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ACTINIDE REDISTRIBUTION IN
MIXED-OXIDE FUELS
IRRADIATED IN A FAST FLUX

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ABSTRACT

Measurements of uranium and plutonium concentrations obtained with an electron-microprobe analyzer are presented for two EBR-II-irradiated fuel elements fabricated with low oxygen-to-metal ratios. The data exhibit a small, but significant, decrease in plutonium concentration near the central void in contrast to results from nearly stoichiometric fuels. These hypostoichiometric fuels were not previously available for examination, and the present data are intended to supplement results presented in an earlier report by the same authors (ANL-7929).

I. INTRODUCTION

Since the appearance of ANL-7929,¹ which described observations of actinide redistribution in mixed-oxide fuels irradiated in a fast flux, a number of additional measurements have been made. These new measurements are significant because they were made in hypostoichiometric fuels not previously available. Although the results of these measurements have been discussed in terms of the effect of the oxygen-to-metal (O/M) ratio on actinide segregation and the responsible mechanism,² these data have not previously been reported.

The introductory remarks and discussion of the phenomenon given in ANL-7929 apply to the present report and will not be repeated. The theoretical description, however, has been greatly extended and can be found in Ref. 3. The experimental procedure and the method of data analysis are identical to those described in ANL-7929, and the results will be presented in the same format.

II. RESULTS

Two pellet-type fuel elements irradiated in EBR-II at moderate power levels were analyzed for actinide redistribution, and the results are summarized in Table I. These two fuel elements differed in their accumulated burnups by about a factor of three, but otherwise they were similar.

One of the multiple data sets from each cross section of both fuel elements is shown in Fig. 1. As in ANL-7929, the straight-line segments in the figure are least-squares averages of the first six to eight data points of all data sets recorded for that cross section.

TABLE I. Summary of Pellet-type Mixed-oxide Fuel Specimens Examined for Uranium and Plutonium Redistribution

Fuel-element Identification	Distance from Bottom of Core, in.	Local Burnup at %	Local Linear Power, MW/l	Initial Fuel O/M Ratio	Fuel type ^a	Initial Fuel Pellet Density, g/cc	Initial Pu/U + Pu, %	Measured Maximum Pu/U + Pu, %	Pu/U + Pu Increase, %	Void Radius, mm	Calculated Centerline Temperature, °C
NUMEC A-4											
Top	11.75	2.8	9.5	1.956	CP-E	83.2	0.20	0.196	-2	16.9	1890
Middle	6.19	5.4	11.6	1.961	MB-E	83.1	0.20	0.203	+2	21.6	2010
Bottom	0.50	2.8	9.5	1.961	MB-E	85.1	0.20	0.192	-5	24.2 ^b	1620
NUMEC C-19											
Top	14.25	6.2	9.4	1.940	CP-E	82.9	0.20	0.195	-3	18.2	2060
Upper middle	7.53-7.67	10.8	12.4	1.940	CP-E	82.9	0.20	0.190	-5	41.5	1800
Lower middle	7.11-6.97	10.9	12.5	1.966	MB-E	83.4	0.20	0.190	-5	41.5	1800
Bottom	0.44	6.9	10.1	1.966	MB-E	83.4	0.20	0.192	-4	25.5	1740

^aCP = encapsulated, MB = mechanically blended, E = encapsulated

^bTypical central void. Trace-1 radius = 18.8 mm, Trace-2 radius = 25.2 mm, Trace-3 radius = 30.6 mm

(a) Fuel Element NUMEC A-4

(b) Fuel Element NUMEC C-19

Fig. 1. Radial Distribution of Plutonium in Two EBR-II-irradiated Fuel Elements Fabricated with Low Oxygen-to-Metal Ratios. Neg. Nos. MSD-59079 and -57837

The specimens were obtained from near the top, middle, and bottom elevations of the 13½-in.-long fuel column of each element, and, except for the middle section of NUMEC C-19, the specimens were transverse (radial) cross sections. The middle section of NUMEC C-19 was a longitudinal section, cut on a diameter, as shown schematically in Fig. 2. Microprobe data were taken both above and below a pellet interface, and this particular pellet interface was between the upper half (O/M = 1.940) and the lower half (O/M = 1.966) of the fuel column; therefore, measurements near the middle of this fuel element were taken in pellets that were originally different. This pellet-interface location was approximately 7.44 in. from the bottom of the core.

Local values of linear power and burnup were calculated, as before, from peak values, which were supplied by Lambert.⁴ The as-fabricated characteristics of the fuels were specified by the manufacturer.⁵

Fig. 2. Diagram of Longitudinal Middle Section of Fuel Element NUMEC C-19 Showing Pellet Interface and Location of Microprobe Traces Neg. No. MSD-89078.

The measured "maximum" plutonium concentrations are shown in Fig. 1 by the intersections of the line segments with the central-void positions, and they are tabulated as fractions in Table I. These concentrations are, in fact, minima in most cases for NUMEC A-4 and C-19; however, the style of Table I in Ref. 1 has been preserved. In both the figures and the table, a large number of data points were combined using a least-squares method. The average standard deviation for a single entry in the percent-increase column (usually a decrease for the present fuels) of Table I is $\sim 5\%$ (e.g., $-2 \pm 5\%$ for the top section of NUMEC A-4). Taking all the data as typical of a fuel with an O/M ratio of ~ 1.955 , these results indicate a decrease of about $3 \pm 2\%$ in plutonium concentration at the edge of the central void. This is a small decrease in plutonium concentration, but it is real and significant. Thus, these results exhibit a reversal of the trend

that has resulted in marked plutonium enrichment near the center of nearly stoichiometric mixed-oxide fuels.

The last two columns in Table I list the measured central-void radii and the calculated centerline temperatures. The bottom section of NUMEC A-4 contained an elliptical central void. The average radius for this central void is listed in Table I, and the radial positions of the void edge, measured along the three radii on which data were taken (data Traces 1-3), are shown in the second footnote to Table I. Centerline temperatures were calculated⁶ for the restructured configuration and local power listed in Table I.

Nonuniform compositions in the outer (cooler) fuel region have been observed frequently, and these variations are seen clearly for fuel element NUMEC C-19 in Fig. 1b and for fuel elements NUMEC C-6 and -11 in Figs. 3e and 3f of Ref. 1. These inhomogeneities were previously thought to be the result of poor mixing during fabrication, and this may be the case. However, several patterns have emerged that may indicate otherwise. Although the irregularities in NUMEC C-11 appear to be random, both NUMEC C-6 and -19 exhibit low plutonium concentrations near the fuel-cladding interface, preceded by a plutonium maximum. In the fuel sections in which this behavior is seen, it is radially symmetric. No correlation of this effect with O/M ratio is apparent; however, these inhomogeneities appear more frequently in fuels with higher burnups. A correlation with

burnup would be rather convincing were it not for NUMEC C-15, which accumulated a high burnup and exhibited consistently smooth concentration profiles. At present, no satisfactory explanation for these low-temperature composition variations is available; however, they do not appear to be related to the high-temperature process of temperature-gradient vapor transport.

III. SUMMARY

Uranium and plutonium concentration data are presented for multiple sections of two fuel elements fabricated with low oxygen-to-metal ratios (~ 1.955). These data exhibit a small, but significant, decrease in plutonium concentration near the central void in contrast to earlier data taken from nearly stoichiometric fuels, which exhibit a marked increase. The present experimental method is identical to that described in Ref. 1, and discussion and analysis based on the experimental data are presented in Refs. 2 and 3.

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