ANL/RA/CP-96362

Paper to be presented at American Nuclear Society Third Topical Meeting DOE Spent Nuclear Fuel and Fissile Material Management, Charleston, South Carolina, September 8-11, 1998

CHARACTERIZATION OF SPENT EBR-II DRIVER FUEL*

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*Work supported by the U.S. Department of Energy, Nuclear Energy Programs under contract W-31-109-ENG-38.

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ABSTRACT

Operations and material control and accountancy requirements for the Fuel Conditioning Facility demand accurate prediction of the mass flow of spent EBR-II driver fuel into the facility. This requires validated calculational tools that can predict the burnup and isotopic distribution in irradiated Zr-alloy fueled driver assemblies. Detailed core-follow depletion calculations have been performed for an extensive series of EBR-II runs to produce a database of material inventories for the spent fuel to be processed. As this fuel is processed, comparison of calculated values with measured data obtained from samples of this fuel is producing a growing set of validation data. A more extensive set of samples and measurements from the initial processing of irradiated driver fuel has produced valuable estimates of the biases and uncertainties in both the measured and calculated values. Results of these comparisons are presented herein and indicate the calculated values adequately predict the mass flows.

1. INTRODUCTION

The EBR-II Spent Fuel-Treatment Demonstration Project will include treatment of 100 spent driver assemblies in the Fuel Conditioning Facility (FCF).[1] The irradiated subassemblies are dismantled; elements containing Zr-alloy fuel are chopped in the element chopper; and segments of the chopped elements are loaded into fuel dissolution baskets for processing in the electrorefiner. Operation of the FCF requires accurate estimates of the masses, compositions, decay heat and activity of these chopped fuel segments. Theoretical or calculated values are used initially for process operations and material control and accountancy. As part of the chopping operation in FCF, selected segments of the chopped irradiated fuel are saved and sent to the Analytical Laboratory for chemical analysis. Typically, this involves saving a single chopped segment (plus an additional adjacent segment retained for backup) taken near the axial midplane of the fueled section of the central fuel element within the subassembly being processed. Routine measurements on these samples might include U and Pu mass, U and Pu isotopic fractions, and lanthanum (La) mass to determine burnup. The measured values are used to confirm or, if necessary, replace the theoretical values being used by FCF operations. This paper focuses on comparisons of calculated and measured values in the characterization of the irradiated EBR-II driver fuel.

2. ANALYSES OF INITIAL IRRADIATED FUEL SAMPLES

In processing the first few irradiated subassemblies in FCF, it was possible to request an increased number of samples and measurements. Nine samples (plus 9 backup samples) were obtained from each of the first two subassemblies (C2954V and C2956V) processed. The nine samples consisted of 3 axial samples (bottom, center, top) from the innermost (closest to the core center) element, 3 axial samples (B, C, T) from the central element, and 3 axial samples (B, C, T) from the outermost (farthest from core center) element. This large number of samples provided a unique opportunity to measure spatial distributions of the nuclide inventories in these assemblies and to compare these distributions with calculated values. These comparisons were particularly helpful in assessing the reproducibility and uncertainties in the measurements, as well as the biases and uncertainties in the calculated values.

A diverse set of measurements was performed on the initial samples of irradiated fuel. These measurements included:

- Sample Mass
- U Mass / Sample Mass
- Pu Mass / Sample Mass
- Zr Mass / Sample Mass
- Na Mass / Sample Mass
- Burnup (via determination of La 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, and ^{95}Zr)

The U and Pu masses and the U and Pu isotopic fractions are experimentally determined by isotopic dilution mass spectroscopic (IDMS) analyses. The measurement of La content is accurately determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) measurements.[2] This technique eliminates the separation of

the fission product (or burnup indicator) by either ion exchange or solvent extraction and subsequent measurement by IDMS methods. The ICP-AES measurements have a number of advantages, including (i) much shorter time and not so labor intensive, (ii) reduced waste generation, and (iii) no mixed waste generation.

Details of the calculations, which are based on the REBUS-3/RCT/ORIGEN-RA methodologies, have been given in Refs. 3 and 4.

Table I gives results of measured and calculated values of the U content, Pu content and burnup (via La content) obtained from the 18 chopped fuel segment samples from the first 2 irradiated driver subassemblies.* It may be noted from these values that mean calculated-to-measured (C/M) values are quite good, although a few discrepant values tend to widen the range of these values. The mean C/M bias for the U content is ~1.3% low; the mean C/M bias for the Pu content is ~2.6% high. These biases are consistent with the observed overprediction of the burnup by ~5.6%. Furthermore, these values are consistent with the validation results presented in Refs. 3 and 4.

It should be emphasized that the observed bias for the calculated values of burnup are very small from a programmatic perspective. The uncertainties in the heavy metal mass flows in FCF have a component based on their pre-irradiation uncertainties and a component based on burnup uncertainties. The significance of a 5-6% bias or uncertainty in burnup is largely mitigated by the fact that the average burnup in the fuel is only ~8%. Given that the initial enrichment uncertainty in the fuel is ±1%, that 92% of the heavy atoms are unchanged by irradiation, and that the uncertainty in the burnup (of the 8% of the fuel which is changed by irradiation) in ±6%, the uncertainty in the discharge enrichment is only increased to ~1.1%. That is, the uncertainty in the isotopic composition of the irradiated fuel is dominated by the assay uncertainty of the fresh fuel.

It is interesting to note the spatial variation of the U, Pu, and burnup values presented in Table I. This is most conveniently done by displaying these data graphically. Consider the calculated and measured values of the U content displayed in Figures 1.A and 1.B, respectively. The distinct axial peaking of the flux and burnup near the midplane is evident by the lower U values in the axial center samples. The measured U values, though exhibiting slightly more variation than the calculated values, consistently display this trend. The radial gradient of the flux (and burnup) across these inner core subassemblies (S/A pitch ~ 2 inches) is only ~ 5%. The resultant radial gradient in the calculated U content is barely observed in the scale of Figure 1.A. Measured values of the U content for the bottom samples of the middle (K449) and outer (K286) elements of C2954V appear too high relative to the other samples (which is confirmed by the low C/M values for these samples).

Consider the calculated and measured values of the Pu content displayed in Figures 2.A and 2.B, respectively. Again the spatial trends are very consistent for both the calculated and measured values. Only the center sample from the inner (K353) element of C2954V appears slightly inconsistent (perhaps 2-3% low).

The calculated and measured values of the burnup are displayed in Figures 3.A and 3.B, respectively. Again most of the measured values display consistent trends. However, in this case, a few of the measured values are clearly inconsistent. For example, the first measured value displayed in this figure (bottom sample of inner element of C2954V) is smaller (by ~12%) than corresponding (bottom) samples from the other two elements. After analysis of these burnup results, burnup measurements were performed on 4 backup samples (corresponding to 4 of the most discrepant values). Results based on these backup samples were not only much more consistent with the calculated values (within ~2-4%) but also much more consistent with the other measured values.

It should be emphasized the measurements on the backup samples were new measurements on new samples (and not a repeat of measurements on the previous samples). The improved agreement of the backup sample results with both the measured and calculated values indicates the measurements on the backup samples are the correct values for these samples, but does not identify the source of the problems with the original values. Those poor results could have resulted from the procedures of the Analytical Laboratory. However, it appeared much more likely these problems resulted from the original samples. There were significant differences in the size, weight, and weight fractions between the original and backup sample. The chopping of the samples and the subsequent handling of the samples appears to be the primary source of variability in the "measured" values.

Measured and theoretical values of the uranium isotopic fractions of the irradiated samples from C2954V and C2956V are shown in Table II. The measurement uncertainties (1σ) are quoted as 0.25% for ^{235}U and ^{238}U,

* Sample results will consistently be presented in all tables and figures in this paper in the following order: 9 samples for C2954V – bottom, center and top for innermost element, B,C, T for central element, and B,C, T for outermost element; followed by 9 samples for C2956V – bottom, center and top for innermost element, B,C, T for central element, and B,C, T for outermost element. This consistent ordering should facilitate observation of trends in radial (Inner, Middle, Outer) and axial (B, C, T) distributions.

1.0% for ^{234}U, and 0.5% for ^{236}U. Agreement between the measured and theoretical values is very good. All of the C/M values for the $^{235}U/U$ fraction are less than or equal to 1.000. These values range from 0.997 – 1.000 with a mean value of 0.999 ± 0.001. All of the C/M values for the $^{238}U/U$ fraction are greater than 1.000. These values range from 1.001 – 1.005 with a mean value of 1.003 ± 0.001. The underprediction of the $^{235}U/U$ fraction and the overprediction of the $^{238}U/U$ fraction by the calculations are consistent with the underprediction of the uranium content in the samples by the calculations (noted above) and with the overprediction of the plutonium content in the samples by the calculations (noted above) – all of which are consistent with an overprediction of the burnup by the calculations.

These comparisons of calculated values with measured values indicate agreement would be improved by lowering the calculated burnup (i.e., reducing the power or flux in the calculations). Renormalized calculations have also been performed by adjusting the flux level in the ORIGEN-RA depletion calculations (by multiplying the one-group flux values by the M/C ratio of the burnup values). This renormalization procedure largely eliminated the C/M bias on burnup (yielding a mean C/M value of 1.0010 for burnup), although there remained some residual bias on the U content (-1.1%) and Pu content (-2.6%). It appears that an adjustment of both the flux level and the ^{238}U capture cross section would provide a means of normalizing the calculated values of burnup and Pu buildup with the measured values.

3. SUMMARY

The availability of calculated and measured results for 18 samples spatially distributed throughout 2 subassemblies has provided a unique opportunity to test the consistency of these values. These comparisons indicate a consistent bias of ~5-6% overprediction of burnup by the calculations. The "sample measurements" predict U and Pu content to ~1-2%, burnup to ~4-5%, and ^{235}U isotopic fractions to ~0.1%.

Further accumulation of measured data and the C/M comparisons for these chopped fuel segment samples as irradiated fuel is processed in FCF will likely improve our understanding of these small C/M biases. The present and continuing validation effort provides confidence both that the calculated values predict input mass flows to FCF accurately for operations and material control and accountancy needs and that the measured values accurately provide verification of those values.

4. REFERENCES

1. R. W. Benedict and S. P. Henslee, "The EBR-II Spent-Fuel Demonstration Project," *Trans. Am. Nucl. Soc.*, 77, 75-76 (1997).

2. P. Goodall and S. G. Johnson, "High-Resolution Inductively-Coupled Plasma-Atomic Emission-Spectrometry for the Determination of Burnup in Spent Nuclear Fuel," *Applied Spectroscopy*, Vol. 51, Issue 2, 182-187 (1997).

3. R. D. McKnight, J. A. Stillman, B. J. Toppel, H. S. Khalil, "Validation and Application of a Physics Database for Fast Reactor Fuel Cycle Analysis," Proc. of Topical Meeting on Advances in Reactor Physics, April 11-15, 1994, Knoxville, TN, Vol. I, 150-159 (April 1994).

4. R. D. McKnight, "Validation of the REBUS-3/RCT Methodologies for EBR-II Core-Follow Analysis," Proc. of Topical Meeting on Advances in Reactor Physics, March 8-11, 1992, Charleston, SC, Vol. 2, 69-80 (1992).

Table I. Measurement and Calculation of U Content, Pu Content and Burnup in Chopped Segment Samples from Driver Assemblies C2954V and C2956V

S/A	Fuel Element	Sample Location*	gms U / gram Sample			mgs Pu / gram Sample			Burnup, a/o (via La)		
			Meas.	Calc.	C / M	Meas.	Calc.	C / M	Meas.	Calc.	C / M
C2954V	K353	B	0.661	0.660	0.999	3.098	3.209	1.036	7.1875	8.4203	1.1715
C2954V	K353	C	0.640	0.651	1.017	3.075	3.240	1.054	8.5579	9.5080	1.1110
C2954V	K353	T	0.673	0.669	0.994	2.652	2.692	1.015	6.6373	7.1941	1.0839
C2954V	K449	B	0.694	0.661	0.953	3.103	3.128	1.008	8.0974	8.2303	1.0164
C2954V	K449	C	0.666	0.651	0.978	3.116	3.171	1.018	9.0546	9.3149	1.0288
C2954V	K449	T	0.675	0.669	0.992	2.565	2.627	1.024	6.5656	7.0348	1.0715
C2954V	K286	B	0.690	0.662	0.960	3.055	3.049	0.998	7.7794	7.9955	1.0278
C2954V	K286	C	0.661	0.653	0.988	3.026	3.092	1.022	8.4611	9.0154	1.0655
C2954V	K286	T	0.673	0.671	0.996	2.552	2.575	1.009	6.4900	6.8301	1.0524
C2956V	K347	B	0.674	0.661	0.982	3.139	3.190	1.016	8.0661	8.2269	1.0199
C2956V	K347	C	0.667	0.653	0.978	3.168	3.203	1.011	8.3905	9.2744	1.1054
C2956V	K347	T	0.670	0.669	0.997	2.608	2.710	1.039	7.1260	7.1486	1.0032
C2956V	K592	B	0.678	0.661	0.975	2.981	3.103	1.041	7.2381	7.8985	1.0912
C2956V	K592	C	0.650	0.653	1.005	2.946	3.089	1.048	9.0957	8.8060	0.9682
C2956V	K592	T	0.673	0.668	0.994	2.516	2.648	1.052	6.4978	6.8685	1.0570
C2956V	K579	B	0.675	0.661	0.980	2.991	3.060	1.023	7.0645	7.6089	1.0771
C2956V	K579	C	0.666	0.654	0.982	2.950	3.003	1.018	8.0958	8.3373	1.0298
C2956V	K579	T	0.669	0.669	1.000	2.513	2.614	1.040	6.4184	6.6102	1.0299
		Min.	0.640	0.651	0.953	2.513	2.575	0.998	6.4900	6.8301	0.9682
		Max.	0.694	0.671	1.017	3.168	3.240	1.054	9.0957	9.5080	1.1715
		Range	0.054	0.020	0.064	0.656	0.665	0.056	2.6057	2.6780	0.2034
		Mean	0.670	0.661	0.987	2.892	2.967	1.026	7.6013	8.0179	1.0561
		Std. Dev.	0.012	0.007	0.016	0.245	0.244	0.017	0.9085	0.9314	0.0472

* Axial Location of Sample: B = bottom, C = center, T = top.

Table II. Measurement and Calculation of U Isotopic Fractions in Chopped Segment Samples from Driver Assemblies C2954V and C2956V

S/A	Fuel Element	Sample Location*	Measured				Calculated				Calculated / Measured			
			^{234}U w/o	^{235}U w/o	^{236}U w/o	^{238}U w/o	^{234}U w/o	^{235}U w/o	^{236}U w/o	^{238}U w/o	^{234}U w/o	^{235}U w/o	^{236}U w/o	^{238}U w/o
C2954V	K353	B	0.74	62.78	2.09	34.38	0.74	62.74	2.07	34.44	0.998	0.999	0.992	1.002
C2954V	K353	C	0.74	62.57	2.11	34.58	0.74	62.40	2.10	34.76	0.997	0.997	0.995	1.005
C2954V	K353	T	0.74	63.49	1.81	33.97	0.74	63.42	1.77	34.07	0.996	0.999	0.978	1.003
C2954V	K449	B	0.74	62.93	2.02	34.31	0.74	62.84	2.03	34.39	0.998	0.999	1.003	1.002
C2954V	K449	C	0.75	62.64	2.07	34.54	0.74	62.50	2.06	34.71	0.984	0.998	0.995	1.005
C2954V	K449	T	0.74	63.51	1.76	33.98	0.74	63.50	1.73	34.03	0.996	1.000	0.985	1.001
C2954V	K286	B	0.74	63.03	1.99	34.24	0.74	62.97	1.98	34.32	0.994	0.999	0.994	1.002
C2954V	K286	C	0.74	62.78	2.03	34.45	0.73	62.64	2.01	34.61	0.993	0.998	0.990	1.005
C2954V	K286	T	0.74	63.60	1.75	33.91	0.73	63.60	1.70	33.97	0.991	1.000	0.973	1.002
C2956V	K347	B	0.74	62.93	2.11	34.22	0.73	62.90	2.07	34.30	0.980	1.000	0.981	1.002
C2956V	K347	C	0.75	62.73	2.11	34.41	0.72	62.60	2.08	34.59	0.965	0.998	0.988	1.005
C2956V	K347	T	0.74	63.59	1.81	33.86	0.72	63.52	1.79	33.96	0.977	0.999	0.989	1.003
C2956V	K592	B	0.74	63.16	2.02	34.07	0.73	63.05	2.03	34.20	0.984	0.998	1.003	1.004
C2956V	K592	C	0.74	62.93	2.03	34.29	0.73	62.79	2.03	34.46	0.983	0.998	0.998	1.005
C2956V	K592	T	0.74	63.72	1.76	33.79	0.73	63.63	1.76	33.88	0.981	0.999	1.000	1.003
C2956V	K579	B	0.74	63.24	2.02	34.01	0.73	63.17	2.01	34.09	0.985	0.999	0.994	1.002
C2956V	K579	C	0.74	63.12	2.00	34.14	0.73	62.98	1.98	34.31	0.984	0.998	0.991	1.005
C2956V	K579	T	0.74	63.73	1.76	33.77	0.73	63.73	1.75	33.80	0.983	1.000	0.992	1.001
		Min.	0.740	62.570	1.750	33.790	0.723	62.399	1.703	33.883	0.965	0.997	0.973	1.001
		Max.	0.750	63.730	2.110	34.580	0.739	63.728	2.100	34.764	0.998	1.000	1.003	1.005
		Range	0.010	1.160	0.360	0.790	0.016	1.329	0.397	0.881	0.033	0.003	0.030	0.004
		Mean	0.741	63.138	1.958	34.162	0.732	63.055	1.942	34.272	0.987	0.999	0.991	1.003
		Std. Dev.	0.003	0.385	0.139	0.255	0.006	0.421	0.143	0.291	0.009	0.001	0.008	0.001

* Axial Location of Sample: B = bottom, C = center, T = top.

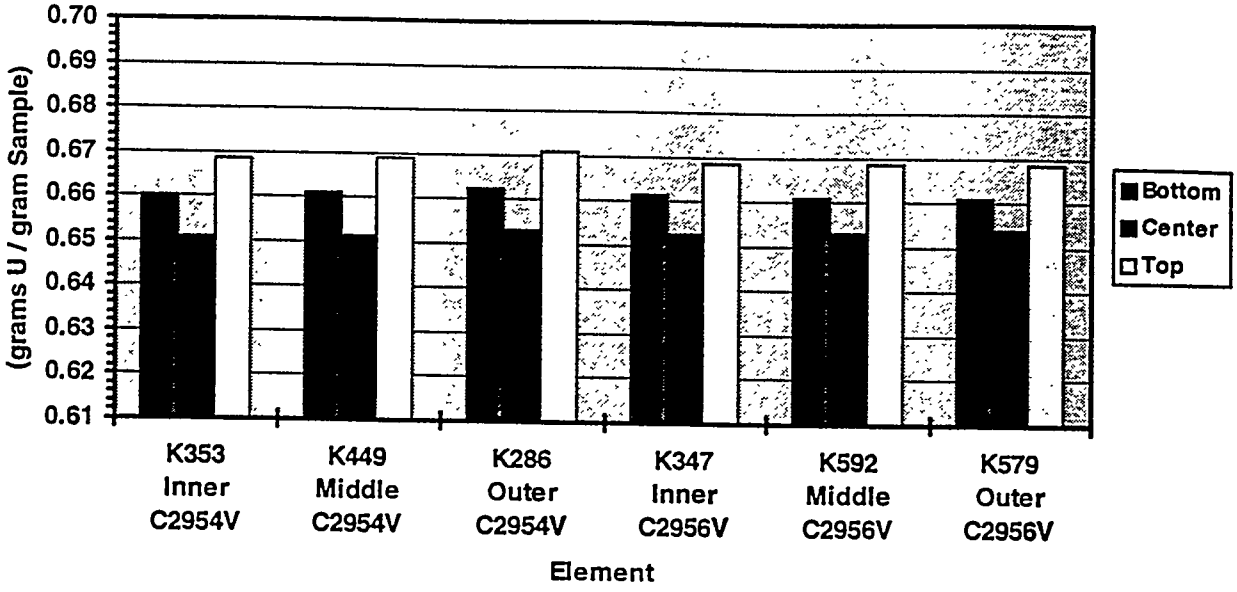

Figure 1.A. Calculated Uranium Content (grams U/gram Sample) in Irradiated Samples from C2954V and C2956V.

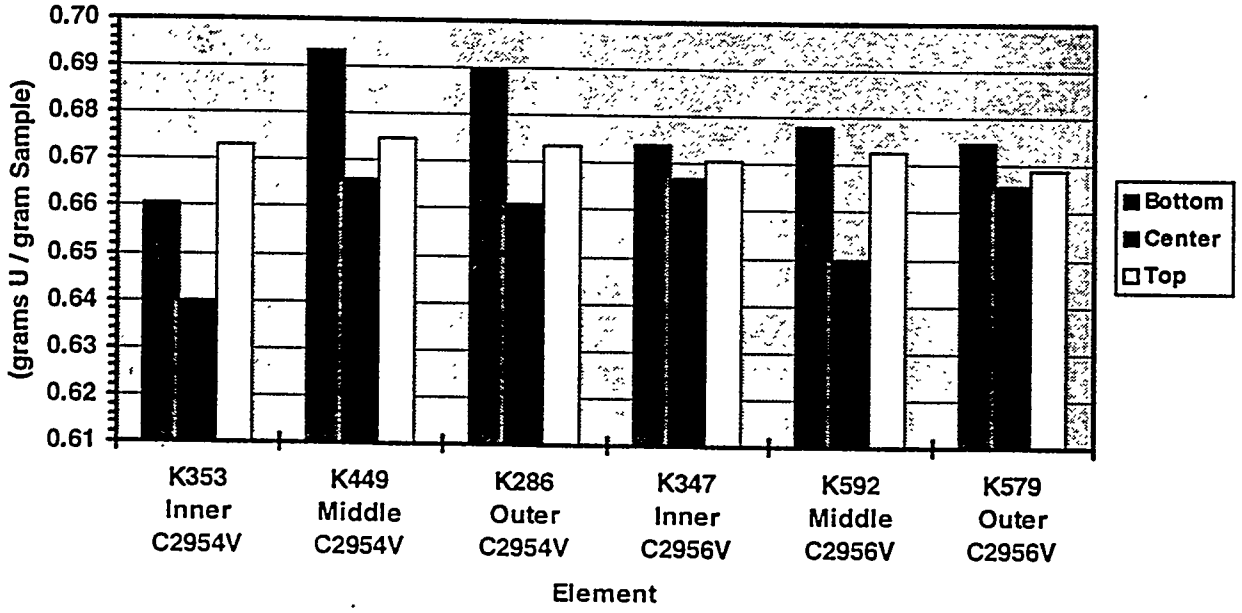

Figure 1.B. Measured Uranium Content (grams U/gram Sample) in Irradiated Samples from C2954V and C2956V.

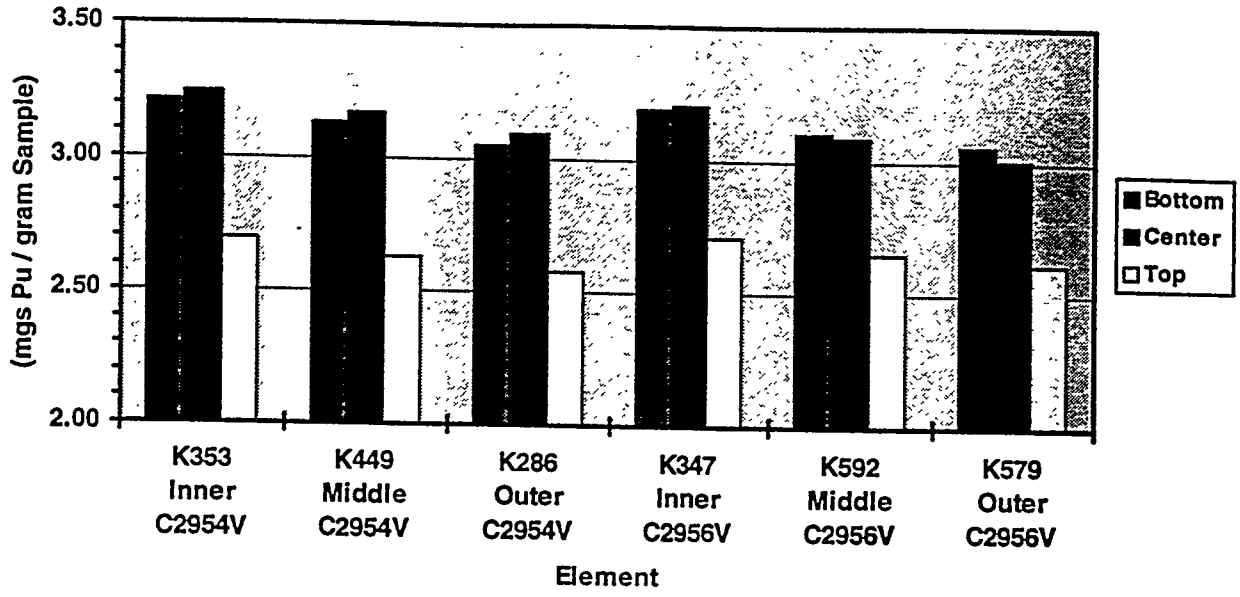

Figure 2.A. Calculated Plutonium Content (mgs Pu/gram Sample) in Irradiated Samples from C2954V and C2956V.

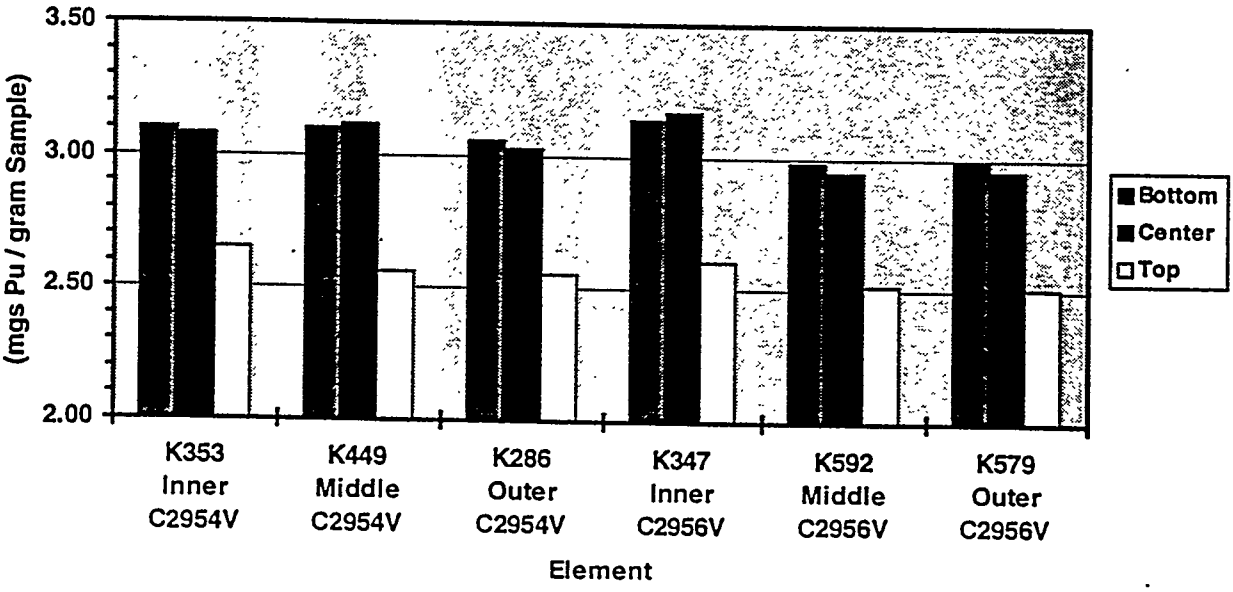

Figure 2.B. Measured Plutonium Content (mgs Pu/gram Sample) in Irradiated Samples from C2954V and C2956V.

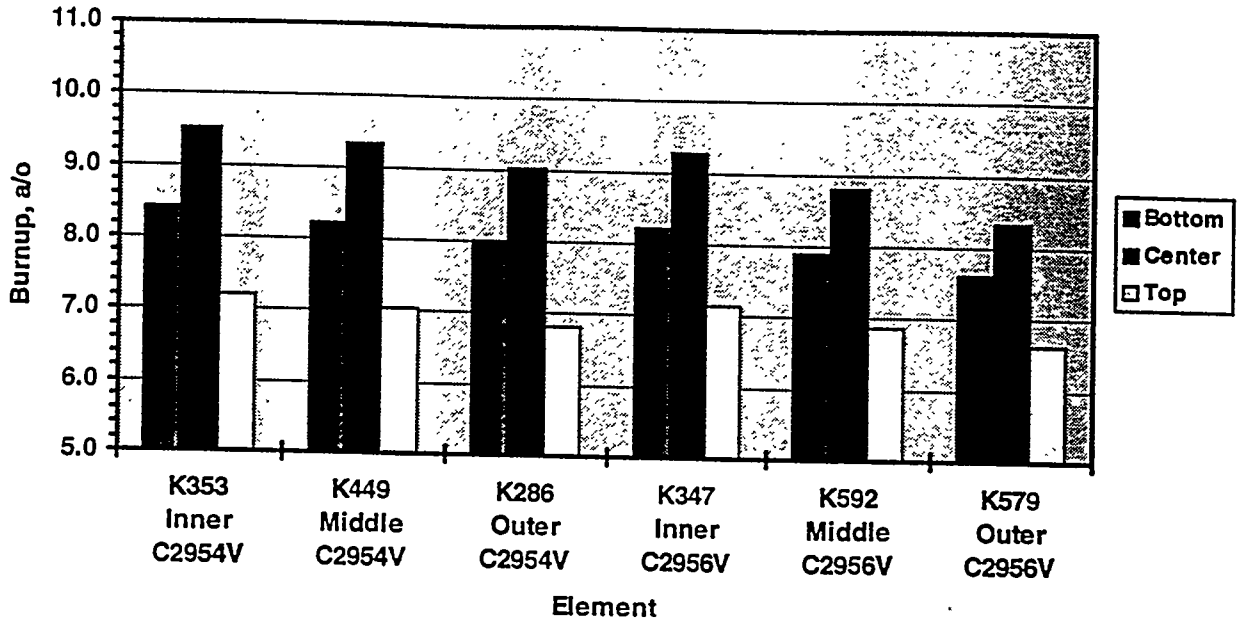

Figure 3.A. Calculated Burnup (atom percent) in Irradiated Samples from C2954V and C2956V.

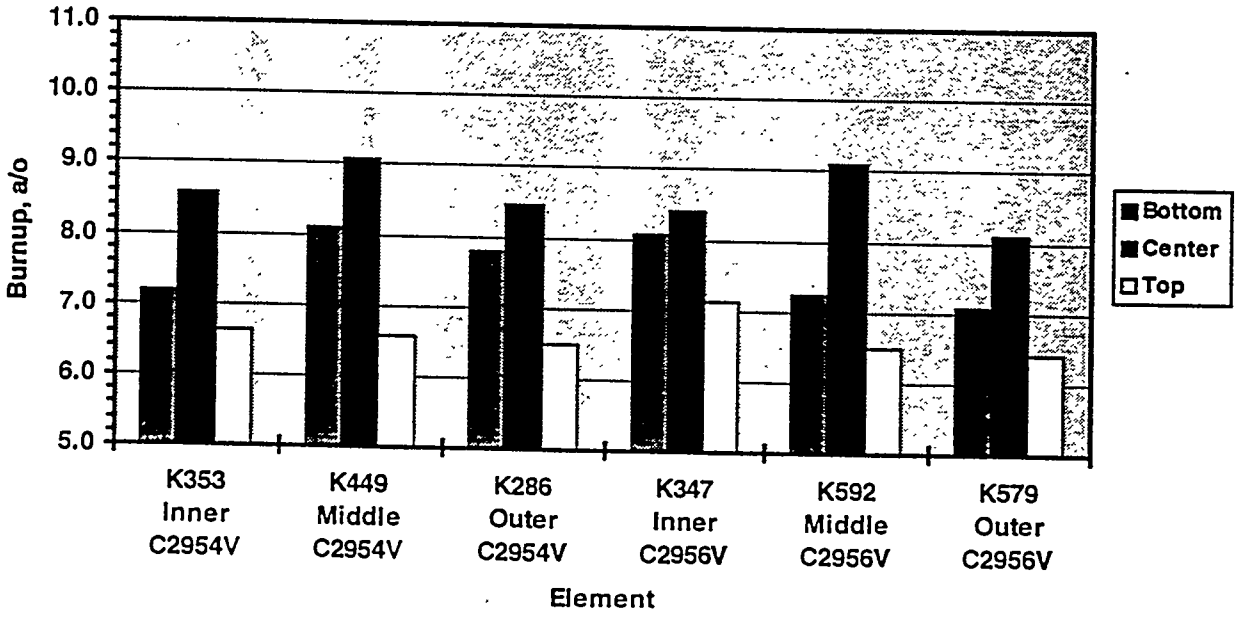

Figure 3.B. Measured Burnup (atom percent) Determined via La in Irradiated Samples from C2954V and C2956V.