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RADIATION MEASUREMENTS OF URANIUM INGOTS FROM THE ELECTROMETALLURGICAL TREATMENT OF SPENT FUEL

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ACRONYMS

ANL-West	Argonne National Laboratory-West
ER	Electrorefiner
EBR-II	Experimental Breeder Reactor-II
FCF	Fuel Conditioning Facility
HUP	High-throughput Uranium Product
SERA	Suited Entry Repair Area
UPS	Uranium Product Storage

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ABSTRACT

Radiation measurements and gamma spectroscopy analyses were made on numerous uranium ingots produced during the treatment of Experimental Breeder Reactor-II (EBR-II) spent nuclear fuel. The objective of these measurements was to provide background data for shielding concerns and potential process optimization. The uranium ingots resulted from the processing of both driver and blanket fuel by the electrometallurgical treatment process. The observed variation in the measurements was traced to the levels of certain fission product residues that remained in the uranium ingots produced during spent fuel treatment. A minor process change to hold the material at an elevated temperature for a specified length of time was found to significantly reduce concentrations of high-activitiy fission products and, thus the radiation field.

1.0 Introduction

Treatment of the Experimental Breeder Reactor-II (EBR-II) spent nuclear fuel is performed by an electrometallurgical process at Argonne National Laboratory-West (ANL-West). The process comprises a series of operations that separate uranium from fission products to produce a low-enriched uranium ingot for disposition or reuse [1-5].

Two types of spent fuel are processed during the treatment of the EBR-II inventory: blanket and driver fuel. Blanket fuel surrounds the core region during reactor operations and is primarily depleted uranium. Driver fuel, an enriched uranium-zirconium alloy, makes up the core and contains significantly more fission products. Both types are either dismantled into elements from assemblies or retrieved from storage containers holding elements from previously dismantled assemblies. Elements are chopped into short segments, approximately 19 mm long and are loaded into perforated baskets that comprise an anode assembly. The anode assemblies are immersed in a salt electrolyte within an electrolytic cell, or electrorefiner (ER). Two electrorefiners are utilized: one for driver operations (Mk-IV) and one for blanket operations (Mk-V). The uranium is separated from the fission products by passing an electric current between the anodic baskets and a cathode, causing the uranium to be deposited at the cathode. The cathode products from both electrorefiners are processed in a vacuum retort to distill off any adhering salt. A casting step following distillation allows for isotopic dilution and sampling of the uranium ingots.

Upon cooling, the uranium ingots are placed into containers and transferred to storage [2,6]. Two different storage containers have been used to date: the uranium product storage (UPS) containers and the high-throughput uranium product (HUP) storage containers. The primary difference between these two container types is the interior height: the HUPs are 11.7 cm tall, whereas the UPSs are 6.7 cm tall.

Measurements of the radiation field around each ingot were made while the ingot was in its storage container through a shielded glovewall. The radiation measurements were taken at several points external to the storage containers with an Eberline RO-7 portable ionization chamber (see Appendix A for a typical survey). Although the primary contribution to the radiation is from gamma energy, both gamma and beta fields were measured.

The radiation measurements were performed to provide data for shielding concerns during storage of the ingots. Once loaded into their storage containers and appropriate waste can, measurements have shown that no additional shielding is required. The measurements of the radiation fields surrounding the storage containers were continued to provide a better understanding of the radiation sources for future processing.

2.0 Background

Early during the processing of driver fuel, correlation of the radiation measurements with the radiochemical results from ingot sampling was performed [7,8]. The radiochemical results from gamma spectroscopy of the ingot samples were converted to radiation dose rates using the ORIGEN nuclear data libraries [9] and the MCNP-4A code [10]. Figure 1 shows the calculated dose rates both axially and radially for one of the uranium ingots inside a UPS container. The

calculated dose rate at 46 cm from the UPS container ranges from ~3 mR/h (at R2) to ~24mR/h (at Z2). These calculations bracket the measured value of 16 mR/h at approximately the same distance. In addition, calculations were performed for a bare ingot without the UPS container since a direct measurement of a bare ingot is not possible due to handling limitations. The dose rate calculated at the surface of the bare ingot was ~1 R/h, which is approximately a factor of 2 greater than that calculated through the UPS container.

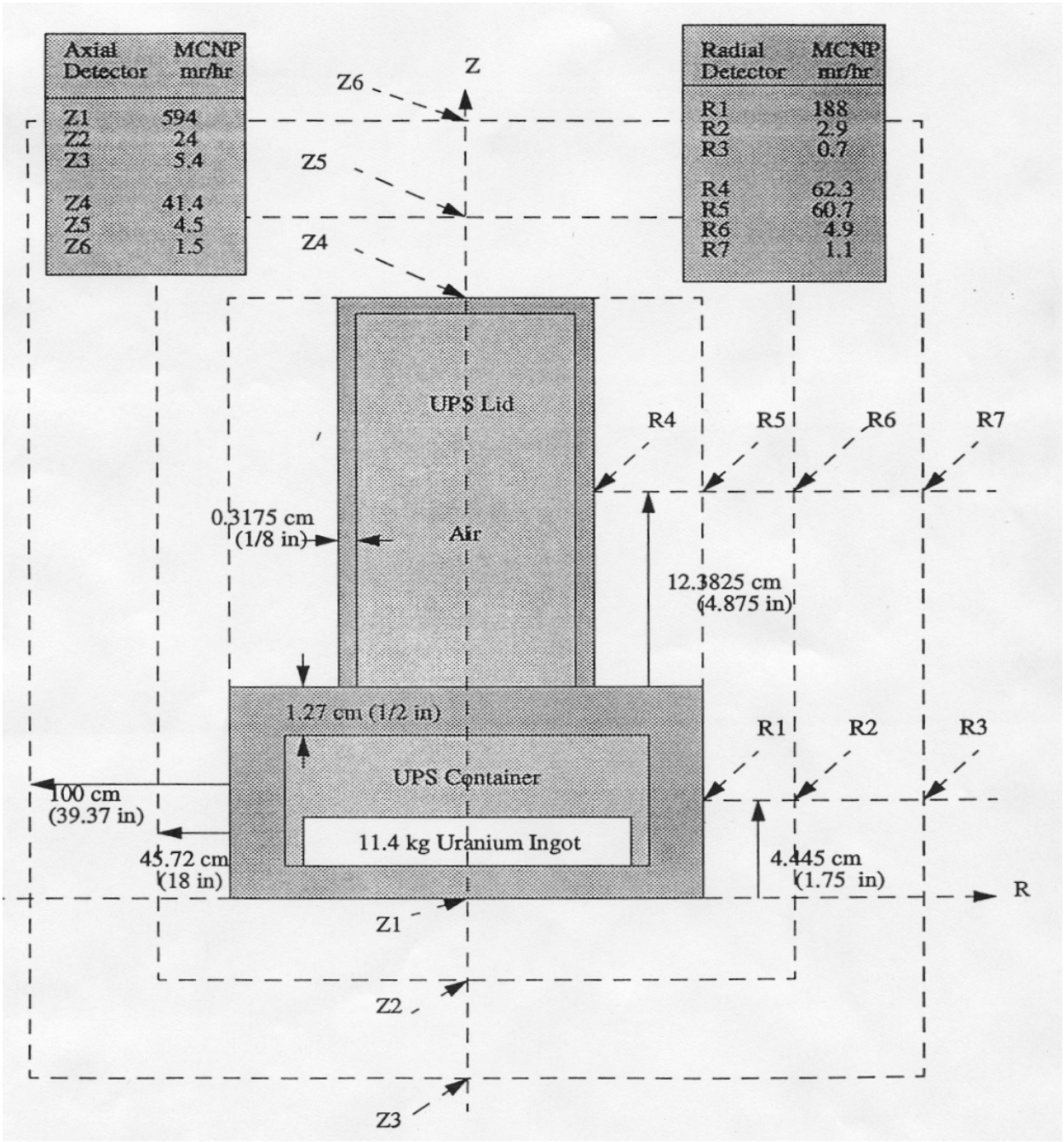

Fig. 1. Calculated Dose Rates for a Uranium Ingot Inside of a UPS Container

Other results from the radiochemistry analyses and the dose calculations concluded that the nuclides Ru-l06 and Sb-l25 were the main contributors to dose in the short term (< 2 years) while Cs-137 was the primary contributor for longer periods (~50 years). These data suggest the primary sources of the radiation field and are the focus of this report.

3.0 Results and Discussion

The radiation measurements for all the driver and blanket ingots through June 2001 are given in Table 1 and Table 2. Data on ingot size, the date of the measurements, loading orientation of the ingots, and relevant gamma-ray spectroscopy data from analyses of ingot samples are included in the tables. For most of the driver ingots, the orientation of the ingots within the containers was not specified, that is, loading the ingot top down or bottom down was considered irrelevant to radiation measurements. However, for driver ingot No. 28, it was determined that loading the top of the ingot down made a significant difference in radiation readings (100 R/h versus 3 R/h). The reasons for this are discussed later in this section. For consistency, ingots loaded after early 1999 were inserted into the UPS/HUP containers top down.

Table 1. Radiation Measurements and Radionuclide Concentrations for Driver Ingots at 5 cm

Driver Ingot	Batch	Ingot Size (kg)	Storage Container	Radiation Field (R/h)	Measurement Date	Loading Orientation	Radionuclide Concentrations			Gamma Scan Date
							Cs-137 (uCi/g)	Ru-106 (uCi/g)	Sb-125 (uCi/g)	
1	BF01a	11.426	UPS001	1.5	5/8/97	Unknown	3.2	21.6	3.7	9/26/96
2	BF01b	9.774	UPS002	0.5	10/22/96	Unknown	0.1	0.3	0.1	11/18/96
3	BF02a	12.768	UPS003	8.0	1/20/97	Unknown	0.6	501.7	72.7	2/20/97
4	BF02b	7.986	UPS004	65.0	1/20/97	Unknown	0.3	1683.3	215.0	2/21/97
5	BF03a	12.256	UPS005	72.0	4/6/97	Unknown	0.3	1768.8	246.7	4/7/97
6	BF03b	13.420	UPS006	36.0	4/6/97	Unknown	1.4	886.7	126.7	4/7/97
7	BF04a	15.277	UPS007	30.0	5/6/97	Unknown	0.3	280.0	40.7	5/7/97
8	BF04b	22.538	UPS008	2.0	5/12/97	Unknown	1.3	2.2	0.5	5/29/97
9	BF04c	19.642	UPS009	1.4	5/22/97	Unknown	0.0	1.1	0.4	6/16/97
10	BF05a	23.984	UPS010	3.0	10/26/97	Unknown	0.3	34.3	9.8	7/16/97
11	BF06a	16.764	UPS011	5.0	10/26/97	Unknown	0.5	20.7	9.4	10/6/97
12	BF06b1	13.701	UPS012	5.0	10/26/97	Unknown	0.1	0.7	0.3	11/21/97
13	BF06c	26.195	UPS013	1.2	11/1/97	Unknown	0.1	0.4	0.5	11/20/97
14	BF08a	27.565	UPS014	1.5	10/26/97	Unknown	0.3	1.0	2.4	11/23/97
15	BF07a	16.570	UPS015	1.0	12/13/97	Unknown	3.1	1.4	0.7	12/3/97
16	BF09a	22.177	UPS016	3.0	1/11/98	Unknown	0.1	0.2	0.7	1/6/98
17	BF09b	16.710	UPS017	40.0	1/10/98	Unknown	0.8	0.0	0.0	1/6/98
18	BF09d	22.256	UPS018	3.3	12/14/97	Unknown	0.2	0.2	0.2	1/7/98

Table 1. (Contd.)

Driver Ingot	Batch	Ingot Size (kg)	Storage Container	Radiation Field (R/h)	Measurement Date	Loading Orientation	Radionuclide Concentrations			Gamma Scan Date
							Cs-137 (uCi/g)	Ru-106 (uCi/g)	Sb-125 (uCi/g)	
19	BF09c	18.881	UPS019	28.0	4/24/98	Unknown	0.0	0.1	0.0	4/4/98
20	BF10a	37.805	HUP001	30.0	7/19/98	Unknown	0.2	0.9	1.4	3/13/98
21	BF12a	18.172	UPS020	1.5	5/2/98	Unknown	2.1	0.9	1.5	6/18/98
22	BF12c	42.518	HUP002	8.0	8/1/98	Unknown	0.5	0.3	0.3	5/29/98
23	BF13a	42.493	HUP003	145.0	5/15/98	Unknown	4.5	0.3	0.6	5/29/98
24	BF12b	40.468	HUP004	0.3	7/20/98	Unknown	0.5	0.1	0.1	6/17/98
25	BF14a	40.465	HUP005	2.5	8/15/98	Unknown	3.4	0.3	0.9	6/18/98
26	BF15a	30.387	HUP006	15.0	8/15/98	Unknown	2.3	0.2	0.9	7/23/98
27	BF15b	30.102	HUP007	0.5	8/14/98	Unknown	1.3	0.2	0.2	7/23/98
28	BF16a	40.421	HUP008	100.0[a]	11/3/98	Top Down	2.5	0.2	0.6	9/24/98
29	BF17a	38.795	HUP009	98.0	8/29/98	Unknown	4.1	0.1	0.4	9/23/98
30	BF18a	28.488	HUP011	143.0	12/13/98	Top Down	1.1	0.3	0.8	2/22/99
31	BF19a	32.708	HUP012	30.0	1/4/99	Top Down	4.2	1.4	1.0	2/5/99
32	BF19b	39.533	HUP013	230.0	1/5/99	Top Down	3.8	0.2	0.3	2/5/99
33	BF20a	41.970	HUP014	330.0	1/21/99	Top Down	2.0	0.3	0.6	2/22/99
34	BF21a	40.548	HUP015	7.5	2/11/99	Top Down	9.3	0.3	0.4	3/23/99
35	BF10b1	16.024	HUP016	2.0	3/14/99	Top Down	1.0	150.0	130.0	3/11/99
36	BF22a	46.899	HUP017	1.9	3/14/99	Top Down	8.0	0.2	0.4	3/24/99

Table 1. (Contd.)

Driver Ingot	Batch	Ingot Size (kg)	Storage Container	Radiation Field (R/h)	Measurement Date	Loading Orientation	Radionuclide Concentrations			Gamma Scan Date
							Cs-137 (uCi/g)	Ru-106 (uCi/g)	Sb-125 (uCi/g)	
37	BF23a	42.506	HUP018	1.6	2/11/99	Top Down	2.2	0.3	0.2	6/29/99
38	BF24a	48.685	HUP022	2.6	5/30/99	Top Down	22.9	0.1	0.2	6/29/99
39	BF25a	52.408	HUP023	1.2	6/6/99	Top Down	8.7	0.3	0.6	6/29/99
40	BF25b	29.254	HUP024	0.4	6/6/99	Top Down	15.2	0.1	0.6	7/7/99

a. Bottom down activity was 3.0 R/h on 11/25/98.

Table 2. Radiation Measurements and Radionuclide Concentrations for Blanket Ingots at 5 cm

Drive Ingot	Batch	Ingot Size (kg)	Storage Container	Radiation Field (R/h)	Measurement Date	Loading Orientation	Radionuclide Concentrations			Gamma Scan Date
							Cs-137 (uCi/g)	Ru-106 (uCi/g)	Sb-125 (uCi/g)	
1	IB01a	14.933	HUP010	2.0	3/14/99	Top Down	3.1	363.3	53.3	3/11/99
2	IB02a	14.443	HUP010	1.9	4/11/99	Top Down	21.1	446.7	71.3	6/7/99
3	UB01a	17.795	HUP019	0.4	4/23/99	Top Down	-	-	-	-
4	IB03a	13.771	HUP010	1.8	5/28/99	Top Down	1.9	283.3	47.3	7/8/99
5	IB08a	32.764	HUP026	9.0	6/13/99	Top Down	0.1	203.3	34.3	7/7/99
6	IB05a	17.937	HUP020	10.0	6/19/99	Top Down	0.4	340.0	52.3	7/29/99
7	IB09a	31.364	HUP021	3.5	7/16/99	Top Down	0.2	213.3	36.0	7/29/99

Table 2. (Contd.)

Drive Ingot	Batch	Ingot Size (kg)	Storage Container	Radiation Field (R/h)	Measurement Date	Loading Orientation	Radionuclide Concentrations			Gamma Scan Date
							Cs-137 (uCi/g)	Ru-106 (uCi/g)	Sb-125 (uCi/g)	
8	IB10a	30.507	HUP027	20.0	7/18/99	Top Down	0.4	256.7	41.0	8/11/99
9	IB11a	29.446	HUP025	19.3	8/8/99	Top Down	0.4	216.7	33.3	8/11/99
10	IB12a	29.567	HUP028	12.7	8/8/99	Top Down	0.4	190.0	33.7	8/11/99
11	IB13a	30.109	HUP029	16.5	8/8/99	Top Down	0.3	233.3	40.0	9/9/99
12	IB14a	29.741	HUP030	16.0	8/22/99	Top Down	2.4	263.3	45.0	9/9/99
13	IB15a	29.040	HUP031	13.7	9/7/99	Top Down	0.4	210.0	35.0	9/9/99
14	IB16a	29.547	HUP032	11.4	9/5/99	Top Down	0.3	190.0	30.7	9/9/99
15	IB17a	21.633	HUP033	17.7	9/5/99	Top Down	1.2	286.7	46.7	9/10/99
16	IB18a	30.526	HUP034	11.0	9/12/99	Top Down	0.3	176.7	32.7	10/12/99
17	IB19a	27.950	HUP035	19.2	11/14/99	Top Down	1.3	230.0	42.3	12/1/99
18	IB20a	25.894	HUP036	17.0	11/14/99	Top Down	1.5	330.0	56.3	12/1/99
19	IB21a	28.583	HUP037	4.0	12/19/99	Top Down	0.1	140.0	27.7	12/23/99
20	IB22a	30.298	HUP038	6.0	12/19/99	Top Down	0.3	263.3	53.0	12/23/99
21	IB04a	16.190	HUP039	11.0	1/2/00	Top Down	1.0	253.3	45.0	12/17/99
22	EB01a	31.959	HUP040	33.0	1/3/00	Top Down	0.6	0.1	0.0	12/17/99
23	IB23a	28.707	HUP042	9.0	1/3/00	Top Down	0.2	170.0	32.0	1/17/00
24	EB02a	25.422	HUP043	4.6	1/3/00	Top Down	0.6	7.2	1.5	1/17/00
25	IB24a	26.786	HUP044	9.2	1/9/00	Top Down	0.7	163.3	35.0	4/27/00

Table 2. (Contd.)

Drive Ingot	Batch	Ingot Size (kg)	Storage Container	Radiation Field (R/h)	Measurement Date	Loading Orientation	Radionuclide Concentrations			Gamma Scan Date
							Cs-137 (uCi/g)	Ru-106 (uCi/g)	Sb-125 (uCi/g)	
26	IB25a	32.275	HUP041	15.9	1/9/00	Top Down	2.8	166.7	38.3	4/27/00
27	EB03a	36.962	HUP045	24.0	1/17/00	Top Down	29.4	0.6	0.3	5/8/00
28	UB02a	22.552	HUP046	1.0	1/23/00	Top Down	0.1	3.2	0.8	5/10/00
29	WILLIT	15.354	HUP047	3.5	11/22/00	Top Down	0.1	0.1	0.0	5/10/00
30	IB26a	29.306	HUP050	9.0	11/22/00	Top Down	0.6	140.0	31.7	4/28/00
31	HB01a	45.620	HUP055	17.0	8/23/01	Top Down	2.3	234.7	83.0	6/8/01
32	IB002	33.246	HUP054	8.5	8/10/01	Top Down	1.2	80.7	28.5	3/6/01

Figures 2 and 3 show the radiation measurements for driver and blanket ingots. All measurements are through the bottom plate (~1 cm thick) of the UPS/HUP containers at approximately 5 cm. For the driver ingots, radiation fields were as high as 330 R/h, although considerably lower (1-2 R/h) dose rates were achieved by the end of the driver program, as described below.

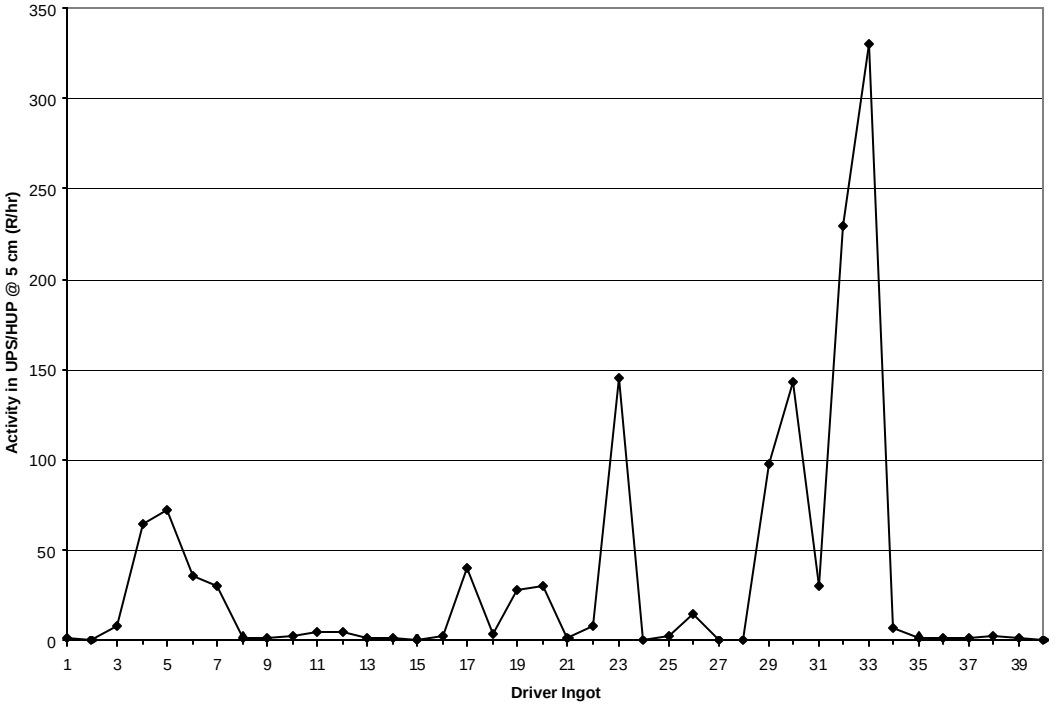

Fig. 2. Driver Ingot Radiation Fields at 5 cm

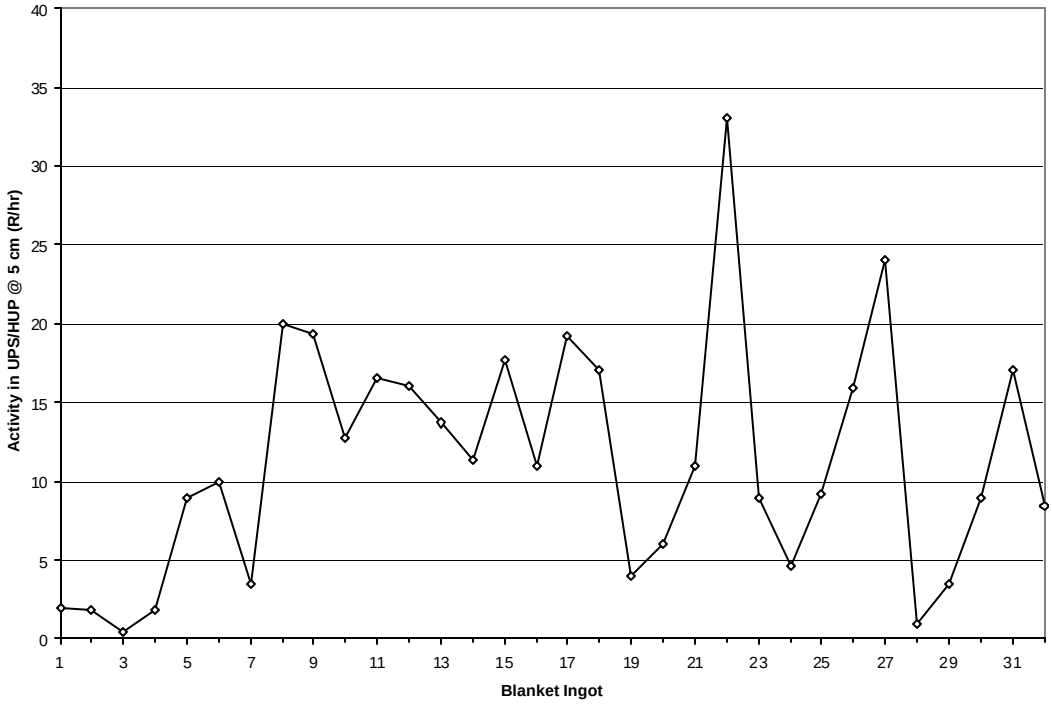

Fig. 3. Blanket Ingot Radiation Fields at 5 cm

The concentrations of the radioactive nuclides Cs-l37, Ru-106, and Sb-l25 are plotted in Fig. 4 and Fig. 5 for both the driver and blanket ingots. These data are a result of molten ingot sampling operations at the casting furnace and thus, represent the ingot in a homogenized state.

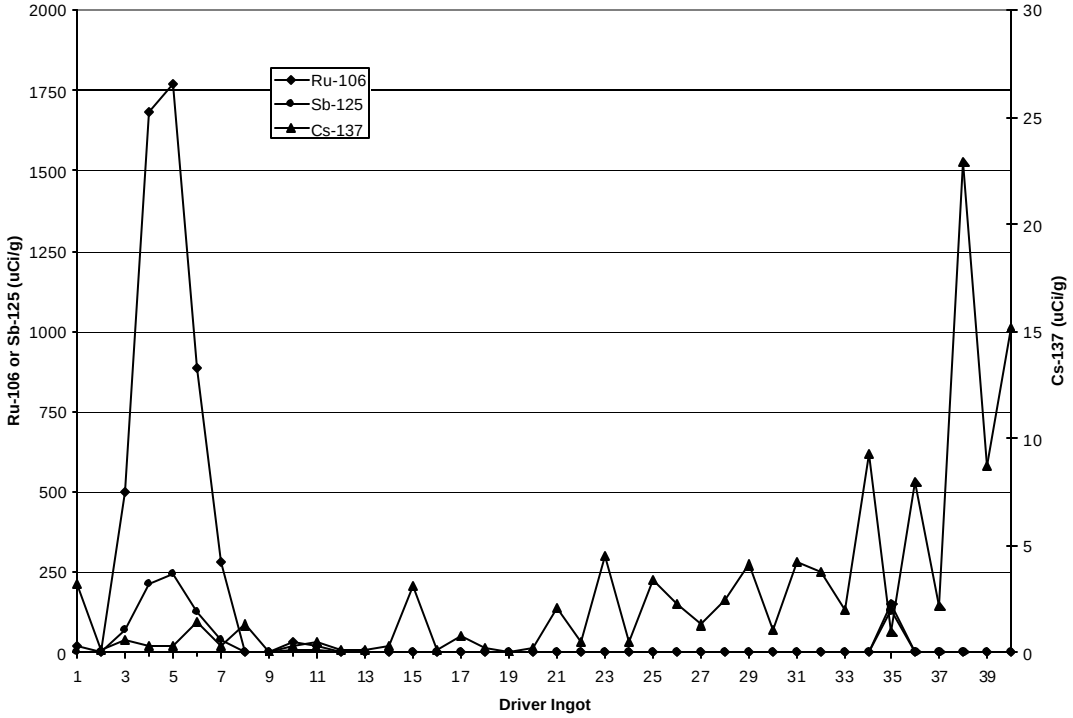

Fig. 4. Concentrations of Selected Isotopes of Driver Ingots

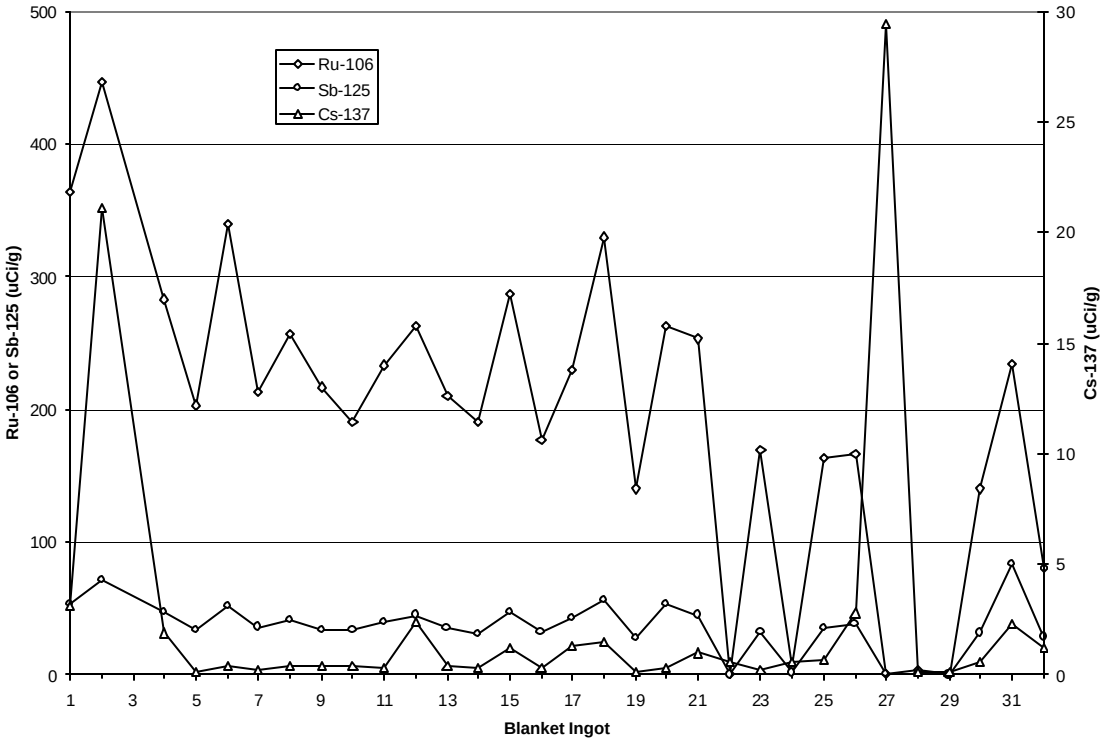

Fig. 5. Concentrations of Selected Isotopes of Blanket Ingots

The Ru-106 and Sb-l25 levels were very high early in the processing of driver fuel due to the conditions of operation for the Mk-IV electrorefiner. Later in the processing, the concentrations were quite low as consistent electrorefining conditions were implemented. Of special note is the data for ingot No. 35, a different anode to cathode arrangement was employed for this material causing the increased levels of Ru-106 and Sb-125. The Cs-l37 shows a gradual increase throughout the processing of driver material, probably coincidence to the increased fission product loading of the Mk-IV electrorefiner salt.

The Ru-106 and Sb-125 activities were generally higher during processing of blanket fuel than during driver processing with a few exceptions. Ingots Nos. 22, 24, and 27 were processed through the Mk-IV electrorefiner instead of the Mk-V. Hence, the depressed levels of Ru-106 and Sb-125 are due to the dissimilar anode/cathode layouts of the Mk-IV and Mk-V electrorefiners. The anode to cathode distances for the Mk-IV and Mk-V electrorefiners differ in that for the Mk-IV, transport of material is through the electrolyte medium to a solid cathode mandrel. For the Mk-V, the distances are much shorter and material is continuously scraped off into a collector residing directly below the anode/cathode area. Ingots Nos. 28 and 29 were special experiments with unirradiated uranium and irradiated salt from the Mk-V and Mk-IV electrorefiners, respectively. For ingot No. 31, slightly higher levels of Ru-106 and Sb-125 were detected, probably because this batch resulted from the removal of heels from the Mk-V collectors followed by processing together in the cathode processor and casting furnace.

The concentrations of Cs-137 in the blanket ingots have been lower than those measured near the end of driver processing because of the significantly reduced levels (approximately 150 uCi/g for the Mk-IV salt to about 2 uCi/g for the Mk-V salt) of Cs-137 in the blanket stream. For ingot No.2, it is suspected that cross-contamination of Cs-137 occurred from the driver stream because both driver and blanket material were processed through the same cathode processor and casting furnace at this time. Again, ingot No. 27 was processed in the Mk-IV electrorefiner and thus, the higher Cs-137 content. Gamma spectroscopy was not performed for ingot No. 3, since this material originated prior to irradiated operations in the Mk-V electrorefiner.

Following the radiation measurements from the top and bottom of driver ingot No. 28, several samples were taken from the ingot for chemical analyses. The sampling locations were from both the top surface as well as below the top surface to distinguish a dross layer, if present. Three surface samples were obtained by barely contacting the top surface of the ingot with the drilling equipment located in-cell and collecting the shavings. A sub-surface sample was taken approximately 5 mm below the top surface by first brushing aside shavings and then redrilling for the sample.

The averaged gamma spectroscopy results for the three surface samples are shown in Table 3. The isotopes Cs-134, Cs-137, Ce-144, Eu-154, and Eu-155 represent the active fission product elements of Cs, Ce, and Eu, respectively. Also given in the table are selected gamma spectroscopy results for the ingot samples taken during casting. When compared with the surface samples, all of the isotopes are at least 100 times larger on the surface than in the ingots and a concentrating effect is occurring according to the relative vapor pressures of Eu ($EuCl_2$), Ce ($CeCl_3$), and Cs (CsCl). A reduced amount of fission products was evident for the sub-surface

sample when compared to the surface samples. This is most likely explained by diffusion/ distillation of salt constituents to the top surface or dross layer.

Table 3. Gamma Spectroscopy Results for Surface and Sub-surface Samples

Isotope	Surface Results (uCi/g)	Ingot Results (uCi/g)	Surface/ Ingot Ratio	Sub-surface Results (uCi/g)
Cs-134	7.4	0.1	102	4.8
Cs-137	328.6	2.5	104	230.0
Ce-144	275.5	1.7	166	130.0
Eu-154	121.0	0.4	316	71.0
Eu-155	483.4	1.7	283	290.0

Following the results of dross sampling of driver ingot No. 28, a hold period at 1100°C was initiated in the cathode processor to enhance the distillation of material (salt for blanket operations or salt and cadmium for driver operations) prior to the melting of uranium at about 1132°C. Holding prior to the uranium melting point is important to minimize the reaction of the salt with molten uranium. The hold at 1100°C was implemented after driver ingot No. 34 to avoid the high radiation readings of the driver ingots from previous batches. Table 4 gives the radiation measurements for selected driver ingots both with and without the 1100°C hold. As shown in Table 3, small quantities of distillate were being carried over to the casting furnace after cathode processing and concentrated on the top surface of the casting ingot. One hour was chosen as the time period based on the amount of distillate being carried over, typically less than 10 g, and the overall length of a cathode processor run [5].

Table 4. Effect of 1100°C Hold Period on Radiation Measurements for Driver Ingots

Driver Ingot	Radiation Measurement (R/h)	Time at 1100°C (h)
23	145	None
32	230	None
33	330	None
36	2	1
37	2	1
38	3	1

Comparing the radiation measurements of the blanket ingots with those of the driver ingots, particularly driver ingots Nos. 35 through 40, the blanket measurements have been consistently

higher, excluding those exceptions previously described. The reason for this is the increased levels of Ru-106 and Sb-125 in the blanket ingots. Because of the anode/cathode arrangement in the Mk-V electrorefiner and subsequent collection method, approximately 40 wt% of the Ru-106 and Sb-125 in the fuel is deposited in the blanket uranium ingots as impurities. As a comparison, much less than 1 wt% of these isotopes are found in the driver ingots. In addition, the specific activities of Ru-106 and Sb-125 are considerably higher than for Cs-l37 (Table 5) and as a result, their contributions to the gamma dose rate (Table 5) are also greater per gram of nuclide [11]. The blanket ingot activities should drop rapidly from the decay of the relatively short-lived Ru-106 ($t_{1/2}$=365 d).

Table 5. Specific Activities and Gamma Doses for Selected Nuclides

Nuclide	Specific Activity (Ci/g)	Gamma Dose Rate (R/h at 1 m)
Ru-106	3309	563
Sb-125	1038	280
Cs-137	87	29

4.0 Conclusions

The following conclusions can be made based on this study:

- The driver ingot dross layer is enriched in fission product chlorides, particularly Cs-137, from incomplete distillation of the salt in the cathode processor.
- An additional distillation hold period of 1 h at 1100°C in the cathode processor reduced the radiation readings of the driver ingots significantly.
- The radiation measurements for the blanket ingots have generally been higher than those for the driver ingots as a result of higher levels of Ru-106 and Sb-125.

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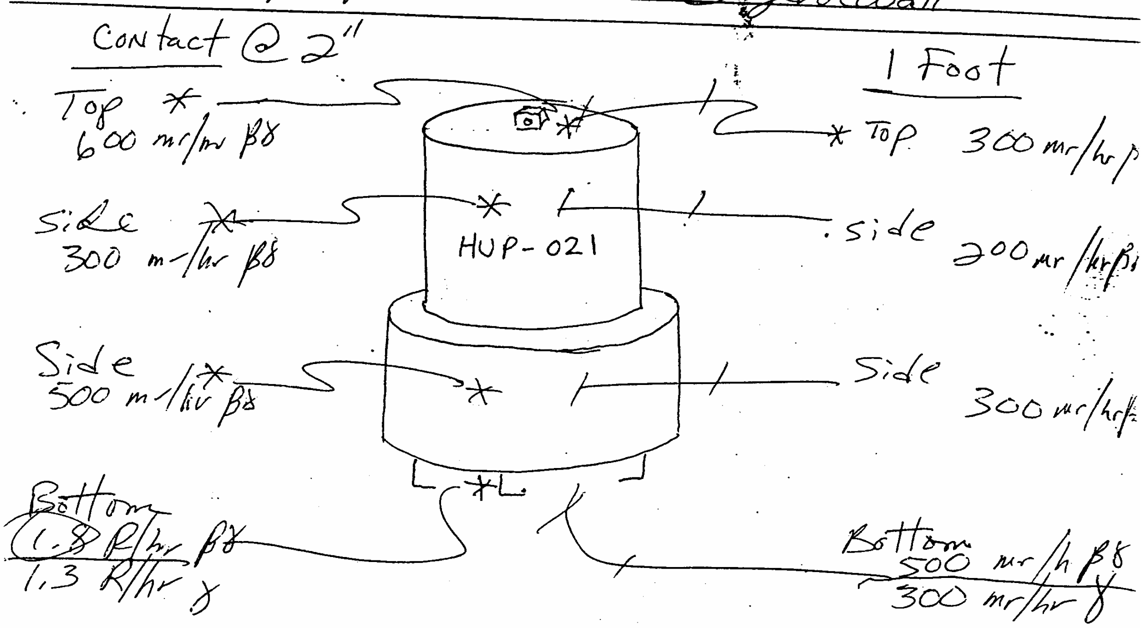
APPENDIX

FCF Special Survey

FCF SPECIAL SURVEY

Log No.: 5-118 Date: 5/28/99 Time: 11 00 Surveyor:
Instrument Type: RO-7 14-C
Instrument Sr #: 850271 801685
Reason for survey: ☐ Routine ☒ RWP # 16398
Survey results: All readings are in mR/hr taken at waist level unless otherwise noted. (* denotes contact readings)
^ represents whatman smears of 100 cm²
Circled numbers indicate large area wipes of approximately 2 square feet counted for βγ with a 260 probe at ½" with the followi
Results:

Remarks: 30 mr/hr βγ General area @ glovewall



Smear #	Type Activity	Gross CPM	DPM	Smear #	Type Activity	Gross CPM	DPM	Smear #	Type Activity	Gross CPM	DPM
	βγ				βγ				βγ		
	α				α				α		
	βγ				βγ				βγ		
	α				α				α		
	βγ				βγ				βγ		
	α				α				α		
	βγ				βγ				βγ		
	α				α				α		
	βγ				βγ				βγ		
	α				α				α		

aler ID: ______ βγ Bkg ____ CPM βγ Eff. ____% α Bkg ____ CPM α Eff. ____% Smear area is 100 cm². For source and background formation see the daily bench counter check sheet. Remarks:

Reviewed By: 5/28/99