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COMPARISON OF MEASURED AND CALCULATED COMPOSITION OF IRRADIATED EBR-II BLANKET ASSEMBLIES*

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ABSTRACT

In anticipation of processing irradiated EBR-II depleted uranium blanket subassemblies in the Fuel Conditioning Facility (FCF) at ANL-West, it has been possible to obtain a limited set of destructive chemical analyses of samples from a single EBR-II blanket subassembly. Comparison of calculated values with these measurements is being used to validate a depletion methodology based on a limited number of generic models of EBR-II to simulate the irradiation history of these subassemblies. Initial comparisons indicate these methods are adequate to meet the operations and material control and accountancy (MC&A) requirements for the FCF, but also indicate several shortcomings which may be corrected or improved.

I. INTRODUCTION

A demonstration of the electrometallurgical treatment of spent fuels[1] is being conducted at ANL-West using fuel assemblies previously irradiated in the EBR-II reactor. The objective of the treatment is to transform sodium-bonded fuel (which is classified as mixed waste) into a uranium product and stable waste forms that contain the key radioactive isotopes and are suitable for disposal in a geologic repository. Execution of the treatment requires that the spent fuel be well characterized with respect to composition, radiation sources, and decay power to ensure that the treatment is conducted safely, efficiently, and in compliance with MC&A accountability requirements. This characterization relies heavily on computational estimates of nuclide inventories which are validated by a limited set of destructive analysis measurements.

Techniques for accomplishing spent fuel characterization have previously been developed for EBR-II driver assemblies, and the accuracy of the calculated inventory has been verified.[2] In this paper, we present a computational approach to address irradiated EBR-II blanket assemblies, and compare calculated inventory predictions to the initial blanket measurements.

II. CALCULATIONAL STRATEGY AND METHODS

In comparison to the driver fuel, characterization of the EBR-II blankets is substantially more challenging because:

- The blankets are irradiated in the peripheral regions which are a transition region for the neutron flux. The blankets act as a shield that rapidly attenuates the flux magnitude, and causes the neutron energy spectrum to soften with penetration. Even advanced-method predictions tend to deteriorate with penetration into the blanket zone, as shown in numerous critical experiments and shielding experiments.[3]

- During the extensive history of EBR-II operation, several major changes in fuel type and basic reactor geometry were introduced, some of which significantly affected the blanket neutronics environment. For example, radial blanket assemblies were positioned directly adjacent to the driver assemblies in the initial loadings until a stainless steel radial reflector was installed between the core and radial blanket in run 56A.

- Many of the EBR-II blankets resided in-core for the entire reactor operating history. The modernized EBR-II analysis techniques employed for the driver evaluation were developed specifically for recent reactor runs.[2] Extension of these procedures to encompass all EBR-II runs and to address blanket analysis requirements is feasible only in principle because of the enormous effort that would be required.

▸ The key blanket inventory components (particularly the fissile inventory, which is dominated by Pu-239) are generated completely in-reactor. Thus, unlike the high enrichment drivers where most of the fissile material is present in the accurate pre-irradiation specification, determination of the blanket inventory relies completely on the depletion computations.

To address these challenges, a new computational approach has been developed to estimate the post-discharge characteristics of blanket assemblies. Its main steps are as follows:

1. All flux calculations utilize the VARIANT nodal transport module[4] in 3-D (hexagonal-z) geometry. A full core geometric model is required to model variations in the outer structure surrounding the fueled assemblies (adjacent to the outermost blanket row). A 28 energy group multigroup cross section set was used throughout the blanket zone. These group constants were collapsed from an ENDF/B-V.2 based 231-group library with a one-dimensional EBR-II model employed for spatial collapse, with a thick blanket region located outside the reflector.

2. The lengthy sequence of actual EBR-II run configurations was simulated with a much smaller sequence of seven "generic configurations" that capture the major core fueling and geometry changes in the EBR-II operating history. Each generic configuration is depleted at the nominal EBR-II power level for an interval that preserves the exposure (MWd) accumulated by actual runs corresponding to that period.

3. For each generic configuration, the driver region is modeled as a non-depleting zone with row-smeared compositions based on an actual core configuration during that time period. Conversely, the blanket depletion level is accumulated from one generic configuration to the next on a row-by-row basis. This strategy effectively assumes that the flux level at any blanket position depends primarily on its proximity to the core (not the neighboring blanket assemblies); and that the core leakage source does not vary significantly for specific core loadings (particularly true when the reflector is interposed between the core and blanket). The blanket depletion calculations utilize five axial depletion zones in each row and solve nuclide transmutation equations which model the major actinides and use a lumped representation of fission products.

4. A flux reconstruction module is used to recover the detailed intra-assembly flux distribution from the nodal VARIANT results. This module yields the multigroup flux as a function of time for each generic model at any blanket pin position.

5. Characterization of a specific blanket subassembly is accomplished using a modified version of the ORIGEN code,[5] taking into account the actual irradiation history of that blanket. This calculation employs one-group flux and cross section values derived from the generic models using the reconstructed multigroup fluxes within each grid location occupied by the blanket assembly during its irradiation life. The ORIGEN-RA analyses employ highly detailed nuclide transmutation chains, and provide the blanket nuclide densities and radiation emission characteristics in detail.

The foregoing procedure was extensively tested through numerical assessments of individual approximations against reference methods. For example, it was confirmed that the VARIANT transport solution corrected the significant flux magnitude errors observed for diffusion theory in previous analyses (e.g., Ref. 6). Similarly, the error in blanket reaction rates that results from neglecting the details of the blanket management and run-by-run loading details was found to be acceptably small.

An assessment of the uncertainty of the space-dependent U-238 capture rate predicted by the "generic configurations" was made based on individual estimates of the reaction rate uncertainty contributed by each of the modeling approximations. These approximations included: (1) homogenization of the driver region by row for each generic configuration; (2) depletion of the blanket region by row; (3) fixing the core size (number of driver assemblies) in a generic configuration; and (4) depletion of the blanket region without fuel management (assembly shuffling). The uncertainty estimates are summarized in Table 1.

The effects of approximations 1 and 2 were determined separately by appropriately homogenizing the driver or blanket regions for a heterogeneous EBR-II configuration and observing the perturbation of the 1-group flux and U-238 capture cross section in each blanket position. The reaction rate uncertainty introduced by approximation 3 was estimated by varying the number of homogenized driver assemblies in the outermost row of drivers (row 7) and observing the effect on the flux and U-238 capture cross section with radial position in the blanket region. The total reaction rate uncertainty was determined assuming that the core size for the actual reactor runs represented by a "generic configuration" varied by ±10 drivers. A comparison of the U-238 capture rate for a generic configuration in which the blanket region was depleted without shuffling and a configuration based on a fuel management history derived from operations data provided the error estimates for approximation 4.

Table 1 Uncertainty Assessment for U-238 Capture Rate in EBR-II Depletion Analyses

EFFECT	ϕ	$\sigma_{(n,\gamma)}$ (U-238)	$R_{(n,\gamma)}$ (U-238)
Driver homogenization	±2.5%	±1%	±3%
Blanket homogenization	±2.3%	±2.5%	±4%
Core size variation from generic model	±0.25% / driver	±0.25% / driver	±4%
Depletion of generic models w/o blanket management			±5%

III. PRELIMINARY RESULTS AND DISCUSSION

The destructive analyses program being conducted for irradiated EBR-II blankets will yield a unique set of data regarding blanket depletion and transmutation. For all other fast power systems worldwide, the vast majority of the blankets reside in core or local storage. Discharge blanket compositions have only been evaluated on a test assembly basis; thus, a comprehensive assessment of blanket inventory predictions has never been conducted.

As described in the previous sections, precise calculations in the blanket region are difficult, and significant methodological errors can be expected. Most critical experiments have observed an underprediction of all reaction rates in the blanket zone which gets worse with penetration; the error compounds from 5-10% at 15 cm to 20-25% at 40 cm depth.[7] One would expect such underpredictions for direct transmutation products such as Pu-239; and bias factors could be employed if a systematic variation is observed. Inventory predictions of secondary products such as Pu-240 or fission products from Pu-239 are expected to be worse because not only are reaction rates underpredicted, but the Pu-239 source isotope inventory is also underpredicted.

Radial blanket subassemblies of EBR-II each contain 19 tightly-packed sodium bonded elements and each element contains five 11-inch-long slugs. In the typical radial blanket subassembly, the five slugs are depleted uranium (DU). Samples have been obtained from three elements from a radial blanket subassembly (U1302) for destructive analysis by wet chemistry and mass spectrometry techniques. Subassembly U1302 was inserted into grid position 13D4 (Row 13) in Run 1 of EBR-II and remained in that location through the final run of the reactor. The three elements sampled spanned the subassembly radially; i.e., the innermost, central and outermost elements (referred to herein as pins 12, 10, and 8, respectively). Ten axial samples were obtained from each element, including 2 adjacent midplane samples. Measurements on this set of 30 samples provides sufficient spatial detail to determine axial distributions and integral values for these three elements and radial distributions for the subassembly. Measurements on these samples include:

* Sample Mass
* U Mass
* Pu Mass
* Burnup (via determination of La or Tc content)
* U Isotopic Fractions (^{234}U/U, ^{235}U/U, ^{236}U/U, ^{238}U/U)
* Pu Isotopic Fractions (^{239}Pu/Pu, ^{240}Pu/Pu)
* Gamma Spectroscopy (^{134}Cs, ^{137}Cs, ^{144}Ce, ^{106}Ru/^{106}Rh, ^{54}Mn, ^{60}Co, ^{95}Nb, ^{125}Sb, ^{154}Eu, ^{155}Eu, ^{182}Ta and ^{95}Zr)

A limited set of the comparisons of calculated values to these measured values is given herein. Of primary interest is the prediction of the buildup of plutonium in the depleted uranium slugs and the burnup. Comparison of the calculated and measured values of the plutonium content in the outermost element (Pin 8) is shown in Fig. 1. It may be noted that the agreement near the midplane is particularly good (~1%). The C/M values for the Pu content for the two midplane samples for this element are 0.995 and 0.981. The C/M values tend to decrease at the lower end (~0.70-0.90) and to increase at the upper end (~1.05-1.15). This trend is observed in comparisons for all three elements. That is, in the midplane region (high buildup), the calculated values are within ~5%, but display an axial tilt relative to the measured values. Further study is necessary to determine the source of these variations. Of principal interest to the operations of the FCF is the prediction of the total Pu content in the elements. Integration of the area under curves (4th order polynomials) fitted to the measured and calculated values of the Pu content in the samples indicate the total Pu content is predicted to within ~6%. C/M values for total Pu content for elements 12, 10, and 8 were 1.058, 1.021, and 0.962, respectively.

Comparisons of the calculated and measured values of the burnup indicators La and Tc in Pin 8 are shown in Fig. 2. It may be noted that measurements of both La and Tc

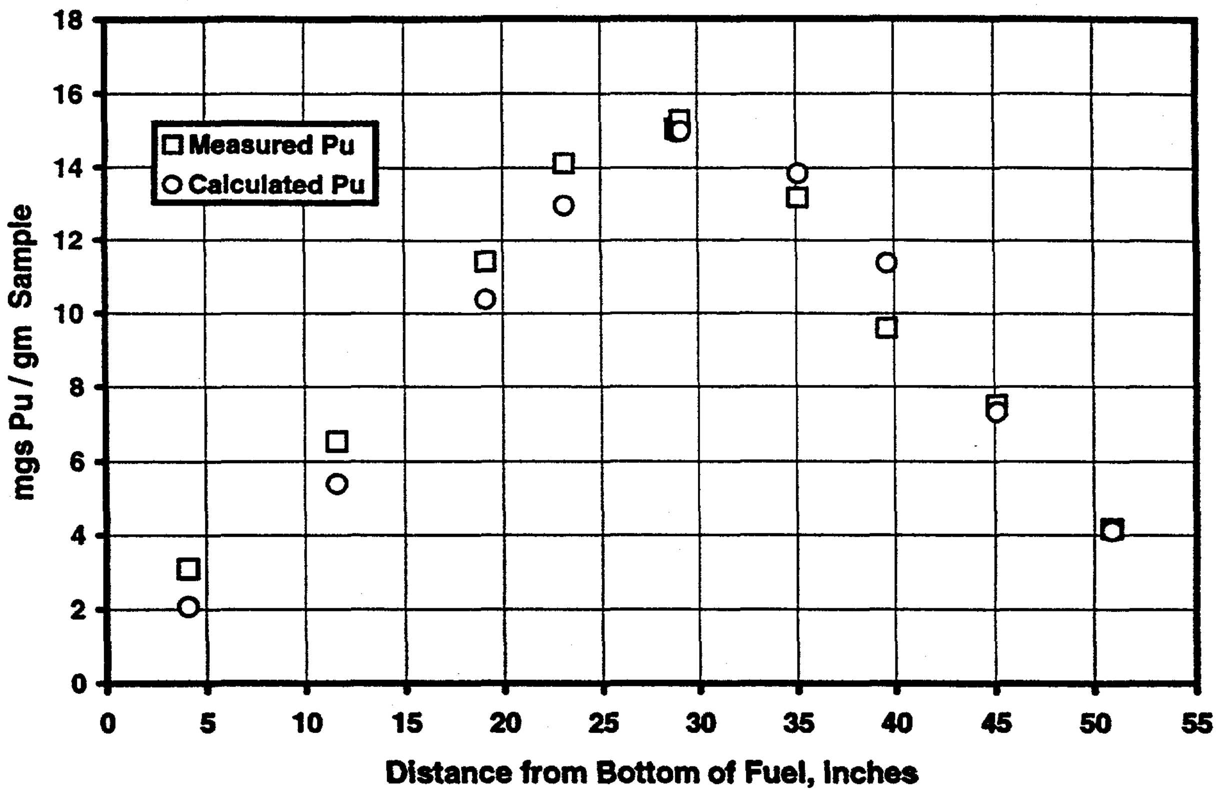

Fig. 1 Pu Content in Pin 8 of Blanket Subassembly U1302

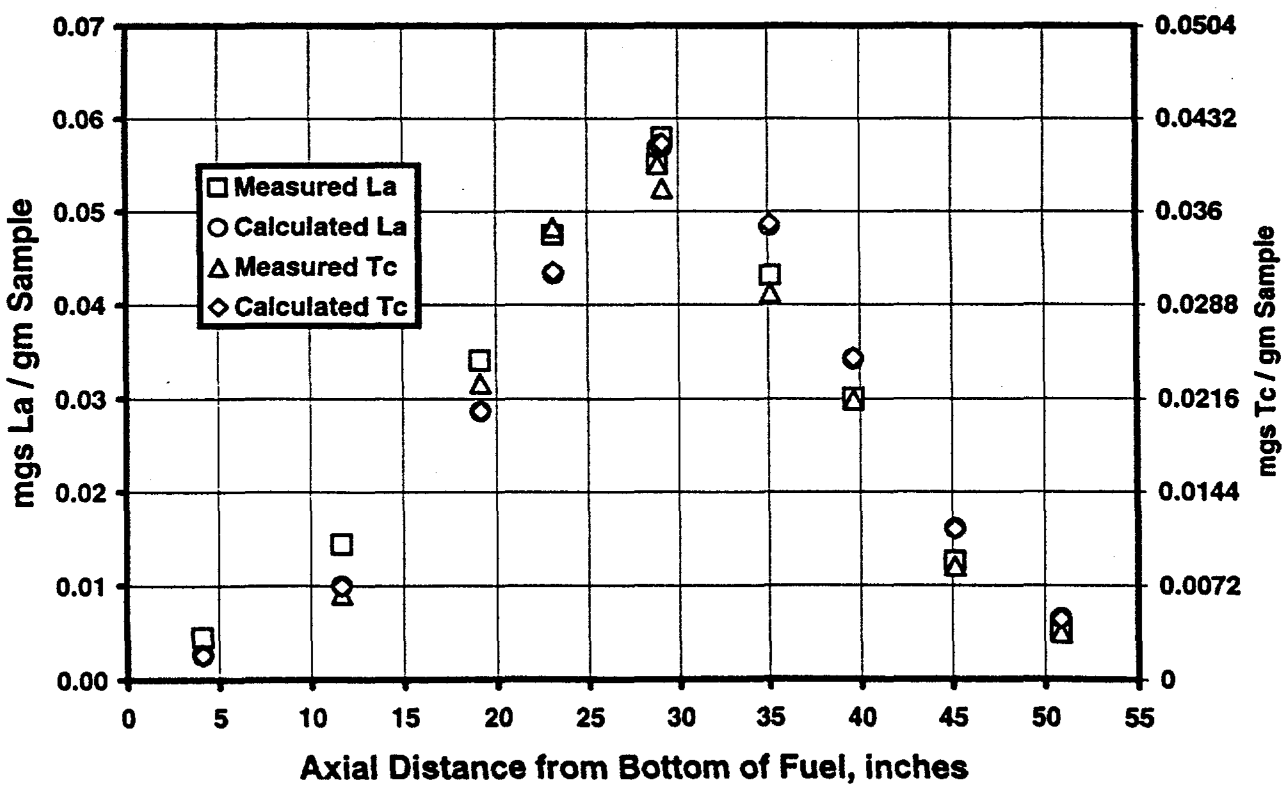

Fig. 2 La and Tc Content in Pin 8 of Blanket Subassembly U1302

Table 2 Plutonium Isotopic Weight Fractions in Blanket Elements in Subassembly U1302

Sample Axial Position (in.)	^{239}Pu/Pu Weight Fraction (%)								
	Pin08			Pin10			Pin12		
	Meas.	Calc.	C/M	Meas.	Calc.	C/M	Meas.	Calc.	C/M
4.105	99.42	99.834	1.0042	99.28	99.731	1.0045	99.08	99.627	1.0055
11.605	99.13	99.546	1.0042	98.89	99.387	1.0050	98.59	99.229	1.0065
19.105	98.68	99.170	1.0050	98.41	98.890	1.0049	97.92	98.610	1.0071
23.105	98.49	98.974	1.0049	98.16	98.629	1.0048	97.65	98.282	1.0065
28.855	98.38	98.820	1.0045	98.05	98.432	1.0039	97.51	98.039	1.0054
29.105	98.38	98.818	1.0045	97.71	98.429	1.0074	97.51	98.036	1.0054
35.105	98.57	98.901	1.0034	98.27	98.528	1.0026	98.13*	98.156	1.0003*
39.605	98.80	99.100	1.0030	98.57	98.791	1.0022	99.13*	98.483	0.9935*
45.105	99.09	99.407	1.0032	98.94	99.156	1.0022	98.57	98.905	1.0034
50.855	99.27	99.640	1.0037	99.15	99.495	1.0035	98.88	99.349	1.0047
		Mean	1.0040		Mean	1.0041		Mean	1.0056
		Std.Dev.	0.0007		Std.Dev.	0.0016		Std.Dev.	0.0012
	^{240}Pu/Pu Weight Fraction (%)								
4.105	0.570	0.1649	0.2893	0.702	0.2673	0.3808	0.895	0.3701	0.4136
11.605	0.852	0.4513	0.5297	1.060	0.6079	0.5735	1.374	0.7637	0.5558
19.105	1.290	0.8233	0.6382	1.550	1.0991	0.7091	2.035	1.3730	0.6747
23.105	1.490	1.0163	0.6821	1.790	1.3546	0.7567	2.300	1.6942	0.7366
28.855	1.590	1.1669	0.7339	1.910	1.5477	0.8103	2.440	1.9303	0.7911
29.105	1.590	1.1687	0.7350	1.890	1.5501	0.8202	2.440	1.9333	0.7923
35.105	1.400	1.0877	0.7769	1.680	1.4533	0.8650	1.820*	1.8168	0.9982*
39.605	1.180	0.8922	0.7561	1.390	1.1958	0.8603	0.844*	1.4978	1.7747*
45.105	0.886	0.5893	0.6652	1.040	0.8366	0.8044	1.390	1.0834	0.7794
50.855	0.710	0.3583	0.5046	0.829	0.5021	0.6057	1.083	0.6457	0.5962
		Mean	0.6311		Mean	0.7186		Mean	0.6675
		Std.Dev.	0.1510		Std.Dev.	0.1553		Std.Dev.	0.1364

*Indicates a suspected error in measured value. These C/M values are not included in the calculation of the mean and standard deviation.

provide useful estimates of the burnup. Again the agreement with calculated values is particularly good near the midplane. For Pin 8, the C/M values for the La and Tc content of the midplane samples were 1.006 and 1.066, respectively. For the three elements these values were generally within 5-10%.

Comparisons have also been made of the U and Pu isotopic fractions in the irradiated blanket elements. Measured and calculated values of the Pu isotopic fractions are given in Table 2. It may be noted that (1) there is a small (0.40%) but very consistent over-prediction of the ^{239}Pu/Pu weight percent in all three elements and (2) there is a large (~20-40%) and rather consistent under-prediction of the ^{240}Pu/Pu weight percent in all three elements. These biases could result from under-prediction of the calculated burnup in these elements. However, given the relatively good comparisons (shown above) for the burnup, it appears these biases result from inadequate one-group constants for the production and destruction of these nuclides. This also will require further study to understand the source of these biases.

Comparisons of calculated values with measured values from blanket subassembly U1302 are encouraging. The initial results indicate proposed depletion methodology based on use of generic EBR-II models to simulate the irradiation history of the blanket subassemblies will be adequate to meet operations and MC&A requirements for processing these blanket subassemblies in FCF.

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