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**HIGH EXPOSURE PLUTONIUM STUDIES
ANALYSES OF SHIPPINGPORT FUEL**

R. C. SMITH AND H. H. VAN TUYL

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ABSTRACT

Detailed analyses on seven samples of Shippingport blanket fuel are presented. The plutonium and uranium isotopic distributions are given for Shippingport blanket fuels varying in estimated exposure from approximately 10,000 to 30,000 MWd/tonne. Calculated amounts of the uranium and plutonium isotopes and several other heavy nuclides of potential commercial use are also presented.

These data are presented as a part of a long-range study to learn the isotopic distribution and radiation exposures to be expected from high exposure plutonium recycled from power reactor fuels. The data fall into consistent patterns. Fuel burnup as calculated from the measured amounts of the various nuclides is compared to ^{137}Cs and ^{148}Nd burnup calculations. Some of the first preliminary dosimetry data from the high exposure plutonium are also included.

INTRODUCTION

Under the Plutonium Utilization Program, Battelle-Northwest has undertaken a study of high exposure plutonium from representative pressurized water and boiling water reactors in the United States. The primary objective of the study is to better define the radiation hazards which fuel fabricators will encounter in the storage and handling of plutonium recovered from spent power reactor fuels. Secondary objectives include: (1) determination of the nuclide composition of long exposure fuels, (2) determination of shielding requirements during fuel fabrication, and (3) improvement in high exposure plutonium handling and fuel fabrication technology. The data evolved

will also be used in the formulation and testing of computer programs for predicting the isotopic composition of fuels from known reactor loadings and irradiation exposure history and shielding requirements and the radiation levels to be expected from plutonium of known isotopic composition.

Analyses of fuel from Shippingport, Yankee, Dresden, and Hanford reactors are presently included in this study and analyses are planned on additional United States reactor fuels with exposures exceeding 20,000 MWd/tonne.

The first fuel obtainable in sufficient quantity with the exposure history required by the objectives of the program was acquired from the Shippingport pressurized water reactor. This document reports by individual nuclides

the analytical data obtained on seven selected samples of fuel ranging in exposure from approximately 10,000 to 30,000 MWd/tonne.

SUMMARY AND CONCLUSIONS

This report is preliminary in nature because the dosimetry, shielding, and fuel fabrication studies with Shippingport fuel are not yet complete.

Due to the preliminary character of this report it is too early to draw many conclusions. It is believed that the data obtained on the Shippingport fuel fall into consistent patterns. If the remaining measurements and the isotopic data from other power reactor fuels correlate as well as the present information, the study will provide very valuable relationships between the isotopic content, radiation hazards in recycled plutonium fuel fabrication, and burnup-isotopic content predictions for power reactor fuels.

Comparisons with fuel from other pressurized water and boiling water reactors now under study should provide data to allow accurate predictions of isotopic contents, expected radiation dose rates for recycle plutonium, and transplutonium element composition of high burnup power reactor fuels of known reactor history.

DISCUSSION

SHIPPINGPORT FUEL DESCRIPTION

The Shippingport reactor is a pressurized water, seed and blanket type. The blanket fuel elements used in this study contained natural uranium as UO_2 pellets fabricated into

approximately 10 in. long rods clad in Zircalloy-2.

The entire first Shippingport blanket, designated as Core I, was left in the reactor for four seed lifetimes. The fuel element arrangement after Seed IV is shown in Figure 1. The blanket elements are arranged both inside and outside of the seed. The fuel samples selected for analysis were taken from three blanket elements in an effort to evaluate typical fluxes and neutron spectra in the blanket and to cover the middle and upper ranges of exposure. The elements selected were No. 45, which had seed on two sides of the element, No. 14, which had seed on one side, and No. 6, which is one element removed from the seed.

Figure 2 shows the numbering system for the 120 rods within the fuel bundle. The bottom right hand corner position does not contain a fuel rod. Seven fuel bundles are stacked vertically to form a fuel element. Analytical samples are identified by three numbers. The first number designates the element, the second number specifies the fuel bundle in the element, and the last number specifies the rod location in the fuel bundle. For example, 45-2-21 corresponds to Fuel Element 45 located in reactor position E6, the second fuel bundle down from the top of the element; and rod 21 which is on the outside row, the second rod from the top right hand corner. In most Westinghouse Bettis literature, the fuel bundles are identified by numbering from the bottom of the element. Their identification would therefore be E6-6-21 for this fuel rod.

	2	3	4	5	6	7	8	9	10	11	12	13	14
B				99	12	102	76	92	125	77			
C			74	101	95	70	85	65	51	48	119		
D		73	75								82	98	
E	59	79			45	23	36	62	39			108	90
F	124	103		25	16	26	15	37	21	44		78	130
G	88	57		38	11	2	17	1	22	19		87	113
H	83	107		29	5	133	123	128	52	114		115	94
I	64	106		13	28	27	20	55	8	14		49	105
J	129	56		40	34	12	6	9	18	24		72	69
J	116	81			32	30	46	42	10			100	118
L		54	68								10	110	
M			109	104	121	71	117	97	61	86	66		
N				96	7	63	58	62	91	111			

*FIGURE 1. Fuel Element Arrangement
After Seed IV*

120	109	98	87	76	65	54	43	32	21	10
119	108	97	86	75	64	53	42	31	20	9
118	107	96	85	74	63	52	41	30	19	8
117	106	95	84	73	62	51	40	29	18	7
116	105	94	83	72	61	50	39	28	17	6
115	104	93	82	71	60	49	38	27	16	5
114	103	92	81	70	59	48	37	26	15	4
113	102	91	80	69	58	47	36	25	14	3
112	101	90	79	68	57	46	35	24	13	2
111	100	89	78	67	56	45	34	23	12	1
110	99	88	77	66	55	44	33	22	11	

*FIGURE 2. Rod Numbering Within a
Fuel Bundle*

There are two characteristics of this fuel that are not typical of most power reactors in the United States. First, the proportion of higher nuclides is indicative of the somewhat unusual character of the natural uranium blanket type reactor. This is brought about because although the average blanket exposure was only 11,000 MWd/tonne, the integrated neutron flux was considerably higher than would have been necessary for the same exposure with enriched fuels. This higher integrated neutron flux is important in the production of the heavier nuclides.

A second characteristic of the Shippingport blanket, and one which promotes uncertainties in calculational correlations, is the large variation in both flux and neutron energy spectra across the blanket. This characteristic is probably best illustrated by the range of burnup values, which extend from about 10,000 to 30,000 MWd/tonne in the selected fuel samples from this one core. Other regions could have been selected to be representative of slightly higher or much lower exposures.

To evaluate the expected range of fuel exposures, burnup was estimated as a function of location within the reactor. The radial power level distribution was deduced from data in WAPD-PWR-TE-141 and translated to a fraction of total core power in a single quadrant of the core. Axial (vertical) power level distribution was estimated based on a plot of the relative thermal flux versus height in the reactor. These data were combined to obtain an approximate average

burnup for each of the 791 fuel bundles within the reactor.

Calculations for the desired exposure ranges were then made with program ISOGEN⁽¹⁾ for the production of actinides in a rather well moderated power reactor, with a fixed ratio of resonance to thermal neutron flux. Most of the cross section data were taken from Westcott's tabulation, but some more recent data were also used. The results from some uranium, plutonium, and transplutonium nuclides are shown in Figures 3, 4, and 5. The calculated values compare well with present samples and with published Westinghouse data from analytical samples taken at the end of Seed II life.

FIGURE 3. Uranium Isotopes in Shippingport Fuel

FIGURE 4. Plutonium Isotopes in Shippingport Fuel

FIGURE 5. Transplutonium Isotopes in Shippingport Fuel

ANALYTICAL DATA

Three primary types of raw data are taken under the scope of this study: nuclide analysis, beta gamma surface dose rates and energy spectra, and neutron dosimetry. This report will mainly be concerned with the isotopic and chemical analysis. Seven samples were analyzed covering an exposure range (as estimated by Bettis from Shippingport reactor and analytical data) of 9500 MWd/tonne to 30,900 MWd/tonne. The chemical and radio-chemical analysis data are summarized in Table I; the analytical methods used and uncertainty levels are listed in Table II.

The analyses were converted from atom percent to weight percent, and then translated to total weight per initial metric ton of uranium. In making this transition it was necessary to determine the amount of uranium lost during irradiation. This was done by assuming the fission of one gram of uranium (or uranium daughters) per megawatt-day exposure, with the approximate exposure for this purpose being determined from the ^{137}Cs ratio to final uranium. The other loss of uranium was in the production of trans-uranics. The total loss was then the sum of the fission product weight and the weight of the transuranics

TABLE I. Shippingport Fuel Analyses Summary

Analyses	45-2-21	6-5-60	45-2-60	14-4-60	45-2-111	14-4-115	14-4-5
Total U, g/liter	183.55	154.52	126.53	103.54	154.40	103.93	131.19
Total Pu, g/liter	1.320	0.963	0.670	0.887	0.962	0.827	1.174
²³⁸ U, at. %	99.678	99.650	99.765	99.747	99.721	99.816	99.856
²³⁶ U, at. %	0.079	0.074	0.088	0.090	0.084	0.095	0.098
²³⁵ U, at. %	0.238	0.271	0.143	0.121	0.191	0.086	0.043
²³⁴ U, at. %	0.0036	0.0042	0.0039	0.0036	0.0040	0.0031	0.0029
²³⁶ Pu, at. %	9.4×10^{-7}	6.2×10^{-7}	3.6×10^{-7}	1.2×10^{-6}	5.7×10^{-7}	8.0×10^{-7}	1.8×10^{-6}
²³⁸ Pu, at. %	0.60	0.47	0.56	0.90	0.66	1.06	1.55
²³⁹ Pu, at. %	65.8	67.8	55.5 55.43R	59.3	60.4	51.2	45.9
²⁴⁰ Pu, at. %	22.4	22.0	31.0 31.1R	25.4	26.2	28.9	30.1
²⁴¹ Pu, at. %	8.60	7.67	8.49 8.43R	10.10	9.11	11.04	11.39
²⁴² Pu, at. %	2.61	2.08	4.44 4.46R	4.31	3.68	7.78	11.12
²⁴¹ Am, g/liter	0.028	0.018	0.013 0.012R	0.019	0.021	0.022	0.030
²⁴² Cm, g/liter	1.68×10^{-5}	9.8×10^{-6}	9.72×10^{-6} R 1.02×10^{-5}	1.66×10^{-5}	1.52×10^{-5}	2.32×10^{-5}	4.0×10^{-5}
²⁴⁴ Cm, g/liter	8.4×10^{-4}	3.19×10^{-4}	3.27×10^{-4} 3.03×10^{-4} R	1.55×10^{-3}	6.36×10^{-4}	2.97×10^{-3}	1.11×10^{-2}
²³⁷ Np, g/liter		0.0139	0.0112	0.00739	0.0151	0.0109	0.0174
¹³⁷ Cs, atoms/liter	3.27×10^{20}	2.22×10^{20}	2.34×10^{20}	2.65×10^{20}	2.85×10^{20}	3.26×10^{20}	5.66×10^{20}
¹⁴⁸ Nd, atoms/liter	9.24×10^{19}					9.13×10^{19}	1.61×10^{20}

"R" indicates rerun on sample.

TABLE II. *Summary of Analytical Methods and Uncertainty Levels*

<u>Determination</u>	<u>Analytical Method</u>	<u>Ref.</u>	<u>Standard Deviation Relative Percent</u>
Total U and Pu	Isotopic dilution	3	±0.8
Total ^{137}Cs	Tetraphenyl boron extraction, then selective gamma counting	4	±1.5
Total ^{241}Am and ^{244}Cm	Ion exchange, then selective alpha counting	5	±4.0
Total ^{242}Cm	Ion exchange, then selective alpha counting	5	±5.0
Total ^{243}Am	Mass spectrometry + total ^{241}Am	5	±5.0
Total ^{237}Np	TTA-TNOA extraction, then selective alpha counting	6	±2.0
<u>Uranium Isotopic Composition</u>			
^{234}U	Mass spectrometry	3	±5.0
^{235}U	Mass spectrometry	3	±0.5
^{236}U	Mass spectrometry	3	±1.0
^{238}U	Mass spectrometry	3	±0.05
<u>Plutonium Isotopic Composition</u>			
^{239}Pu	Mass spectrometry	3	±0.1
^{240}Pu	Mass spectrometry	3	±0.2
^{241}Pu and ^{242}Pu	Mass spectrometry	3	±0.5
^{236}Pu	Alpha energy analysis	3	±20.0
^{238}Pu	Alpha energy analysis	3	±2.0
<u>Americium Isotopic Composition</u>			
^{241}Am and ^{243}Am	Mass spectrometry	7	±2.0

produced. This evaluation is not exact, but it is sufficiently close for the present purposes. The corrected amounts converted to a basis of initial metric tons of uranium are shown in Table III.

In Table IV, estimated exposures of the various fuel samples are compared. Calculations made using the common burnup nuclides, such as ^{137}Cs and ^{235}U , were compared with calculations from several of the other nuclides

TABLE III. Heavy Nuclide Content per Initial Tonne of Shippingport Fuel

Sample Number	%U, Remain	Total Pu	Amount of Nuclide per Initial Tonne of Uranium - Grams												
			^{234}U	^{235}U	^{236}U	^{236}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{237}Np	^{241}Am	^{242}Cm	^{244}Cm
6-5-60	98.5	6138	41	2636	723	3.8×10^{-5}	29	4153	1353	474	129	88.6	112.2	0.0625	2.033
45-2-21	98.1	7056	35	2305	768	6.5×10^{-5}	42	4633	1584	611	186		157.0	0.0935	4.675
45-2-111	98.2	6116	39	1852	818	3.4×10^{-5}	40	3684	1605	560	227	96.0	132.9	0.0967	4.045
45-2-60	98.3	5204	35	1165	930	1.8×10^{-5}	29	2881	1616	444	233	87.0	101.8	0.0792	2.540
45-2-60R								2813	1585	431	230		96.3	0.0755	2.354
14-4-60 ^(a)	97.5	8318	34	1540	870	1.0×10^{-4}	74	4920	2116	845	362	69.6	175.1	0.1563	14.60
14-4-115 ^(a)	97.2	7732	30	825	916	6.0×10^{-5}	80	3904	2213	849	601	103.9	206.7	0.2170	27.78
14-4-5	96.4	8628	28	408	937	1.5×10^{-4}	133	3944	2597	987	968	129.0	217.5	0.2939	81.56

(a) Sample numbers 14-4-60 and 14-4-115 were reversed in initial data due to mislabeling

TABLE IV. Estimated Exposures of Shippingport Samples

Sample Number	Estimated Exposure, Thousands of MWd/MTU										
	¹³⁷ Cs	¹⁴⁸ Nd	²³⁴ U	²³⁵ U	²³⁶ U	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴¹ Am	²⁴² Cm	²⁴⁴ Cm
6-5-60	9.3		10.0	8.8	8.2	8.8	9.3	9.3	10.0	9.3	10.3
45-2-21	12.0	11.7	16.0	10.1	9.3	10.7	11.9	10.9	13.7	11.2	12.5
45-2-111	11.9		12.1	12.2	11.0	10.8	10.9	12.0	11.7	11.4	12.1
45-2-60	12.0		12.9	15.0	12.7	10.9	9.0	12.1	9.2	10.3	10.9
14-4-60 ^(a)	16.2		16.0	16.7	13.2	16.7	17.1	15.0	16.0	14.9	16.4
14-4-115 ^(a)	19.9	19.9	21.1	20.0	(b)	(b)	(b)	20.0	(b)	18.5	19.4
14-4-5	26.6	27.6	23.5	26.8	(b)	(b)	(b)	27.0	(b)	22.2	26.2

(a) Sample numbers 14-4-60 and 14-4-115 were reversed in initial data due to mislabeling.

(b) Omitted because it is not a reliable exposure indicator.

determined in this study. The integrated exposure for the Shippingport samples was estimated from the previously described calculations and the analytical results of the selected samples.

The most reliable exposure indicators are ^{137}Cs , ^{148}Nd , ^{235}U , ^{242}Pu , and ^{244}Cm . Estimates based on the ^{239}Pu and some of the ^{236}U , ^{240}Pu , ^{241}Pu , and ^{241}Am analyses (especially at higher exposure) are less certain because of only slight variations in nuclide content with burnup. Exposure estimates based on ^{234}U analyses are uncertain because of the analytical uncertainty of about 5% in the amount of ^{234}U remaining. For samples 45-2-111, 14-4-115, and 14-4-5, all of the good indicators give exceptionally close agreement to estimated exposure values. In general, the analytical data fall into quite consistent patterns and provide good correlations both throughout the present study and with previously reported data from Westinghouse on the blanket fuel.

For samples 6-5-60, 45-2-21, and 14-4-60, the amounts of ^{235}U , ^{239}Pu , ^{241}Pu , and the transplutonium elements are generally larger than the values calculated on the basis of the ^{137}Cs analyses. Based on the calculated values for these samples, the ^{235}U corresponds to a lower exposure than the average, and the others correspond to a higher exposure than the average. This is to be expected if the resonance flux is high relative to the thermal flux for these samples. The effective reactor cross sections for ^{238}U , ^{240}Pu , and ^{242}Pu are primarily

dependent on the resonance flux; therefore, a high resonance to thermal ratio would favor the formation of ^{239}Pu , ^{241}Pu , and the transplutonium isotopes. Correspondingly, the cross sections of ^{235}U , ^{239}Pu , and ^{241}Pu are primarily dependent on the thermal flux. The result is best illustrated by the tabulation below showing the effect of increasing the resonance flux at a constant thermal flux.

Factor	^{235}U	^{238}U	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
"Decay" Constant	NC	UP	NC	UP	NC	UP
Formation Rate	NA	NA	UP	UP	UP	UP
Final amount	NC	down	UP	~NC	UP	~NC

NC = No change NA = Not applicable

UP = increased

The "decay" constant referred to here may be better called a transmutation constant or destruction rate constant.

For a constant integrated thermal flux, the amount of ^{235}U remaining is about constant, and the number of ^{235}U fissions is also about constant. This value is affected only slightly by the resonance to thermal flux ratio. The amount of ^{238}U destroyed, and thus the amount of ^{239}Pu formed, is quite dependent on the resonance to thermal flux ratio. A high resonance flux will therefore cause more ^{239}Pu to be present at the end of the irradiation. As a consequence, more ^{239}Pu will be destroyed during the irradiation, more plutonium fissions will occur, and more ^{240}Pu will be formed. The increased plutonium fissions together with a constant amount of ^{235}U fissions will result

in a higher burnup value as compared with the same thermal flux and a lower resonance flux.

Just as the rate of destruction of ^{238}U increases with increasing resonance flux, so also does the rate of destruction of ^{240}Pu increase. This increased destruction results in only a slight increase in final ^{240}Pu content even though a significant increase in ^{239}Pu is obtained. This occurs because much of the increased formation of ^{240}Pu is offset by the increased rate of destruction. The same pattern of cross sections exists with ^{241}Pu and ^{242}Pu , so that ^{242}Pu increases only slightly while ^{241}Pu increases substantially. When considered on the basis of equal burnup, the result is larger amounts of ^{235}U , ^{239}Pu , and ^{241}Pu at higher resonance to thermal flux ratio, and smaller amounts of each of these nuclides at lower resonance to thermal flux ratio.

Samples 6-5-60, 45-2-21, and 14-4-60 are consistent with a higher resonance to thermal flux ratio than was used in the calculations, and sample 45-2-60 is consistent with a lower resonance to thermal flux ratio than was used in the calculations. This variation in flux ratios should be related to location within the reactor.

Previously published analyses (by Westinghouse)⁽²⁾ for blanket elements from Seed I and Seed II show increased plutonium production per MWd and lowest integrated exposure at the center of a fuel bundle. The patterns exhibited by the fuel analyses appear to verify the Westinghouse data and the calculational assumptions made in this report. However, one pair of

samples deviated from the general trend. Sample 45-2-60 shows the type of variation noted but opposite to the proposed direction, and sample 45-2-21 is not located in the fuel bundle center location. An interchange in sample numbers would provide a perfect correlation, and the patterns for low integrated exposure at the bundle center and high resonance to thermal flux ratio at the center would both be maintained. There is some basis for doubt concerning the sample numbering; however, despite this uncertainty, it is believed the data for these samples fall into a reasonably consistent pattern.

The apparently scattered data for ^{236}Pu fit into a similar pattern. The amount of ^{236}Pu is dependent on the fast neutron flux which causes an (n,2n) reaction with ^{237}Np . The fast flux should vary in the same direction as the resonance flux. The pattern noted with respect to the ^{235}U , ^{239}Pu , etc., holds true on this basis because samples 6-5-60, 45-2-21, and 14-4-60 have a high ^{236}Pu content relative to their integrated exposures.

Finally, ^{237}Np analyses were made and are listed in summary Table I. These analyses were included in this study because of the potential value of this by-product isotope. Also, as it is a precursor to ^{236}Pu , we hope to find the data to use in related ^{236}Pu studies. As yet, no attempt has been made to analyze or classify these data.

DOSIMETRY DATA

A portion of the Shippingport fuel from the three elements used in the analytical studies was used to obtain

a purified plutonium product from which source disks of about 10^6 dis/min were made. Gamma and X-ray spectral data were obtained by analysis of these sources with a 4096 channel analyzer and a Ge(Li) detector. Good spectral data were obtained for all photo peaks above 20 KeV. Rerun analyses made with a Si(Li) detector to resolve the low energy photo peaks of 20 KeV and below were completed too late to be included in this report.

Table V is a listing of the photo peaks resolved in the three samples. The counts tabulated are those in the

peak channels only. A computer program was evolved and is being tested to read out the total count under the photo peaks. The total count will, of course, be larger than listed in these preliminary data.

It should be noted that the 59.6 KeV peak from ^{241}Am is the most prominent by a large margin. Monthly analyses are made to plot the ^{241}Am growth with time.

The preliminary dosimetry data presented at this time are not intended to be used for absolute values because improved equipment now undergoing

TABLE V. Photopeaks Resolved in Shippingport Plutonium Product Samples

Bundle No. 6		Bundle No. 14		Bundle No. 45	
Counts (a)	Energy, KeV	Counts (a)	Energy, KeV	Counts (a)	Energy, KeV
3307	17.98	3922	17.33	3612	17.48
2169	20.80	2429	21.43	2259	20.63
1319	24.62	1423	26.77	1448	26.22
1761	43.55	1996	44.36	1814	43.53
2902	51.66	2873	50.46	2932	50.42
90532	59.6	81436	59.6	83959	59.6
700	93.6	1052	95.66	1010	93.68
1513	97.1	1726	97.74	1773	97.71
1257	100.22	1504	100.34	1724	100.16
745	111.33	555	110.74	579	109.77
350	113.3	674	113.34	666	113.44
163	147.46	312	147.48	281	147.71
223	206.72	275	206.4	314	207.14

(a) Counts in photopeak channel only. Total count under the photopeaks will be much larger.

final testing will be used for final analysis. The present data will, however, indicate the trends and show the ^{241}Am to be by far the greatest factor above 20 KeV in the gamma dose rate hazard from this plutonium. Correspondingly, if the plutonium is stored for periods of one year or longer, the (α, n) reaction from the ^{241}Am buildup will be a significant neutron contributor. Additional and refined data will be obtained to correlate experimental and calculated dose rate hazards from this source.

Isochem, Inc. processed the Shippingport blanket fuel during the last quarter of CY-1966 (approximately 13 1/2 tons). Three samples were drawn from the product stream at approximately even intervals during the processing campaign. Three plutonium oxide sources were made from these samples, each containing about 70 g of plutonium. The plutonium oxides were packaged in plastic, and surface dose rate measurements were made. Each of the plutonium oxide dose rates (as extrapolated through the plastic to obtain true surface dose rates) was approximately 3.3 Rad/hr. All three samples were of approximately the same isotopic composition; typically 0.5 ^{238}Pu , 65.3 ^{239}Pu , 23.1 ^{240}Pu , 7.5 ^{241}Pu , and 3.5 ^{242}Pu in atomic percent.

Larger samples of Shippingport metal and oxide have been received and are being used for additional dosimetry and shielding studies.

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