

# **Post-Irradiation Examination of Array Targets—Part I**

**December 2003**

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## **ABSTRACT**

During FY 2001, two arrays, each containing seven neptunium-loaded targets, were irradiated at the Advanced Test Reactor in Idaho to examine the influence of multi-target self-shielding on  $^{236}\text{Pu}$  content and to evaluate fission product release data. One array consisted of seven targets that contained 10 vol %  $\text{NpO}_2$  pellets, while the other array consisted of seven targets that contained 20 vol %  $\text{NpO}_2$  pellets. The arrays were located in the same irradiation facility but were axially separated to minimize the influence of one array on the other. Each target also contained a dosimeter package, which consisted of a small  $\text{NpO}_2$  wire that was inside a vanadium container. After completion of irradiation and shipment back to the Oak Ridge National Laboratory, nine of the targets (four from the 10 vol % array and five from the 20 vol % array) were punctured for pressure measurement and measurement of  $^{85}\text{Kr}$ . These nine targets and the associated dosimeters were then chemically processed to measure the residual neptunium, total plutonium production,  $^{238}\text{Pu}$  production, and  $^{236}\text{Pu}$  concentration at discharge. The amount and isotopic composition of fission products were also measured. This report provides the results of the processing and analysis of the nine targets.



## **1. INTRODUCTION**

During FY 2001, two arrays, each containing seven neptunium-loaded targets, were irradiated at the Advanced Test Reactor (ATR) in Idaho. One array consisted of seven targets that contained 10 vol %  $\text{NpO}_2$  pellets, while the other array consisted of seven targets that contained 20 vol %  $\text{NpO}_2$  pellets. The arrays were located in the same irradiation facility but were axially separated to minimize the influence of one array on the other. This irradiation, which has also been referred to as Phase III irradiations in earlier reports,<sup>1</sup> was performed to examine the influence of multi-target self-shielding on  $^{236}\text{Pu}$  content and to evaluate fission product release data. At the completion of the irradiation, the targets were shipped to Oak Ridge National Laboratory (ORNL) for post-irradiation examination.

To date, 9 of the 14 targets have been punctured for the measurement of the internal pressure and fission product gases. The results of these measurements have been reported separately.<sup>2</sup> The nine targets were subsequently dissolved and analyzed in order to evaluate the  $^{238}\text{Pu}$  and  $^{236}\text{Pu}$  yields, as well as the nonvolatile fission product yields. This report summarizes the results of the dissolutions. Because efforts are under way to enhance the gas analysis capability, the remaining five targets will be punctured for pressure measurement and then dissolved at a later date. Consequently, these targets will be the subject of a later report.

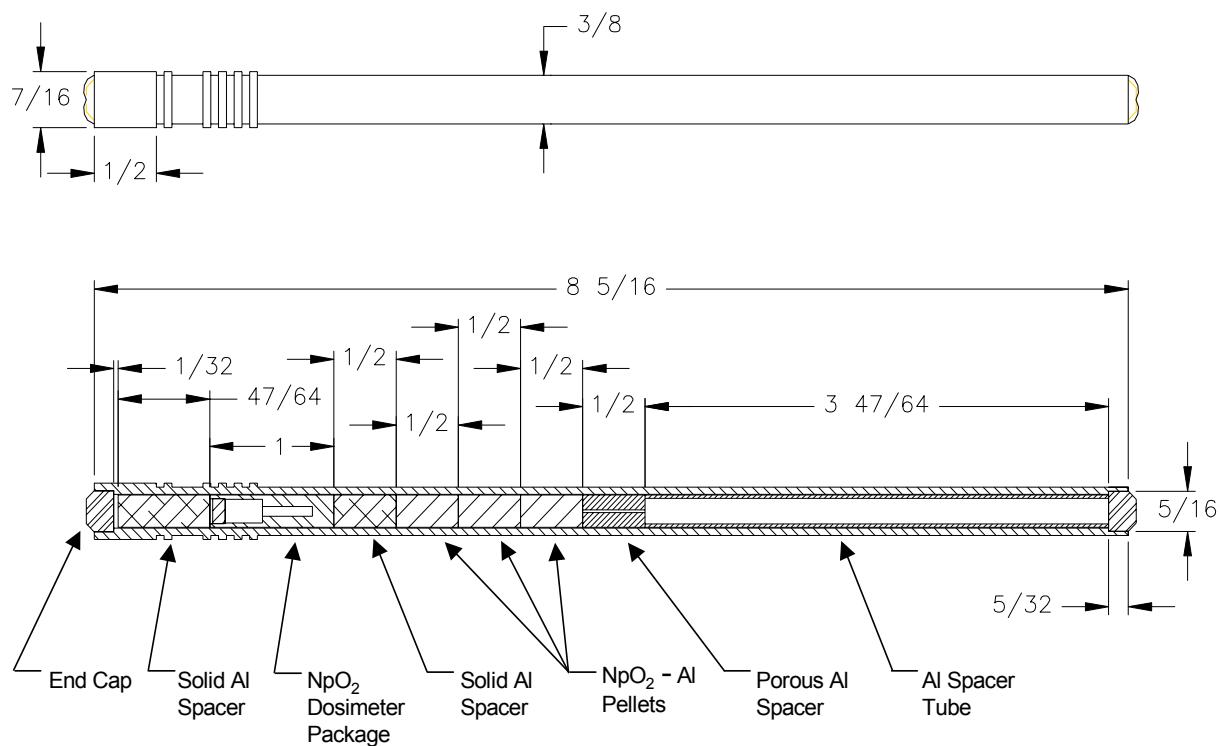
In the following sections, the array targets, ATR irradiation, and target processing flowsheets are described. Finally, the results of target dissolution are presented.

## 2. ARRAY TARGET DESCRIPTION

The array targets consisted of aluminum tubing that contained three  $\text{NpO}_2\text{-Al}$  pellets and a dosimeter package. The development work for the fabrication of the targets is described in detail in Ref. 1. The pellets were prepared by mixing the desired amount of  $\text{NpO}_2$  with aluminum powder and then pressing the mixture to the desired dimensions and density. Pellets that contained 10 and 20 vol %  $\text{NpO}_2$  were prepared and then uniaxially pressed to 90% theoretical density. After pressing, the pellets were fired at 350°C, under vacuum, for 3 h to remove stearic acid, which served as a lubricant in the die press. The dosimeter package consisted of a  $\text{NpO}_2$  wire that was contained within a vanadium can.

A schematic of the target configuration is shown in Fig. 2.1. Each target had a pellet section, containing three pellets, and a dosimeter section, containing one dosimeter package. Seven of the targets were loaded with 10 vol %  $\text{NpO}_2$  pellets, while the other seven were loaded with 20 vol %  $\text{NpO}_2$  pellets. The pellet and dosimeter sections, which were separated by an aluminum spacer, were located in the top half of the target. The bottom half of the target contained an aluminum tube spacer that acted as a plenum for fission gas expansion. The plenum was separated from the pellets by a porous aluminum spacer, which allowed fission product gases to expand into the plenum. The pellet and dosimeter sections could be removed from a target by cutting through the solid spacers, which resulted in a pellet section, a dosimeter section, a top aluminum spacer section, and aluminum tube spacer section.

After assembly, a closure weld was performed and the weld area was radiographed. The targets were then hydrostatically compressed at 20,000 psig to ensure that the target tube was in contact with the pellets. The final assembly was then radiographed, and a helium leak test and a dye penetrate check were performed on each target. Tables 2.1 and 2.2 provide summary details about the pellet loading and dimensions for each of the targets irradiated in the ATR.



**Fig. 2.1. Schematic of aluminum-clad target that contains three NpO<sub>2</sub>-Al pellets and a dosimeter package.**

**Table 2.1. Summary of fabrication data for 10 vol % NpO<sub>2</sub> targets**

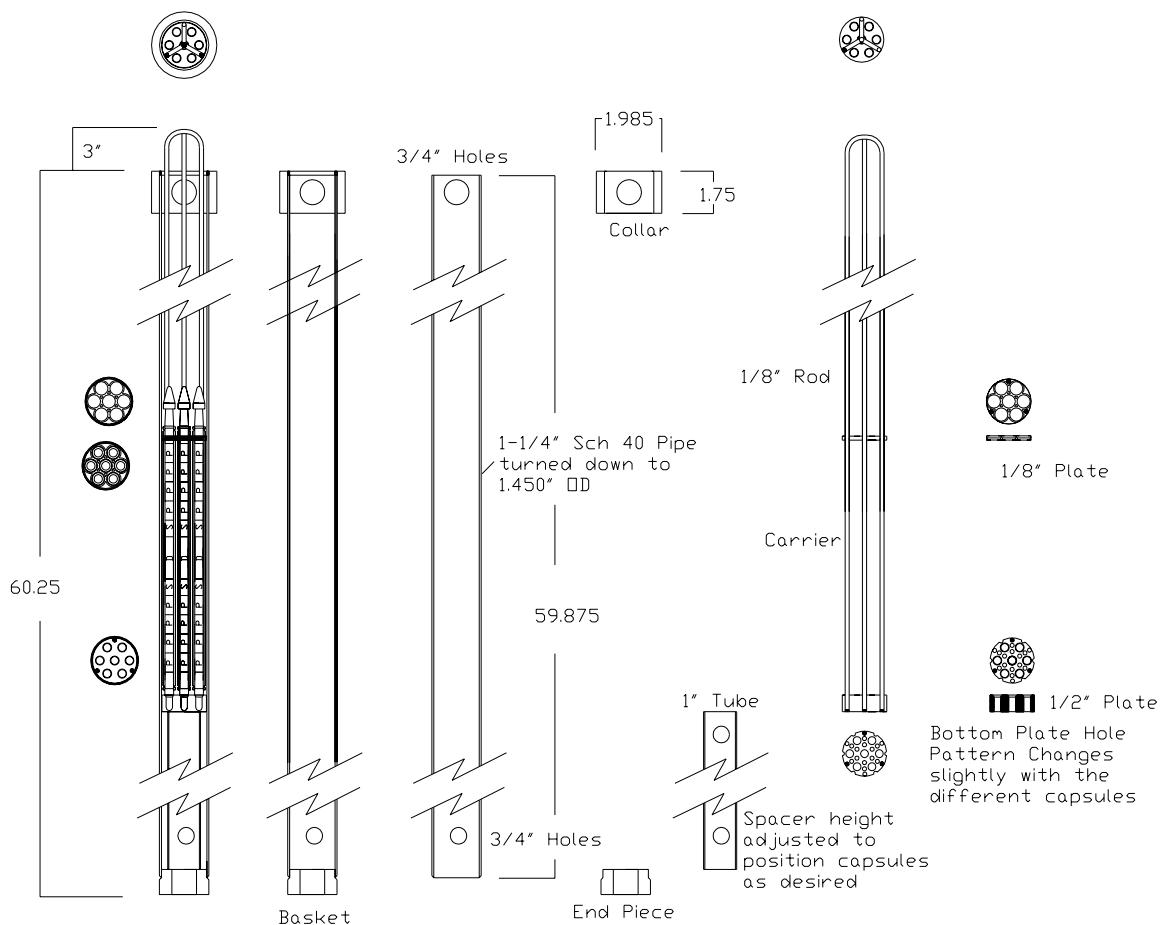
Target number	Pellet number	Pellet density (g/cm <sup>3</sup> )	Fraction of theoretical density	NpO <sub>2</sub> (vol %)	Pellet dimensions		NpO <sub>2</sub> (g)	Np metal (g)
					Diameter (in.)	Length (in.)		
MK-10.0	BNP-17	3.13	0.895	10.00	0.25	0.503	0.3902	0.3351
	BNP-1	3.16	0.900	10.00	0.25	0.5	0.3902	0.3351
	BNP-3	3.11	0.891	10.00	0.25	0.505333	0.3901	0.3351
	Dosimeter 0-1						0.001888	0.001668
MK-10.1	BNP-2	3.16	0.905	10.01	0.25	0.497	0.3906	0.3355
	BNP-14	3.14	0.895	10.00	0.25	0.502667	0.3902	0.3351
	BNP-7	3.14	0.896	10.01	0.25	0.502333	0.3907	0.3356
	Dosimeter 0-2						0.001712	0.001512
MK-10.2	BNP-4	3.15	0.897	10.00	0.25	0.501667	0.3902	0.3351
	BNP-16	3.13	0.893	10.00	0.25	0.504167	0.3903	0.3352
	BNP-10	3.12	0.899	10.00	0.25	0.5005	0.3902	0.3351
	Dosimeter 0-3						0.001731	0.001529
MK-10.3	BNP-25	3.15	0.900	10.01	0.25	0.500167	0.3905	0.3354
	BNP-26	3.14	0.900	10.02	0.249833	0.500833	0.3909	0.3357
	BNP-27	3.15	0.903	10.00	0.25	0.498167	0.3902	0.3351
	Dosimeter 0-11						0.001131	0.000999
MK-10.4	BNP-11	3.16	0.904	10.01	0.2495	0.500167	0.3905	0.3354
	BNP-18	3.14	0.894	10.02	0.25	0.503333	0.391	0.3358
	BNP-5	3.15	0.900	10.01	0.2495	0.501833	0.3902	0.3351
	Dosimeter 0-8						0.001744	0.001541
MK-10.5	BNP-6	3.14	0.896	10.00	0.25	0.502167	0.3901	0.3351
	BNP-12	3.13	0.895	9.99	0.25	0.503	0.3899	0.3349
	BNP-8	3.13	0.893	10.03	0.25	0.504167	0.3913	0.3361
	Dosimeter 0-9						0.001993	0.001761
MK-10.6	BNP-15	3.14	0.896	10.00	0.2495	0.504333	0.3901	0.3351
	BNP-23	3.14	0.897	10.00	0.25	0.501667	0.3901	0.3351
	BNP-20	3.13	0.894	10.00	0.25	0.503367	0.3903	0.3352
	Dosimeter 0-10						0.001969	0.001739

**Table 2.2. Summary of fabrication data for 20 vol % NpO<sub>2</sub> targets**

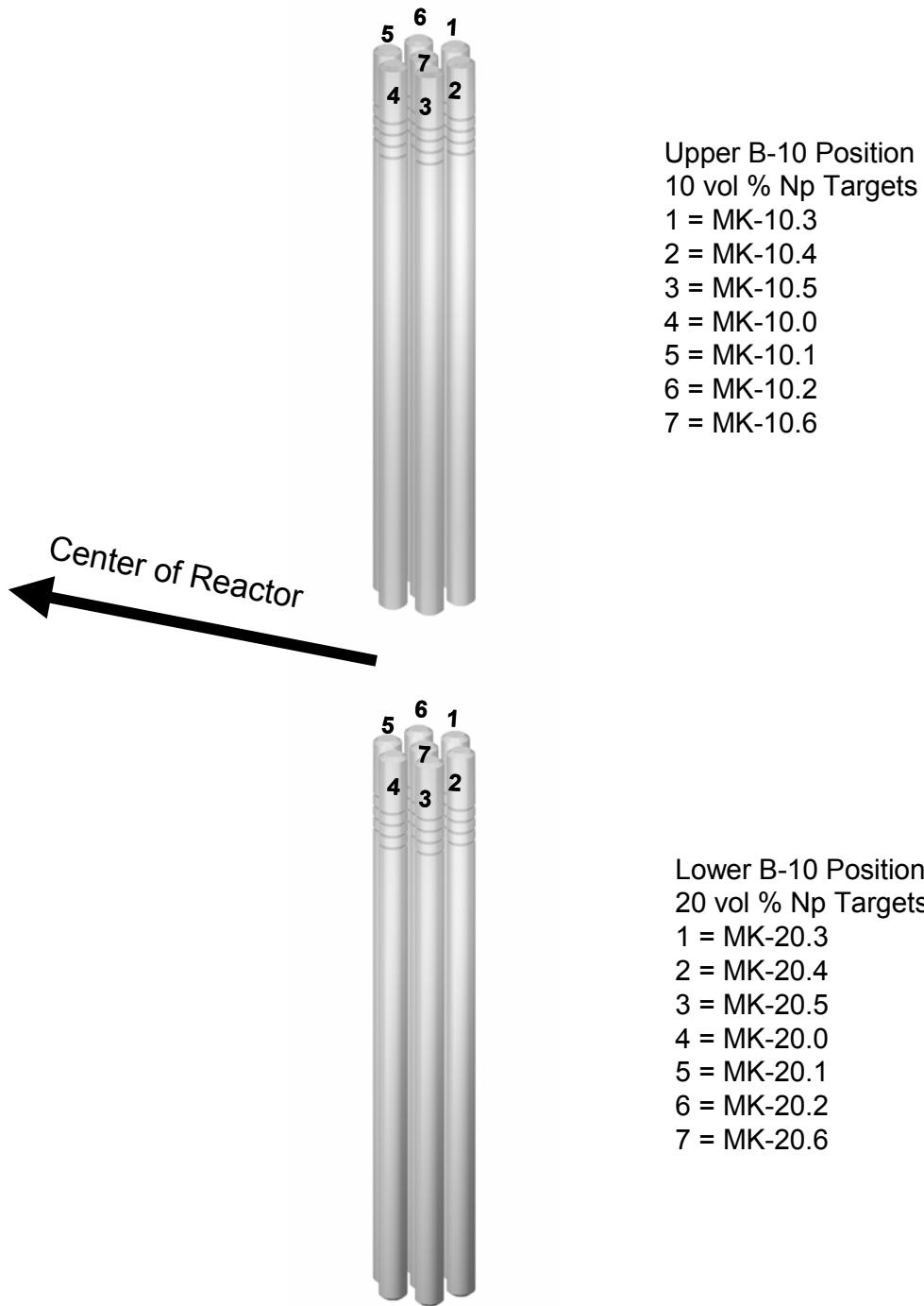
Target number	Pellet number	Pellet density (g/cm <sup>3</sup> )	Fraction of theoretical density	NpO <sub>2</sub> (vol %)	Pellet dimensions			NpO <sub>2</sub> (g)	Np metal (g)
					Diameter (in.)	Length (in.)			
MK-20.0	BNP-40	3.82	0.886	20.02	0.25	0.50783	0.7815	0.6712	
	BNP-47	3.82	0.887	20.00	0.2505	0.50516	0.7801	0.6700	
	BNP-58	3.85	0.889	19.99	0.25	0.5065	0.7802	0.6701	
	Dosimeter 0-12						0.001264	0.001117	
MK-20.1	BNP-41	3.83	0.890	20.03	0.25	0.506	0.7816	0.6713	
	BNP-53	3.83	0.890	19.98	0.25	0.50553	0.7794	0.6694	
	BNP-59	3.83	0.892	20.00	0.25	0.50483	0.7806	0.6704	
	Dosimeter 0-13						0.002198	0.001942	
MK-20.2	BNP-43	3.78	0.878	20.01	0.25	0.51266	0.7808	0.6706	
	BNP-52	3.82	0.888	19.99	0.25	0.5065	0.7797	0.6697	
	BNP-49	3.83	0.892	20.00	0.25	0.50466	0.7804	0.6703	
	Dosimeter 0-14						0.00166	0.001466	
MK-20.3	BNP-48	3.84	0.890	20.00	0.25	0.50583	0.7807	0.6705	
	BNP-42	3.79	0.880	20.00	0.2495	0.51333	0.7806	0.6704	
	BNP-60	3.86	0.895	19.99	0.25	0.50283	0.7798	0.6698	
	Dosimeter 0-15						0.002073	0.001831	
MK-20.4	BNP-50	3.83	0.889	20.01	0.25	0.50666	0.781	0.6708	
	BNP-57	3.87	0.898	20.00	0.25	0.50116	0.7807	0.6705	
	BNP-44	3.84	0.892	19.99	0.25	0.50466	0.7801	0.6700	
	Dosimeter 0-16						0.001633	0.001443	
MK-20.5	BNP-45	3.84	0.892	20.00	0.25	0.50483	0.7804	0.6703	
	BNP-51	3.84	0.891	20.00	0.2505	0.50316	0.7804	0.6703	
	BNP-55	3.80	0.884	19.99	0.2505	0.50716	0.7798	0.6698	
	Dosimeter 0-17						0.001411	0.001246	
MK-20.6	BNP-54	3.82	0.888	20.01	0.2505	0.50466	0.7806	0.6704	
	BNP-61	3.84	0.890	19.99	0.25	0.5055	0.7799	0.6698	
	BNP-56	3.80	0.881	20.01	0.2505	0.509	0.781	0.6708	
	Dosimeter 0-18						0.001762	0.001557	

### 3. ATR IRRADIATION

The 14 targets were shipped to the Idaho National Engineering and Environmental Laboratory (INEEL), where they were loaded into an aluminum basket (similar to that depicted in Fig. 3.1), thereby forming two arrays as depicted in Fig. 3.2. The seven 10 vol % targets comprised the upper array, while the seven 20 vol % targets formed the lower array. Note that, as shown in Fig. 3.2, the targets were aligned along an axis relative to the center of the reactor. This orientation was selected to aid in the evaluation of self-shielding by the targets. The arrays were irradiated for two cycles in position B-10 of the ATR (Fig. 3.3), which is a 1.5-in.-diam hole near two control drums on the east side of the reactor. The irradiation data for the two cycles are summarized in Table 3.1.



**Fig. 3.1. Depiction of aluminum basket similar to that used in the array target irradiations.**



**Fig. 3.2. Depiction of configuration of the 10 vol % and 20 vol % NpO<sub>2</sub> arrays relative to the center of the ATR core.**

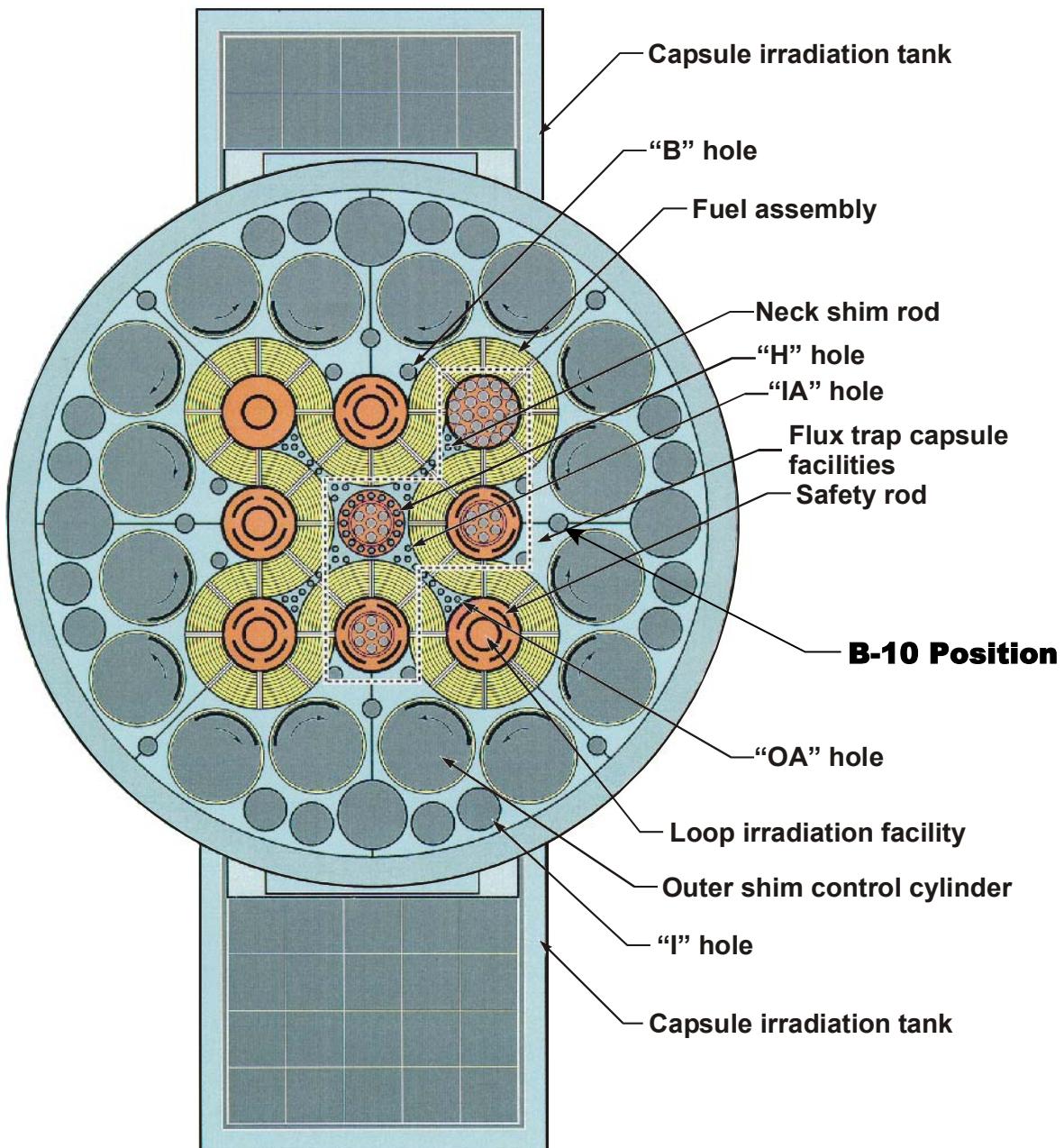


Fig. 3.3. Cross section of the ATR core showing position B-10, which was used in the array target irradiation.

**Table 3.1. Summary of data for the two ATR cycles used to irradiate the array targets**

	Cycle	
	125B	126A
Start date and time	07:00, 23JUL01	12:00, 24SEP01
Finish date and time	14:00, 11SEP01	15:00, 03NOV01
Cycle length, days	50.292	40.125
Core burnup, MWd	5337.60	4139.80
East quadrant burnup, MWd	1321.02	1044.45
Average core power, MW(t)	106.13	103.17
Average east quadrant power, MW(t)	26.27	26.03

## 4. TARGET PROCESSING

Nine of the targets (four from the 10 vol % array and five from the 20 vol % array) were processed using a two-stage dissolution process, which consisted of first dissolving the aluminum in a caustic solution and then dissolving the remaining actinides and fission products in an acid solution. The caustic decladding solution and acid product solution were sampled and analyzed by a gamma scan, gross beta scan, gross alpha scan, and inductively coupled plasma (ICP)–mass spectroscopy. Analysis for  $^{236}\text{Pu}$  was performed by using a 2-thenoyltrifluoroacetone (TTA) extraction of the acid product solution, which was then followed by a 96-h alpha count. Based on the analytical results, a material balance was performed to determine the amounts of  $^{238}\text{Pu}$ ,  $^{236}\text{Pu}$ , total Pu,  $^{237}\text{Np}$ , and fission products that were recovered in the dissolution process. The following subsections describe the equipment and chemical processing steps used in the target processing.

### 4.1 TARGET-CUTTING JIG

A target-cutting jig, shown in Fig. 4.1, was designed to remove the pellet and dosimeter sections from the target body. The apparatus was constructed as a stainless steel trough, with slots cut into the trough to guide a saw blade through the aluminum spacers in the target. A target was placed in the trough and then held in place by using five push clamps mounted on the side of the jig. A fine-tooth hacksaw, fabricated with manipulator grips, was placed in the cutting slots, and the target was cut into four sections. Each cut was made through a spacer that separated the various sections of the target (see Fig. 2.1).



**Fig. 4.1. Photograph of stainless steel saw jig used to section targets.**

## 4.2 DISSOLVER EQUIPMENT

The pellet dissolution were performed in a hot cell using a Teflon dissolver as shown in Fig. 4.2. The Teflon dissolver was developed after previous dissolution with a glass dissolver vessel revealed that the silica competed with the actinides for the fluoride ions, which promoted the degradation of the glass and produced a gelatinous material in the bottom of the dissolver.

The Teflon dissolver was designed with three penetrations for a condenser, an input/output tube, and a thermometer. The dissolver was fitted with a reflux condenser that was cooled with chilled water from a constant-temperature recirculation bath. The dissolver solution was effectively refluxed during both the caustic and acid dissolution. The dissolver off-gas was passed through a condensate trap and then scrubbed with about 300 mL of 2 M NaOH to neutralize any acid fumes. The inlet/outlet tube was a 1/4-in. stainless steel tube containing a 2- $\mu\text{m}$  stainless steel filter.



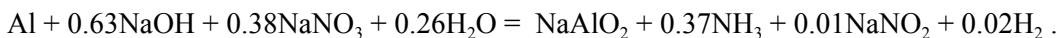
**Fig. 4.2. Photograph of dissolver used for pellet dissolution.**

A peristaltic pump was connected to the dissolver inlet/outlet line to transfer solution into and out of the dissolver. Tests indicated that the pump would transfer the caustic solutions through the 2- $\mu\text{m}$  filter in a reasonable amount of time and that the filter would not allow actinide solids to pass through.

The dissolver was placed on a hot plate that also had a stirrer motor. Mixing during the caustic dissolution was very important in breaking up the pellet as the aluminum matrix was being dissolved. In previous dissolutions carried out in a glass dissolver, it was observed that a hard skeleton of the pellet remained as the aluminum was dissolved from the pellet core. This pellet skeleton was easily broken up when a magnetic stirring bar was used.

#### 4.3 CAUSTIC DISSOLUTION OF TARGET SECTIONS

The caustic dissolution (also referred to as aluminum dejacketing) dissolves the aluminum target tube and aluminum matrix in the pellet core. This dissolution was performed by placing a target section in the dissolver with  $\text{NaNO}_3$  solution. (The  $\text{NaNO}_3$  reacts with  $\text{H}_2$  to produce  $\text{NH}_3$  and thereby suppresses the  $\text{H}_2$  concentration in the off-gas.) The solution was heated to  $\sim 96^\circ\text{C}$  and mixed using a stirring bar with the magnetic stirrer. Sodium hydroxide was then metered into the solution. The overall chemical reaction is shown by the following equation:<sup>3</sup>



The amount of aluminum in each pellet section was calculated based on the aluminum mass in the target tube, spacers, and three pellets. The pellet sections of the target were estimated to contain  $\sim 8.5$  g of aluminum. An excess of  $\text{NaOH}$  was used to promote complete dissolution of the aluminum metal.

Figure 4.3 is a typical flowsheet for the caustic dissolution. The caustic dissolved all of the aluminum associated with one pellet section, which contained three pellets. The pellet section was first placed in the dissolver, and 200 mL of 2.2  $M$   $\text{NaNO}_3$  was then added using the pump. The solution was heated to a gentle reflux with temperatures ranging from 90 to  $96^\circ\text{C}$ . Approximately 60 mL of 10  $M$   $\text{NaOH}$  was then added at a rate of  $\sim 1$  mL/min.\* The solution was gently refluxed for 4 h after the  $\text{NaOH}$  addition at a temperature of  $\sim 103^\circ\text{C}$ . If the dissolution reaction were instantaneous, then  $\sim 125$  sccm

---

\* At first, the 10  $M$   $\text{NaOH}$  solution was added at 5-min intervals, waiting for the reaction to slow before adding additional caustic. However, no problems with overheating or uncontrolled off-gas flow occurred; therefore, the  $\text{NaOH}$  was added continuously at a 1-mL/min rate without interruption.

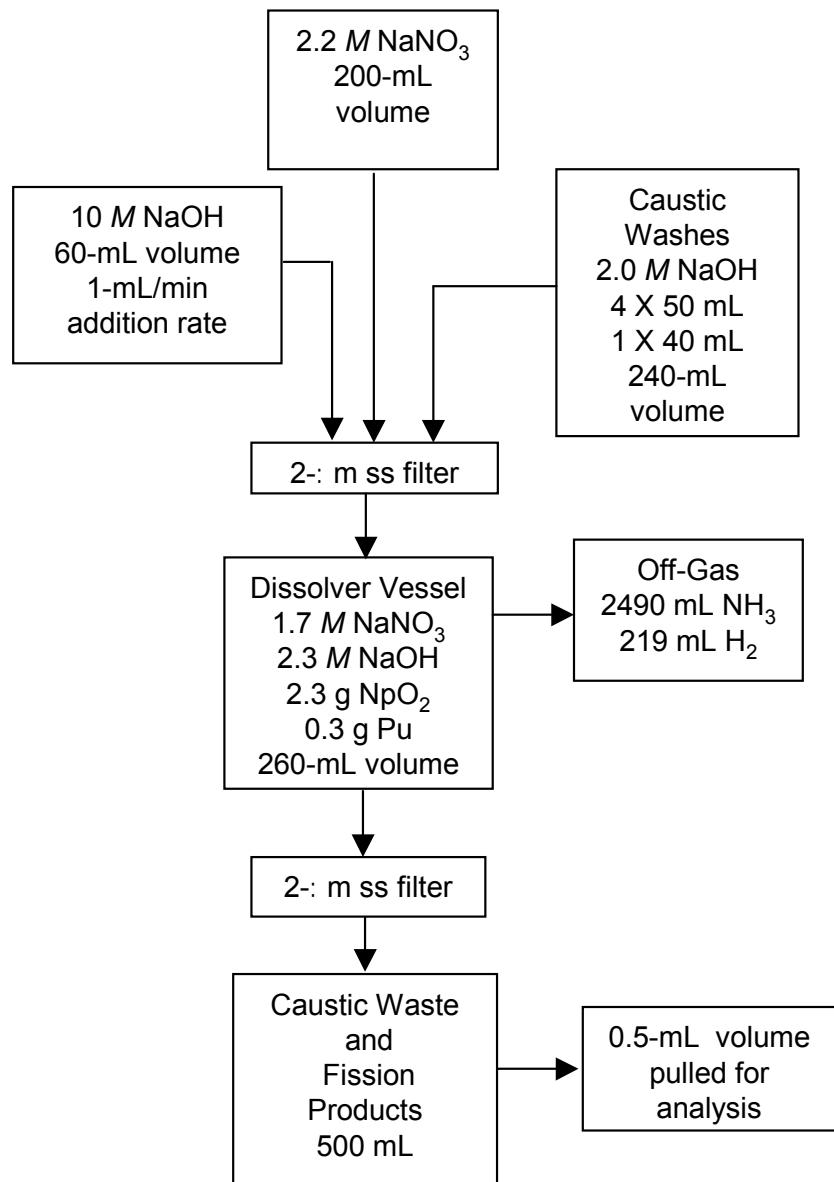


Fig. 4.3. Typical flowsheet for the caustic dissolution.

of gas would evolve during the addition of NaOH. During the dissolution, the pump was left in operation, which continuously purged the dissolver with air from the hot cell.

After a 4-h digestion period, the solution was allowed to settle and cool for 4 h. The solution was then slowly transferred out of the dissolver through the 2- $\mu\text{m}$  stainless steel filter into a 500-mL volumetric flask. The dissolver and solids were then washed with 2  $M$  NaOH solution, which also flushed any solids on the filter back into the dissolver vessel. The dissolver was washed at least twice using ~100 mL of 2  $M$  NaOH or until the volumetric flask reached a volume of 500 mL. The 500-mL caustic solution was sampled using a pipette to pull a 0.5-mL sample, which was then submitted for analysis.

#### 4.4 ACID DISSOLUTION

The acid dissolution of the neptunium and plutonium oxides was carried out using HNO<sub>3</sub> with a small amount of added HF. Figure 4.4 depicts a typical flowsheet for the acid dissolution. A 150-mL acid solution containing 8.0  $M$  HNO<sub>3</sub> and 0.02  $M$  HF was added to the dissolver. It should be noted that Al(NO<sub>3</sub>)<sub>3</sub> was not added to the acid solution, since the quantity of fluoride was so small that it was not necessary to bind any possible excess fluoride. The solution was heated to gentle reflux and continuously stirred for 3 h while the temperature was maintained between 106 and 108°C. The solution was usually allowed to cool to <50°C overnight.

The acid product solution was then pumped from the dissolver, through a 2- $\mu\text{m}$  filter, and into a 500-mL volumetric flask. After the initial product removal, a series of acid flushes using 8  $M$  HNO<sub>3</sub>–0.02  $M$  HF were performed to wash any residual product or solids from the dissolver and filter. The acid flushes were then transferred to the 500-mL volumetric flask. The first acid flush contained about 50 mL of solution. The second, consisting of about 150 mL, was heated to a gentle reflux for ~30 min. This hot acid flush was pulled up into the filter several times. This step was performed to contact the filter surfaces and thus dissolve any residual product present on the surfaces. Three more 50-mL acid flushes of the dissolver were carried out after the hot acid flush. The 500-mL volumetric product flask was then sampled by pipetting a 0.5-mL sample volume, which was submitted for analysis.

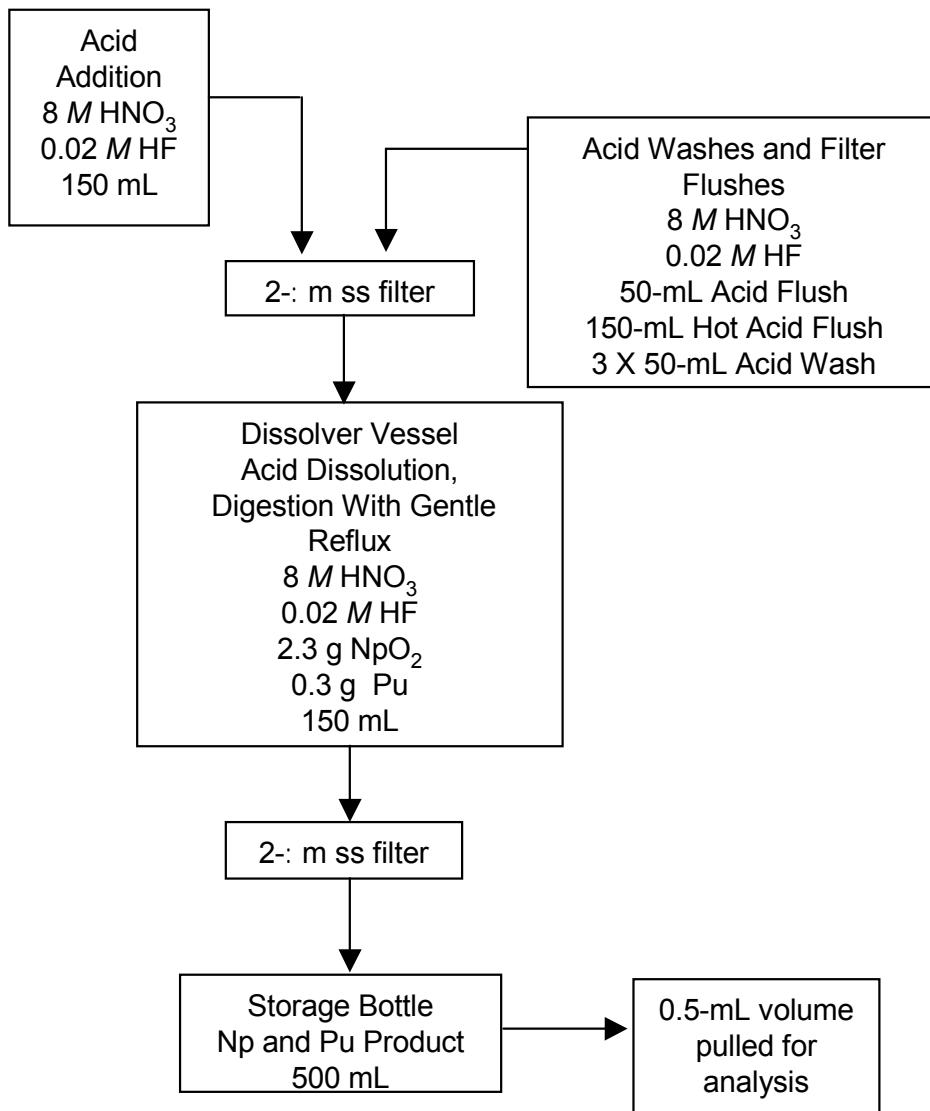


Fig. 4.4. Typical flowsheet for the acid dissolution.

#### **4.5 DOSIMETER DISSOLUTION**

The dosimeter within the array target consisted of  $\text{NpO}_2$  wire that was placed in a vanadium can and then seal welded. The vanadium can was placed in a zircaloy cylinder, which was then sealed with a press-fit seal. The target section containing the dosimeter packages was precisely cut through the top 0.375 in. of the zircaloy package, a process that usually resulted in the vanadium can sliding out of the target section.

The dissolution process for the dosimeters was carried out using a two-step acid dissolution in a 10-mL Teflon screw-top vial. In order to handle the vials using in-cell manipulators and to heat the vials on a hot plate, an aluminum vial holder was fabricated. The vial holder was slotted to allow the dissolution and liquid level to be visually observed. The receptacle, which was fabricated to hold up to seven vials and holders, was a rectangular-shaped aluminum block into which seven slotted cavities had been machined. Each cavity held a vial and a vial holder, and the slots allowed the vial to be visually inspected during the dissolution process. A thermometer or thermocouple well was drilled into the receptacle to allow the temperature of the dissolution process to be monitored. Figure 4.5 is a photograph of the Teflon vials, vial holders, and receptacle.

The vanadium cans were placed in a 10-mL Teflon vial, where the vanadium was dissolved using ~6–7 mL of 3 *M*  $\text{HNO}_3$  during a 12–18-h digestion period. As the vanadium dissolved, the solution became emerald green in color. After the digestion period, the dissolver solution was carefully pipetted out of the vial, while leaving the solids. The solids were washed three times using ~6 mL of 3 *M*  $\text{HNO}_3$ .

The solids remaining from the first acid dissolution were then dissolved using ~6 mL of an 8 *M*  $\text{HNO}_3$ –0.1 *M* HF acid solution. The temperature of the solution was maintained above 90°C for at least 6 h; however, the dissolution was allowed to digest overnight. The acid product was then carefully pipetted out of the vials, and the vials were washed three times with 6 mL of clean 8 *M*  $\text{HNO}_3$ –0.1 *M* HF solution. The acid product and the washes were sampled by pipetting out about 0.4 mL of the solution.



**Fig. 4.5. Photographs of vials, vial holders, and receptacle used in the dosimeter dissolution.**

## 5. RESULTS AND DISCUSSION

Table 5.1 provides a list of the targets that were processed. The two target dissolution numbers reflect the two-step dissolution process that was performed. The single dosimeter dissolution number reflects essentially the one-step acid dissolution of the  $\text{NpO}_2$  dosimeter. The analytical results from each dissolution sample were used to calculate the amount of neptunium converted to plutonium, the amount of  $^{238}\text{Pu}$  recovered, and the amount of  $^{236}\text{Pu}$  produced. The results of the analyses are summarized in Table 5.2, while the detailed results for each set of target pellets and dosimeters are presented in Appendix A. To provide some insight into the effect of position and target self-shielding, results are presented in terms of the relative power. The relative power reported for each target was calculated by dividing the neutron flux at a target by the average flux in the region. Note that this calculation was performed for one point in time during the irradiation and is therefore representative of only a particular control drum configuration. Calculations are being performed to establish the relative power based on the total fluence at each target.<sup>4</sup> However, it is not expected that the relative powers will change significantly. The quantities reported in Table 5.2 are briefly defined in the following paragraph.

The neptunium recovery in both the caustic and acid dissolutions is provided, and the sum of these two quantities divided by the initial quantity of neptunium is reported as the residual neptunium. For the plutonium acid recovery, the plutonium product is reported for both  $^{238}\text{Pu}$  and total plutonium. The weight percentage of  $^{238}\text{Pu}$  (on the basis of total plutonium) is also shown. The conversion of neptunium to both plutonium and fission products is calculated by dividing the moles of each of these quantities by the initial moles of neptunium. The values for neptunium and plutonium loss to waste are the weight percentages of the recovered neptunium and plutonium in the caustic dissolution stream. The concentration of  $^{236}\text{Pu}$  reported is the amount (in parts per million on a total plutonium basis) present at discharge of the target from the reactor. This amount was calculated from the measured concentration of the processed target and the time since discharge.

The isotopic distributions of the fission products are shown for the caustic dissolution and the acid dissolution streams in Tables 5.3 and 5.4, respectively. These amounts are reported in terms of parts per million of the  $^{238}\text{Pu}$  produced and can therefore be used to estimate fission product concentrations in waste streams for a particular  $^{238}\text{Pu}$  production rate. A summary of the data measured for the dosimeters is presented in Table 5.5. Samples were taken and analyzed for the acid dejacket solution of two dosimeters. The analysis for the MK-10.1 dosimeter revealed that 1.4 and 2.4 wt % of the Np and  $^{238}\text{Pu}$  were in the dejacket solution, respectively. For the MK-20.4 dosimeter, these quantities were 0.4 and 0.5 wt %.

Figures 5.1–5.3 provide graphical representations of the data as a function of the relative power. In Fig. 5.1, the percentages of neptunium converted to plutonium are shown. The percentages of total plutonium that consist of  $^{238}\text{Pu}$  are shown in Fig. 5.2. Finally, the  $^{236}\text{Pu}$  concentration (parts per million on a total plutonium basis) are presented in Fig. 5.3.

**Table 5.1. List of the array targets that were processed**

Target number	Reactor Position	Target dissolution numbers <sup>a</sup>	NpO <sub>x</sub> (vol %)	Dosimeter dissolution number <sup>b</sup>
MK-10.1	U-5	DJ-19/AD-19	10	ADD-25
MK-10.2	U-6	DJ-23/AD-23	10	<i>c</i>
MK-10.4	U-2	DJ-20/AD-20	10	ADD-26
MK-10.6	U-7	DJ-21/AD-21	10	ADD-27
MK-20.1	L-5	DJ-16/AD-16	20	ADD-21
MK-20.2	L-6	DJ-22/AD-22	20	ADD-22
MK-20.4	L-2	DJ-17/AD-17	20	ADD-23
MK-20.5	L-3	DJ-24/AD-24	20	<i>c</i>
MK-20.6	L-7	DJ-18/AD-18	20	ADD-24

<sup>a</sup> DJ = dejacket caustic dissolution; AD = acid dissolution.

<sup>b</sup> ADD = acid dissolution of dosimeter.

<sup>c</sup> Not analyzed.

**Table 5.2. Summary of results for array target analysis**

Target number	Relative Power	Initial Np (g)	Np recovery (g)		Residual Np (wt %)	Pu acid recovery (g)		$^{238}\text{Pu}/\text{Total Pu}$ (wt %)	Conversion (mol %)		Loss to waste (wt %)		$^{236}\text{Pu}$ at discharge (ppm) <sup>b</sup>
			Caustic	Acid		$^{238}\text{Pu}$	Total Pu		Np to Pu	Np to FP <sup>a</sup>	Np	Pu	
MK-10.4	0.49	1.0063	0.0119	0.965	97.08	0.101	0.112	89.99	11.14	0.06	1.22	0.29	$3.26 \pm 8.5\%$
MK-10.6	0.91	1.0054	0.0115	0.870	87.67	0.113	0.129	87.81	12.76	0.09	1.30	0.12	$3.86 \pm 5.1\%$
MK-10.2	1.32	1.0054	0.0073	0.855	85.76	0.118	0.137	86.06	13.52	0.09	0.84	0.07	$3.30 \pm 11.2\%$
MK-10.1	1.7	1.0062	0.0059	0.890	89.04	0.140	0.165	84.80	16.34	0.18	0.66	0.05	$2.96 \pm 6.2\%$
MK-20.4	0.46	2.0113	0.0030	1.825	90.89	0.184	0.204	90.09	10.08	0.05	0.17	0	$6.24 \pm 2.5\%$
MK-20.5	0.72	2.0104	0.0026	1.715	85.44	0.215	0.246	87.36	12.18	0.08	0.15	0.0077	$4.58 \pm 6.3\%$
MK-20.6	0.81	2.0110	0.0021	1.810	90.11	0.209	0.236	88.51	11.69	0.08	0.12	0	$5.36 \pm 3.7\%$
MK-20.2	1.25	2.0106	0.0113	1.780	89.09	0.259	0.297	87.17	14.69	0.10	0.63	0.07	$3.58 \pm 2.7\%$
MK-20.1	1.72	2.0111	0.0007	1.675	83.32	0.242	0.285	84.68	14.11	0.15	0.04	0	$3.88 \pm 5.1\%$

<sup>a</sup> FP = fission products.<sup>b</sup> Relative error based on 2 standard deviations as derived from the counting statistics.

**Table 5.3. Fission products in caustic dejacket stream (ppm) relative to  $^{238}\text{Pu}$  production**

Isotope	Target number (Relative power) <sup>a</sup>								
	MK-10.4 (0.49)	MK-10.6 (0.91)	MK-10.2 (1.32)	MK-10.1 (1.7)	MK-20.4 (0.46)	MK-20.5 (0.72)	MK-20.6 (0.81)	MK-20.2 (1.25)	MK-20.1 (1.72)
$^{106}\text{Ru}$	57	76	73	105	40	29	39	46	36
$^{134}\text{Cs}$	11	18	19	33	9	12	14	15	28
$^{137}\text{Cs}$	1430	2199	2379	3328	1270	1517	1709	1743	2766

<sup>a</sup> The value for relative power is listed parenthetically following the target number.

**Table 5.4. Fission products in acid dissolution stream (ppm) relative to  $^{238}\text{Pu}$  production**

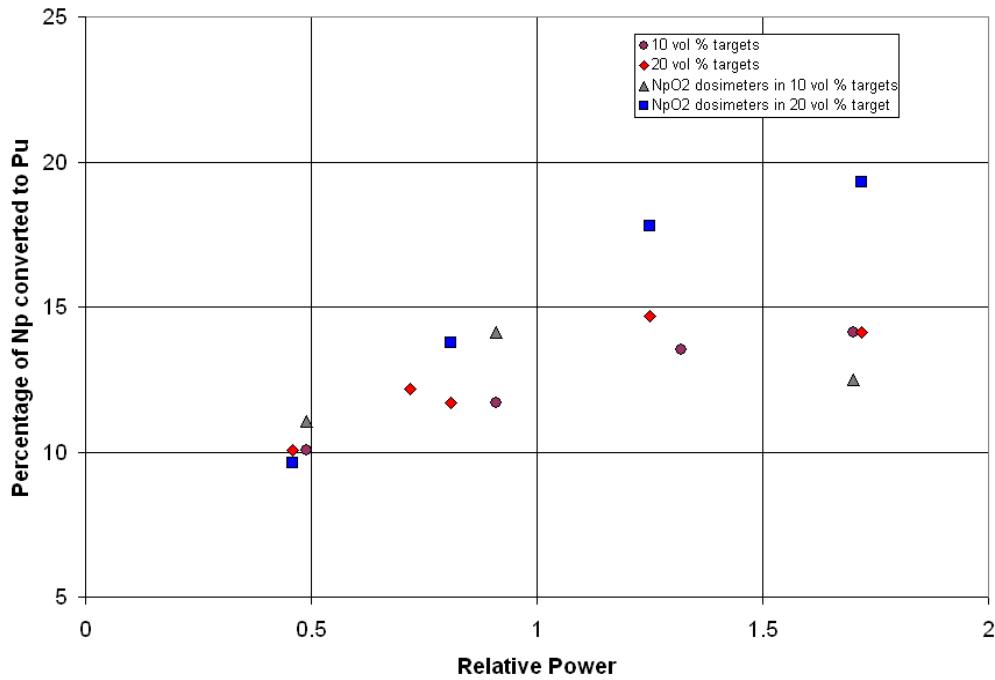
Isotope	Target number (Relative power) <sup>a</sup>								
	MK-10.4 (0.49)	MK-10.6 (0.91)	MK-10.2 (1.32)	MK-10.1 (1.7)	MK-20.4 (0.46)	MK-20.5 (0.72)	MK-20.6 (0.81)	MK-20.2 (1.25)	MK-20.1 (1.72)
$^{95}\text{Zr}$	2	2	1	4	3	1	4	2	7
$^{106}\text{Ru}$	275	362	348	526	290	323	391	380	711
$^{125}\text{Sb}$	23	< 10	< 10	< 10	23	25	26	24	37
$^{134}\text{Cs}$	7	9	10	14	9	13	10	12	26
$^{137}\text{Cs}$	907	1086	1243	1331	1185	1662	1338	1502	2444
$^{144}\text{Ce}$	353	488	470	698	416	395	508	476	879
$^{154}\text{Eu}$	< 20	< 23	< 25	< 26	< 15	< 16	< 18	< 16	< 23
$^{155}\text{Eu}$	23	33	37	42	21	31	36	25	57

<sup>a</sup> The value for relative power is listed parenthetically following the target number.

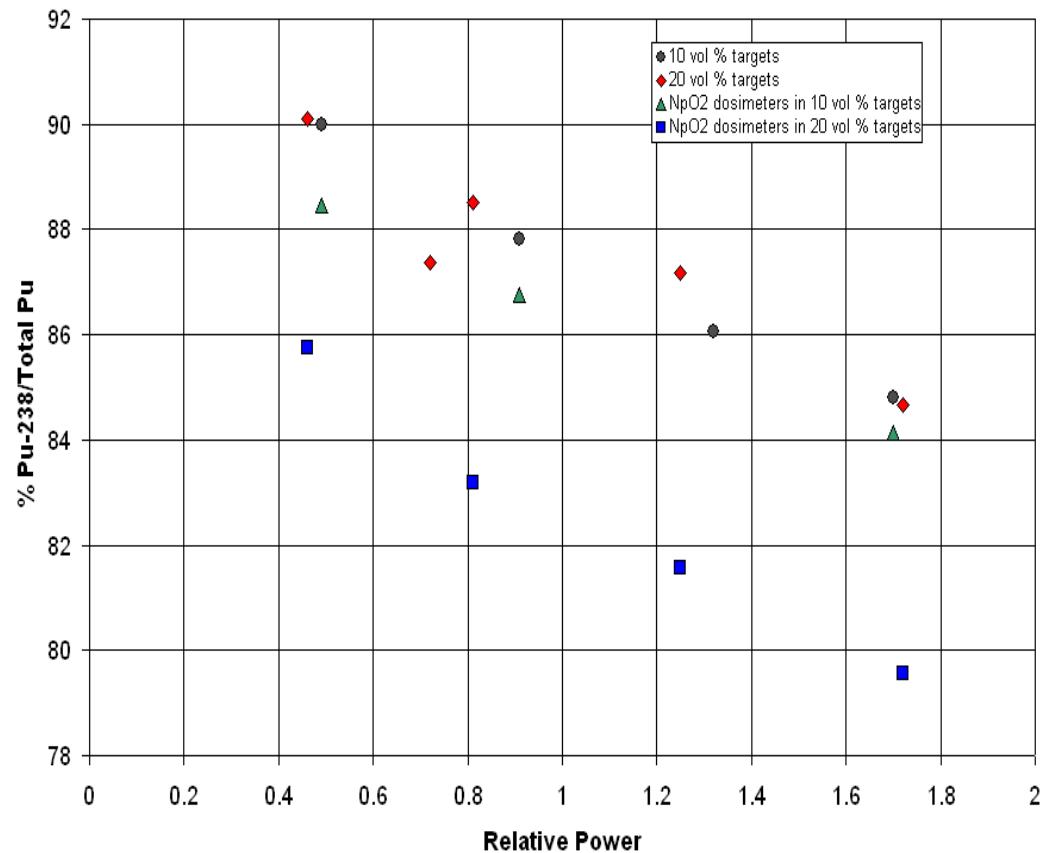
**Table 5.5. Summary of dosimeter data**

Target ID for dosimeter	Relative power	Initial Np (g)	Np recovery (g)	Residual Np (wt %)	$^{238}\text{Pu}$ recovery (g)	Total Pu recovery (g)	$^{238}\text{Pu}/\text{Total Pu}$ (wt %)	Conversion of Np to Pu (mol %)	$^{236}\text{Pu}$ at discharge (ppm) <sup>a</sup>
MK-10.4	0.49	0.0015	0.0012	79.14	0.00015	0.00017	88.46	11.07	$2.76 \pm 6.2\%$
MK-10.6	0.91	0.0017	0.0014	82.21	0.00021	0.00024	86.75	14.11	$2.39 \pm 5.0\%$
MK-10.2	1.32	b	b	b	b	b	b	b	b
MK-10.1	1.7	0.0015	0.0009	60.42	0.00016	0.00019	84.12	12.47	$3.79 \pm 4.2\%$
MK-20.4	0.46	0.0014	0.0007	51.59	0.00012	0.00014	85.76	9.62	$3.55 \pm 5.2\%$
MK-20.5	0.72	b	b	b	b	b	b	b	b
MK-20.6	0.81	0.0016	0.0010	61.09	0.00018	0.00022	83.18	13.75	$4.24 \pm 4.5\%$
MK-20.2	1.25	0.0015	0.0011	76.01	0.00021	0.00026	81.56	17.81	$3.33 \pm 6.2\%$
MK-20.1	1.72	0.0019	0.0014	72.05	0.00030	0.00038	79.56	19.29	$4.20 \pm 3.7\%$

<sup>a</sup> Relative error based on 2 standard deviations as derived from the counting statistics.<sup>b</sup> Not analyzed.



**Fig. 5.1.** Percentage of neptunium converted to plutonium as a function of relative power for the targets and dosimeters.



**Fig. 5.2.** Plutonium-238 production (percentage of total plutonium production) as a function of relative power for the targets and dosimeters.

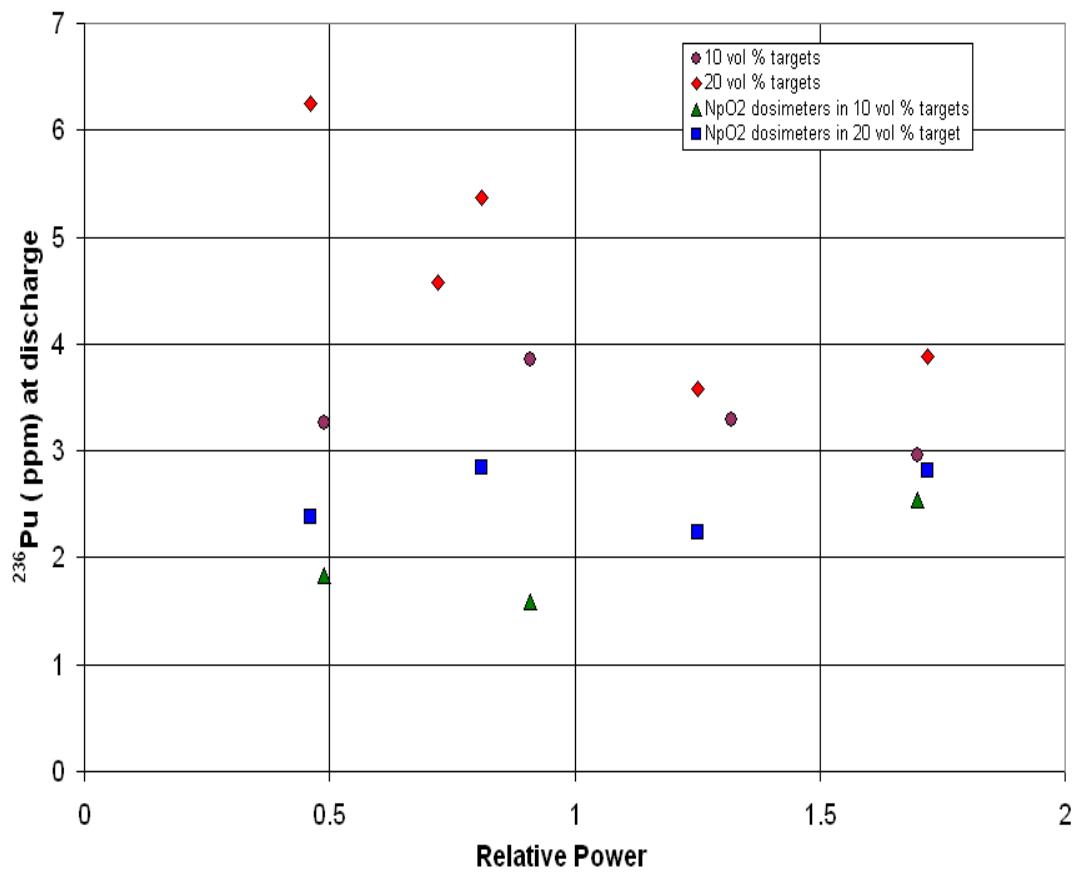


Fig. 5.3. Plutonium-236 content at discharge from the ATR as a function of relative power for the targets and dosimeters.

Several trends that reflect the different flux (and target self-shielding) at the various target positions are evident in the figures. The percentage of neptunium that is converted to plutonium increases with relative power—ranging from about 10 to 16 mol % for the pellets. The results for the 10 and 20 vol % targets are similar. The  $^{238}\text{Pu}$  production, as a percentage of total plutonium, generally decreases with increasing relative power. This value ranged from about 85 to 90 wt % for the pellets. The specification for  $^{238}\text{Pu}$  production is 82 wt %, as of the date of processing. Again, the 10 and 20 vol % targets provided similar results. In Fig. 5.3, the  $^{236}\text{Pu}$  production at discharge tends to decrease with increasing relative power. Additionally, the  $^{236}\text{Pu}$  production is higher in the 20 vol % pellets than in the 10 vol % targets. The  $^{236}\text{Pu}$  production in the pellets ranged from about 3 to 6 ppm. The specification for  $^{236}\text{Pu}$  is 2 ppm, as of the date of processing. Hence, these targets would require a cooling period of 1.7 to 4.5 years.

The processing of the remaining five targets, which will be the subject of a future report, will help to further establish the trends that are observed for the various target positions. Additionally, the preparation of the final calculations for the relative power at each position will provide better insight into the flux depression caused by each of the targets and the role of target self-shielding in the production of  $^{238}\text{Pu}$  and  $^{236}\text{Pu}$ .

## **6. SUMMARY**

Fourteen targets were irradiated in two arrays during two cycles at the ATR. One array consisted of a set of seven targets that contained 10 vol %  $\text{NpO}_2$  pellets, while the other array consisted of seven targets that contained 20 vol %  $\text{NpO}_2$  pellets. The pellets consisted of a mixture of  $\text{NpO}_2$  and aluminum powder. Each target also contained a dosimeter package, which consisted of a small  $\text{NpO}_2$  wire that was inside a vanadium container.

After completion of irradiation and shipment back to ORNL, nine of the targets (four from the 10 vol % array and five from the 20 vol % array) were punctured for pressure measurement and measurement of  $^{85}\text{Kr}$ . These nine targets and the associated dosimeters were then chemically processed to measure the residual neptunium, total plutonium production,  $^{238}\text{Pu}$  production, and  $^{236}\text{Pu}$  concentration at discharge. The amount and isotopic composition of fission products were also measured. These data will be useful in designing the production targets, as well as in planning for the handling of waste streams.

The five remaining targets will be punctured for pressure measurement after additional gas analysis equipment is brought on-line. Finally, calculations on the expected yield are being performed by INEEL personnel. When available, these calculations will be compared with the experimental results to confirm the model used for the ATR and to identify the appropriate cross-section data set for  $^{236}\text{Pu}$  production.

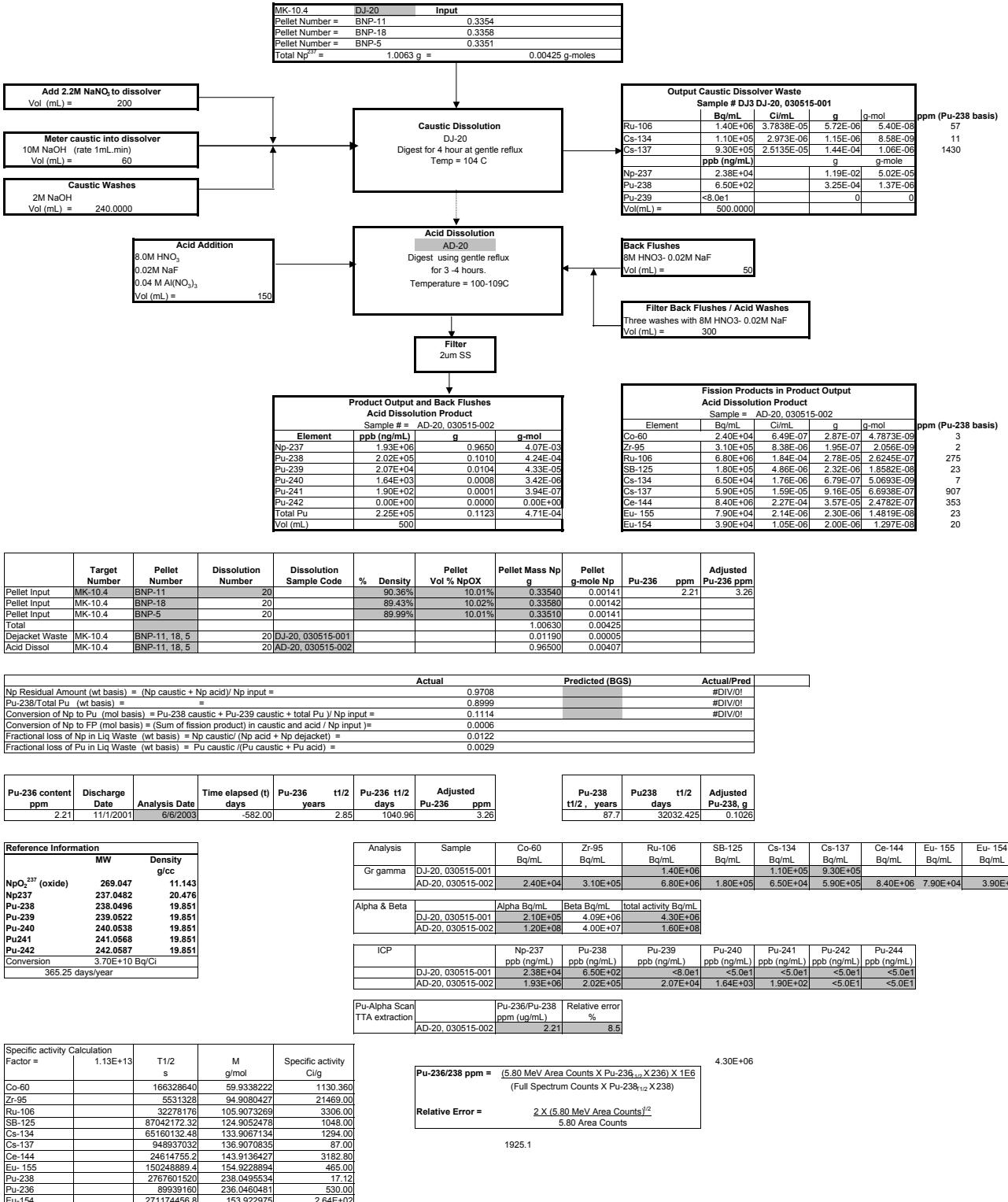
## **REFERENCES**

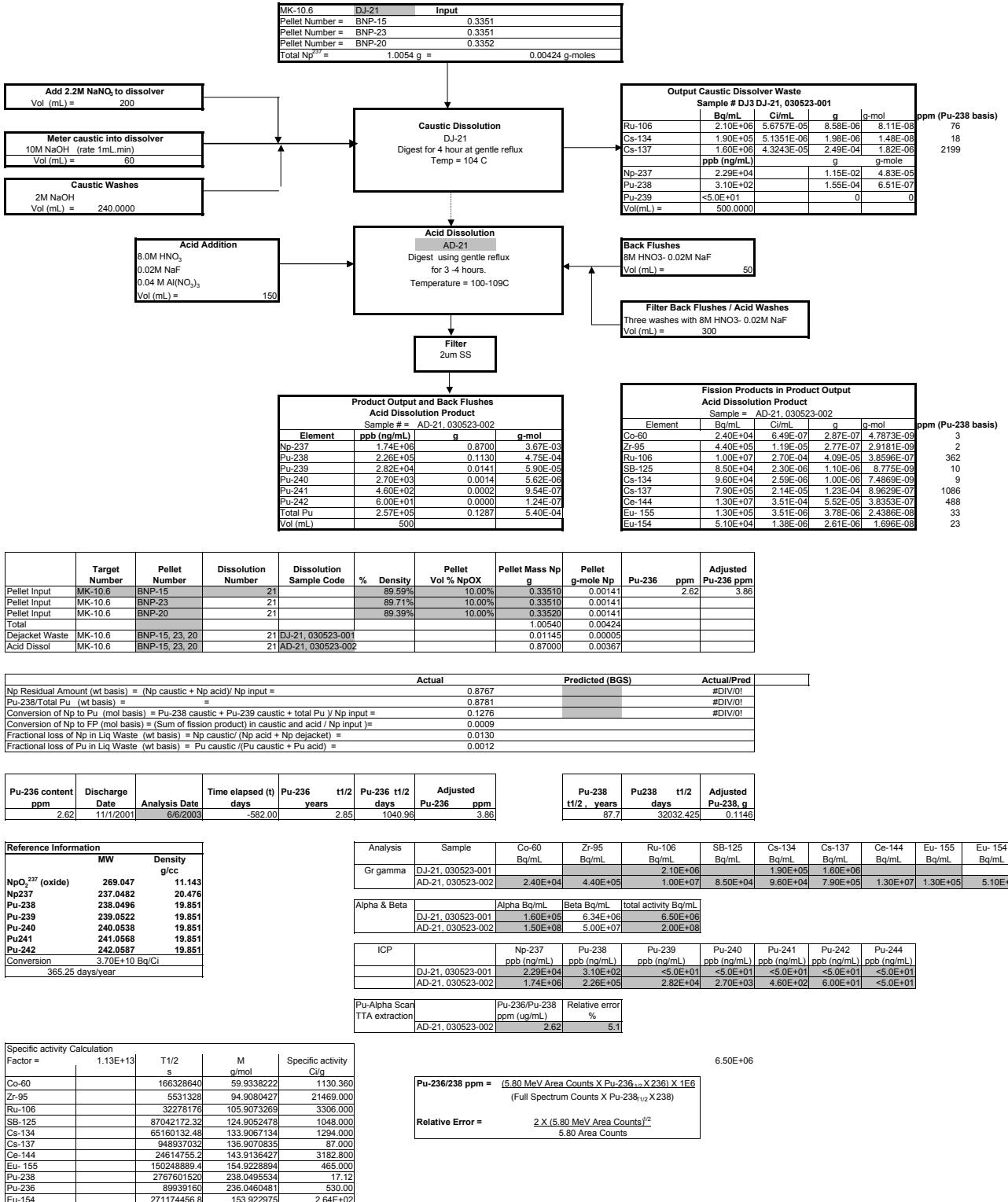
1. *Interim Status of Array Target Fabrication Report for the Advanced Test Reactor (ATR)*, Oak Ridge National Laboratory, Oak Ridge, Tennessee, DRAFT, September 30, 2001.
2. “Results of Np Target Capsule Testing,” memo from R. N. Morris, Oak Ridge National Laboratory, to R. M. Wham, Oak Ridge National Laboratory, August 9, 2001.
3. L. J. King, *TRU Operating Manual*, Section 1333, Radiochemical Engineering Development Center, Oak Ridge National Laboratory, January 1975.
4. Personal communication from B. G. Schnitzler, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, October 2003.

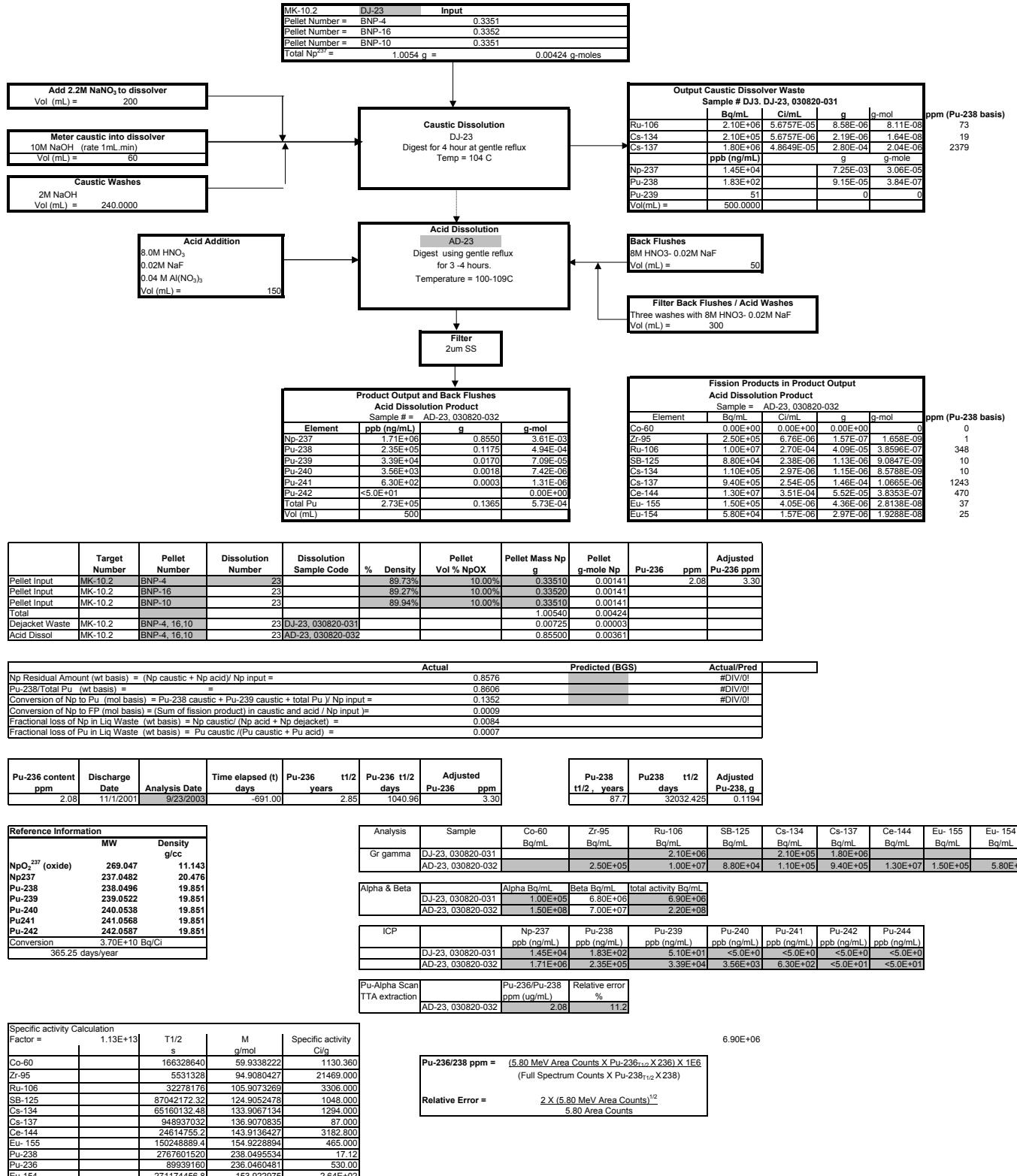


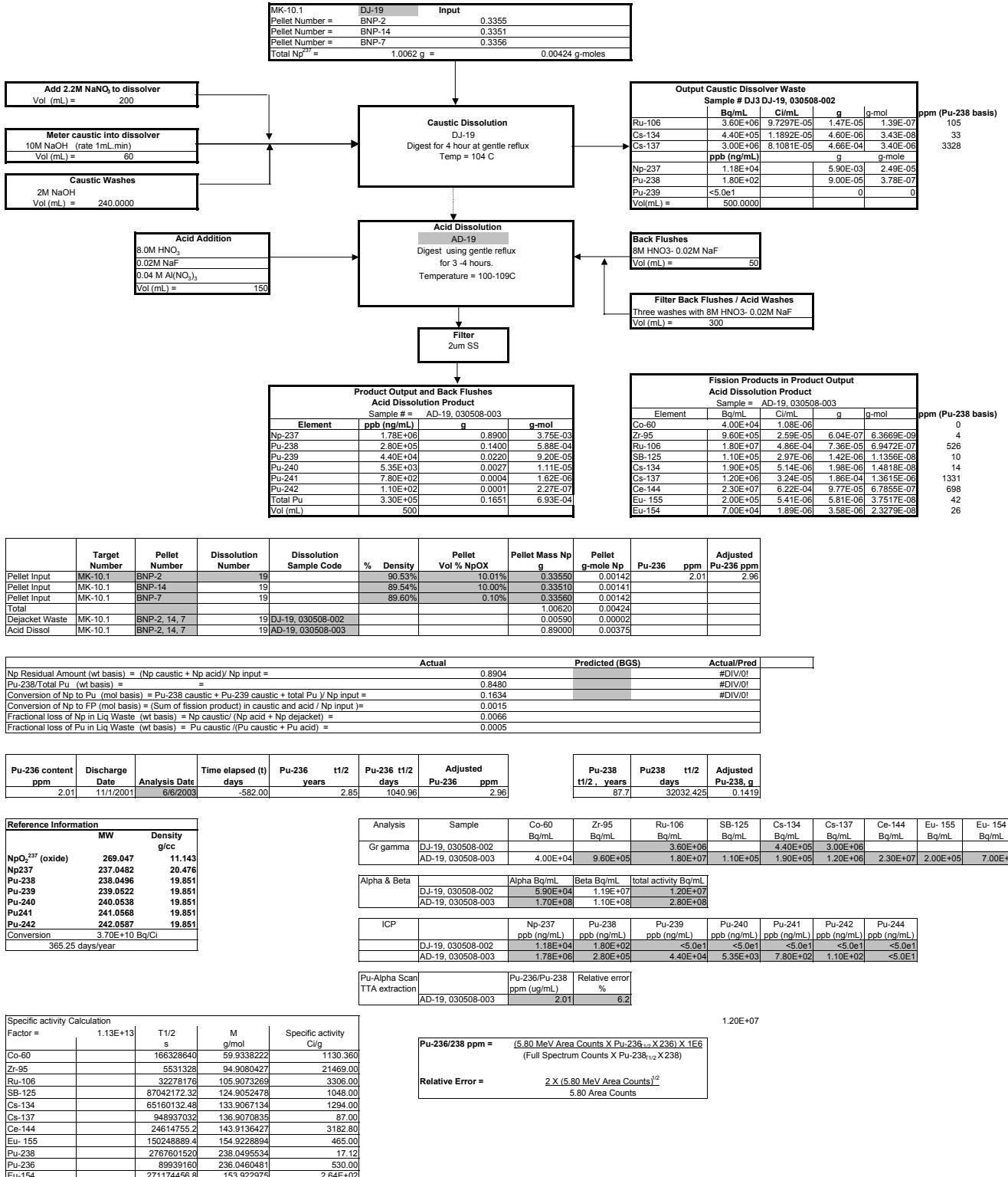
**APPENDIX A: Detailed Data Sheets for Targets and Dosimeters**

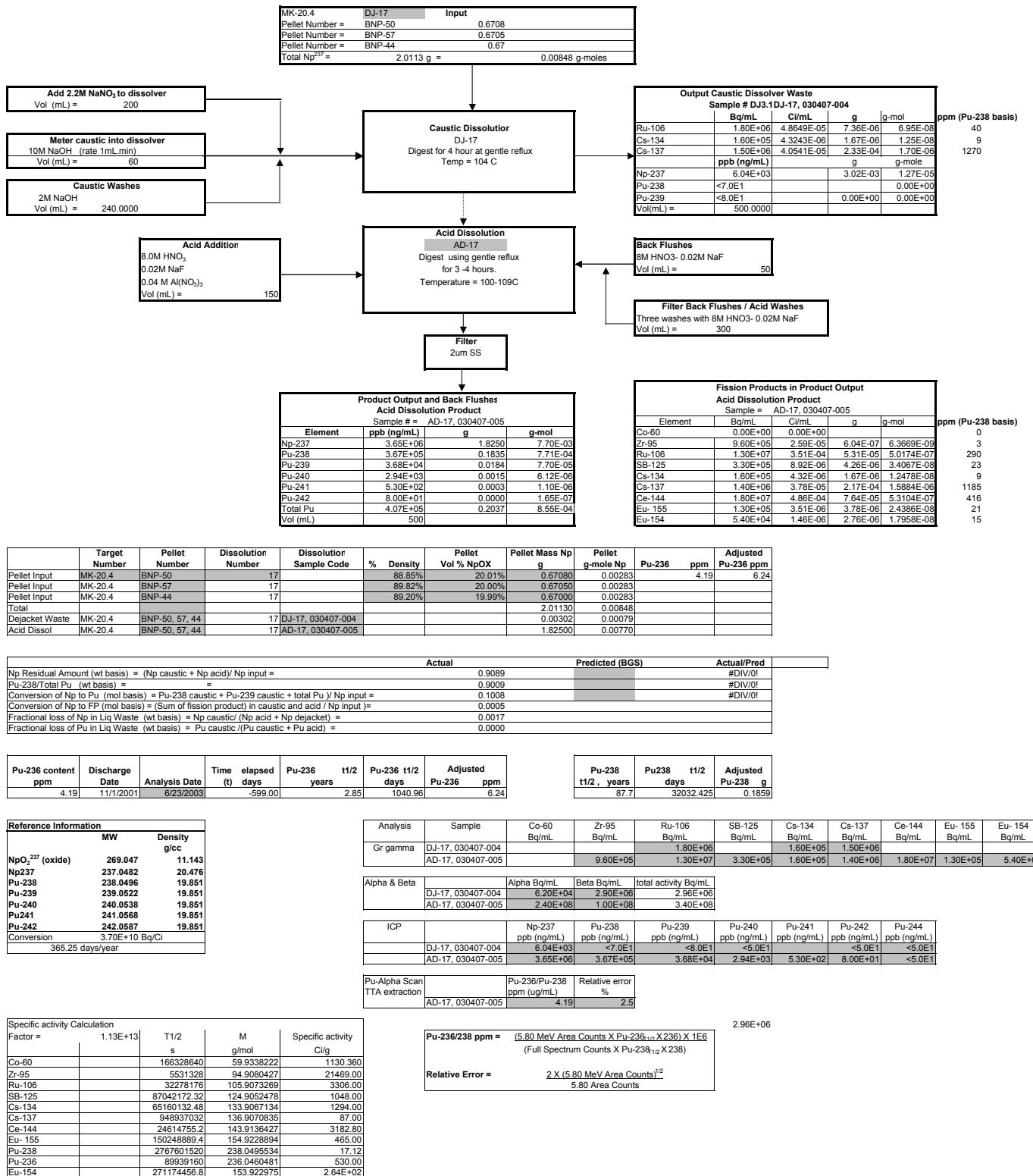


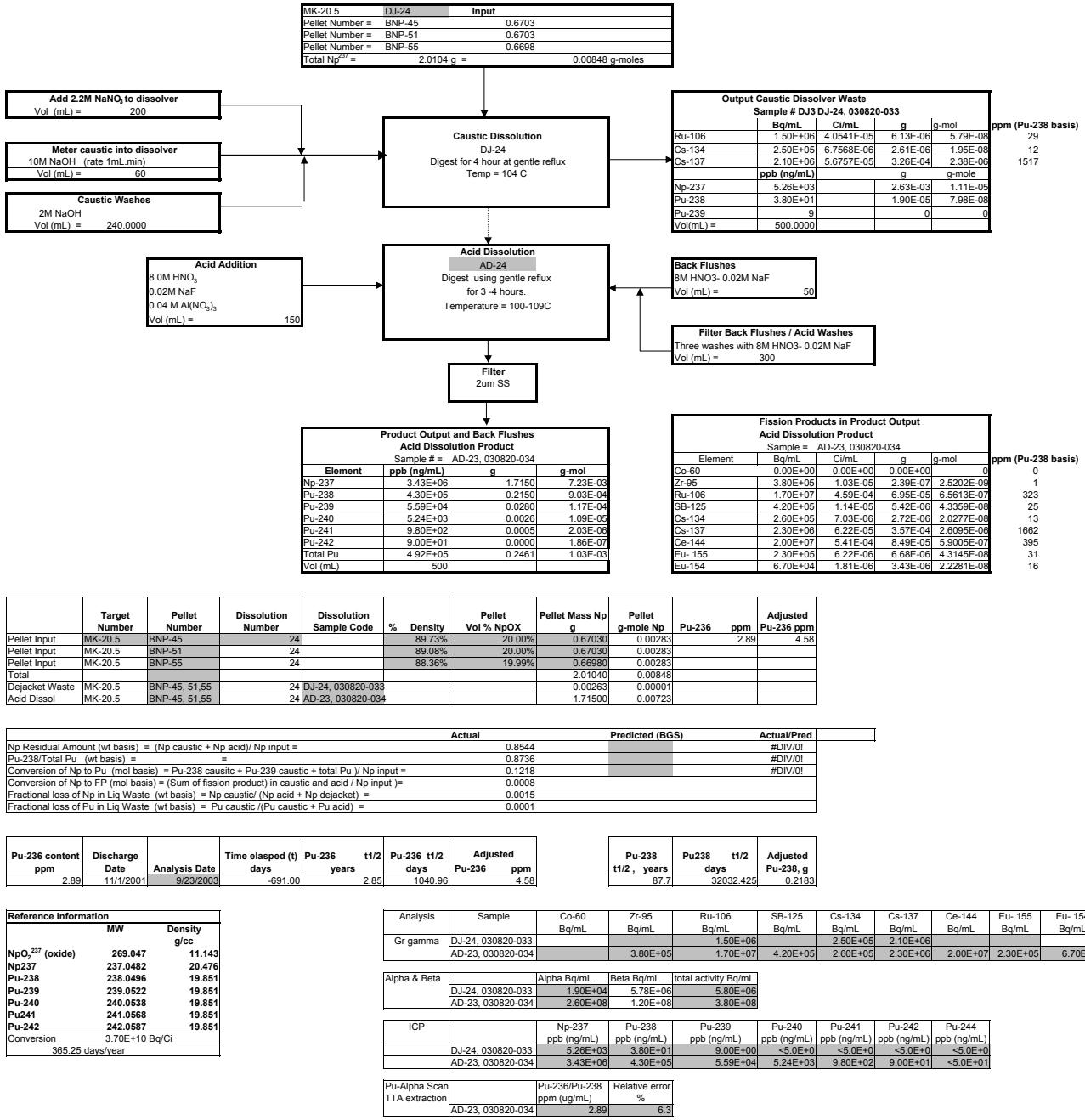








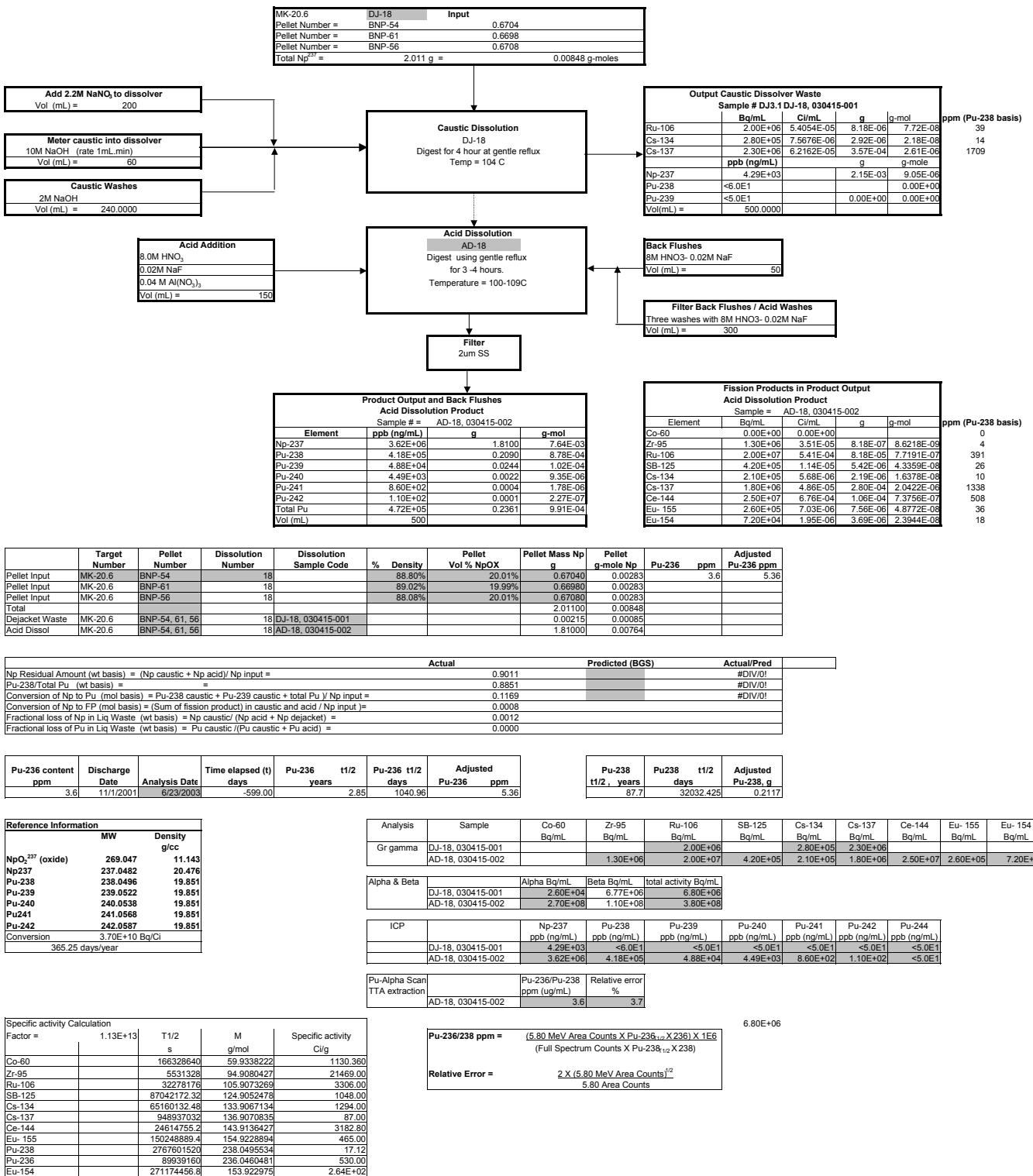


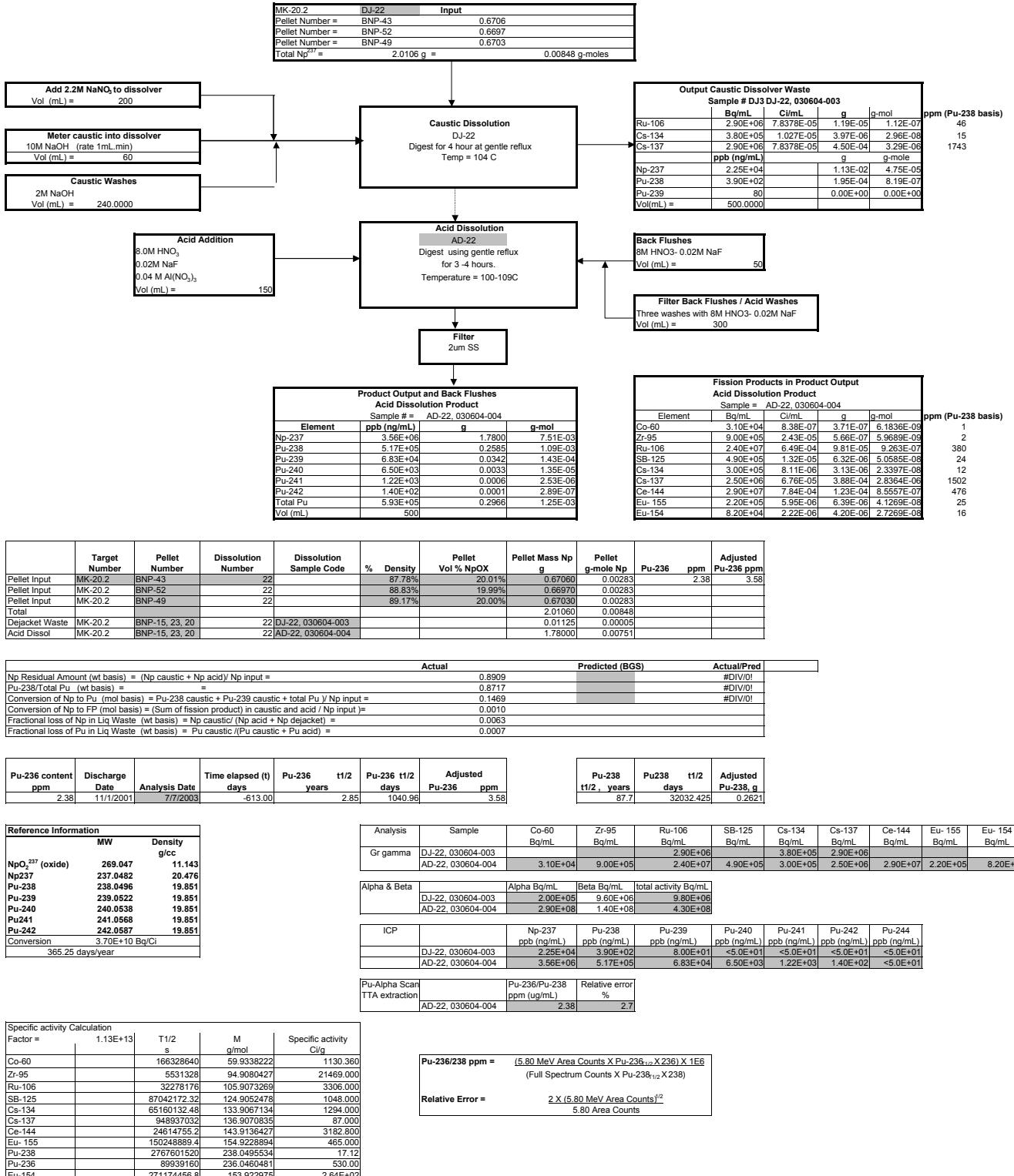


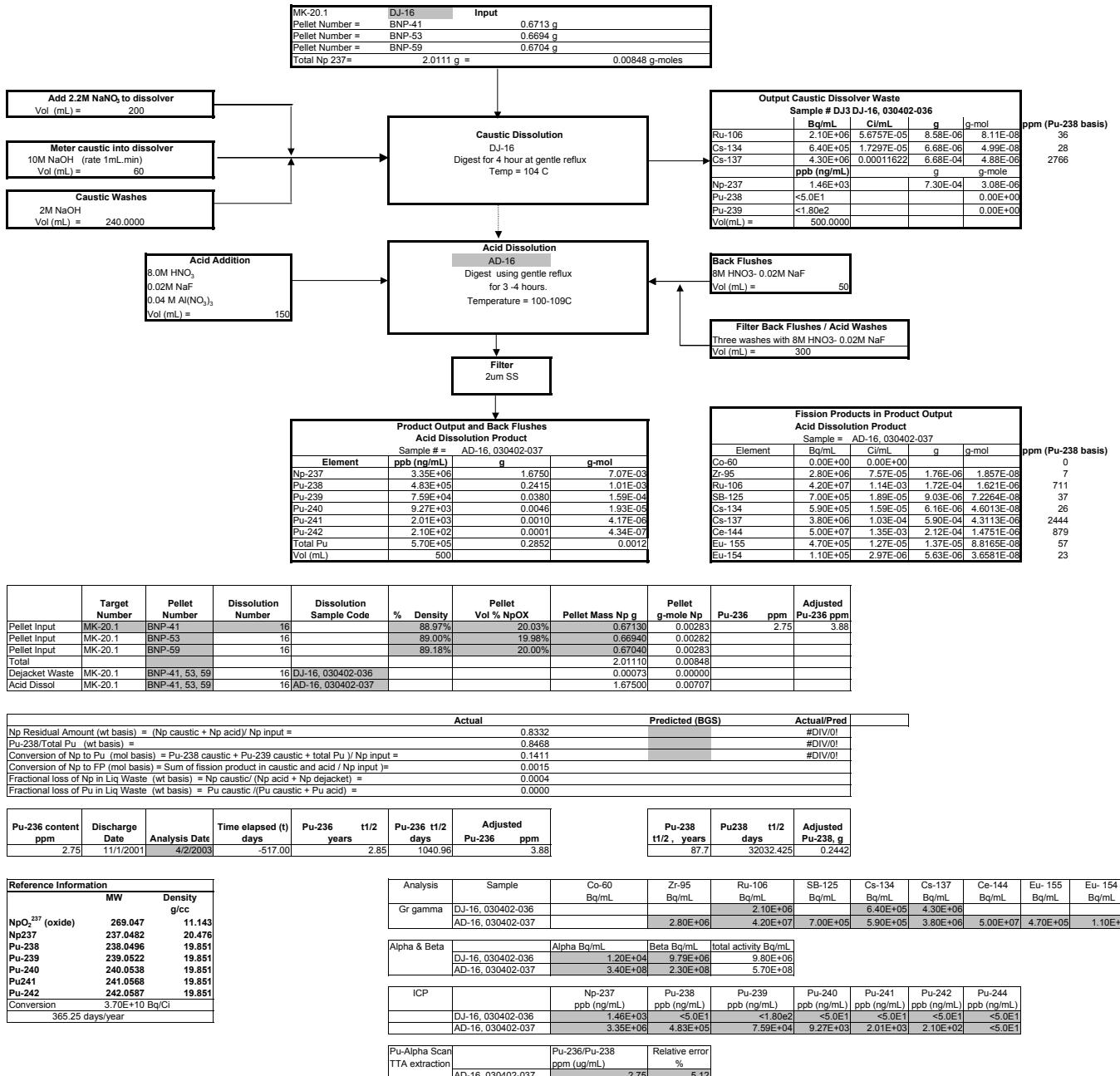
$$\text{Pu-236/238 ppm} = \frac{(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{1/2} \times \text{X238})}{(\text{Full Spectrum Counts} \times \text{Pu-238}_{1/2} \times \text{X238})} \times 10^6$$

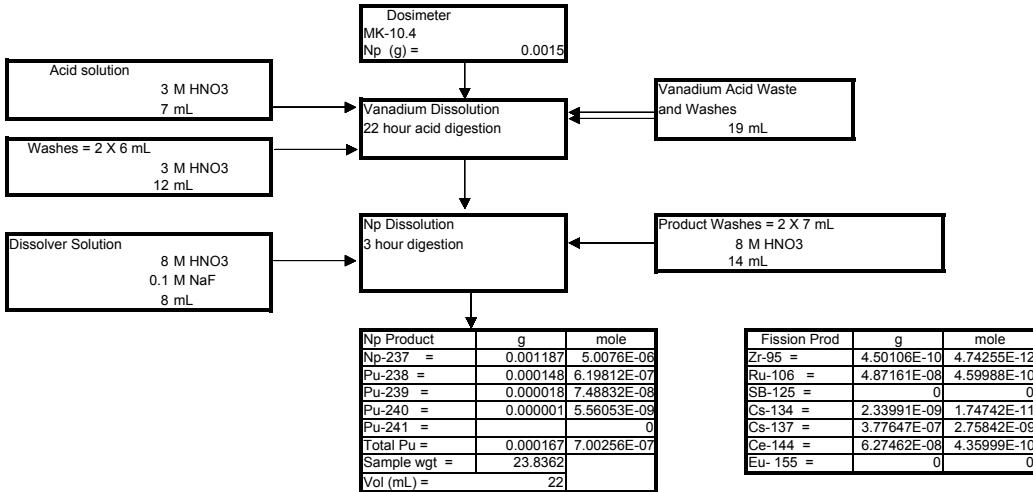
$$\text{Relative Error} = \frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$$

5.80E+06









Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu-236 ppm	Total Pu g
MK-10.4	ADD-26	0.001500	6.33E-06	27.1444	50.9806	23.836200	0.001187	5.01E-06	0.000148	1.840000	2.764341	0.000167
	DDJ-26			27.1495	45.4386	18.2891						

	Actual	Predicted (BGS)	Actual/Pred
Np Residual Amount (wt basis) Np prod / Np in	= 0.7914		#DIV/0!
Pu-238/Total Pu (wt basis)	= 0.8847		#DIV/0!
Conversion of Np to Pu (mol basis) = Pu total/ Np in	= 0.1107		#DIV/0!
Conversion of Np to FP (mol basis) = Fp prod/Np in	= 0.0006		
Fractional loss of Np in Liq Waste (wt basis)	= NA		
Fractional loss of Pu in Liq Waste (wt basis)	= NA		

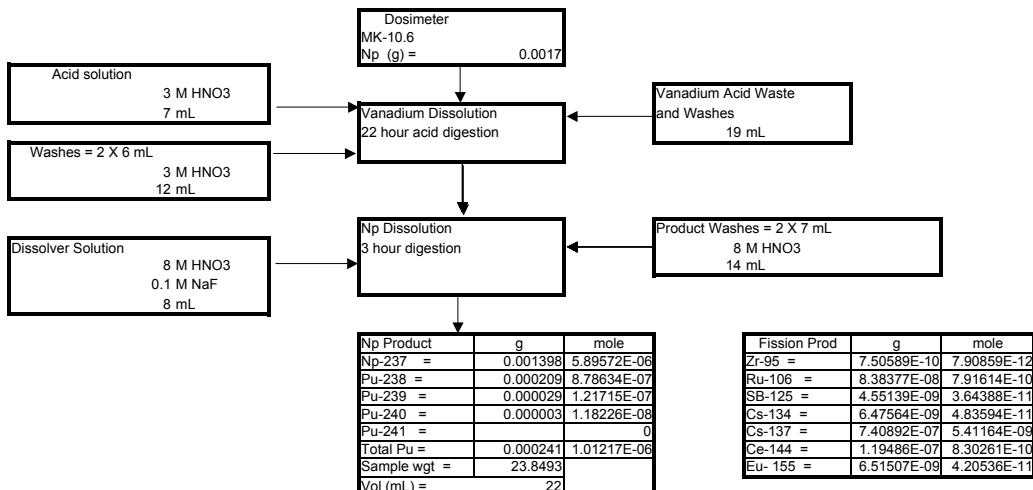
Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years	t1/2 days	Adjusted Pu-236 ppm	Analysis Date	Time elapsed (t) days	Pu-238 t1/2 years	Pu-238, t1/2 days	Adjusted Pu-238, g
1.84	11/1/2001	7/7/2003	-613.00	2.86	1043.88	2.76	5/28/2003	-573.00	87.7	32032.425	0.000149

Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu-155 Bq/g
Gr gamma	ADD-26, 030429-059	1.50E+04	2.50E+05	4.70E+03	5.10E+04	3.10E+05			
<hr/>									
Alpha & Beta									
Sample		Alpha Bq/g	Beta Bq/mL	Total activity Bq/m	alpha PHA	Pu238, Bq			
ADD-26, 030429-059		3.80E+06	Beta Bq/g	total activity Bq/g					
<hr/>									
ICP	Sample	Np-237 ppb (ng/g)	Pu-238 ppb (ng/g)	Pu-239 ppb (ng/g)	Pu-240 ppb (ng/g)	Pu-241 ppb (ng/g)	Pu-242 ppb (ng/g)	Pu-244 ppb (ng/g)	
	ADD-26, 030429-059	4.98E+04	6.19E+03	7.51E+02	5.60E+01	<4.00E+01	<4.00E+01	<4.00E+01	
<hr/>									
Pu-Alpha Scan TTA extraction	Sample	Pu-236/Pu-238 ppm (ug/mL)	Relative error %						
	ADD-26, 030429-059	1.84	6.2						

Reference Information			Reference Information			
	MW	Density g/cc	Specific activity Calculation Factor = 1.13E+13	T1/2 s	M g/mol	Specific activity Ci/g
NpO2	269.047	11.143				
Np237	237.0482	20.476				
Pu238	238.0496	19.851				
Pu239	239.0522	19.851				
Pu240	240.0538	19.851				
Pu241	241.0568	19.851				
Pu242	242.0587	19.851				
Conversion	3.70E+10 Bq/Ci					
	365.25 days/year					
N = N0 * exp(-(ln2/t1/2 * t))						

$$\text{Pu-236/238 ppm} = \frac{(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{1/2} \times 236) \times 1E6}{(Full Spectrum Counts \times \text{Pu-238}_{1/2} \times 238)}$$

$$\text{Relative Error} = \frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$$



Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu-236 ppm	Total Pu g
MK-10.6	ADD-27	0.001700	7.17E-06	26.9450	50.7943	23.849300	0.001398	5.90E-06	0.000209	1.590000	2.388751	0.000241
	DDJ-27			27.2825	46.4323	19.1498						

	Actual	Predicted (BGS)	Actual/Pred
Np Residual Amount (wt basis) Np prod / Np in	= 0.8221		#DIV/0!
Pu-238/Total Pu (wt basis)	= 0.8675		#DIV/0!
Conversion of Np to Pu (mol basis) = Pu total/ Np in	= 0.1411		#DIV/0!
Conversion of Np to FP (mol basis) = Fp prod/Np in	= 0.0010		
Fractional loss of Np in Liq Waste (wt basis)	= NA		
Fractional loss of Pu in Liq Waste (wt basis)	= NA		

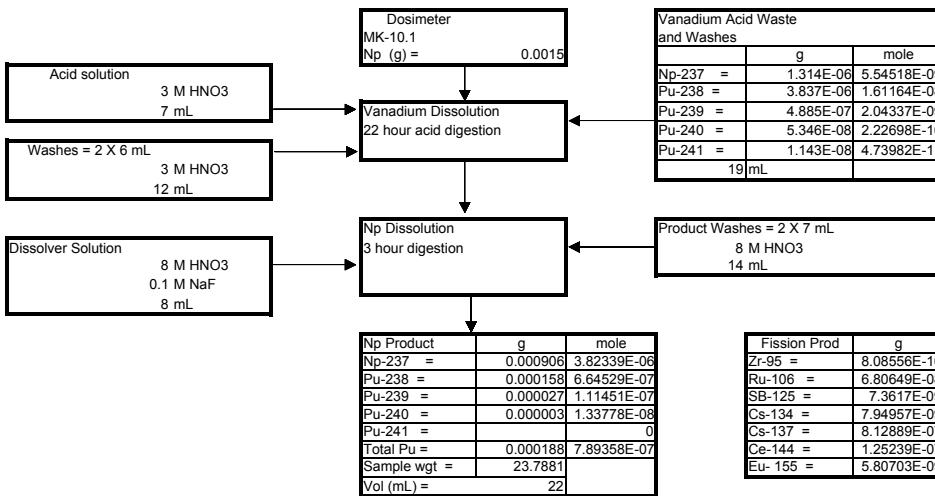
Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years	t1/2 days	Adjusted Pu-236 ppm	Analysis Date	Time elapsed (t) days	Pu-238 t1/2 years	Pu238, t1/2 days	Adjusted Pu-238, g
1.59	11/1/2001	7/7/2003	-613.00	2.86	1043.88	2.39	5/28/2003	-573.00	87.7	32032.425	0.000212

Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu- 155 Bq/g
Gr gamma	ADD-27, 030429-060	2.50E+04	4.30E+05	7.40E+03	1.30E+04	1.00E+05	5.90E+05	4.70E+03	
<hr/>									
Alpha & Beta									
Sample									
ADD-27, 030429-060									
Alpha Bq/g									
5.30E+06									
Beta Bq/g									
2.70E+06									
total activity Bq/g									
8.00E+06									
alpha PHA									
Pu238, Bq									
<hr/>									
ICP									
Sample									
ADD-27, 030429-060									
Np-237 ppb (ng/g)									
5.86E+04									
Pu-238 ppb (ng/g)									
8.77E+03									
Pu-239 ppb (ng/g)									
1.22E+03									
Pu-240 ppb (ng/g)									
1.19E+02									
<3.97E+01									
Pu-241 ppb (ng/g)									
<3.97E+01									
Pu-242 ppb (ng/g)									
<3.97E+01									
<hr/>									
Pu-Alpha Scan TTA extraction									
Sample									
ADD-27, 030429-060									
Pu-236/Pu-238 ppm (ug/mL)									
1.59									
Relative error %									
5									

Reference Information		Reference Information				
	MW	Density g/cc	Specific activity Calculation Factor = 1.13E+13	T1/2 s	M g/mol	Specific activity Ci/g
NpO2	269.047	11.143				1130.670
Np237	237.0482	20.476				21469.000
Pu238	238.0496	19.851				3306.000
Pu239	239.0522	19.851				1048.000
Pu240	240.0538	19.851				1294.000
Pu241	241.0568	19.851				87.000
Pu242	242.0567	19.851				
Conversion	3.70E+10	Bq/Ci				
	365.25	days/year				
N = N <sup>0</sup> * exp(-(ln2/t <sub>1/2</sub> ) * t)						

$$\text{Pu-236/238 ppm} = \frac{(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{1/2} \times 236)}{(Full Spectrum Counts \times \text{Pu-238}_{1/2} \times 238)}$$

$$\text{Relative Error} = \frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$$



Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu-236 ppm	Total Pu g
MK-10.1	ADD-25	0.001500	6.33E-06	27.0241	50.8122	23.788100	0.000906	3.82E-06	0.000158	2.540000	3.790738	0.000188
	DJJ-25			27.1424	48.1069	20.964500	0.0000131					0.000919

Actual		Predicted (BGS)		Actual/Pred	
Np Residual Amount (wt basis) Np prod / Np in	=	0.6042		#DIV/0!	
Pu-238/Total Pu (wt basis)	=	0.8412		#DIV/0!	
Conversion of Np to Pu (mol basis) = Pu total/ Np in	=	0.1247		#DIV/0!	
Conversion of Np to FP (mol basis) = Fp prod/Np in	=	0.0012			
Fractional loss of Np in Liq Waste (wt basis)	=	NA			
Fractional loss of Pu in Liq Waste (wt basis)	=	NA			

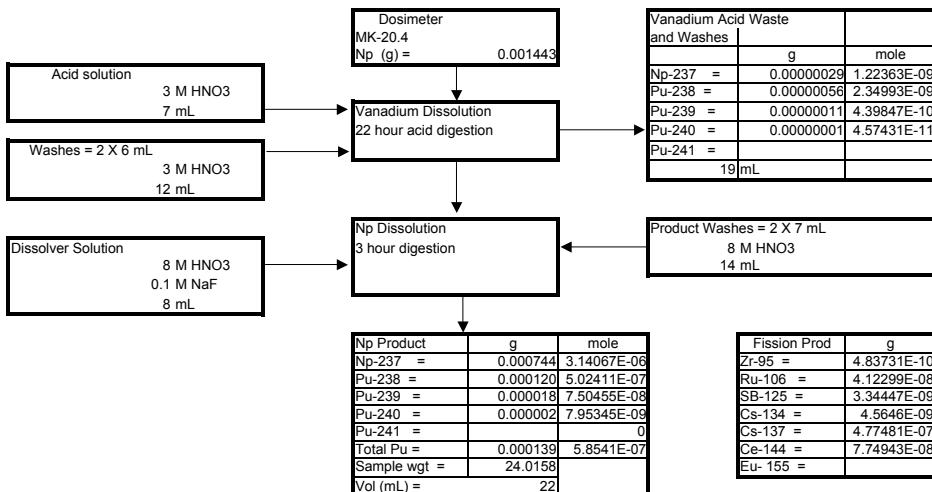
Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years		Adjusted Pu-236 ppm	Analysis Date	Time elapsed (t) days	Pu-238 t1/2 years	Pu238, t1/2 days	Adjusted Pu-238, g
				t1/2	years						
2.54	11/1/2001	6/27/2003	-603.00	2.86	1043.88	3.79	5/28/2003	-573.00	87.7	32032.425	0.000160

Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu-155 Bq/g
Gr gamma	ADD-25, 030429-058		2.70E+04	3.50E+05	1.20E+04	1.60E+04	1.10E+05	6.20E+05	4.20E+03
	DDJ-25, 030820-037	7.30E+03		3.20E+04		2.00E+03	6.00E+03		
Alpha & Beta	Sample	Alpha Bq/g	Beta Bq/g	total activity Bq/g	alpha PHA	Pu238, Bq			
	ADD-25, 030429-058	4.30E+06	2.50E+06	6.80E+06					
	DDJ-25, 030820-037	1.10E+05	2.00E+04	1.30E+05					
ICP	Sample	Np-237 ppb (ng/g)	Pu-238 ppb (ng/g)	Pu-239 ppb (ng/g)	Pu-240 ppb (ng/g)	Pu-241 ppb (ng/g)	Pu-242 ppb (ng/g)	Pu-244 ppb (ng/g)	
	ADD-25, 030429-058	3.81E+04	6.65E+03	1.12E+03	1.35E+02	<3.97E+01	<3.97E+01	<3.97E+01	
	DDJ-25, 030820-037	6.27E+02	1.83E+02	2.33E+01	2.55E+00	5.45E-01	<4.55E-01	<4.55E-01	
Pu-Alpha Scan TTA extraction	Sample	Pu-236/Pu-238 Relative error ppm (ug/mL)	%						
	ADD-25, 030429-058	2.54	4.2						

Reference Information		Reference Information				
	MW	Density g/cc	Factor = 1.13E+13	T1/2 s	M g/mol	Specific activity Ci/g
NpO <sub>2</sub>	269.047	11.143				
Np-237	237.0482	20.476				
Pu-238	238.0496	19.851				
Pu-239	239.0522	19.851				
Pu-240	240.0538	19.851				
Pu-241	241.0568	19.851				
Pu-242	242.0587	19.851				
Conversion	3.70E+10	Bq/Ci				
	365.25	days/year				
N = N <sup>o</sup> * exp -(ln2/t <sub>1/2</sub> * t)						
Pu-238						

$$\text{Pu-236/238 ppm} = \frac{(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{\text{t1/2}} \times \text{Pu-236}) \times 10^6}{(\text{Full Spectrum Counts} \times \text{Pu-238}_{\text{t1/2}} \times \text{Pu-238})}$$

$$\text{Relative Error} = \frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$$



Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu-236 ppm	Total Pu g
MK-20.4	ADD-23	0.001443	6.09E-06	27.0334	51.0492	24.015800	0.000744	3.14E-06	0.000120	2.380000	3.551951	0.000139
	DDJ-23			27.1932	44.4586	17.265400	0.0000029					

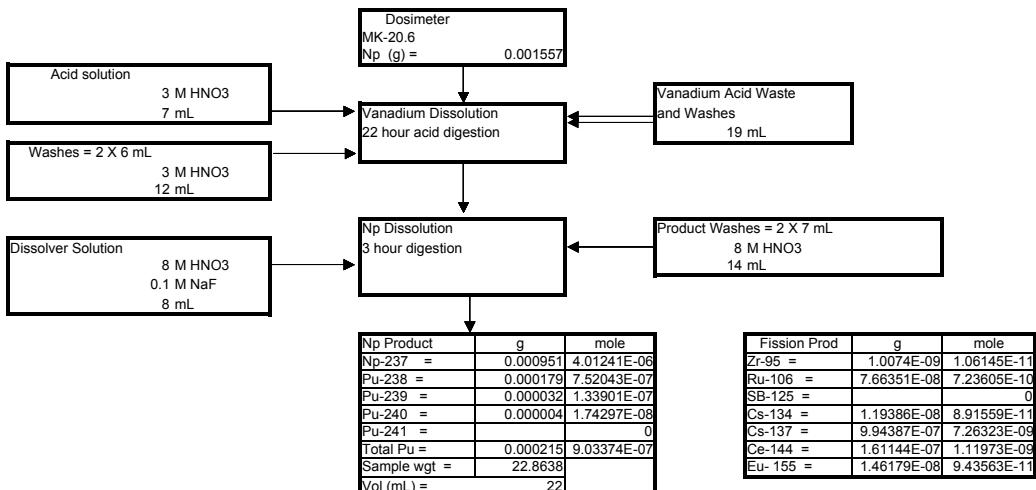
Actual		Predicted (BGS)		Actual/Pred	
Np Residual Amount (wt basis) Np prod / Np in	=	0.5159		#DIV/0!	
Pu-238/Total Pu (wt basis)	=	0.8577		#DIV/0!	
Conversion of Np to Pu (mol basis) = Pu total/ Np in	=	0.0962		#DIV/0!	
Conversion of Np to FP (mol basis) = Fp prod/Np in	=	0.0007			
Fractional loss of Np in Liq Waste (wt basis)	=	NA			
Fractional loss of Pu in Liq Waste (wt basis)	=	NA			

Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years	t1/2 days	Adjusted Pu-236 ppm	Analysis Date	Time elapsed (t) days	Pu-238 t1/2 years	Pu238, t1/2 days	Adjusted Pu-238, g
2.38	11/1/2001	6/27/2003	-603.00	2.86	1043.88	3.55	5/28/2003	-573.00	87.7	32032.425	0.000121

Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu-155 Bq/g
Gr gamma	ADD-23, 030429-056	1.60E+04	2.10E+05	5.40E+03	9.10E+03	6.40E+04	3.80E+05	3.80E+03	
	DDJ-23, 030820-036	7.30E+03	1.80E+04		2.10E+03	4.70E+03			
Alpha & Beta	Sample	Alpha Bq/g	Beta Bq/g	total activity Bq/g	alpha PHA	Pu238, Bq			
	ADD-23, 030429-056	2.90E+06	1.80E+06	4.70E+06					
	DDJ-23, 030820-036	1.90E+04	2.51E+05	2.70E+05					
ICP	Sample	Np-237 ppb (ng/g)	Pu-238 ppb (ng/g)	Pu-239 ppb (ng/g)	Pu-240 ppb (ng/g)	Pu-241 ppb (ng/g)	Pu-242 ppb (ng/g)	Pu-244 ppb (ng/g)	
	ADD-23, 030429-056	3.10E+04	4.98E+03	7.47E+02	7.95E+01	<3.97E+01	<3.97E+01	<3.97E+01	
	DDJ-23, 030820-036	1.68E+02	3.24E+01	6.09E+00	6.36E-01	<4.55E-01	<4.55E-01	<4.55E-01	
Pu-Alpha Scan TTA extraction	Sample	Pu-236/Pu-238 ppm (ug/mL)	Relative error %						
	ADD-23, 030429-056	2.38	5.2						

Reference Information		Reference Information			
		Specific activity Calculation Factor = 1.13E+13			
NpO2	269.047	11.143	T1/2 s	M g/mol	Specific activity Ci/g
Np237	237.0482	20.476	Co-60	166328640	59.9338222 1130.670
Pu238	238.0496	19.851	Zr-95	5531328	94.9080427 21469.000
Pu239	239.0522	19.851	Ru-106	32278176	105.9073269 3306.000
Pu240	240.0538	19.851	SB-125	87042172.32	124.9052478 1048.000
Pu241	241.0568	19.851	Cs-134	65160132.48	133.9067134 1294.000
Pu242	242.0587	19.851	Cs-137	948937032	136.9070835 87.000
Conversion	3.70E+10	Bq/Ci	Ce-144	24614755.2	143.9136427 3182.800
	365.25 days/year		Eu-155	150248889.4	154.9228894 465.000
N = N <sup>0</sup> * exp -(ln2/t <sub>1/2</sub> * t)			Pu-238	2767601520	238.0495534 17.119

Pu-236/238 ppm =  $(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{1/2} \times \text{X} 236) \times 10^6$   
 (Full Spectrum Counts X Pu-238<sub>1/2</sub> X 238)  
 Relative Error =  $\frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$



Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu-236 ppm	Total Pu g
MK-20.6	ADD-24	0.001557	6.57E-06	27.1896	50.0534	22.863800	0.000951	4.01E-06	0.000179	2.840000	4.238463	0.000215
	DJJ-24			27.1381	46.6526	19.5145						

Actual		Predicted (BGS)		Actual/Pred	
Np Residual Amount (wt basis)	Np prod / Np in	=	0.6109	#DIV/0!	
Pu-238/Total Pu (wt basis)		=	0.8318	#DIV/0!	
Conversion of Np to Pu (mol basis) = Pu total / Np in		=	0.1375	#DIV/0!	
Conversion of Np to FP (mol basis) = Fp prod/Np in		=	0.0014		
Fractional loss of Np in Liq Waste (wt basis)		=	NA		
Fractional loss of Pu in Liq Waste (wt basis)		=	NA		

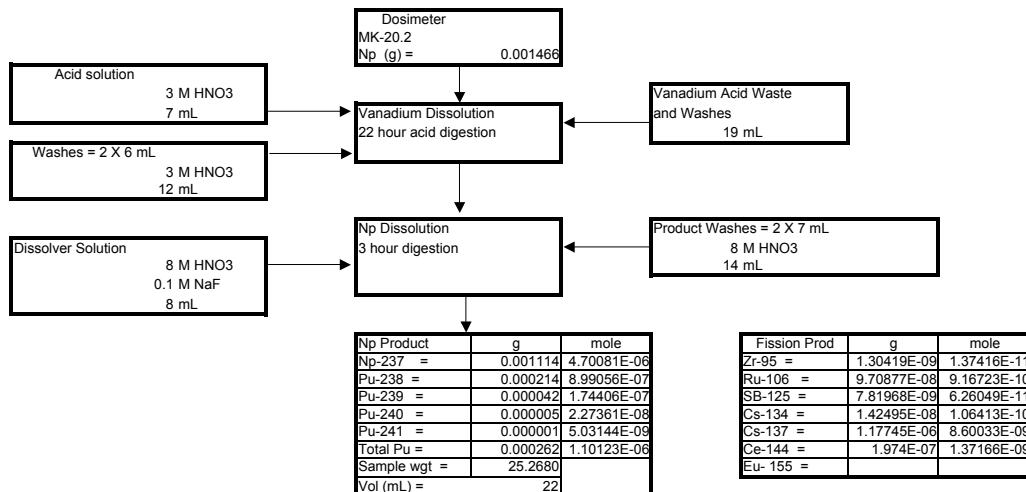
Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years	t1/2 days	Adjusted Pu-236 ppm	Analysis Date	Time elapsed (t) days	Pu-238 t1/2 years	Pu238, t1/2 days	Adjusted Pu-238, g
2.84	11/1/2001	6/27/2003	-603.00	2.86	1043.88	4.24	5/28/2003	-573.00	87.7	32032.425	0.000181

Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu- 155 Bq/g
Gr gamma	ADD-24, 030429-057	3.50E+04	4.10E+05		2.50E+04	1.40E+05	8.30E+05	1.10E+04	
Alpha & Beta									
Sample Alpha Bq/g Beta Bq/g total activity Bq/g alpha PHA Pu238, Bq									
ADD-24, 030429-057 4.90E+06 3.60E+06 8.50E+06									
ICP									
Sample Np-237 ppb (ng/g) Pu-238 ppb (ng/g) Pu-239 ppb (ng/g) Pu-240 ppb (ng/g) Pu-241 ppb (ng/g) Pu-242 ppb (ng/g) Pu-244 ppb (ng/g)									
ADD-24, 030429-057 4.16E+04 7.83E+03 1.40E+03 1.83E+02 <3.98E+01 <3.98E+01 <3.98E+01									
Pu-Alpha Scan TTA extraction									
Sample Pu-236/Pu-238 Relative error ppm (ug/mL) %									
ADD-24, 030429-057 2.84 4.5									

Reference Information					
	MW	Density g/cc	Specific activity Calculation Factor = 1.13E+13	T1/2 s	M g/mol Specific activity Ci/g
NpO2	269.047	11.143			
Np237	237.0482	20.476			
Pu238	238.0496	19.851	Co-60	166328640	59.9338222, 1130.670
Pu239	239.0522	19.851	Zr-95	5531328	94.9080427, 21469.000
Pu240	240.0538	19.851	Ru-106	32278176	105.9073269, 3306.000
Pu241	241.0568	19.851	SB-125	87042172.32	124.9052478, 1048.000
Pu242	242.0587	19.851	Cs-134	65160132.48	133.9067134, 1294.000
Conversion	3.70E+10	Bq/Ci	Cs-137	948937032	136.9070835, 87.000
	365.25	days/year	Ce-144	24614755.2	143.9136427, 3182.800
N = N° * exp -(ln2/t1/2 * t)					
			Eu- 155	150248889.4	154.9228894, 465.000
			Pu-238	2767601520	238.0495534, 17.119

$$\text{Pu-236/238 ppm} = \frac{(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{1/2} \times 236) \times 1E6}{(Full Spectrum Counts \times \text{Pu-238}_{1/2} \times 238)}$$

$$\text{Relative Error} = \frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$$



Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu- 236 ppm	Total Pu g
MK-20.2	ADD-22	0.001466	6.18E-06	27.1953	52.4633	25.268000	0.001114	4.70E-06	0.000214	2.230000	3.328089	0.000262
	DDJ-22			26.9513	46.8505	19.99200						

	Actual	Predicted (BGS)	Actual/Pred
Np Residual Amount (wt basis) Np prod / Np in	=	0.7601	#DIV/0!
Pu-238/Total Pu (wt basis)	=	0.8157	#DIV/0!
Conversion of Np to Pu (mol basis) = Pu total/ Np in	=	0.1781	#DIV/0!
Conversion of Np to FP (mol basis) = Fp prod/Np in	=	0.0018	
Fractional loss of Np in Liq Waste (wt basis)	=	NA	
Fractional loss of Pu in Liq Waste (wt basis)	=	NA	

Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years	t1/2 days	Adjusted Pu-236 ppm		Analysis Date	Time elapsed (t) days	Pu-238 t1/2 years	Pu238, t1/2 days	Adjusted Pu-238, g
2.23	11/1/2001	6/27/2003	-603.00	2.86	1043.88	3.33		5/28/2003	-573.00	87.7	32032.425	0.000217

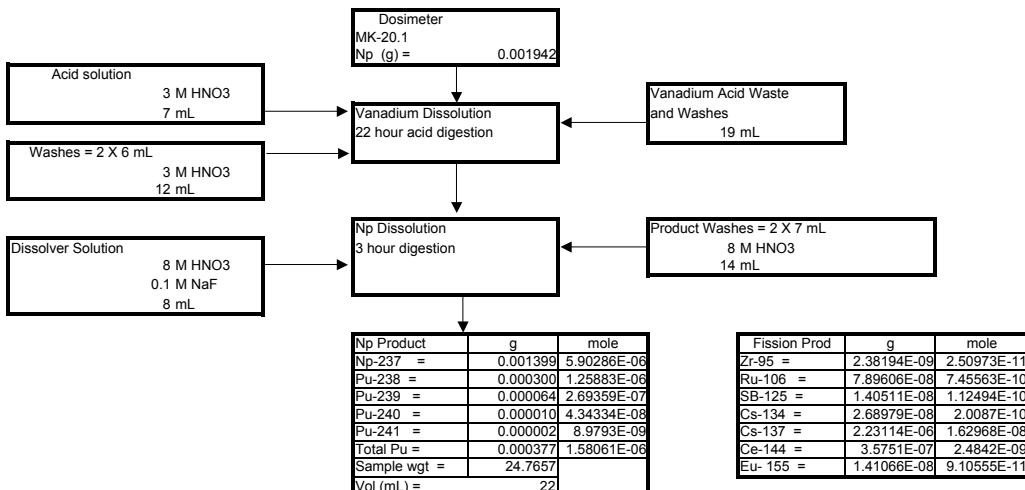
Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu- 155 Bq/g
Gr gamma	ADD-22 030429-055	4.10E+04	4.70E+05	1.20E+04	2.70E+04	1.50E+05	9.20E+05		

Alpha & Beta	Sample	Alpha Bq/g	Beta Bq/g	total activity Bq/g	alpha PHA	Pu238, Bq
	ADD-22, 030429-055	5.40E+06	3.80E+06	9.20E+06		

ICP	Sample	Np-237 ppb (nG/g)	Pu-238 ppb (nG/g)	Pu-239 ppb (nG/g)	Pu-240 ppb (nG/g)	Pu-241 ppb (nG/g)	Pu-242 ppb (nG/g)	Pu-244 ppb (nG/g)
ADD-22, 030429-055		4.41E+04	8.47E+03	1.65E+03	2.16E+02	4.80E+01	<4.0 E+01	<4.0 E+01

Pu-Alpha Scan TTA extraction	Sample	Pu-236/Pu-238 ppm (ug/mL)	Relative error %
	ADD-22_030429-055	2.23	6.2

Pu-236/238 ppm =	$\frac{(5.80 \text{ MeV Area Counts} X \text{Pu-236}_{1/2} X 236) X 1E6}{(\text{Full Spectrum Counts} X \text{Pu-238}_{1/2} X 238)}$
Relative Error =	$\frac{2 X (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$



Target	Diss #	Dosimeter Np input g	Np input mole	Vial Tare g	Solution & Vial g	Solution weight g	Np Prod. g	Np Prod. mole	Pu-238 g	Pu-236 ppm	Adjusted Pu- 236 ppm ppm	Total Pu g
MK-20.1	ADD-21	0.001942	8.19E-06	26.9772	51.7429	24.765700	0.001399	5.90E-06	0.000300	4.197451	0.000377	
	DDJ-21			27.0216	45.8164	18.7948						

	Actual	Predicted (BGS)	Actual/Pred
Np Residual Amount (wt basis) Np prod / Np in	= 0.7205	#DIV/0!	
Pu-238/Total Pu (wt basis)	= 0.7956	#DIV/0!	
Conversion of Np to Pu (mol basis) = Pu total/ Np in	= 0.1929	#DIV/0!	
Conversion of Np to FP (mol basis) = Fp prod/Np in	= 0.0024		
Fractional loss of Np in Liq Waste (wt basis)	= NA		
Fractional loss of Pu in Liq Waste (wt basis)	= NA		

Pu-236 content ppm	Discharge Date	Analysis Date	Time elapsed (t) days	Pu-236 t1/2 years	t1/2 days	Adjusted Pu-236 ppm	Analysis Date	Time elapsed (t) days	Pu-238 t1/2, years	Pu-238, t1/2 days	Adjusted Pu-238, g
2.82	11/1/2001	6/23/2003	-599.00	2.86	1043.88	4.20	5/28/2003	-573.00	87.7	32032.425	0.000303

Analysis	Sample	Co-60 Bq/g	Zr-95 Bq/g	Ru-106 Bq/g	SB-125 Bq/g	Cs-134 Bq/g	Cs-137 Bq/g	Ce-144 Bq/g	Eu- 155 Bq/g
Gr gamma	ADD-21, 030429-054	7.64E+04	3.90E+05	2.20E+04	5.20E+04	2.90E+05	1.70E+06	9.80E+03	

Alpha & Beta	Sample	Alpha Bq/g	Beta Bq/g	total activity Bq/g	alpha PHA	Pu238, Bq
	ADD-21, 030429-054	7.60E+06	5.40E+06	1.30E+07		

ICP	Sample	Np-237 ppb (ng/g)	Pu-238 ppb (ng/g)	Pu-239 ppb (ng/g)	Pu-240 ppb (ng/g)	Pu-241 ppb (ng/g)	Pu-242 ppb (ng/g)	Pu-244 ppb (ng/g)
	ADD-21, 030429-054	5.65E+04	1.21E+04	2.60E+03	4.21E+02	8.74E+01	<3.97E+01	<3.97E+01

Pu-Alpha Scan TTA extraction	Sample	Pu-236/Pu-238 Relative error ppm (ug/mL)	%
	ADD-21, 030429-054	2.82	3.7

Reference Information			Reference Information				
	MW	Density g/cc	Specific activity Calculation	Factor =	T1/2 s	M g/mol	Specific activity Ci/g
NpO2	269.047	11.143		1.13E+13			
Np237	237.0482	20.476			166328640	59.9338222	1130.670
Pu238	238.0496	19.851			5531328	94.9080427	21469.000
Pu239	239.0522	19.851			32278176	105.9073269	3306.000
Pu240	240.0538	19.851			87042172.32	124.9052478	1048.000
Pu241	241.0568	19.851			65160132.48	133.9067134	1294.000
Pu242	242.0587	19.851			948937032	136.9070835	87.000
Conversion	3.70E+10	Bq/Ci			24614755.2	143.9136427	3182.800
	365.25 days/year				150248889.4	154.9228894	465.000
	N = N <sup>0</sup> * exp -(ln(2/t <sub>1/2</sub> ) * t)				2767601520	238.0495534	17.119

$\text{Pu-236/238 ppm} = \frac{(5.80 \text{ MeV Area Counts} \times \text{Pu-236}_{\text{t1/2}} \times 236) \times 10^6}{(\text{Full Spectrum Counts} \times \text{Pu-238}_{\text{t1/2}} \times 238)}$   
 $\text{Relative Error} = \frac{2 \times (5.80 \text{ MeV Area Counts})^{1/2}}{5.80 \text{ Area Counts}}$

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