CONF-8310176--6

FUEL DEVELOPMENT ACTIVITIES OF THE U.S. RERTR PROGRAM*

CONF-8310176--6

DE84 003620

by

James L. Snelgrove, R. F. Domagala, T. C. Wiencek

Argonne National Laboratory
Argonne, Illinois

and

George L. Copeland

Oak Ridge National Laboratory
Oak Ridge, Tennessee

to be presented at the

INTERNATIONAL MEETING ON REDUCED ENRICHMENT

FOR RESEARCH AND TEST REACTORS

Tokai Research Establishment

Japan Atomic Energy Research Institute

24-27 October 1983

*Work supported by the U.S. Department of Energy

FUEL DEVELOPMENT ACTIVITIES OF THE U.S. RERTR PROGRAM*

J. L. Snelgrove, R. F. Domagala, and T. C. Wiencek
Argonne National Laboratory
Argonne, Illinois, U.S.A.

and

G. L. Copeland
Oak Ridge National Laboratory
Oak Ridge, Tennessee U.S.A.

ABSTRACT

Progress in the development and irradiation testing of high-density fuels for use with low-enriched uranium in research and test reactors is reported. Swelling and blister-threshold temperature data obtained from the examination of miniature fuel plates containing UAl_x, U_3O_8, U_3Si_2, or U_3Si dispersed in an aluminum matrix are presented. Combined with the results of metallurgical examinations, these data show that these four fuel types will perform adequately to full burnup of the ^{235}U contained in the low-enriched fuel. The exothermic reaction of the uranium-silicide fuels with aluminum has been found to occur at about the same temperature as the melting of the aluminum matrix and cladding and to be essentially quenched by the melting endotherm. A new series of miniature fuel plate irradiations is also discussed.

INTRODUCTION

Since its establishment in 1978, the U.S. Reduced Enrichment Research and Test Reactor (RERTR) Program has pursued the development of the high-density fuels needed to provide an economic fuel cycle with low-enriched uranium (LEU, <20% enriched). For use in plate-type fuel elements, uranium-aluminide (UAl_x), uranium-oxide (U_3O_8), and uranium-silicide (U_3Si_2, U_3Si, and U_3SiAl) fuels dispersed in an aluminum matrix have been studied in the U.S. and in several other countries, including Germany, France, and Argentina. The primary means of testing the irradiation behavior of the fuels being developed and of providing the basic data needed for fuel qualification has been the irradiation and post-irradiation examination (PIE) of miniature fuel plates (miniplates). Miniplates produced by Argonne National Laboratory (ANL), Oak Ridge National Laboratory

*Work supported by the U.S. Department of Energy.

(ORNL), and EG&G Idaho, Inc. in the U.S., by NUKEM in Germany, and by the Comision Nacional de Energia Atomica (CNEA) in Argentina have been irradiated in the Oak Ridge Research Reactor (ORR)+. During the past year, the initial series of miniplate irradiations and the PIE of a number of miniplates have been completed. As a consequence of a much improved understanding of the irradiation behavior of the uranium-silicide dispersion fuels,[1] a new series of miniplate irradiations has been planned. A study of the exothermic reaction of uranium-silicide fuel particles with the aluminum matrix of the fuel meat has also been completed. The results of these activities are described below.

MINIPLATE IRRADIATIONS AND PIE

The initial series of miniplate irradiations in the ORR ended on June 13, 1983. Since the beginning of the irradiations on July 18, 1980, 132 miniplates in 11 modules have been irradiated to ^{235}U depletions (fission + capture) ranging from 34% to at least 89%, as shown in Table 1. The ^{235}U depletions listed were calculated on the basis of a flux that does not change with burnup and were normalized to measured burnup data from Module 3 plates. However, since the flux must increase somewhat as the fuel in the miniplates burns, the calculations underestimate the actual burnup, except for Module 3. A number of samples from miniplates have been analyzed for total fissions at ORNL, ANL, and General Electric Company - Vallecitos Nuclear Center (GE-VNC) using the ^{148}Nd method of ANSI/ASTM E321-79. The results appear to give too high a fission density in all cases, requiring close to 100% ^{235}U depletion in some cases. Very preliminary comparisons made between measured and calculated U and Pu isotopic compositions indicate that the actual ^{235}U depletions lie between the values listed in Table 1 and those obtained from the ^{148}Nd results. Evaluation of the results is continuing. For this paper, the quoted fission densities for the high-burnup plates were obtained by multiplying the calculated ^{235}U depletions by 1.07.

During the past year, PIE's have been completed at ORNL on eight UAl_x miniplates fabricated by EG&G, which ranged in uranium density from 1.88 to 2.30 Mg/m^3. Four plates contained medium-enriched uranium (MEU, 40.2%) and four plates contained low-enriched uranium (LEU, 19.9%). Swelling of the fuel meat ranged between -0.3 and 2.9% for fission densities between 0.8×10^{27} and 1.8×10^{27} $fissions/m^3$. Two of the MEU plates were fabricated with 100% fine (<44 μm) fuel particles. These plates showed the lowest net swelling even though their fission densities were 1.5×10^{27} $fissions/m^3$, probably because of a high initial void content of the fuel meat. Blister-threshold temperature tests on one LEU plate and on one high-fines MEU plate produced no blisters even at the highest temperature, 550°C. These data are consistent with the 561°C blister-threshold temperature measured for similar plates in previous tests.[2] Metallographic examinations were made of sections of one LEU and two MEU (including a high-fines) miniplates and showed each plate to be in excellent condition. There were no indications of actual or incipient failures. A photomicrograph of a section from the MEU miniplate with the highest fission density is shown in Fig. 1. The few large voids seen may be remnants of the voids initially in the fuel meat. No other gas bubbles are evident in the fuel particles. PIE's of UAl_x miniplates fabricated by NUKEM and the CNEA have also been completed, and detailed results are

+In lieu of miniplate irradiations, full-sized plates fabricated by CERCA (France) have been irradiated in the SILOE reactor at the Centre d'Etudes de Grenoble in cooperation with the RERTR Program.

Table 1. ORR Miniplate Irradiation Summary - September 15, 1983.

Module No.	Fabricator	Fuel Type(s)	No. of Plates	Date of Last Irradiation	Irradiation Time, fpd	Calculated ^{235}U Depletion, at.%	Current Status
1	EG&G	UAl_x	8*	10/24/82	470	84	PIE Complete, ORNL
	ORNL	U_3O_8	4				PIE Complete, ORNL
2	ORNL	U_3O_8	9	8/31/81	305	77	PIE Complete, ORNL
	ANL	U_3SiAl	3				PIE Complete, ORNL
3	ANL	U_3SiAl, U_3Si	7,5	10/05/80	75	34	PIE Complete, ANL
4	EG&G	UAl_x	4	5/27/82	420	80	PIE Complete, ORNL
	ORNL	U_3O_8	4				PIE Complete, ORNL
	ANL	U_3SiAl, U_3Si	2,2				PIE Complete, ANL
5	ORNL	U_3O_8	12	7/10/81	268	75	PIE Complete, ORNL
6	CNEA	UAl_2, U_3O_8, U_3Si	5,4,3	6/13/83	324	77	Poolside Cooling, ORNL
7	ANL	U_3Si_2, U_3SiAl, U_3Si	4,4,4	11/15/81	300	83	PIE Complete, ANL
8	NUKEM	UAl_x, U_3O_8	4,8	3/25/82	262	77	PIE Complete, GE-VNC
9	CNEA	UAl_x, U_3O_8	6,4	10/24/82	352	87	PIE Complete, ORNL
	ORNL	U_3O_8	2				PIE Complete, ORNL
10	ANL	U_3SiAl, U_3Si	8,4	12/07/83	279	68	Poolside Cooling, ORNL
13	ANL	U_3SiAl	12	6/13/83	284	71	Poolside Cooling, ORNL
14	ANL	U_3Si_2+	2	6/13/83	385	89	Poolside Cooling, ORNL

*Four plates removed for fission-product-release tests after 275 fpd with an estimated ^{235}U depletion of 63%.

+Two plates from Module 7 which were reinserted for further irradiation.

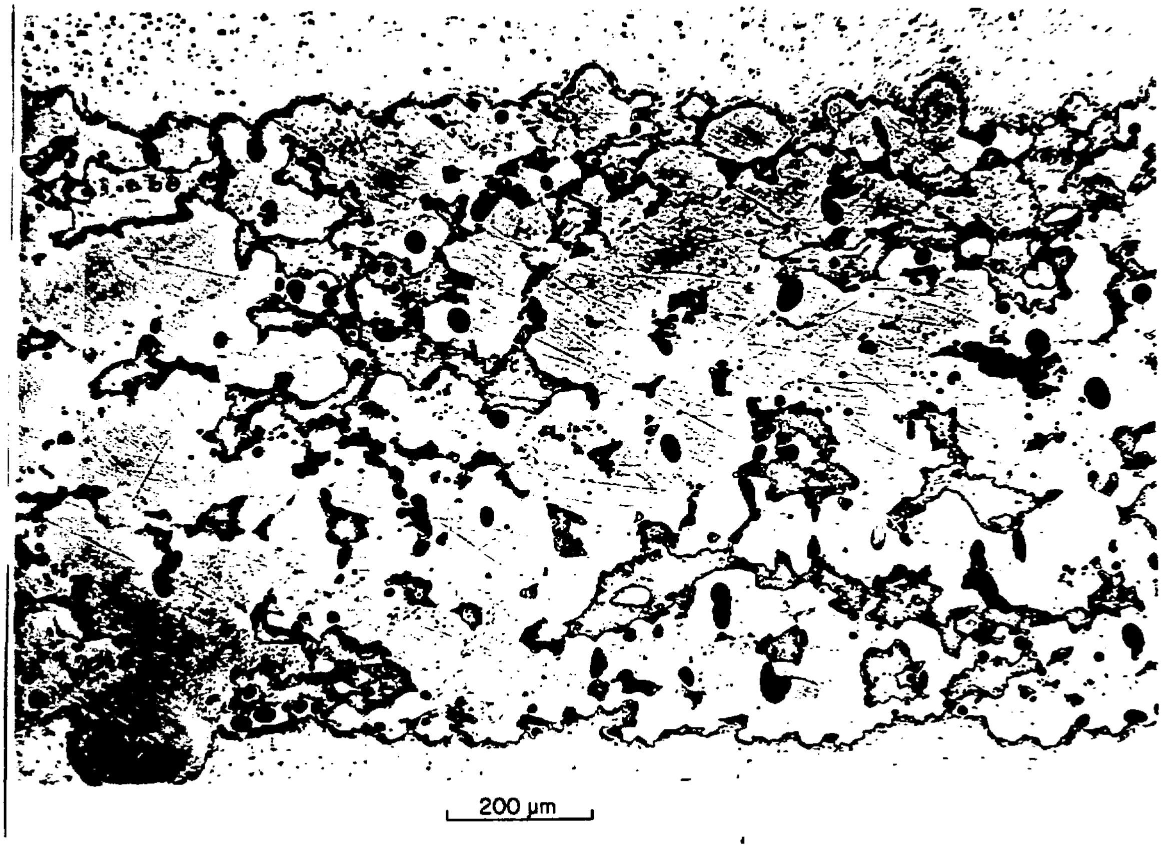

Fig. 1. Longitudinal Section of 2.3-MgU/m^3 UAl_x Miniplate E-117 after Irradiation to 1.8 x 10^{27} Fissions/m^3. (ORNL Photo R-78319)

reported in separate papers.[3,4] Based upon all of the results obtained, UAl_x appears to be an extremely well-behaved fuel for LEU and MEU applications requiring uranium densities of up to 2.5 Mg/m^3.

Substantial progress has been made in understanding the irradiation behavior of uranium-silicide dispersion fuels.[1] In summary, it has been established that the presence of aluminum in the uranium-silicide fuel particles enhances the mobility of the fission gases to the point where large bubbles can form, leading to breakaway swelling at high burnups. This phenomenon has been proved to be the cause of the large scale breakaway swelling (pillowing) in core last year of one 4.62-MgU/m^3 U_3SiAl (U + 3.5 wt% Si + 1.5 wt% Al) miniplate in Module 4 after ~94% ^{235}U depletion (2.1 x 10^{27} fissions/m^3) and is presumed to be the cause of pillowing of a 6.98-MgU/m^3 U_3SiAl miniplate in Module 13 after ~76% ^{235}U depletion (2.4 x 10^{27} fissions/m^3). Therefore, all further work on this fuel has been abandoned by the RERTR Program. The other two silicides, U_3Si_2 and U_3Si, have behaved very stably up to ^{235}U depletions of ~94% at densities of 3.75 and 4.8 MgU/m^3 (1.6 x 10^{27} and 2.2 x 10^{27} fissions/m^3), respectively. Some small regions of the U_3Si particles, where aluminum has diffused in from the matrix, behave similarly to U_3SiAl, but the overall effect on the fuel swelling is small. On the basis of the examinations performed to date, it is expected that the swelling rate will continue to be well-behaved for higher uranium loadings in the fuel meat and for higher fission densities in the fuel particles.

PIE's of the NUKEM U_3O_8 miniplates from Module 8 and the CNEA U_3O_8 miniplates from Module 9 have been completed, as have swelling measurements on seven additional ORNL U_3O_8 miniplates.[3,4] A summary of the swelling and blister-threshold temperature data for all but the U_3SiAl miniplates examined to date is given in Table 2. It should be mentioned that when the variation in initial void content of the various plates is taken into account, the swelling behavior of the plates is much more consistent than is apparent from the net swelling values tabulated. As was stated earlier, the UAl_x appears to be an extremely well-behaved fuel for both LEU and MEU applications. As was reported last year,[5] the U_3O_8 fuel appears to be adequate for use in LEU applications at uranium densities up to ~3.2 Mg/m^3, for which the fission density attainable for 100% ^{235}U depletion cannot exceed 1.6 x 10^{27} fissions/m^3. All of the new data support this position. There is some indication that the swelling rate begins to increase at around this fission density and that the initial characteristics of the U_3O_8 powder may influence the fission density at which the rate increase begins. Study of this phenomenon is continuing. The U_3Si_2 and U_3Si fuels also appear to be well-behaved up to the uranium densities and fission densities achieved in the test plates.

EXOTHERMIC REACTION OF URANIUM-SILICIDE WITH ALUMINUM

Since it has been reported that the uranium-silicide fuels react rapidly with aluminum at approximately 620°C,[6] measurements of the heat generated in such a reaction in fabricated fuel plates were made during the past year. The differential thermal analysis (DTA) technique has been used, and the study has covered the temperature range from room temperature to 1300°C. The studies were conducted using 4.1-mm-diam punchings from 1.53-mm-thick miniplates with nominally 0.51-mm-thick fuel meat. Fuel loadings of 30 and 45 vol% were tested for U_3Si_2 (3.3 and 5.0 MgU/m^3) and U_3Si (4.4 and 6.6 MgU/m^3). The fuels contained depleted uranium. The DTA instrument was calibrated using samples of materials having well-known heats of fusion, the principal ones in the temperature range of interest being Al and Sb. The samples were heated in flowing He gas at a rate of 10°C per minute.

Specimens of each sample type were typically heated to 840-850°C, and at least one specimen of each type was heated to 1300°C. No evidence of any reaction was found except in the vicinity of the aluminum-melting endotherm. In order to obtain sufficient data to unfold the exothermic reaction from the aluminum-melting endotherm, most of the specimens were taken through three heating and cooling cycles. Approximately 5% of the total energy was liberated during the second heating. Fig. 2 shows segments of the T and ΔT curves for the first and second heatings of a 45 vol% U_3Si_2 specimen. Using data from the three heating cycles, reaction energies of 363 ± 69, 320 ± 21, 496 ± 26, and 363 ± 59 kJ/kg of fuel have been determined for 30 and 45 vol% loadings of U_3Si_2 and U_3Si, respectively. The uncertainties quoted are only the standard deviations of the three measurements made for each sample type. The U_3Si_2 has a lower energy release than U_3Si, and, for each fuel type, the energy release per unit fuel weight is lower at the higher volume loading. Although the exothermic energy release from the reaction of the uranium-silicides with aluminum is larger than that from U_3O_8 and aluminum,[7] the overlapping of the reaction temperature with that of aluminum melting effectively quenches the reaction. Depending on the volume loading of fuel and the cladding thickness, the net effect would range from a substantial endotherm to a small exotherm. The thermograms also indicate that most of the reaction occurs at a temperature somewhat higher than 620 °C.

Table 2. Summary of Swelling and Blister-Threshold Temperature Data for High-Density Dispersion Fuels (From PIE of Miniature Fuel Plates).

Fuel Type	Fabricator*	Density Range, Mg/m^3 Low	High	Enrichment	No. of Plates	Fission Density Range, $10^{27}/m^3$ Low	High	Swelling Range, % $\Delta V/V_m$ Low	High	Blister-Threshold Temperature, °C
UAl_x	C	1.47		45.1	1	1.3		4.3		-
UAl_x	E	1.88	1.95	40.2	4	1.1	1.5	-0.3	0.6	550 - 565
UAl_x	E,N	2.13	2.31	39.8 - 40.2	6	1.3	1.8	1.9	3.4	550 - 561
UAl_x	E	1.88	1.99	19.9	3	0.8	0.9	0.7	2.9	>550
UAl_x	E,N,C	2.14	2.33	19.9 - 27.3	6	1.0	1.1	-1.7	4.0	>550
UAl_x	C	2.48	2.52	20.2	2	1.1		-3.9	-3.3	>550
U_3O_8	O,N	2.40	2.46	39.7 - 45.0	3	1.7	2.0	2.9	9.7	470
U_3O_8	O	2.77		45.0	1	2.3		P+		
U_3O_8	O	3.10		45.0	3	2.1	2.5	11.2	P+	
U_3O_8	O,N,C	2.30	2.48	19.5 - 27.3	9	0.8	1.1	0.0	2.0	490 - >550
U_3O_8	O	2.76	2.79	19.5	11	0.9	1.2	-0.7	1.3	>550
U_3O_8	O,N,C	2.91	3.13	19.5 - 27.3	15	1.0	1.6	-3.8	12.6	478 - 550
U_3Si_2	A	3.72	3.76	19.9	4	1.6		3.7	4.9	530
U_3Si	A	4.79	4.83	19.9	5	0.7		0.1	0.8	510
U_3Si	A	4.77	4.81	19.9	6	2.0	2.2	8.9	11.8	500

*Fabricators: ANL, EG&G Idaho, ORNL, NUKEM, CNEA.

+Indicates that plates "pillowed" during irradiation.

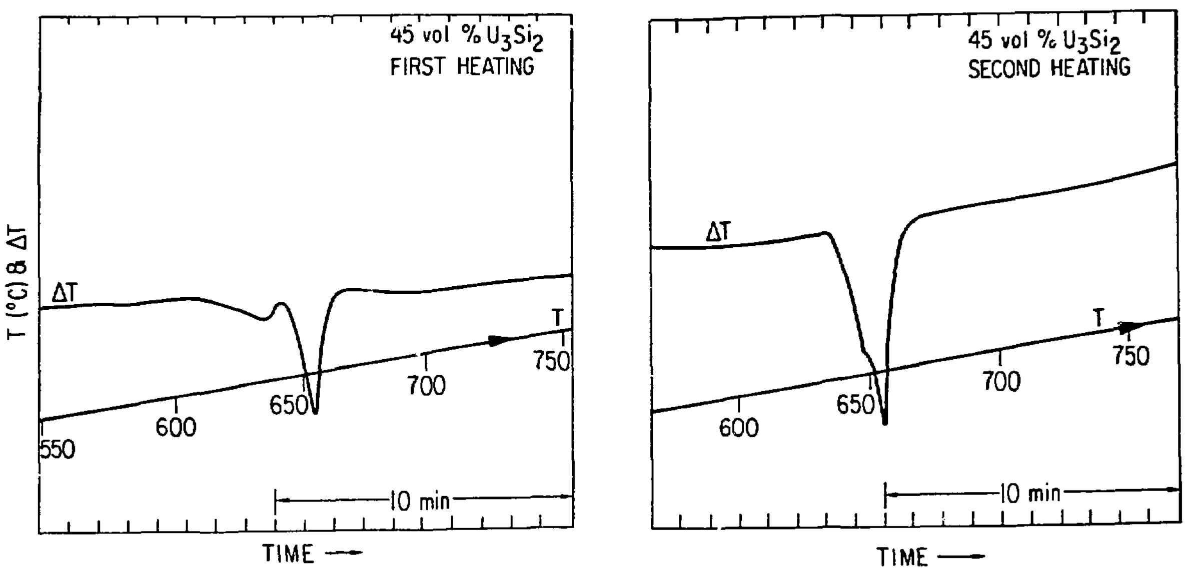

Fig. 2. Differential Thermograms for First and Second Heatings of a Single Specimen of 45 vol% U_3Si_2 Fuel. (Exotherm, +; endotherm, -)

NEW SERIES OF MINIPLATE IRRADIATIONS

A new series of miniplate irradiations has been planned to establish the performance limits of U_3Si and U_3Si_2 fuels dispersed in aluminum, since a majority of the uranium-silicide miniplates irradiated thus far have contained U_3SiAl. The uranium-loading goals are 5.5 and 7.0 MgU/m^3 (50 and 48 vol%) for U_3Si_2 and U_3Si, respectively. The size distribution of the fuel powder has been set at 85 wt% between 44 and 149 μm and 15 wt% less than 44 μm. As in the initial irradiation series, the plate thicknesses will be 1.27 and 1.52 mm. Most of the plates will have a nominal meat thickness of 0.51 mm; however, some plates with high ^{235}U loadings will have reduced meat thicknesses to reduce the heat generation rate, and one set of U_3Si_2 plates will be fabricated with 0.76-mm-thick meat. In order that these miniplates be more representative of full-sized plates, an O-temper anneal will not be performed and the plates will be left in the cold-rolled condition.

Since U_3Si_2 dispersed in aluminum appears to be very stable under irradiation, the new series of irradiations will be used to assess the uranium-loading and fission-density limits for its use in plate-type fuels. In the initial miniplate irradiation series, four plates in the density range of 3.7 to 3.8 MgU/m^3 were irradiated. Four full-sized elements with a uranium density of approximately 4.8 Mg/m^3 have also been irradiated in the ORR to somewhat lower burnup.[8] It is planned to irradiate two miniplates at this same loading to establish a correspondence between the behavior of full-sized and miniature plates. Four miniplates containing 5.5 MgU/m^3 will be irradiated to establish satisfactory behavior of LEU fuel at the upper limit of fabricability. Since it appears that the U_3Si_2 fuel might perform acceptably at much higher fission densities than could be achieved in LEU fuel and because an MEU backup fuel may be needed if sufficiently high uranium densities do not prove feasible, four MEU (40.2%-enriched) miniplates will be tested at 4.0 and 5.5 MgU/m^3 and two high-enriched uranium (HEU, 93%-enriched) miniplates will be tested at 1.7 MgU/m^3. Data from the combination of LEU, MEU, and HEU miniplates will be used to establish the fission density limits for the various fuel loadings, and, therefore, the margins to failure for U_3Si_2 fuels with LEU and MEU.

As reported earlier, U_3Si fuel dispersed in aluminum also appears to be stable except, possibly, in those areas where aluminum has diffused into the fuel particles. Its overall behavior is good enough to justify an irradiation program as described above for U_3Si_2. The densities to be tested are 6.0 and 7.0 MgU/m^3 with LEU, 4.0 and 6.0 MgU/m^3 with MEU, and 1.7 MgU/m^3 with HEU. Work is also underway to modify U_3Si to inhibit the diffusion of aluminum into the fuel particles and/or to decrease the mobility of fission gases. If any of these modifications show promise, a similar series of miniplates will be fabricated for irradiation.

Based upon the promising performance of highly-loaded UAl_2 dispersion fuel with HEU,[9] it is probable that UAl_2 could provide an MEU backup for some reactors which might require very high LEU fuel densities. In order to confirm this, it is planned to irradiate two MEU (45%-enriched), 3.0-MgU/m^3 UAl_2 miniplates. Some preirradiation characterization work on U_6Fe dispersed in aluminum has been performed by the German reduced enrichment program.[10] In order to test its potential under irradiation, a limited number of U_6Fe miniplates with densities of 7.0 and 8.0 MgU/m^3 will be irradiated in the upcoming series. There are no plans to pursue further the development of U_6Fe, however.

Fabrication of the new miniplates to be irradiated is underway at ANL. In addition, NUKEM will fabricate six U_3Si miniplates for testing, and the CNEA has expressed interest in testing a few additional miniplates. It is anticipated that the irradiations will begin early in 1984 and will continue for 1.5 to 2 years.

REFERENCES

1. G. L. Hofman and L. A. Neimark, "Irradiation Behavior of Uranium-Silicide Dispersion Fuels," International Meeting on Reduced Enrichment for Research and Test Reactors, Tokai-mura, Japan, October 24-17, 1983.

2. T. Shibata, K. Kanda, K. Mishima, J. L. Snelgrove, D. Stahl, J. E. Matos, A. Travelli, F. N. Case, and J. C. Posey, "Release of Fission Products from Irradiated Aluminide Fuel at High Temperature," in Proceedings of the International Meeting on Research and Test Reactor Core Conversions from HEU to LEU Fuels, Argonne, Illinois, November 8-10, 1982, ANL/RERTR/TM-4, CONF-821155 (September 1983).

3. M. Hrovat and H. W. Hassel, "Recent Status of Development and Irradiation Performance for Plate Type Fuel Elements with Reduced ^{235}U Enrichment at NUKEM," International Meeting on Reduced Enrichment for Research and Test Reactors, Tokai-mura, Japan, October 24-27, 1983.

4. E. E. Perez, C. A. Kohut, D. R. Giorsetti, G. L. Copeland, and J. L. Snelgrove, "Irradiation Performance of CNEA UAl_x and U_3O_8 Miniplates," International Meeting on Reduced Enrichment for Research and Test Reactors, Tokai-mura, Japan, October 24-27, 1983.

5. G. L. Copeland and J. L. Snelgrove, "Examination of Irradiated High-U-Loaded U_3O_8-Al Fuel Plates," in Proceedings of the International Meeting on Research and Test Reactor Core Conversions from HEU to LEU Fuels, Argonne, Illinois, November 8-10, 1982, ANL/RERTR/TM-4, CONF-821155 (September 1983).

6. C. E. Weber, "Progress on Dispersion Elements," in Progress in Nuclear Energy, Ser. V, Vol. 2, Pergamon Press, New York, N.Y., 1959.

7. A. E. Pasto, G. L. Copeland, and M. M. Martin, A Quantitative Differential Thermal Analysis Study of the U_3O_8-Al Thermite Reaction, Oak Ridge National Laboratory Report ORNL-5659 (June 1980).

8. J. L. Snelgrove and G. L. Copeland, "Irradiation Testing of Full-Sized, Reduced-Enrichment Fuel Elements," International Meeting on Reduced Enrichment for Research and Test Reactors, Tokai-mura, Japan, October 24-27, 1983.

9. K. R. Brown, "Irradiation Program of Highly Loaded UAl_2-Al Fuel Plates," in Proceedings of the International Meeting on Research and Test Reactor Core Conversions from HEU to LEU Fuels, Argonne, Illinois, November 8-10, 1982, ANL/RERTR/TM-4, CONF-821155 (September 1983); and K. R. Brown, personal communication, September 29, 1983.

10. G. H. Thamm, "Status of the German AF-Programme Considerations with Respect to INFCE Recommendations and Criteria," and S. Nazaré, "Development of U_6Fe-Al Dispersions for the Use of LEU in Research and Test Reactors," in Proceedings of the International Meeting on Research and Test Reactor Core Conversions from HEU to LEU Fuels, Argonne, Illinois, November 8-10, 1982, ANL/RERTR/TM-4, CONF-821155 (September 1983).