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40. REPROCESSING RERTR FUELS*

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ABSTRACT

The Reduced Enrichment Research and Test Reactor Program is one element of the United States Government's nonproliferation effort. High density, low enrichment aluminum-clad dispersed uranium compound fuels may be substituted for the highly enriched aluminum-clad aluminum-uranium alloy fuels now in use. Savannah River Laboratory has performed studies which demonstrate reprocessability of spent RERTR fuels at Savannah River Plant. Results of dissolution and feed preparation tests with both unirradiated and irradiated (up to approximately 90% burnup) fuels will be presented.

INTRODUCTION

Conversion of research and test reactors from highly enriched (>70%) to low-enriched (<20%) uranium fuels is an important part of the United States Government's nonproliferation policy. To be accepted, such conversions should be accomplished with little or no adverse impact on reactor performance, fuel element configuration, and fuel cycle cost. The goal of the Reduced Enrichment Research and Test Reactor (RERTR) program is the technical demonstration of fabrication, irradiation, and reprocessing of low-enriched fuels.[1]

Uranium silicide, oxide, and aluminide fuels dispersed in an aluminum matrix have total uranium densities at low enrichments high enough to meet reactor requirements. However, concerns about reprocessability[2,3,4] have made acceptance of these fuels uncertain. Aluminum clad uranium aluminide and oxide fuels have been successfully reprocessed within the Department of Energy complex with little difficulty.[3,5] However, these fuels had lower total uranium loadings and, therefore, were not dispersed in the aluminum matrix by advanced powder metallurgy techniques. Savannah River Laboratory (SRL) was requested to determine the compatibility of spent RERTR fuels with the present reprocessing capabilities of Savannah River Plant (SRP). Studies at SRL have already demonstrated the key reprocessing steps of dissolution, clarification, and solvent extraction with both unirradiated and 30% burnup silcide RERTR fuel compositions.[6,7]

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Recent SRL studies reported here addressed remaining reprocessing uncertainies. These include the effect of trace fluoride and high burnup on dissolving behavior and demonstration of dissolution of aluminide and oxide RERTR fuel compositions.

SUMMARY

Aluminum clad RERTR dispersed fuels can be reprocessed at SRP. Studies with both unirradiated and irradiated fuel have now demonstrated dissolution, clarification, and solvent extraction steps.

All candidate RERTR fuels were successfully dissolved in mercury catalyzed nitric acid in bench-scale tests. The dispersed fuels phase dissolved at least as fast as the aluminide matrix and cladding. High burnup aluminide, oxide, and silicide fuels were dissolved in times comparable to unirradiated and 30% burnup silicide fuels, as well as fuels now processed at SRP. No significant advantage in dissolving behavior was found to adding trace fluoride. Solids remaining after dissolution were amorphous silica containing negligible uranium and plutonium.

DISCUSSION

Reprocessing studies were conducted at SRL with both unirradiated and irradiated RERTR miniplates (see Figure 1) provided by Argonne National Laboratory (ANL). These fuel elements were originally prepared for fabrication and irradiation studies. Depleted miniplates were produced during fabrication and uranium loading studies at ANL. Enriched miniplates were produced at ANL and irradiated in the Oak Ridge Research Reactor (ORR) for postirradiation examination studies at ANL. Important properties of the high burnup fuels studied are shown in Table 1.

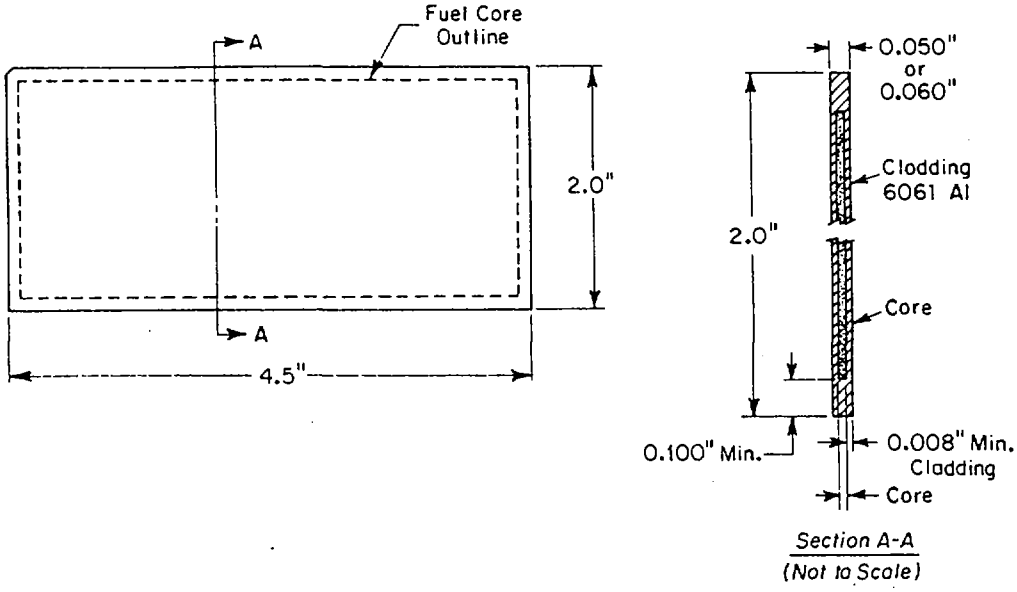

FIGURE 1. RERTR Miniplate

Table 1. Properties of High Burnup RERTR Fuels

Fuel Composition	Miniplate Designation (ORNL Plate #)	Original Uranium Density (gm/cm^3)	Estimated Burnup ($^{235}U\%$)	Dissolution Time (Hours)
U_3Si_2	A-32	3.75	87.5	4.3
U_3Si	A-23	4.77	83	6.9
U_3SiAl	A-26	4.64	83	8.1
U_3O_8	O-58-4	3.10	75	7.9
UAl_x	E-061	1.88	>88	6.6

Dissolution Studies

Bench-scale studies demonstrated that candidate RERTR fuels show no significant difference in dissolution from aluminum clad fuels now reprocessed at SRP. Dissolution procedures for RERTR fuels are based on current SRP practices and previous SRL studies on dissolution of aluminum clad fuels.[5,8-12]

Dissolution studies were conducted in bench-scale glassware. Studies with the intensely radioactive irradiated fuel were conducted in SRL's heavily shielded High Level Caves (HLC) facility. The dissolver apparatus consisted of an electrically heated flask fitted with a water cooled reflux condenser. The flask was also fitted with a burette for metering in reagents during dissolution. The acid consumed during the course of each dissolution was replenished periodically with 15.6M HNO_3. A two-hour boiling digestion period dissolved any fuel particles remaining after all large cladding fragments were dissolved. This digestion also precipitated most of the dissolved silicon as amorphous silica.

Boiling 3M HNO_3-0.002M Hg^{+2} completely dissolved sections of highly irradiated miniplates of all candidate RERTR compositions. This dissolving recipe produced vigorous initial dissolution without uncontrollable foaming. The range of total dissolution times (Table 1) of 4-8 hours compares favorably with times for fuels now processed at SRP. The dispersed fuel particles dissolved as fast or faster than the aluminium matrix and cladding in all tests, which is a necessary condition to ensure that no unsafe accumulation of fissile solids will occur in plant operations. Average composition of the unclarified dissolver solutions is shown in Table 2. Plutonium isotopics shown in Table 3 reasonably confirm previously predicted values.[13]

Burnup does not have a significant adverse effect on dissolution time. Figure 2 shows that increasing burnup may actually decrease time slightly.

Table 2. Average Composition of Dissolved High Burnup RERTR Fuel

Fuel	H^+ (M)	U (gm/l)	NO_3^- (M)	Al^{+3} (M)	Dissolved Si (ppm)	Pu (gm/l)
U_3Si_2	2.1	6.4	6.8	0.95	290	0.13
U_3Si	2.1	8.1	7.1	0.98	330	0.16
U_3SiAl	1.9	9.5	8.3	0.69	300	0.10
UAl_x	2.4	4.5	8.1	0.78	410	0.11
U_3O_8	2.1	8.2	8.2	0.79	348	0.10

Table 3. Plutonium Isotopics
(Atom %)

Fuel	Burnup (^{235}U atom %)	^{238}Pu (atom %)	^{239}Pu (atom %)	^{240}Pu (atom %)	^{241}Pu (atom %)	^{242}Pu (atom %)
U_3Si_2	87.5	2.89	56.7	21.1	12.7	5.7
U_3Si	83	2.90	56.5	21.1	12.7	5.8
U_3SiAl	83	3.21	55.4	21.4	12.9	6.2
U_3O_8	75	1.70	63.8	19.7	11.7	5.3
UAl_x	>88	3.57	54.0	21.8	13.1	6.9

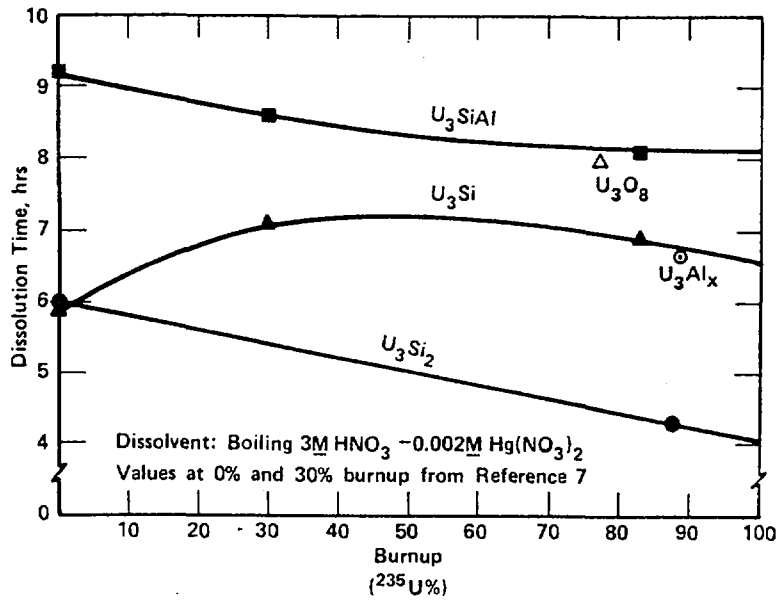

FIGURE 2. Effect of Burnup on Dissolution Time

Adding trace fluoride does not provide any significant advantage to the dissolution recipe. Figure 3 shows that only a slight decrease in dissolution time results from trace fluoride. Significant corrosion and waste handling problems result from using even trace fluoride.

A number of dissolutions of unirradiated fuel were made with reduced (0.0002M) Hg^{+2}. This change increased the average dissolution time by 50%, but would result in a tenfold decrease in mercury to high level waste storage.

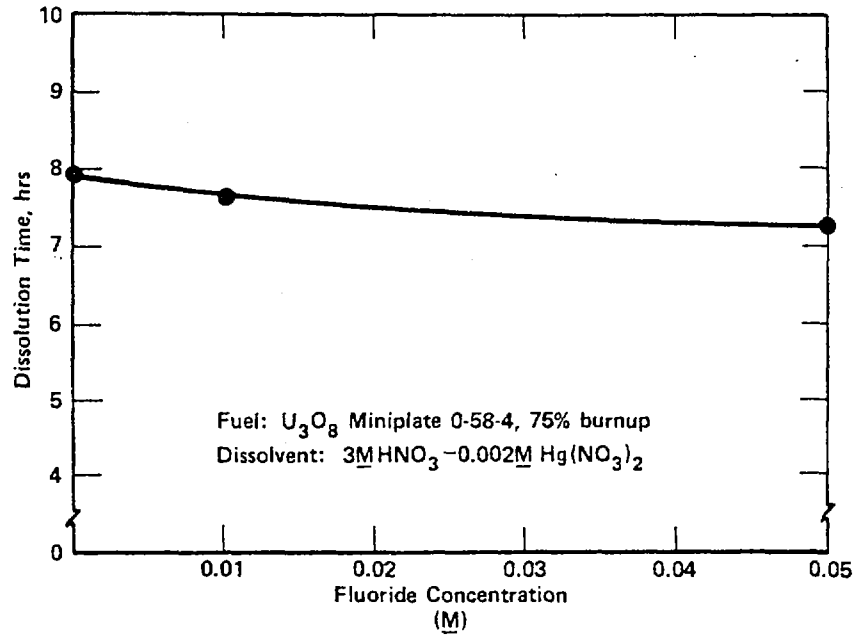

FIGURE 3. Effect of Trace Fluoride on Dissolution Time

Residual Solids

Solids remaining after dissolution contained negligible uranium (see Table 4). These solids appeared to be amorphous silica. X-ray diffraction analysis of unirradiated silicide fuel dissolver residues in previous tests[6] showed no crystal structure. The solids from irradiated silicide fuel (both 30% and up to 88% burnup) were darker and more granular in appearance, but again appeared to be precipitated silica. Both high burnup oxide and aluminide fuels produced much less dissolver solids than the silicides. These solids were much darker and finer. Plasma torch spectroscopy showed negligible uranium in these solids also, along with a high, but undetermined level of silicon.

Clarification Studies

Studies at SRL[6] have shown that the present SRP clarification steps of reverse permanganate and gelatin strike will adequately remove dissolved silicon from dissolved RERTR fuel. Even for silicide fuels, the dissolved silicon content dropped to 70 ppm or below (average 40 ppm) following the gelatin strike. Previous studies[7] also showed that clarified RERTR fuel solution may be processed within present operating standards in existing SRP solvent extraction equipment with no hydraulic disruption.

Table 4. Composition of Dissolver Residue

Fuel Composition	Insoluble Fraction (Wt. % of Original)	U (Wt.%)	Pu (Wt.%)
U_3Si_2	4.28	nd	0.085
U_3Si	2.72	nd	0.012
U_3SiAl	2.55	nd	0.042
U_3O_8	0.07	nd	0.007
UAl_x	*	*	*

*Negligible solids remained.
nd - not detected

CONCLUSION

These studies demonstrated that spent aluminum-clad oxide, aluminide, and silicide RERTR fuels can be successfully reprocessed at SRP. Head-end dissolving and feed preparation rates are compatible with currently projected campaign schedules. Because the dispersed fuel phase dissolved at least as fast as the aluminum matrix, no nuclear safety problems are anticipated. Trace fluoride is not necessary to reliably dissolve these fuels in reasonable times. A gelatin strike is recommended to ensure solvent extraction operability.

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