DEVELOPMENT OF VERY-HIGH-DENSITY FUELS BY THE RERTR PROGRAM*

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ABSTRACT

The RERTR program has recently begun an aggressive effort to develop dispersion fuels for research and test reactors with uranium densities of 8 to 9 g U/cm^3, based on the use of γ-stabilized uranium alloys. Fabrication development teams and facilities are being put into place, and preparations for the first irradiation test are in progress. The first screening irradiations are expected to begin in late April 1997 and the first results should be available by the end of 1997. Discussions with potential international partners in fabrication development and irradiation testing have begun.

INTRODUCTION

Fuel development has been a cornerstone of the U.S. Reduced Enrichment for Research and Test Reactors (RERTR) program since its inception in 1978. This development work, performed cooperatively with many partners internationally, resulted by 1987 in the qualification of several dispersion fuels having significantly increased densities: UAl_x-Al at 2.3 g U/cm^3, U_3O_8-Al at 3.2 g U/cm^3, and U_3Si_2-Al at 4.8 g U/cm^3. About 20 reactors have been converted from high-enriched uranium (HEU) to low-enriched uranium (LEU), and several new LEU reactors have been built using these fuels. In most cases the uranium silicide fuel has been used.

Although a fuel with a uranium density of 4.8 g U/cm^3 is sufficient to convert approximately 90% of the research reactors which used HEU of U.S. origin in 1978, conversion of the remaining reactors, which use a significant quantity of HEU, require fuels having considerably higher densities. Therefore, the RERTR program continued to develop methods to fabricate plates containing higher loadings of uranium silicide fuels until funding constraints forced an end to this work at the end of September 1989. CERCA, one of the RERTR program's key partners in the development of uranium silicide dispersion fuel, continued its development efforts and announced in 1993 that it had developed an advanced fabrication process which allowed the loading of U_3Si_2-Al fuel to be increased to 6.0 g U/cm^3 [1].

Even though development of higher-density fuels was stopped, analysis of the irradiation experiments performed earlier in the RERTR program continued, resulting in a better understanding of the fundamental behavior of these fuels. Also, work to better understand and refine dispersion fuel fabrication techniques continued. Papers reporting these results have been presented annually at the RERTR international meetings. During this period of decreased fuel development activity within the RERTR program, development of uranium silicide dispersion fuel in the U.S. continued under the auspices of Oak Ridge National Laboratory's Advanced Neutron Source (ANS) project, with Argonne National Laboratory (ANL) designated as the lead fuel development laboratory [2]. A series of three irradiation experiments in the High Flux Isotope Reactor (HFIR) provided data on fuel behavior at high temperatures and high fission rates. Results from the most recent of these experiments prompted the new analysis of U_3Si_2-Al fuel behavior which will be reported later at this meeting [3]. Irradiation performance modeling was also emphasized since fuels could not be tested at or beyond ANS conditions. The advances made during this interim period have helped provide a good basis for further development of dispersion fuels. Perhaps even rivaling the technical advances in long-term importance, the combination of work for the RERTR program and for the ANS project allowed ANL to keep together the nucleus of its dispersion fuel development team, thereby preserving the possibility to restart serious fuel development within the RERTR program.

In 1994 the Department of Energy announced its intention to resume the development of very-high-density fuels to address needs of existing and new research and test reactors. However, owing to pressures of a shrinking federal budget, funding for this work could not be made available until March of this year, as reported earlier at this meeting [4]. The present paper describes the plans and schedule of this new development work.

GOAL AND APPROACH

We have set a goal of achieving a uranium density in a dispersion fuel meat of 8 to 9 g U/cm^3. This goal is consistent with assessments of uranium densities needed to convert the 10% of reactors not convertible using 4.8 g U/cm^3 U_3Si_2 fuel (see, for example, Ref. 5) and, at least on paper, appears to be achievable from the fabrication point of view. Irradiation behavior and fabrication costs will ultimately determine the success of any proposed fuel, however.

We think that, from the fabrication-cost perspective, the best chance of success lies with extension of the current aluminum-based dispersion fuel concept. Since CERCA's experience with highly loaded U_3Si_2-Al [1] and UN-Al [6] fuels indicates that one is not likely to achieve a fuel volume loading greater than 55% in a commercially viable process, fuel dispersants with very-high uranium densities, ≥15 g U/cm^3, must be used. Table 1 lists the uranium compounds with densities greater than or equal to that of U_3Si_2. With the exception of U_6Fe and U_6Mn, which were tested earlier in the RERTR program and shown to be subject to breakaway swelling at relatively low burnups [7, 8], and similar compounds also expected to exhibit similar poor swelling properties, none of these compounds meets our density requirement.

Therefore, the only fuels with sufficiently high uranium density are pure uranium metal and alloys of uranium and small amounts of other metals. Pure uranium is a notoriously poor performer under irradiation, but a series of alloys designed to maintain uranium in a metastable γ phase have shown good irradiation performance in bulk form under fast reactor conditions. Examples of such alloys are listed in Table 2.

The key issues which must be addressed are the reaction of these alloys with the matrix and the irradiation behavior of the dispersion, which includes both fuel swelling and fuel-matrix interaction. In the event that reaction of these alloys with an aluminum matrix is excessive, we will investigate the use of a magnesium matrix while retaining the aluminum cladding, since magnesium does not form compounds with uranium. We have also considered using zirconium as both matrix and cladding material, but we do not currently plan to pursue this option owing to concerns about fabrication cost.

Table 1. Uranium Compounds with Uranium Densities Higher than That of U_3Si_2

Compound	Density, g/cm^3	U Density, g/cm^3
U_3Si_2	12.2	11.3
UB_2	12.7	11.6
UCo	15.4	12.3
UC	13.6	13.0
UN	14.3	13.5
U_2Ti	15.1	13.7
U_2Mo	16.6	13.8
U_2Tc	16.8	13.9
U_2Ru	16.9	13.9
U_3Si	15.5	14.6
U_6Co	17.7	17.0
U_6Ni	17.6	16.9
U_6Fe	17.7	17.0
U_6Mn	17.8	17.1

Table 2. Densities of Representative Uranium Alloys

Alloy*	Density, g/cm^3	U Density, g/cm^3
U-9Mo	17.0	15.5
U-5Mo	17.9	17.0
U-3Zr-9Nb	16.2	14.3
U-4Zr-2Nb	17.3	16.3

*Alloying element amount given in wt.%.

PLANS AND SCHEDULE

Fabrication

During the early stages of the RERTR program, when development of only three different fuels was being pursued, we had fabrication teams working at ANL's Illinois site (ANL-East), at Oak Ridge National Laboratory (ORNL), and at the Idaho National Engineering Laboratory (INEL). However, by the mid-1980s only the ANL development team remained, and by the mid-1990s that group had dwindled to one person. Therefore, a major increase in our fabrication development capacity was required to support an aggressive fuel development effort. In order to utilize best the resources available within ANL, we decided to reestablish a fabrication development team at ANL-East and to establish a new fabrication development laboratory and team at ANL's Idaho site (ANL-West). The procurement process for most of the equipment needed at ANL-West is underway, and detailed installation plans are being prepared. In addition, we are close to reaching the currently required staffing levels at both sites. At ANL-West we have already begun to produce fuel alloys and expect to begin producing fuel powder, by mechanical means, and compacts for small test plates within the next two months. We anticipate that the development laboratory will be ready for full use by the end of CY 1997, when the installation of the longest-lead item, the rolling mill, will have been completed. In the meantime, we will carry out all rolling operations at ANL-East. In particular, the first rolling experiments have already been performed and fuel plates for the initial irradiation tests (discussed below) are scheduled to be rolled during the first quarter of CY 1997.

We plan to follow the development pattern that worked so successfully for the uranium silicide fuel, that is, to limit our fabrication development to the study of basic issues and to the production of small fuel plates for irradiation testing and to depend on the commercial fabricators to adapt and extend our results to the fabrication of full-sized fuel plates or tubes. We have held preliminary discussions with both Babcock and Wilcox (B&W) and CERCA. Both are interested in participating but will need to assess further the commercial potential of the proposed fuels before deciding the amounts of company resources which can be committed to the required development work.

As was mentioned in an earlier paper [4], ANL and KAERI are in the process of setting up a cooperation agreement under which the use of atomized fuels will be explored. KAERI has already done some work with U-Mo alloys [9], and has agreed to provide a small quantity of powder for fabrication development work at ANL.

Materials Properties Studies

In order to address the key issues stated above, a number of studies are planned on the phase structure of the uranium alloys and diffusion of the matrix materials into the alloys. We expect to make substantial progress in this work over the next year. ANL is also negotiating a cooperation agreement with the A. A. Bochvar All-Russian Research and Development Institute of Inorganic Materials in Moscow, under which phase studies of interacting materials is one area of possible collaboration.

Irradiation Behavior Experiments

Little is known about the irradiation behavior of the proposed fuel alloys when they are dispersed in aluminum or about the behavior of the fuel itself at the high burnups typical of research and test reactors. Our highest priority is to begin irradiation screening tests of the proposed dispersions. We are planning to perform these irradiations in the Advanced Test Reactor (ATR) at INEL and to perform the postirradiation examinations at ANL-West. Discussions are underway with the ATR staff, and an irradiation rig is being designed.

Because of space limitations in irradiation holes near the core, the fuel plates (which we are calling microplates) must be much smaller than the miniplates irradiated previously by the RERTR program in the Oak Ridge Research Reactor (ORR). The current design calls for microplates with outside dimensions 76 mm x 22 mm x 1.3 mm, compared to the 114 mm x 51 mm x 1.3 mm dimensions of a typical ORR miniplate. The microplates will be produced using 9.5-mm-diameter cylindrical compacts, resulting in an elliptical-shaped fuel zone with nominal dimensions of 57 mm x 9.5 mm x 0.5 mm, whereas the fuel zone of the typical miniplate was 102 mm x 46 mm x 0.5 mm. Nevertheless, mechanical analyses have shown that the fuel zone area of the microplate is large enough to behave in the same manner as a much-larger plate.

Since the first tests will focus on fuel particle-matrix interactions and fuel particle swelling, it is not necessary to test plates with uranium densities approaching the 8 to 9 g U/cm^3 goal. In fact, it is much more important to obtain results as quickly as possible. Therefore, in order to minimize heat removal problems in the design of the irradiation rig and to minimize fuel plate fabrication problems, we have chosen a density of 4.8 g U/cm^3.

The uranium alloy fuels to be tested are listed in Table 3. Since the amount of γ-phase stabilization increases as the amount of alloying additions increase, this test matrix is expected to establish the types and minimum amounts of additions necessary. We plan to test a U_2Mo-Al dispersion, since that compound may be formed during the irradiation of the U-Mo alloys, and U_3Si_2-Al, in order to compare the performance of the microplates with that of the previously

Table 3. Fuel Alloys Being Prepared for Irradiation Screening Tests
(Alloying element amount given in wt.%)

U-10Mo	U-10Mo-0.05Sn	U-9Nb-3Zr
U-8Mo	U-4Mo-0.1Si	U-2Mo-1Nb-1Zr
U-6Mo	U-5Nb-3Zr	U-2Mo-1Nb-1Zr-1V
U-4Mo	U-6Nb-4Zr	U-6Mo-2Ru

irradiated miniplates and full-sized plates. The current design allows 12 microplates to be irradiated in one module and three modules to be irradiated in one irradiation position. We currently plan to begin the irradiation near the end of April 1997. We plan to irradiate two sets of plates—one to about 40% burnup and the other to about 80% burnup. The first set will require two 45-day ATR cycles. Assuming that we meet this schedule, postirradiation examinations would be expected to start at about the time of next year's international RERTR meeting.

After the screening tests, we expect to test one or more successful candidate fuels in full-sized plates, irradiated either individually or in full elements. Since the U.S. has no facilities for performing such irradiations, we must look to our international partners for such tests. Very preliminary discussions have been held with representatives of the CEA in France and BATAN in Indonesia. It may also be feasible to irradiate single plates in KAERI's HANARO.

Out-of-reactor irradiation tests have provided valuable insight into the changes taking place in the fuel particles in the very early stages of irradiation (see, for example, Ref. 10). These studies have included neutron irradiations at ANL's Intense Pulsed Neutron Source (IPNS) and ion-beam irradiations at ANL's High-Voltage Electron Microscope (HVEM) facility. We plan to continue such work with the new fuels. A recent improvement in the ability to measure aluminum diffusion rates at the HVEM facility was recently funded by the RERTR program and should prove particularly useful.

Irradiation Behavior Modeling

As mentioned previously, modeling has become a very important part of our development work, since it is impossible to anticipate all conditions under which the fuels we develop might be used. We are currently modifying the DART code [11] to incorporate swelling models for uranium alloy dispersion fuels, based on data available from previous irradiations of bulk fuel. We will continue to use the model as a tool to help us understand the observed irradiation behavior. We will be pursuing improvements to our model, especially in the area of mechanical deformations, under the agreement with the Bochvar Institute in Moscow, mentioned earlier.

OTHER FUELS

The RERTR program maintains an interest in several fuels which do not have sufficiently high uranium densities to qualify as very-high-density fuels, namely, U_3Si_2, UO_2, and UN. However, to simplify management of our overall fuels work we will integrate our work on these other fuels with that on the very-high-density fuels. Current activities are briefly discussed below.

U_3Si_2-Al

As discussed in another paper at this meeting, there now appear to be limits to the applicability of U_3Si_2-Al fuel in certain high-temperature, high-fission rate situations [3]. The problem, of course, is that we have insufficient data to determine those limits. We recently discussed our analyses with a representative of the French CEA, since the CEA is currently irradiating some 5.8 and 6.0 g U/cm^3 plates in SILOE. We proposed a collaboration with the CEA in performing postirradiation examinations of these plates.

The ANL-KAERI cooperation agreement also calls for the irradiation in HANARO of miniplates fabricated in the U.S., some containing atomized U_3Si_2 powder produced by KAERI and some containing the traditional comminuted U_3Si_2 powder in order to demonstrate the acceptable irradiation characteristics of the atomized fuel. A miniplate irradiation rig for HANARO is currently being designed.

UO_2

Considerable experience in the behavior of U_3O_8-Al dispersion fuel was obtained during the early phases of the RERTR program. Our interest in UO_2-Al fuel has been kindled by our collaboration with the Russian RERTR program, in which that fuel is being tested up to 3.8 g U/cm^3 for use in converting certain Russian-designed research reactors to use LEU. Our contribution thus far is to revise the DART code to include a model for uranium oxide, based on the U_3O_8 data. This work will be presented in a separate paper at this meeting [12]. We expect to continue to work with our Russian colleagues to evaluate the results of irradiation tests planned to begin within the next year.

UN

In 1994, CERCA reported that uranium densities of about 7.0 g U/cm^3 could be achieved in UN-Al dispersion fuel by a commercially acceptable process [6]. Although this achievement represented a significant advance in uranium density, a study performed at ANL and reported in another paper at this meeting [13] has shown that the rather-large thermal neutron absorption cross section of nitrogen substantially reduces the advantage of UN-Al fuel over U_3Si_2-Al fuel at the same fuel volume loading. We understand, however, that concerns about problems presented by uranium silicide fuels in the back end of the fuel cycle have prompted some consideration of the use of UN as a substitute for U_3Si_2. One major problem which will be encountered when conducting a feasibility study for the use of this fuel system is the lack of irradiation behavior data. However, should one of our international partners decide to pursue the use of UN, the RERTR program would offer to collaborate in obtaining and evaluating the needed irradiation data.

CONCLUSIONS

The RERTR program has recently begun an aggressive effort to develop dispersion fuels for research and test reactors with uranium densities of 8 to 9 g U/cm^3, based on the use of γ-stabilized uranium alloys. Fabrication development teams and facilities are being put into place, and preparations for the first irradiation test are in progress. The first screening irradiations are expected to begin in late April 1997 and the first results should be available by the end of 1997. Discussions with potential international partners in fabrication development and irradiation testing have begun.

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