flow rate. The behavior of alloy 1100 (99% min. Al) at temperatures up to 95°C is summarized in Fig. 23.[91] Figure 24 summarizes the corrosion behavior of the same alloy over a wider range of temperatures.[92] In evaluating the relevance of the data in Figs. 23 and 24 to particular cases, it should be noted that 1 mdd ($\text{mg}/\text{cm}^2/\text{day}$) is equivalent to 13.5 $\mu\text{m}/\text{yr}$ (0.53 mils/yr).

The data summarized in Figs. 23 and 24 were obtained in high-purity water (except for the acid and alkali used for pH control) and in the absence of reactor radiation. Chlorides and heavy metals, e.g., Ni, Cu, Co, Pb, Sn, and Hg, in concentrations above ~0.1 ppm (above 0.02 ppm for Cu), particularly in solutions of pH below 6, lead to severe pitting.[91,93] At temperatures up to ~150°C, water velocities up to 7.6 m/sec (25 ft/sec) do not adversely affect corrosion behavior.[92] At higher temperatures, the effects of flow rate and

surface/volume ratio are complex.[94] Corrosion rate increases slightly with temperature in the range of interest for research reactors, and also increases with flow rate in an unsaturated (with respect to corrosion product) system.[95] The corrosion rate of freshly exposed alloy is rapid, but it decreases parabolically to a lower constant rate as corrosion product builds up.[96] This corrosion product is a multi-layered aluminum oxide and there is evidence that dissolution of at least one of these layers can have a significant effect on the corrosion rate.[97-101] The dissolution rate is a function of at least three different variables (temperature, pH, and refreshment rate), but there is no generally accepted model of the detailed behavior.

Alloys with improved corrosion behavior at higher temperatures have been developed.[92,94,102] However, it is expected that the commonly used aluminum alloys, e.g., 6061 (0.25 Cu, 0.6 Si, 1.0 Mg, 0.25 Cr) and 5052 (2.5 Mg, 0.25 Cr) will behave similarly to 1100 and these improved alloys under research reactor conditions.[92] Although quantitative data are limited, there is evidence that reactor irradiation does not adversely affect the corrosion behavior of aluminum alloys.[102] For example, specimens of 6061 aluminum exposed in the core of the ORR at 57°C (135°F) for one year showed a very low corrosion rate of less than 1.3 $\mu\text{m}/\text{yr}$ (<0.1 mils/yr).[103]

5.5.1.3. Conclusions and recommendations

Based upon the data presented above, it is clear that research reactors can be operated in such a manner that corrosion of the aluminum cladding is not a practical limit on fuel element lifetime. The recommended conditions to achieve the desired long service life are pH between ~5 and ~6.7, preferably ~6, and chloride and heavy metal concentrations as low as can be achieved, but not more than ~0.02 ppm for Cu and below 0.1 ppm for the other ions. It is emphasized that these recommendations apply to periods of downtime as well as to those periods when the reactor is operating. The effect on uniform corrosion rate of periods of exposure at pH outside the recommended range can be estimated from the data in Figs. 23 and 24. Increased concentrations of chlorides and/or heavy metals will lead to severe pitting, including complete penetration of the cladding, in a relatively short time.

Also, care should be taken during storage in air to keep humidity or contamination (e.g., from fingerprints) from plate surfaces, particularly if the plates have not been cleaned and passivated.

5.5.2. Corrosion of fuel dispersions

The available information on the corrosion of aluminum dispersion fuels is sparse and consists mainly of boiling water testing of plates and rods with artificial defects. $\text{U}_3\text{O}_8\text{-Al}$ mini-plate specimens at 3.0 g U/cm^3 , clad with 0.38-mm (15-mil)-thick 6061 aluminum alloy containing a 3.2-mm (1/8-in.) drilled hole, were exposed for up to 168 h in boiling deionized water under reflux conditions.[104] No fuel loss or swelling was observed. However, some weight gain from the oxidation of the cladding was found. A similar test was performed with $\text{U}_3\text{Si-Al}$ and $\text{U}_3\text{SiAl-Al}$ mini-plates at ~4.8 g/ cm^3 , clad with 0.38 mm (15 mil) thick 6061 aluminum alloy. No fuel loss was detected, as determined by the lack of radioactivity in the water after the test. No swelling was detected and weight changes were insignificant.

Out-of-reactor defect tests were also carried out at CRNL with extruded rods of $U_3SiAl-Al$ and $U-Al$ alloy.[30] The tests were conducted at 150 and 180°C in static autoclaves. The defect size was 1 mm. The reaction with the $U-Al$ alloy was insignificant; however, some reaction with the silicide fuel occurred, as evidenced by internal pits. This reaction may be due to the lack of a bond at the fuel-cladding interface.

5.5.3. Corrosion and quench testing of $U-ZrH_x$ fuel

Hydride fuel has excellent corrosion resistance in water. Bare fuel specimens have been subjected to a pressurized water environment at 300°C (570°F) and 8.5 MPa (1230 psi) during a 400-h period in an autoclave.[87] The average rate of corrosion (weight gain) was 350 mg/cm²-month, accompanied by a conversion of the surface layer of the hydride to an adherent oxide film. The maximum extent of corrosion penetration after 400 h was less than 50 µm (2 mils).

In the early phases of development of the TRIGA fuel, water-quench tests were carried out from elevated temperatures.[87] Fuel rods 25 mm (1 in.) in diameter were heated to 800°C and end-quenched to test for thermal shock and corrosion resistance. No deleterious effects were observed. Later, these tests were extended to temperatures as high as 1200°C, in which tapered fuel rods were dropped into tapered aluminum cans in water. Although the samples cracked and lost hydrogen, no safety problem arose in these tests. Recently, the low-enriched TRIGA fuels have been subjected to water-quench safety tests at General Atomic Company at temperatures of 800 to 1200°C. Quench testing at 800 and 1000°C resulted in no unusual occurrences for hold times of up to 3 min at temperature. However, tests at 1100°C resulted in some localized melting of the fuel when the Inconel-sheathed thermocouple came into direct contact with it. One sample at 1200°C contacted and reacted with the tantalum susceptor used to heat the fuel. However, a second sample at 1200°C exhibited satisfactory behavior.

5.6. Phase stability

5.6.1. U_3O_8-Al thermite reaction

As noted previously, 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.[105] 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 MJ/kg fuel. This reaction was also studied by workers at ANL using various compositions, pretreatments, environments, and particle size distributions.[106,107] However, a slow heat-up rate (25°C/min) was used in this test, 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 that 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 reactor experiments on U_3O_8 -Al HFIR fuel plates were performed in the Transient Reactor Test Facility (TREAT) at ANL.[108] 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. Since earlier studies showed that the peak energy release occurs in the 55 to 75 wt% U_3O_8 range, which is the region of interest for the current developmental high-uranium-loaded fuel, an investigation was conducted of ignition temperature and maximum heat release for these compositions using differential thermal analysis and heating of miniature fuel plates.[109] The observed ignition temperature of around 900°C (1650°F) agreed well with previous studies. The energy releases, however, were much lower than predicted and in general are low compared with the energy required to initiate them. The heating of miniature fuel plates to as high as 1400°C (2550°F) produced no violent thermal effects, explosions, or observable gas release. Therefore, it is concluded from these tests that increasing the U_3O_8 loading does not add a significant chemical reaction or thermal hazard to the other considerations of safe reactor operation.

5.6.2. Stability of aluminide fuels

Studies on aluminides [110] 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.

5.6.3. Stability of silicide fuels

Few data are available from the literature. One set of experiments dealt with reactions of U_3Si , U_3Si_2 , UC, and UC_2 with aluminum at 620°C for up to 48 h, for comparison with the rapid reaction rate obtained with UO_2 . [110] No reaction occurred with UC_2 ; U_3Si , U_3Si_2 , and UC were found to react with aluminum, but at a slower rate than UO_2 .

At ANL, aluminum-dispersion samples of U_3Si , U_3SiAl and U_3Si_2 were prepared and annealed at 300° and 450°C for up to 1000 h. The samples were prepared and roll bonded into a single 6061 aluminum alloy cover/frame/cover assembly. After hot rolling at 500°C and cold rolling to a final thickness of 1.27 mm, the resultant plates were sectioned to provide separate specimens for testing. The fuel zones were oval in shape, with major and minor axis dimensions of 75 and 10 mm. Thickness measurements in the fuel zones were determined periodically. After 1000 h at 300°C , no change was observed for U_3Si_2 , and only about a 20- μm thickness increase was observed for the U_3Si and U_3SiAl fuels. At 450°C , swelling was more pronounced for all fuels. There appears to be a shallow plateau after 500 h at about 350 μm for the U_3Si and U_3SiAl fuel and about 150 μm for the U_3Si_2 fuel. Between 700 and 1000 h, rapid swelling occurs which may be due to entrapped gases and cladding creep.

After the 450°C test, the specimens were sectioned and the reaction products were analyzed by the standard x-ray (Debye-Scherrer) method. The only reaction product evident was UAl_3 with some silicon substituted for aluminum. (Diffraction lines for Al were also present in the U_3Si and U_3SiAl samples.)

This result is consistent with phase equilibria work just completed at ANL.[111] The U-Al-Si system was studied in the composition range U- UAl_4 - USi_3 . More than 260 binary and ternary alloys were investigated by metallography and x-ray powder diffraction methods. The 900°C isothermal section is shown in Fig. 25. One, or possibly two ternary compounds exist at or near the U_2AlSi_2 composition. The crystal structure has not been determined.

The binary silicides U_3Si , U_3Si_2 , and USi can hold very small amounts of Al in solid solution, but the higher silicides U_3Si_5 , USi_{2-x} , and USi_3 dissolve increasingly larger amounts of Al. UAl_2 can hold more than 12 at.% Si in solid solution. UAl_3 forms a continuous solid solution with USi_3 at 900°C and above. UAl_4 disappears with less than 3 at.% Si substitution.

U_3Si exhibits considerable ductility, which can be enhanced by partial substitution of Ge or Ga for Si. All other intermetallic compounds in the system are brittle.

5.7. Mechanical properties

5.7.1. Dispersion fuels

Buckling due to axial compressive loads developed from either thermal stresses or irradiation growth stresses has not been detected in ATR fuel elements or plates.[8] In ETR fuel elements in which flat fuel plates were pinned instead of roll swaged to side plates so that axial slippage was restricted, some buckling was observed.

It is standard operating practice to require that the calculated thermal stress, for the operating conditions in the center half-span of the fuel plates, be at some margin below the unirradiated and irradiated yield strength of the fuel plate composite throughout the cycle. Some typical limiting strength values are given in Table 4. The compressive yield strength values from the composite plates with thinner cladding and the fuel cores with higher wt% UAl_3 tend to be higher. The initial effect of irradiation is to reduce the strength of the composite cold-worked plates (Type 1100-H12), and increase the strength of the Type 6061-0 plates. The limiting strength values should be conservative since they include effects of irradiation, thick annealed cladding, and a fuel core with low UAl_3 content.

Table 4. Typical Values Selected for Limiting Thermal Stress^a

Material	Temperature K (°F)	Limiting Strength (MPa)	Measured Compressive Yield Strength, MPa	
			Unirradiated	Irradiated ^b
ATR type (6061-0 clad)				
100-mil plate, 35 wt% UAl ₃	367(200)	96	98.5	---
	422(300)	90	91.0	---
	478(400)	79	81.1	---
	533(500)	44	44.8	---
50-mil plate, 51 wt% UAl ₃	478(400)	---	---	120.6
	533(500)	47	47.3	82.7
	589(600)	---	---	68.9
ETR type (1100-H12 clad)				
50-mil plate, 35 wt% UAl _x	422(300)	48	107	48.3
	478(400)	48	83	48.3
	533(500)	42	61	42.0

^aModified from Ref. 8 with permission.^bATR plates irradiated to 3-14x10²⁰ fiss/cm³; ETR plates to 2-14x10²⁰ fiss/cm³.

The tensile properties of U₃O₈-Al dispersions were recently determined at IPEN, Sao Paulo, Brazil.[112] The concentration of U₃O₈ was varied from 0 to 55 wt%. The dispersion densities varied from 2.696 (for pure Al-1100 powder) to 3.945 (at 55 wt% U₃O₈). The maximum uranium density was 1.84 g/cm³. The yield and ultimate strength as well as ductility decreased with increasing U₃O₈ content, as shown in Figs. 26 and 27.[112] The ultimate strength of the material was related to the volume fraction of the ceramic phase according to the following relationship:

$$\sigma = \sigma_0 (1 - KV^{2/3}),$$

where

- σ = ultimate tensile strength of the cermet,
- σ_0 = ultimate tensile strength of the aluminum matrix,
- K = constant = 1.21,
- V = volume fraction of U₃O₈.

Hence, the ceramic bond to the matrix is weak or non-existent and strength is a function of the area of the matrix only. It is evident from the data on yield strength of aluminide fuels (Table 4) that the aluminide increases the yield strength of the fuel. This is likely to be the case for silicide fuel as well.

5.7.2. UO_2

The data on the mechanical properties of UO_2 are voluminous, [35,47,48] and only selected data are included here. The effect of fuel density and temperature on Young's modulus is shown in Fig. 28.[48] The fracture strength as a function of temperature is shown in Fig. 29;[48] the large scatter is attributed to differences in pore morphology.

The creep of UO_2 has been measured in both out-of-reactor and in-reactor tests. The creep rate was found to be proportional to both the applied stress and the fission rate. Figure 30 [35] summarizes the data of various investigators, normalized to standard stress and radiation conditions. The effect of radiation on the creep rate is twofold. At high temperatures the creep rate is greater than that observed in the absence of radiation but exhibits the same temperature dependence. This phenomenon is termed fission-enhanced creep and its effect on high-temperature low-stress creep is to increase the creep rate by approximately a factor of 4. At low temperatures, where thermal creep is negligible, a temperature-independent (or "athermal") component of creep is observed. This form of creep, which is due solely to the radiation field, is called fission-induced creep. Fission-induced creep is represented by the broad band at low temperature in Fig. 30.

5.7.3. U-ZrH_x

The mechanical properties of ZrH_x are difficult to measure because of its brittle nature. However, at elevated temperatures ZrH_x exhibits significant ductility and creep deformation. The creep strength is markedly influenced by the structure, as shown in Fig. 31.[33] The beta phase has a much lower creep strength than the delta phase. The dynamic elastic modulus of $\text{ZrH}_{1.6}$ was measured at room temperature as 63,000 MPa. This value decreases to about 41,000 MPa at 650°C.

The tensile strength and elongation of zirconium hydride as a function of at.% H are shown in Fig. 32.[3] The presence of minor constituents such as carbon and oxygen also has a marked effect on the mechanical properties.

5.8. Fission product release

5.8.1. Dispersion fuels

5.8.1.1. Introduction

Information on the release of fission products from plate-type fuels is relatively scarce. Many of the early data are for uranium metal or UO_2 fuels;[113,114] however, some data are available for uranium-aluminum alloy fuels and UAl_x dispersion fuels. Data on U_3O_8 dispersion-type fuels are limited. As discussed in the following paragraphs, the available out-of-reactor data for all types of plate-type aluminum-based fuel show a consistent pattern, namely that significant amounts of fission products are not released below the melting point of the aluminum alloy cladding. Once the melting point is exceeded by 100°C, however, the release of the volatile fission products (noble gases and iodines) is largely complete.

Available data from reactor accidents indicate that, in cases where water coolant is present in the core when fuel melts, only the noble gases are released in any quantity. The iodines and less volatile fission products are effectively retained by the water.

5.8.1.2. Releases during controlled out-of-reactor experiments

Workers at ORNL have studied fission-product release in U-Al alloy plate fuels.[115,116,117] Punched segments of plates which had undergone a trace (~60-h) irradiation were heated by induction within quartz tubes under a flowing air atmosphere. These segments were held at 700 and 800°C for one hour, but the hold time was later found to be insignificant and was reduced to two minutes at temperature. For the initial trace-irradiation experiments, the fractional release of iodine and noble gases was insignificant (<1%) at 700°C; 4.8 and 1.8%, respectively, at 800°C; and essentially complete above 900°C. The releases of cesium and tellurium were small (~6%) at a temperature of 900°C and increased slowly with increasing temperature. The release of ruthenium was small (<1%) over the entire temperature range (700-1085°C) tested. Further experiments showed that release data for fuel plates of intermediate (3.2 and 9.0%) burnup were similar to those for fuel at 23.6% burnup, indicating the presence of a saturation effect at a relatively low burnup level. High flow rate or the mixing of steam with the air had no significant effect on release fractions. No data were taken at temperatures below 700°C, where significant release of the noble gases and moderate releases of iodine and cesium would be expected.

An irradiation program was carried out in the ETR and MTR on 1.27-cm-diameter high-density pellets of UAl_3 , UAl_4 , and 20 wt% U-Al fuel.[118] After irradiation to a burnup of 18-35% of the ^{235}U , the pellets were subjected to slow heating to permit the collection of fission products and the monitoring of noble gas release by counting of ^{85}Kr . No activity release was found below 600°C. The alloy fuel released most of its fission gas at about the eutectic temperature of 640°C. The UAl_4 released a large fraction of its fission gas at about 730°C, the UAl_4 peritectic temperature, with small releases at higher temperatures. For UAl_3 , some fission gas was released at 1280°C, but the major release occurred at about 1350°C, the UAl_3 peritectic temperature. Similar results were obtained for these fuels at a burnup of ~60% of the ^{235}U . [119] In addition, it was noted that the major release of cesium occurred at 730°C for UAl_4 and 1350°C for UAl_3 . Smaller releases occurred at temperatures slightly above and below these peritectic temperatures. The rate of release of cesium was slower than that of the noble gases for UAl_4 and the U-Al alloy, but was as rapid as the noble gases for the UAl_3 . [118] These releases, however, are indicative of the bulk fuel and hence are meaningful for the alloy only. For a dispersion fuel the largest release comes from the recoil matrix region surrounding the particles and not from the particles themselves. [119] Additionally, post irradiation microprobe studies show that the structural character of the particle greatly affects retention. [120,121] Indications are that the defect structure of UAl_4 retains more gas than that of UAl_3 or U_3O_8 .

One study was found in the literature [122] which compares the fission-gas release from two plates of UAl_3 -Al fuel (~1.3-1.4 g U/cm³) and one plate of U_3O_8 -Al fuel (~1.2 g U/cm³). The burnup was 18-26% of the ^{235}U . The

irradiation plates (with meat dimensions of $0.25 \times 25 \times 140$ mm) were heated under vacuum in Vycor tubes at 600, 630, then 675°C, with a 30-minute hold at each temperature. The temperature at which fission gases began to evolve was in a narrow range between 575 and 600°C for both types of plates. No fission gases were detected after heating of the plates at 500°C for 40 minutes. Since the solidus temperature for type 6061 aluminum cladding is 582°C, it was inferred that fission-gas release began at this temperature. The bulk of the fission gas was released by 630°C, with the UAl_3 fuel releasing gas at a faster rate and at a lower temperature than the U_3O_8 fuel. Fission-product cesium was released from the fuels, but was deposited in copper tubing upstream from the gas collection reservoir. No cesium was found in the gas analysis.

A summary of some of the above data is presented in Fig. 33. The conclusion is that no fission products are released below the melting (eutectic) point of the cladding (582°C) and that essentially all of the fission gas is released by 675°C. As shown in the figure, fission products such as iodine are released more slowly. Recently, sensitive tests performed at ORNL have indicated that some fission gases are released from aluminide fuels just below the melting point of the cladding. However, photographic evidence indicated that local melting of the blistered surfaces might have occurred. The analysis of these results is in process.

5.8.1.3. Releases during reactor accidents

The release of fission products during reactor accidents has also been examined. Several accidents involved U-Al fuel which had undergone melting owing to flow blockages caused by extraneous material. Partial melting of one subassembly occurred in each case and the fission products, except for the noble gases, were largely contained within the coolant water. Similar results were obtained as a result of a 30-MJ excursion of the ISIS reactor.[123] Releases into the water and air were 0.06% and 0.001%, respectively, each significantly less than 1 Ci. Controlled overpower excursion tests at CABRI using a special control drive assembly produced significant (70%) melting of the control fuel plate with the release of 100% of the noble gas and <10% release of iodine and tellurium from the molten fuel.[124] However, both these elements were retained in the water and only between 0.0005 and 0.005% was released to the reactor room.

More extensive damage and somewhat greater releases occurred in the SPERT-1 destructive test and the SL-1 accident.[125,126] The nuclear energies and peak powers generated were 31 and 130 MJ, and 2.3×10^2 and 1.9×10^4 MW, respectively. In the SPERT-1 test, all the fuel plates underwent at least partial melting; greater than 50% of the plates melted completely. Fission-product release, mainly due to fission gases, was estimated to be about 0.7% of the total inventory. Iodine release to the atmosphere was estimated to be less than 0.01%. The reactor building was only moderately contaminated and re-entry occurred about four hours after the test. As a result of the SL-1 accident, the building was badly contaminated, with an extensive low-level deposition of ^{131}I . Total fission product release was estimated to be 5-10% of the total inventory, and release to the environment was estimated to be only about 0.01%. The iodine release to the environment was estimated to be less than 0.5%.

5.8.2. UO₂

The fission-gas and fission-product release behavior of UO₂ is similar to that exhibited by dispersion fuels, but release begins at higher temperatures for UO₂ [35], as seen in Figs. 34 and 35.[35] Figure 34 shows fractional gas release from both the matrix (dashed curves) and traps (solid curves) as a function of temperature. For long time steps (2016 h), the matrix release fraction is less than 0.011 for temperatures below 1600 K and approaches 1.0 at temperatures above 2000 K. For time steps less than 200 h, the matrix release fraction approaches 1.0 at ~2500 K. The release of fission gas from traps shows similar behavior; however, at temperatures in the range of 1700 to 2100 K, the trap release fraction is larger than the matrix release fraction.

5.8.3. U-ZrH_x

A number of experiments have been performed to measure the retention of fission products by UZrH (TRIGA) fuel. Experiments on fuel with a uranium density of 0.5 g/cm³ (8.5 wt% U) were conducted over a period of 11 years and under a variety of conditions. Results prove that only a small fraction of the fission products are released, even in completely unclad UZrH fuel. The release fraction varies from 1.5×10^{-5} for an irradiation temperature of 350°C to $\sim 10^{-2}$ at 800°C.[127]

The experiments show that two mechanisms are involved in the release of fission products from UZrH fuel, each of which predominates over a different temperature range. The first mechanism is that of fission fragment recoil into the gap between the fuel and clad. This effect predominates in fuel at temperatures up to ~400°C; the recoil release rate is dependent on the fuel surface-to-volume ratio but is independent of fuel temperature. Above ~400°C, the controlling mechanism for fission product release from UZrH fuel is a diffusion-like process, and the amount released is dependent on the fuel temperature, the fuel surface-to-volume ratio, the time of irradiation, and the isotope half-life.

The results of the UZrH experiments, and measurements by others of fission product release from SNAP fuel, have been compared and found to be in good agreement.

The fractional release, ϕ , of fission-product gases into the gap between fuel and clad from a full-size standard UZrH fuel element is given by

$$\phi = 1.5 \times 10^{-5} + 3.6 \times 10^3 e^{-1.34 \times 10^4 / (T + 273)},$$

where T = fuel temperature, °C. The first term of this function is a constant for low-temperature release; the second term is the high-temperature portion. Figure 36 [127] shows a theoretical plot of fission-product release for a fuel element that has been irradiated for a sufficiently long time that all fission-product activity is at equilibrium. The measured release fractions in Fig. 36 have all been normalized to a standard ratio of fuel-clad gap to fuel diameter, although individual measurements were made with different geometries. The measured values of fractional release fall well below the curve. Therefore, for safety considerations, this curve gives conservative values for the high-temperature release of fission products from UZrH fuel.

The results of recent studies [128] in the TRIGA reactor at General Atomic Company on fission-product release from fuel elements with high uranium loadings (up to 3.7 g U/cm^3 or 45 wt% U) agree well with the data just presented above.

VI. CONCLUSIONS

A review of past and current data on nuclear fuels for steady-state thermal research and test reactors has been provided. The major accomplishments noted were the extensions in loadings of existing fuels and the development of uranium oxide caramel fuel and uranium silicide dispersion fuels. All fuels are undergoing extensive irradiation screening as short plates or partial elements, with tests of full-sized elements in progress or to follow. Some blistering in-reactor and during post-irradiation blister testing has been noted for some mini-plates containing highly loaded fuels at a burnup of $\sim 2 \times 10^{21} \text{ fissions/cm}^3$. However these blisters occurred during the screening irradiations in which cladding restraint was at a minimum. No blisters have been found in these fuels during testing of prototypic full-sized plates or elements. It is likely that these fuels will enable the reduction of uranium enrichment in research and test reactors, thereby meeting the objectives of the RERTR program.

In addition, the encouraging early results of dissolution studies on dispersion-type LEU fuels suggest that reprocessing of these more highly loaded fuels will not be a problem.

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Fig. 1

Schematic of plate-type fuel element from Oak Ridge Reactor. Modified from IAEA Data Bank.

Fig. 2. Dimensional specifications for mini-plates to be used in the Oak Ridge Reactor irradiation test assembly. (Conversion factor: 1 in. = 25.4 mm.)

Fig. 3. Microstructure of clad dispersion of 75 wt % U_3O_8 -Al. (a) Cross section; (b) longitudinal section. Taken from Ref. 20.

Fig. 4
Void content of U_3O_8 -Al cores as a function of U_3O_8 concentration and core thickness. Taken from Ref. 20.

Fig. 5
Uranium density in fuel meat versus
volume fraction of dispersed phase
for plate-type fuels.

Fig. 6
The U-Si system.

20 μm

(a)

(b)

Fig. 7. Microstructures of U_3Si (U + 4 wt % Si).
(a) As cast; (b) heat treated (72 h at 800°C).

20 μm

(a)

(b)

Fig. 8. Microstructures of U_3Si_2 (U + 7.4 wt % Si).
(a) As cast; (b) heat treated (72 h at 800°C).

20 μm

(a)

(b)

Fig. 9. Microstructures of U_3SiAl (U + 3.5 wt % Si + 1.5 wt % Al).
(a) As cast; (b) heat treated (72 h at 800°C).

I
200 μm

(a)

(b)

Fig. 10. (a) U_3Si (U + 4 wt % Si) powder, -100 +325 mesh;
(b) Alcan MD-101 Al powder, -100

Fig. 11. Mini-plate hardware before assembly (bottom left) and after rolling (top), and finished mini-plate (bottom right).

Fig. 12

Representative cross sections of silicide mini-plates. (a) Longitudinal mini-plate section (30 vol.% U_3Si) near trailing edge; (b) transverse mini-plate section (30 vol.% U_3Si); (c) transverse mini-plate section (30 vol.% U_3SiAl).

Fig. 13. Polished sections of -100 mesh $USiAl$ powder produced by machining bar stock. U_3Si_2 particles (white) and UAl_2 particles (dark) are visible in the U_3Si matrix. Taken from Ref. 70 with permission.

Al-55 wt % USiAl

Al-75 wt % USiAl

Fig. 14. Longitudinal sections of extruded USiAl rods.
Taken from Ref. 30 with permission.

Fig. 15

Thermal conductivity of 95% theoretically dense USiAl-Al cermet vs USiAl composition, calculated using Maxwell's equation (100-200°C). Taken from Ref. 30 with permission.

Fig. 16

Caramel fuel assembly and its components.
Taken from Ref. 74.

Fig. 17

Thermal conductivity of U-Al alloy at 65°C.

Fig. 18

Thermal conductivity versus temperature for uranium-aluminum alloy fuels with various uranium loadings. Open symbols denote results obtained in three different runs; solid symbols denote results for pure aluminum

Fig. 19

Thermal conductivity of U_3O_8 -Al dispersions.

Fig. 20

Model prediction for thermal conductivity of 0.98% TD UO_2 compared with data from specimens with densities in the range of 0.975 to 0.985% TD. Taken from Ref. 48.

Fig. 21

Thermal conductivity of $U-ZrH_x$ as a function of temperature. Curves A-D show heat-flow data for fuels with $x = 1.58, 1.65, 1.7$, and 1.8 , respectively. Lines UL, LL, and MP indicate the upper limit, lower limit, and most probable value, respectively, for the thermal conductivity of fuel with $x = 1.6$. Line A shows pulse-heating data for fuel with $x = 1.6$, which do not agree with curve MP. Taken from Ref. 84 with permission.

Fig. 22

Density diagram for $U-ZrH_x$ fuel. Taken from Ref. 84 with permission.

Fig. 23

Effect of pH and other variables on the aqueous corrosion of 1100 aluminum. Taken from Ref. 91.

Fig. 24
Aqueous corrosion of 1100 aluminum. Taken from Ref. 92.

Fig. 25. The U-Al-Si system at 900°C. Taken from Ref. 111.

Fig. 26

Stress-strain curves for U_3O_8 -Al dispersions at ambient temperature. Taken from Ref. 112 with permission.

Fig. 27

Elongation at failure vs vol.% U_3O_8 -Al in U_3O_8 -Al dispersions. Taken from Ref. 112 with permission.

Fig. 28

Young's modulus for stoichiometric UO_2 fuel as a function of temperature and density. Taken from Ref. 48.

Fig. 29

Out-of-reactor fracture strength data for UO_2 (normalized to $10\text{-}\mu\text{m}$ grain size and 95% TD). Taken from Ref. 48.

Fig. 30

Composite in-reactor creep of UO_2 (normalized to $\sigma = 2.4 \times 10^4 \text{ kN/m}^2$ and $F = 1.2 \times 10^{13} \text{ fissions cm}^{-3} \text{ sec}^{-1}$). Taken from Ref. 35.

Fig. 31
Creep properties of β -, σ -, and $(\sigma + \epsilon)$ -phase zirconium hydride. Taken from Ref. 33 with permission.

Fig. 32
Ultimate tensile strength and elongation of zirconium hydride at 600°C, as a function of atom percent H. Taken from Ref. 33 with permission.

Fig. 33
Fission-product release from plate-type fuels (burnup = 20-25% of ^{235}U).

Fig. 34

Fission-gas release from UO_2 as a function of temperature. Taken from Ref. 48.

Fig. 35

Calculated release of iodine and cesium from UO_2 as a function of temperature. Taken from Ref. 48.

Fig. 36

Release of gaseous fission products from $UZrH$ fuel (experimental values above $400^\circ C$ corrected to infinite irradiation). Taken from Ref. 127 with permission.

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