QUALIFICATION STATUS OF LEU FUELS

J. L. Snelgrove
Argonne National Laboratory
9700 S. Cass Avenue
Argonne, IL 60439 USA

ABSTRACT

Sufficient data has been obtained from tests of high-density, low-enriched fuels for research and test reactors to declare them qualified for use. These fuels include $UZrH_x$ (TRIGA fuel) and UO_2 (SPERT fuel) for rod-type reactors and UAl_x, U_3O_8, U_3Si_2, and U_3Si dispersed in aluminum for plate-type reactors. Except for U_3Si, the allowable fission density for LEU applications is limited only by the available ^{235}U. Several reactors are now using these fuels, and additional conversions are in progress.

The basic performance characteristics and limits, if any, of the qualified low-enriched (and medium-enriched) fuels are discussed. Continuing and planned work to qualify additional fuels is also discussed.

INTRODUCTION

The U.S. Reduced Enrichment Research and Test Reactor (RERTR) Program has been developing, testing, and demonstrating reduced-enrichment fuels for the past nine years. Our goal has been to qualify the highest density fuels possible in order to make feasible the conversion of the maximum number of reactors to the use of low-enriched uranium (LEU) fuels. We consider a fuel qualified when we believe that sufficient data exists for regulatory authorities to approve the use of the fuel in the reactors they regulate.

Many organizations, listed in Table 1, have participated in the fuel development and testing efforts of the RERTR Program. A number of these--B&W, CERCA, CNEA, NUKEM, CEA, JRC/ECN, Studsvik, and, informally thus far, CEN/SCK--have entered into cooperative agreements with the RERTR Program to provide fabrication, irradiation, and/or postirradiation examination services without cost to the Program. The U.S. Government has provided enriched uranium and will provide for disposition of the spent fuel. Our partners have greatly contributed to the achievements discussed in this paper.

The RERTR Program has concentrated on the development of plate-type fuels since they account for most of the HEU consumption. We have, however, contributed to the qualification of two fuels for use in rods or pins. These will be discussed briefly before proceeding with a more extensive discussion of plate-type fuels. Although we have been primarily concerned with LEU fuels, i.e., with uranium enrichments less

Table 1. Fuel Development Participants

Devel./Fab.	Irrad./PIE
ANL	ANL
B&W	CEA (SILOE, OSIRIS)
CERCA	CEN/SCK (BR2)*
CNEA	JRC/ECN (HFR-Petten)
GA	ORNL (ORR)
INEL (EG&G)	Studsvik (R2)
NUKEM	
ORNL	
TI	

*Irradiation contract not yet negotiated.

than 20%, qualification of some fuels with medium-enriched (40- to 45%-enriched) uranium (MEU) is included in the work of the Program in case adequate LEU fuels cannot be developed for all reactors. Notwithstanding the title of this paper, these MEU fuels will briefly be discussed.

ROD- OR PIN-TYPE FUELS

$UZrH_x$ (TRIGA) Fuel

At about the time the RERTR Program was being formed, GA began the development of $UZrH_x$ fuels with uranium densities significantly in excess of those in use at that time. The RERTR Program provided the long-term, high-burnup irradiation tests of LEU TRIGA fuels with uranium densities of 1.3, 2.2, and 3.7 Mg/m^3 (20, 30, and 45 wt% U, respectively). The pins were irradiated in the ORR to peak burnups as high as 80% of the originally contained ^{235}U. The fuel behavior was excellent and in agreement with predictions.[1] The NRC has recently granted generic approval for use of the 20- and 30-wt% fuels as a replacement for the 8.5-wt% fuel currently used in most TRIGA reactors.[2] Specifically, operation of a whole core of the fuel has been licensed for GA's TRIGA Mark F. Other potential users must provide analyses covering their specific conditions of operation, but they will not have to address the behavior of the fuel. It should be pointed out that a whole core of the 20-wt% fuel is already in operation in Bangladesh and will soon be in operation in the Philippines.[3]

UO_2 (SPERT) Fuel

Approximately 9000 stainless steel-clad, 4.81%-enriched, UO_2 fuel pins were produced in the mid-1960s for use in the Special Power Excursion Reactor Test (SPERT) Program. Although the fuel pins had been saved, the fabrication inspection and certification records had been destroyed. Rennselaer Polytechnic Institute (RPI) chose to use these pins for the conversion of its Critical Facility,[4] and two other universities have been considering using them. In all applications being considered, burnup would be so low that irradiation testing was not needed. However, in order to confirm that the as-fabricated attributes of the pins remained within the range of the fabrication specifications and that no deterioration of the pins had occurred, the RERTR Program performed a series of requalification inspections.[5] The pins were successfully requalified, and the NRC granted generic approval for their use in non-power reactors, subject to possible constraints in specific situations.[6] The RPI Critical Facility has now been licensed to operate with this fuel.

PLATE-TYPE FUELS

General Remarks

The RERTR Program chose to concentrate its efforts on further development of aluminum-matrix dispersion fuels in order to take advantage of the large commercial base of equipment for and experience in fabrication of such fuels. Both UAl_x and U_3O_8 were being used as dispersants in HEU fuels at the beginning of the RERTR Program, with maximum uranium densities of 1.7 and 1.3 Mg/m^3, respectively. Both of these fuels have been developed and qualified to their practical fabrication limits. The much-higher-density fuels needed have been provided through the use of uranium-silicon alloys. One of these, U_3Si_2, has proven to be a very stable and attractive fuel.

The first irradiation tests of the higher-density fuels were performed using miniature fuel plates (miniplates) in order to screen the candidate fuels and provide basic irradiation behavior data. Approximately 240 miniplates have been irradiated and examined thus far. The acceptable performance of successful candidate fuels under typical operating conditions has been confirmed by the testing of full-sized fuel elements in the reactors listed in Table 1. Thus far, 39 test elements have been irradiated or are scheduled for irradiation. In most cases some elements of each type were irradiated to burnups well in excess of those typical of test reactors. Average burnups of from 70 to 82% have been achieved, with peak burnups ranging up to 98%. Extensive PIEs have confirmed the expected behavior of all fuels. In two whole-core demonstrations the good behavior to normal burnup of statistically significant numbers of commercially fabricated elements has been demonstrated. In these demonstrations 130 elements have been irradiated to at least partial burnup. As evidence of the acceptance of the fuel performance data generated by the RERTR Program, approximately 750 more reduced-enrichment fuel elements have been ordered on a commercial basis by reactor operators--for testing, for conversion, or for fueling of new reactors.

To provide the data needed for qualification, the fuels were extensively characterized both before and after irradiation. The fuel compounds; the fuel meat porosity, heat capacity, and thermal conductivity; the compatibility of the fuel with the matrix and cladding; the corrosion behavior of the fuel; and any exothermic reaction between the fuel and aluminum were studied prior to irradiation. Following irradiation the primary attributes studied have been the volumetric swelling of the fuel meat, the microstructural behavior of the fuel, and the blister threshold temperature. The swelling and microstructure studies have resulted in a good understanding of the fundamental irradiation behavior of dispersion fuels. The basic fuel swelling mechanisms of dispersions of intermetallic fuels and U_3O_8 in aluminum are discussed elsewhere.[7,8] In this paper I will summarize the swelling data for the dispersion fuels tested by the RERTR Program.

What is usually of most interest to the reactor operator is the net swelling of the fuel meat or fuel plate. However, it is the swelling of the fuel particles which I wish to discuss first. The fuel meat volume of an unirradiated fuel plate is occupied by the fuel particles, the aluminum matrix, and pores created when matrix aluminum does not flow around all fuel particles and into the cracks produced in fuel particles during the rolling of the plate. Such pores can occupy from approximately 4% to greater than 12% of the volume of high-density fuel meats. During the early stages of irradiation, irradiation-induced sintering results in some consolidation of the pores, and the volume of the fuel meat can actually decrease. As the fuel particles begin to swell from the buildup of solid and gaseous fission products, the pores begin to be filled. The fuel meat exhibits a net positive volume change only after the volume of the fuel particles has increased by approximately the amount of the original pore volume. Since the amount of as-fabricated porosity is influenced by many factors, the effect of the porosity must be removed to ascertain the swelling behavior of the fuel itself.

The swelling of the fuel particle as a function of the fission density in the particle is shown in Fig. 1 for UAl_x[9] and three of the uranium silicide compounds. The swelling of the fuel meat was measured by an immersion technique. The original volume of as-fabricated porosity was added to the volume change of the meat, the sum was divided by the original volume of fuel particles in the meat, and the result was converted to percent to obtain the fuel particle swelling. The fission density in the meat, based on the measured burnup, was divided by the original fuel volume fraction to obtain the fission density in the fuel particle. It is seen that UAl_x, USi, and U_3Si_2 fuel particles swell linearly to fission densities well beyond those achievable with LEU (indicated by the ends of the solid parts of the curves). The data are scattered in narrow bands about the USi and U_3Si_2 curves. The scatter is considerably greater for UAl_x, owing at least partially to the fact that UAl_2 and UAl_3 react with Al to form UAl_4 during fabrication, obscuring the original fuel particle and porosity volumes. Within the accuracy of the data, the swelling of UAl_x and U_3Si_2 fuel particles per unit fission density are equal.

U_3Si exhibits an entirely different behavior. As has been described previously,[10] the U_3Si particles in very highly loaded fuel plates tend to swell rapidly (breakaway swelling) at high fission densities under the influence of fission gas pressure. The curve shown represents the upper limit of U_3Si fuel particle swelling. In lower-loaded fuel plates, where the larger amounts of matrix aluminum restrain the swelling fuel particles and prevent interparticle linkage of fission gas bubbles, the fuel particle swelling is considerably less. The data for such plates lie to the right of the U_3Si curve. It is obvious that a fission density limit is required to maintain an adequate margin to breakaway swelling. At the highest loadings, represented by the curve, a limit of 4.0×10^{27} f/m^3 (1.8×10^{27} f/m^3 in the fuel meat for a 45 vol% fuel loading) will provide more than a 25% margin. This limit corresponds to a peak burnup of 60% of the originally contained ^{235}U. For lower loadings the particle fission density limit will be higher. For example, 2.0 Mg U/m^3 miniplates performed acceptably at 12.6×10^{27} f/m^3 in the U_3Si particles. It is interesting to note that this corresponds to 1.7×10^{27} f/m^3 in the meat, so a limit of approximately 1.8×10^{27} f/m^3 in the meat may be generally applicable to U_3Si dispersion fuel.

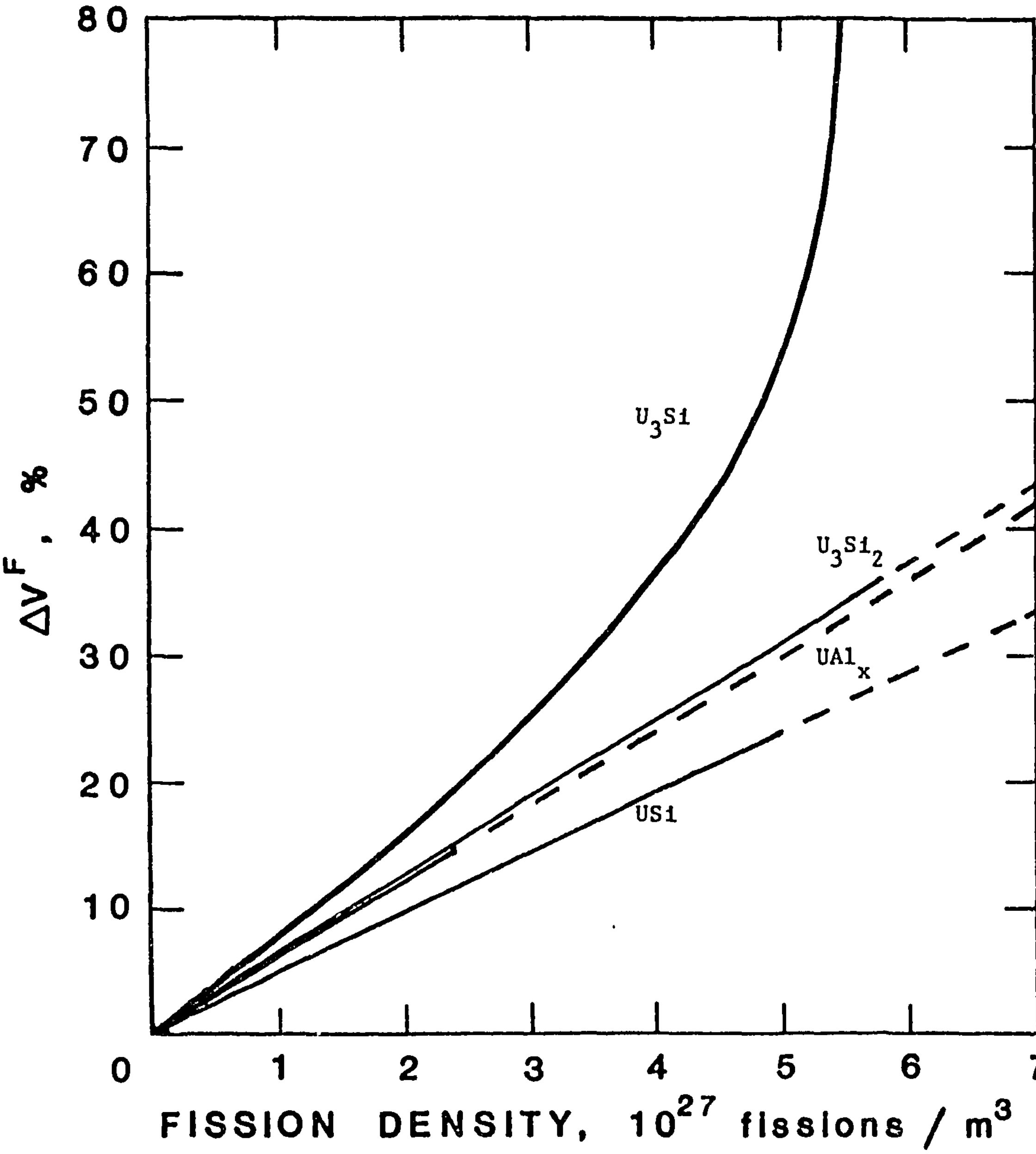

Fig. 1. Swelling of Uranium Silicide and UAl_x Fuel Particles vs. Fission Density in the Particle. Dashed Lines Indicate Fission Densities Not Attainable in LEU Fuel.

Although the swelling mechanism of U_3O_8 dispersion fuel differs somewhat from that of U_3Si dispersion fuel, the data are very similar. The same curve represents both the U_3Si and U_3O_8 swelling, except that, because of its much lower density, a fission density of 3.6×10^{27} f/m^3 cannot be exceeded in low-enriched U_3O_8. This fission density is low enough that no additional limit need be applied for low-enriched U_3O_8 dispersion fuel.

Qualified Dispersion Fuels

The limits for LEU plate-type dispersion fuels considered qualified by the RERTR Program are listed in Table 2. Full-sized elements loaded to the densities listed have been tested. These densities, incidentally, represent the practical limits for commercial fabrication (50 vol% for UAl_x, 45 vol% for U_3O_8, and 43 vol% for U_3Si_2). It should be noted that the actual densities of the nominally 4.8-Mg U/m^3 U_3Si_2 elements ranged from 4.6 to 5.2 Mg U/m^3, depending principally on the amount of porosity in the meat. As discussed in the previous section, the linear swelling behavior of UAl_x and U_3Si_2 and the relatively low uranium density of U_3O_8 obviate the need for a fission density limit for LEU applications. The expected fuel meat swelling, which can be used to calculate the plate thickness change, is given as a range to account for uncertainties in the fuel particle swelling data and variations in as-fabricated porosity. The swelling indicated for each fuel is acceptable. Blister threshold temperatures are acceptably high, being comparable to those measured for the currently used highly enriched uranium (HEU) fuels. Our tests have shown that full-sized fuel plates tend to blister at somewhat higher temperatures than do miniplates. (The 500°C temperature for U_3Si was measured for miniplates.) We have found that the blister threshold temperature is quite insensitive to both fuel loading and burnup. Because the first release of fission gas occurs during blistering,[11] we can also state that the threshold temperatures for release of fission gas from the high-density fuels are comparable to those of the currently used dispersion fuels.

Table 2. Limits for Qualified LEU Fuels

Fuel Type	Maximum Density, Mg U/m^3	Fission Density Limit,* 10^{27}/m^3	Meat Swell.,† %	Blister Temp., °C
UAl_x	2.3	None (1.2)	0-3	>550
U_3O_8	3.2	None (1.7)	2-5	475->550
U_3Si_2	4.8	None (2.5)	5-7	515->550
U_3Si	6.0	1.8 (3.1)	5-10	500

*The values in parentheses are for 100% burnup of the ^{235}U in LEU fuel, including non-^{235}U fissions, and, therefore, are physical fission density limits.

†Expected swelling at the maximum uranium density and at the fission density limit.

The principal fuel developed by the RERTR Program, U_3Si_2, will make the conversion of most research and test reactors technically feasible. Two major reports[12,13] covering the qualification of this fuel are currently being reviewed by the U.S. Nuclear Regulatory Commission (NRC). Generic approval for use of this fuel in NRC-licensed reactors is expected within a few months. The major conclusions of the qualification report are:

1. The fuel is compatible with the Al matrix and cladding.

2. The fuel meat thermal conductivity is similar to that of UAl_x and U_3O_8 dispersion fuels.

3. The magnitude and rate of the U_3Si_2-Al exothermic reaction are low enough to mitigate its consequences in an accident.

4. U_3Si_2 swells stably under irradiation, with fission gas contained in submicron-sized bubbles.

5. Minor amounts of other phases which might be present in nominal U_3Si_2 (U_3Si, U_{ss}, or USi) are acceptable. The U_{ss} (or free uranium) apparently converts to UAl_x, which, as noted above, is very stable under irradiation.

6. Blister threshold temperatures are at least as high as those of UAl_x and U_3O_8 dispersion fuels.

7. Fuel elements irradiated to well beyond normal burnup were dimensionally stable.

8. Release fractions for volatile fission products were not measured but cannot be significantly above the 25 to 70% values measured for U-Al alloy or U_3O_8-Al fuels.

A relatively small effort has been made to qualify MEU dispersion fuels. Early in the Program some 1.7-Mg U/m^3 UAl_x and U_3O_8 elements were tested to provide the means for an intermediate enrichment reduction prior to adequate LEU fuels being qualified. Irradiation of medium-enriched UAl_2 miniplates has shown that this fuel behaves well. Further experience with highly enriched UAl_2 miniplates[14] leads us to declare medium-enriched UAl_x qualified even though no full-sized elements have been irradiated. There can be no doubt that the fuel will behave properly. Medium-enriched U_3O_8 dispersion fuel is qualified for loadings up to 1.7 Mg U/m^3 with no fission density limit (1.8×10^{27} f/m^3 is the maximum achievable at that loading with 45%-enriched uranium).

Acceptance of the qualification status of the fuels discussed above is indicated by the use of whole cores of UAl_x, U_3O_8, or U_3Si_2 fuels in several reactors. Work leading to the conversion of additional reactors is actively in progress.

Continuing and Future Work

We now believe that we understand the mechanism underlying the poor swelling behavior of U_3Si.[7] There is some possibility that modifications of the fuel alloy might give some marginal improvement in the fission density limit.[15] If any such modifications are identified;

irradiation tests will be performed. The prospects for economic commercial fabrication of fuel plates with densities much in excess of 6.0 Mg U/m^3 (say, above 6.3 Mg U/m^3) are not good because of homogeneity problems.

There continues to be an interest in uranium silicide alloys with composition between U_3Si_2 and U_3Si. Miniplate ($U_3Si_{1.5}$) irradiations have been completed, one full-sized; 4.7-Mg U/m^3 $U_3Si_{1.7}$ element under irradiation in OSIRIS has reached 60% burnup; and three full-sized elements (4.8 to 5.5 Mg U/m^3) are being fabricated with compositions in the range $U_3Si_{1.5-1.6}$ for irradiation in the HFR-Petten. The results thus far indicate a somewhat higher fission density limit than for U_3Si.

Since it appears likely that the uranium density achievable with U_3Si will be lower than that needed for use of LEU fuel in a few reactors, we intend to qualify medium-enriched U_3Si_2 dispersion fuel. The very good behavior of MEU and HEU miniplates indicates excellent prospects for acceptable behavior of medium-enriched U_3Si_2 fuel plates with loadings in the range 4.0 to 4.8 Mg U/m^3 to fission densities of at least 2.5×10^{27} f/m^3 in the fuel meat.

SUMMARY AND CONCLUSION

With the help of our many partners, the RERTR Program has qualified several high-density fuels for use in LEU conversions of research and test reactors. Conversions have already been accomplished using TRIGA ($UZrH_x$) and SPERT (UO_2) rod-type fuels and UAl_x plate-type fuel. Work leading to the conversion of a number of plate-type reactors using UAl_x or U_3Si_2 dispersion fuels is underway. Generic NRC approval for use of U_3Si_2 dispersion fuel is expected soon. One new reactor recently began its operation using high-density, low-enriched U_3O_8 dispersion fuel.

Work to qualify higher-density dispersion fuels (U_3Si and U_3Si_x) continues. In order to provide the technical means to convert all research and test reactors, the RERTR Program intends to pursue qualification of U_3Si_2 with MEU.

With the fuels qualified with LEU to date, however, the RERTR Program has already provided the technical means to convert most of the free world's research and test reactors.

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