# QUALIFICATION OF URANIUM-MOLYBDENUM ALLOY FUEL—CONCLUSIONS OF AN INTERNATIONAL WORKSHOP

J. L. SNELGROVE

*Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, Illinois 60439-4815 U.S.A.*

and

A. LANGUILLE

*CEA/DEC Centre de Cadarache, 13108 St. Paul Lez Durance CEDEX - France*

## ABSTRACT

Thirty-one participants representing 21 reactors, fuel developers, fuel fabricators, and fuel reprocessors in 11 countries discussed the requirements for qualification of U-Mo alloy fuel at a workshop held at Argonne National Laboratory on January 17-18, 2000. Consensus was reached that the qualification plans of the U.S. RERTR program and the French U-Mo fuel development program are valid. The items to be addressed during qualification are summarized in the paper.

## 1. Introduction

The U.S. RERTR program and the French U-Mo fuel development program, conducted jointly by the CEA, CERCA, COGEMA, Technicatome, and Framatome, are developing very-high-density dispersion fuels based on γ-phase U-Mo alloys [1-4]. The common goal of both programs is to develop and qualify a fuel with a uranium density ≥8.0 g/cm$^3$ in order to (1) make possible the conversion to LEU of reactors that still require HEU, (2) make possible the design of new high-performance LEU reactors, and (3) provide a fuel with an available back-end solution as an alternative for those reactors currently using $U_3Si_2$ fuel. The requirement of the U.S. spent fuel return program that only fuel removed from the core by May 12, 2006, is eligible for return adds a special urgency to qualify a fuel to replace $U_3Si_2$ fuel at an early date.

The definitive qualification document for $U_3Si_2$ fuel is NUREG-1313, issued by the U.S. Nuclear Regulatory Commission (NRC) in July 1988 [5]. It consists of NRC's evaluation of qualification data presented by the RERTR program in two documents, incorporated as Appendix A [6] and Appendix B [7]. We have adopted the qualification of $U_3Si_2$ fuel as a model for the qualification of U-Mo fuel, and both programs are acquiring similar qualification data. However, in view of changes in the regulatory environment since 1988, we felt it prudent to reexamine qualification requirements while there was still time to accommodate new requirements without substantial delay in our respective qualification programs. Therefore, a workshop was held at Argonne National Laboratory on January 17-18, 2000, to obtain input from reactor operators currently using $U_3Si_2$ fuel, fuel developers, fuel fabricators, and those designing or procuring new reactors.* In addition, the workshop explored means to facilitate early implementation of the new fuels in order to address the May 2006 deadline.

This paper summarizes the conclusions of the workshop.

* Workshop participants are listed in the Appendix of this paper.

## 2. Reprocessing of U-Mo Fuel

The central problem for most current users of $U_3Si_2$ fuel is the lack of an available back-end solution. As stated above, one requirement for a high-density fuel is that it can be reprocessed by the one remaining commercial reprocessor of research reactor fuels, COGEMA. A key feature of the French program is that COGEMA is technically and financially involved in the U-Mo development to make sure that reprocessability criteria are met. To date, COGEMA has shown that U-Mo reprocessing is feasible. The internationally accepted specification for vitrified waste will be met. Research and development are ongoing at COGEMA to confirm and adjust operating conditions [8].

## 3. U-Mo Fuel Qualification

The types and amounts of data used to qualify $U_3Si_2$ fuel and planned for qualifying the U-Mo fuel were discussed. It was agreed that only those items related to the fuel meat should be addressed, since the cladding will not be changed. The items to be addressed are summarized in Table 1.

In general, the qualification program for the U-Mo fuel is, not unexpectedly, quite similar to that for $U_3Si_2$ fuel. A few significant differences deserve comment. Qualification of $U_3Si_2$ fuel was based on basic irradiation-performance data obtained from relatively few (15) miniplates. This basic behavior was confirmed by the irradiation of a number of full-sized elements and by a whole-core demonstration. At the time of $U_3Si_2$ fuel qualification, we were only beginning to study the mechanisms of fuel particle and dispersion irradiation behavior; in the intervening years our understanding of what is important has increased markedly. Now we are able to focus our tests on those issues of primary importance and to apply our increased knowledge of fuel modeling.

Table 1. Items Addressed in Qualifying $U_3Si_2$ Fuel and To Be Addressed in Qualifying U-Mo Fuel

| Item | Qualification of $U_3Si_2$ | Qualification of U-Mo |
|---|---|---|
| **Fuel Powder Properties** | | |
| Fuel Phases | From literature; composition specification established to eliminate free uranium and $U_3Si$. | Phase composition of fuel particles produced mechanically or by atomization and phase stability during fabrication are being determined. Phase composition and microstructural damage/dislocation density will be related to irradiation performance. |
| Method Used to Produce Fuel Powder | Comminution from ingot only method used at time of qualification. | Powder can be produced mechanically and by atomization. Determine general specifications applicable to both powder types, considering information obtained from fuel phase and irradiation studies. |
| Particle Size Distribution | Determined by fabricator and reported. | To be determined by fabricator and reported. Optimum amount of 'fines' could be different than for $U_3Si_2$ because of fuel/Al reaction. |
| Density | Measured. | Measured and calculated. |
| Heat Capacity | From literature. | From literature and will be derived from fuel meat heat capacity measurements. |
| Thermal Expansion Coefficient | From literature. | From literature. |

| | | |
|---|---|---|
| **Fabrication Considerations** | Discussed problems associated with high fuel-volume loadings (dogbone, min. clad, stray fuel particles) and fuel particle oxidation. | Same considerations apply to U-Mo fuel; apply $U_3Si_2$ experience to U-Mo. |
| **Fuel Meat Properties (Unirradiated)** | | |
| Porosity | Measured. | Will be measured for miniplates and full-sized plates. Porosity will be lower because fuel particles are not brittle. Amount of porosity is controlled by fabrication process and should not be specified. |
| Heat Capacity | From literature by combination of values for $U_3Si_2$ and Al. | Will be measured during reaction enthalpy measurements. Can also be calculated from literature values. |
| Thermal Conductivity | Measured for unirradiated fuel. Inferred from data for irradiated fuel based on fraction of Al remaining in meat. | Will be measured for unirradiated fuel. Some measurements will be made on annealed samples to verify relationship of thermal conductivity and fraction of Al remaining in meat. No plan to make measurements on irradiated fuel. |
| Fuel/Aluminum Compatibility | Measured qualitatively during long-term anneals at 400°C. No reaction-rate data obtained. | Measure reaction rates and fuel-meat-volume change as a function of fuel composition and temperature. |
| Exothermic Energy Releases | Measured enthalpy and kinetics of fuel/Al reaction. | Will measure enthalpy and kinetics of fuel/Al reaction as a function of fuel alloy composition. |
| Corrosion Behavior | Measured corrosion resistance in boiling water. Now appears not to be a good indicator of corrosion resistance during irradiation. | Although some measurements will be made, these data should not be considered necessary for qualification, based on $U_3Si_2$ experience. A substantial body of U-Mo corrosion literature exists. |
| Mechanical Properties | Not discussed. | Results of tensile tests on extruded meats will be reported. |
| Meat Thermal Expansion Coefficient | Not discussed. | No measurements planned. Any discussion in qualification report to be based on data for other fuel/Al meats. |
| **Fuel Meat Irradiation Behavior** | | |
| Test Samples—Small plates | Data from 15 LEU, four MEU, and two HEU miniplates irradiated in the ORR discussed in qualification report. Densities from 1.66 g/cm$^3$ for HEU to 5.67 g/cm$^3$ for LEU. Burnups to 98%. Temperatures 75-125°C. | Twenty-four microplates and 24 nanoplates irradiated to date. All LEU. PIE in progress. Ground and atomized samples with Mo content between 6 and 10 wt.%. Burnups to 73%. Temperatures ~60°C for microplates and 140-235°C for nanoplates. Miniplate experiment being planned will contain 24 or more LEU plates. |

| | | |
|---|---|---|
| Test Samples—Full-Sized Plates | Thickness increase data from four plates irradiated in SILOE. Densities ranged from 2.0-5.4 g/cm$^3$. Average burnups 73-78%. | Two LEU plates currently being irradiated in OSIRIS. Thickness increases being monitored. Two LEU and two MEU plates will be irradiated in HFR-Petten. Fuel loading is 50 vol.%. Average burnups ~50% in OSIRIS, 70-80% in HFR. PIE planned for all plates. |
| Test Samples—Full-Sized Elements | Six in ORR. Nominal loading 4.8 g/cm$^3$; actual loadings 4.6-5.2 g/cm$^3$. One in SILOE, but results not discussed. Average burnups 35-82%. At least five plates from each ORR element were examined. | Up to four ~8-g/cm$^3$ elements in OSIRIS and HFR. Up to four ~6-g/cm$^3$ elements in HFR. PIE of one or more plates from each element planned. |
| Whole-Core Demonstration | Sixty-eight 4.8-g/cm$^3$ fuel elements and eight 3.5-g/cm$^3$ fuel followers irradiated in ORR to various burnups. Good behavior reported, but PIE data not available. | No whole-core demonstration is planned. Irradiation of a number of prototype elements is anticipated. |
| Fuel Meat Swelling | Measured by immersion density. | Measured by quantitative metallography for microplates and nanoplates. Will also be measured by immersion density for miniplates. Thicknesses of full-sized plates will be measured. |
| Fuel Meat and Fuel Particle Microstructures | Examined. | Will be examined. Behavior of mechanically produced and atomized powders will be compared. Effect of Mo content of alloy on performance will be determined. |
| Fuel/Aluminum Interaction | Not discussed. Micrographs indicated that the amount of interaction was not significant. In subsequent years a detailed study was made and a correlation was developed as a function of fission density and temperature. | Detailed data will be available from miniplate, microplate, and nanoplate irradiations. A similar correlation is being developed. |
| Blister Threshold Temperature | Measured on selected miniplates and full-sized plates from test elements. | Measurements planned on selected microplates, miniplates, and full-sized plates. |
| Threshold Temperature for Fission Product Release | Measurements on $UAl_x$, $U_3O_8$, and $U_3Si$ plates showed first fission products are released when plate blisters. No measurements made on $U_3Si_2$. | Will be assumed that first release occurs when plate blisters. No additional measurements are planned. |
| Fission Product Release from Defected Plate | No measurements were reported in the qualification report. French authorities required such a measurement in SILOE. | No tests are planned. However, data are available on corrosion of bulk U-Mo fuel under irradiation. |

Therefore, the RERTR program has placed increased emphasis on irradiation tests of small fuel plates to focus on particular behavior issues. In addition, the requirement for significantly increased fuel-volume loadings in LEU fuels required the fuel developers and fabricators to place special emphasis on such issues as dogbone, homogeneity, and stray fuel particles. The whole-core demonstration allowed the fabricators to demonstrate their ability to consistently manufacture these high-density fuels. During the intervening years, fabrication techniques have been continually improved so that, now, there is no question about the ability to manufacture high-quality, high-volume-loaded fuel plates. Hence, we do not plan a whole-core demonstration for the U-Mo fuel. It is expected, however, that prototype elements will be irradiated in a number of reactors.

To facilitate qualification of U-Mo fuel, the ANL and French participants in the workshop have agreed to collaborate in the following areas: (1) interpretation of results of irradiation behavior tests, (2) modeling of U-Mo fuel, and (3) obtaining approval from their respective national licensing authorities for the use of U-Mo fuel. As stated earlier, both programs share the mutual goal of qualifying fuels with a density of $\geq 8$ g/cm$^3$. The French program foresees reaching that goal by the end of 2004 [4], and the RERTR program expects to achieve it about one year later. However, the RERTR program has adopted an intermediate goal of qualifying 6-g/cm$^3$ U-Mo fuel before the end of 2003 in order to provide as much lead-time as possible for reactor operators needing to replace all $U_3Si_2$ fuel by May 2006.

## 4. Implementation of U-Mo Fuel

The workshop participants discussed various issues related to implementing the U-Mo fuel, including safety analyses and the irradiation of prototype elements. The IAEA guidebook on conversion [9] covers the issues that should be discussed in the safety analysis. The focus should be on those things that change when changing from $U_3Si_2$ fuel to U-Mo fuel. Preliminary calculations indicate that only negligible changes in reactor performance should be expected when replacing a $U_3Si_2$ element with the equivalent U-Mo element. From a thermal-hydraulic point of view, the lower fuel volume loading in the U-Mo element will result in higher thermal conductivity and, hence, lower fuel meat temperature.

The practice of many reactors has been to irradiate prototype elements before conversion to a new fuel type. It was generally agreed that the usual practice of waiting until regulatory approval was granted in the U.S. (or France) before ordering prototypes could not be followed for the U-Mo fuel by those reactors faced with the May 2006 deadline. This time, efforts must be made to convince funding and regulatory authorities to allow early introduction of prototypes. To do this, however, the irradiation performance data being acquired by the RERTR and French programs must be published much more quickly than before. Another useful method of data dissemination would be the preparation, regular updating, and circulation of a draft qualification document similar to Ref. [6]. The RERTR program is willing to coordinate the production and updating of such a draft qualification document. In order to encourage the irradiation of prototype elements, the RERTR program will make available a limited quantity of LEU for these elements, thereby reducing the financial risk of ordering the elements before all of the qualification data are available.

## 5. Conclusion

The workshop confirmed that the qualification plans described by the ANL and French participants in the workshop, modeled on the qualification of $U_3Si_2$ fuel, were valid. No items were added, and it was agreed that a few items could be addressed on the basis of previous data and experience. It was also established that a significant need of reactor operators wishing to initiate early irradiation of prototypes is timely publication of qualification data.

## 6. References

[1] J. L. Snelgrove, G. L. Hofman, M. K. Meyer, C. L. Trybus, and T. C. Wiencek, "Development of very-high-density low-enriched-uranium fuels," *Nucl. Eng. and Design* **178** (1997), p. 119.

[2] J. L. Snelgrove, G. L. Hofman, M. K. Meyer, S. L. Hayes, T. C. Wiencek, and R. V. Strain, "Progress in Developing Very-High-Density Low-Enriched-Uranium Fuels," Trans. Intl. Conf. Research Reactor Fuel Management (RRFM'99), Bruges, Belgium, March 28-20, 1999, p. 44.

[3] A. Languille, J. P. Durand, and A. Gay, "New High Density MTR Fuel--The CEA-CERCA-COGEMA Development Program," Trans. Intl. Conf. Research Reactor Fuel Management (RRFM'99), Bruges, Belgium, March 28-30, 1999, p. 61.

[4] C. Joly, P. Sacristan, H. Vacelet, and A. Languille, "First Results on Full-sized Plates Under Irradiation in the OSIRIS Reactor," these proceedings.

[5] U.S. Nuclear Regulatory Commission, "Safety Evaluation Report related to the Evaluation of Low-Enriched Uranium Silicide-Aluminum Dispersion Fuel for Use in Non-Power Reactors," NUREG-1313 (July 1988).

[6] J. L. Snelgrove, R. F. Domagala, G. L. Hofman, T. C. Wiencek, G. L. Copeland, R. W. Hobbs, and R. L. Senn, "The Use of $U_3Si_2$ Dispersed in Aluminum in Plate-Type Fuel Elements for Research and Test Reactors," Argonne National Laboratory Report ANL/RERTR/TM-11 (October 1987).

[7] G. L. Copeland, R. W. Hobbs, G. L. Hofman, and J. L. Snelgrove, "Performance of Low-Enriched $U_3Si_2$-Aluminum Dispersion Fuel Elements in the Oak Ridge Research Reactor," Argonne National Laboratory Report ANL/RERTR/TM-10 (October 1987).

[8] J. P. Durand, B. Maugard, and A. Gay, "Technical Ability of New MTR High Density Fuel Alloys Regarding the Whole Fuel Cycle," Trans. Intl. Conf. Research Reactor Fuel Management (RRFM'98), Bruges, Belgium, March 29-31, 1998, p. 55.

[9] International Atomic Energy Agency, "Research reactor core conversion guidebook," IAEA-TECDOC-643 (April 1992).

## Appendix—Workshop Attendees

Reactor Representatives: H. Krohn (BER-II/HMI, Germany), P. Gubel (BR2/SCK-CEN, Belgium), M. Bagger Hansen (DR-3/Risø National Laboratory, Denmark), J. C. Lee (FNR/Univ. of Michigan, U.S.A.), H.-R. Kim (HANARO/KAERI, Republic of Korea), F. J. Wijtsma (HFR-Petten/NRG, The Netherlands), J. W. DeVries (HOR/IRI, The Netherlands), K. Fujiki (JMTR/JAERI, Japan), Y. Murayama (JRR-3/JAERI, Japan), W. J. Garland (MNR/McMaster University, Canada), A. Chabre (OSIRIS/CEA-Saclay, France), R. Calabrese (RA-3/CNEA, Argentina), P. S. Bull (Replacement Reactor/ANSTO, Australia)

Fuel Developers, Fabricators, and Reprocessors: D. Sears (AECL, Canada), M. M. Bretscher, G. L. Hofman, J. E. Matos, M. K. Meyer, I. Prokofiev, J. Rest, J. L. Snelgrove, A. Travelli, T. C. Wiencek (ANL, U.S.A.), B. W. Pace (BWX Technologies, U.S.A.), A. Languille (CEA-Cadarache, France), R. Romano, S. J.-P. Naccache, H. Vacelet (CERCA, France), P. Adelfang (CNEA, Argentina), J. Thomasson (COGEMA, France), C.-K. Kim (KAERI, Republic of Korea)