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MOLYBDENUM-99 TECHNOLOGICAL DEVELOPMENT**

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**Development of Frontend Processing to Allow Use of High-Density  
LEU Foil Targets in Current Mo-99 Production Facilities**

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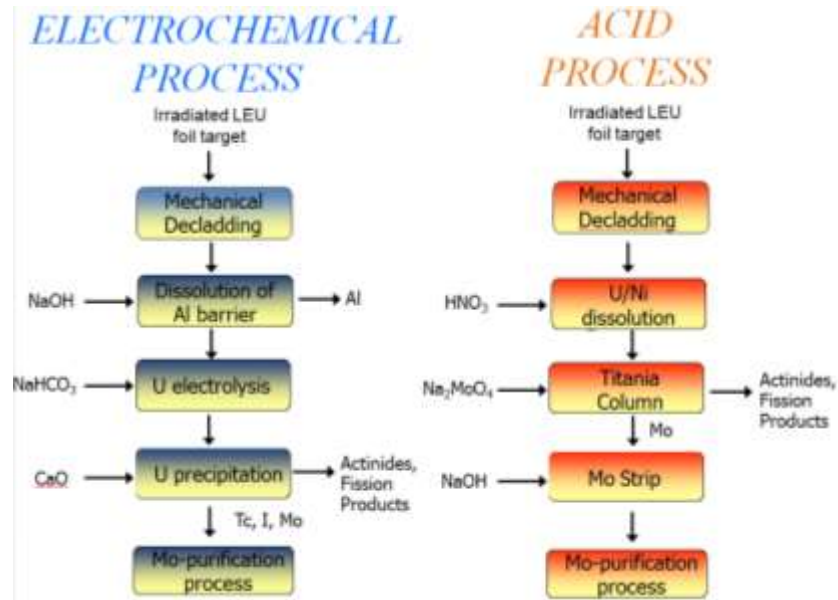
**ABSTRACT**

In order to meet the Mo-99 yields produced by highly-enriched uranium (HEU) fission while maintaining the scope of the Global Threat Reduction Initiative (GTRI), high-density low enriched elemental uranium (LEU) targets could offer a competitive edge. This denser fuel material (as compared to uranium-aluminide, -oxide, or -silicide) will allow similar or increased U-235 content in an annular foil target than the current HEU target, which would benefit the neutron economy and fission yields during irradiations. Two frontend processes for treating irradiated LEU targets were developed and tested at Argonne. The product of these frontend processes is a solution that could be fed into current Mo-purification processes. One process investigated the ambient pressure dissolution of irradiated LEU foil with nitric acid. Following the dissolution, the acidic dissolver solution containing uranium and all activation and fission products was fed to a column containing a TiO<sub>2</sub> sorbent that is specific for Mo. The Mo held on the column was stripped from the column using alkaline solution, which is a perfect feed for current purification processes. In the second frontend process, LEU foils were electrochemically dissolved by anodic oxidation in a bicarbonate solution. The cationic actinides and fission products were precipitated using CaO, leaving the anionic Mo in an alkaline solution. These processes were demonstrated using low-burnup uranium foils.

**1. Introduction**

In light of the GTRI, which addresses nuclear-proliferation issues, the production of the medical isotope Tc-99m and its parent isotope Mo-99 should not rely on the use of (HEU). However, to produce HEU equivalent amounts of Mo-99 with LEU, an approximate five-fold increase of uranium is needed to match the thermal neutron fission yields [1]. Consequently, increasing the amount of uranium by five times and changing the form of uranium in a target will have a profound impact on the current chemical processing of the irradiated targets and new chemical

treatments must be devised. The two frontend processes developed as part of this project are: (1) the dissolution of irradiated LEU foil (up to 250 grams in a single batch) and nickel fission recoil barrier in nitric acid at ambient pressure followed by separation of Mo on a titania column and (2) the electrochemical dissolution of LEU foil followed by actinide and fission product precipitation. Both these frontend processes produce an alkaline (basic) solution feed for Mo purification. The flow diagrams for these processes are shown in Figure 1.



**Figure 1: A flow-diagram of two frontend processes for LEU-foil dissolution and Mo-99 recovery.**

## 2. Nitric Acid Dissolution

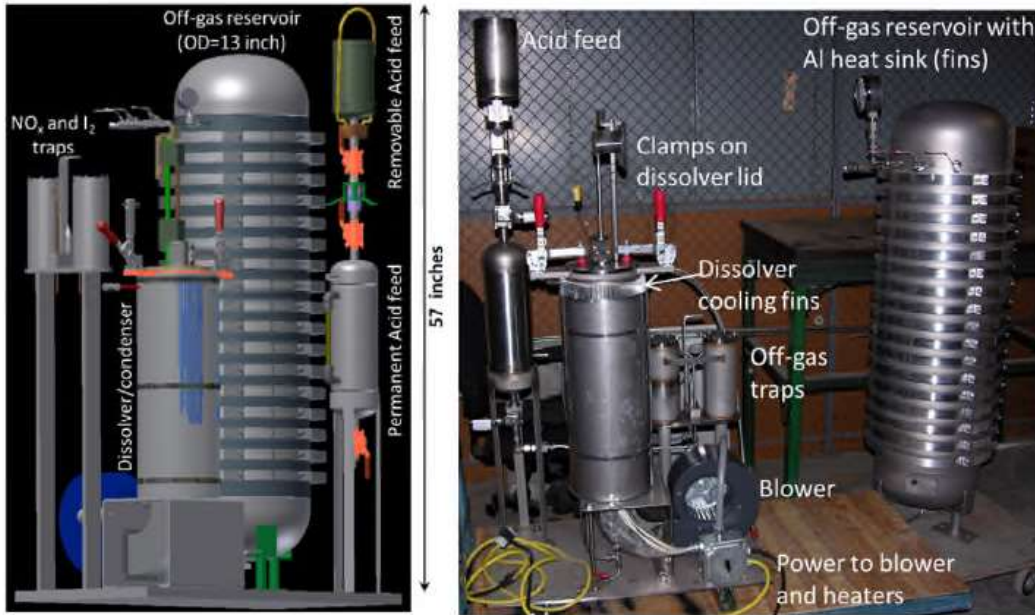
An ambient pressure, nitric-acid-dissolver system designed for the dissolution of up to 250 grams of irradiated LEU-foil and associated fission recoil barrier metal (e.g., Ni) has been tested using irradiated and non-irradiated foils (Figure 2). The design, fabrication and previous performance tests for this system are described in more detail in previous reports [2-5]. A separate report discusses in more detail the chromatographic separation of Mo from the acid solution and its recovery in alkaline solution [6].

### 2.1 Design & Testing of Foil Dissolution

The dissolver system components were tested at full-scale. The key design criteria of the acid dissolver system are summarized below:

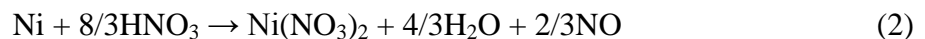
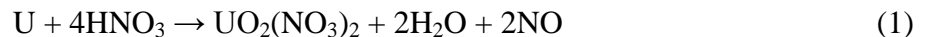
- All water vapor, reaction products, and fission gases must be contained within the dissolver system at a maximum temperature of 125°C and 2 atmospheres of pressure (absolute) under both normal and off-normal (loss of cooling during reaction) conditions.
- The acid-feed system must be designed so that the thermally hot LEU foil (hot from decayheat) can be immersed in nitric acid without losing solution due to instantaneous boiling.

- Gas-trap components must be designed to trap/neutralize all nitrogen oxide and acid gases (NO, NO<sub>2</sub>, HNO<sub>2</sub>, HNO<sub>3</sub>) as well as trap iodine gas.
- Noble fission gases are passively contained within the system.
- All dissolver system components must be designed for remote operation in a hot cell facility.



**Figure 2: Dissolver system design drawings and a photograph of the layout.**

The volume and concentration of nitric acid used for a given dissolution run depends on the mass of the metal being dissolved as well as the desired final acid concentration of the “product” solution (i.e. the solution produced by dissolution experiment). Controlling the final acid concentration is important for optimizing the Mo-99 extraction step that comes after dissolution. The volumes and concentrations of acid as well as the amount of nitrogen oxide gas (NO<sub>x</sub>: NO, NO<sub>2</sub>, N<sub>2</sub>O<sub>4</sub>) that will be produced are determined by the following general reactions:



In the presence of oxygen, the NO(g) produced in these dissolution reactions is rapidly converted to NO<sub>2</sub>(g):



When water vapor and oxygen are present, NO<sub>2</sub> is readily converted to both nitrous and nitric acid vapors [HNO<sub>2</sub>(g) and HNO<sub>3</sub>(g)], which will dissolve in condensed water and flow back down into the dissolver. One of the main purposes of the experiments discussed below are to determine how much acidity is lost from the dissolver solution as a result of the loss of NO<sub>x</sub> and acid gases from the condenser section of the dissolver.

Three experiments were performed as part of this study: (1) 197.26 grams of depleted uranium was dissolved discontinuously, that is the test was interrupted to determine the extent of dissolution after 20 minutes, 40 minutes, 80 minutes and 90 minutes. The acid dissolver solution was not changed out during the interruptions of this test. (2) 202.02 grams of depleted uranium was dissolved with minimal heat input to measure the temperature profile produced by the heat of dissolution of the foil. (3) 242.4 grams of depleted uranium foil was combined with 6.84 grams of irradiated LEU foil and dissolved with the full heat load. Nickel metal was added to each test in an amount consistent with its presence as a 7 – 10 micron thick fission recoil barrier in the actual target.

The starting acid concentrations for these experiments were 9.7 molar for the 197 and 202 gram U tests and 11.5 molar for the 250 gram U test. These concentrations are in excess of the starting concentrations required to end up with 1.0 molar nitric acid if the dissolution follows the stoichiometry described in Reactions (1) and (2) above. The higher than stoichiometric starting acid concentrations were used to counter of the loss of acid through the loss of  $\text{NO}_x$  gas during the tests. Using the potentiometric titration method of Motojima and Izawa it was found that for all three tests there was only minor (if any) decrease in acidity due to the loss of  $\text{NO}_x$  gas during the dissolution [7]. The acidities of the final solutions were as follows: 1.8 M for the 197 gram U test, 2.1 M for the 202 gram U test and 2.1 M for the 250 gram U test. These results indicate that the dissolution is close to following the stoichiometry shown in Reactions (1) and (2). The final solution from the 250 gram U test was adjusted using NaOH to 1.0 M acidity prior to transferring it for the Mo-99 extraction step.

## *2.2 Mo Column Recovery and Strip*

The dissolved irradiated target was followed by Mo recovery on a titania column. In this step ~650 mL (899 g, ~350gU/L) of irradiated target solution was loaded on a 3.5 x 5 cm column containing 63 g of S40 sorbent at 25 mL/min at 60° C. Stable Mo,  $\text{Na}_2\text{MoO}_4$ , was added to the feed to increase the concentration of Mo to ~5.4 mM. The loading was followed by ~260 mL 1 M  $\text{HNO}_3$  wash and 75 mL  $\text{H}_2\text{O}$  wash. Mo was stripped from the column with 0.1 M NaOH at 5 cm/min at 25° C. The strip was divided into 4 fractions containing 224, 292, 293 and 120 mL of solution, respectively. The Mo recovery process was designed using VERSE simulation program (Purdue University, IN) to determine intraparticle diffusivity and column process designs. Isotherm parameters, solution, sorbent and mass transfer parameters were determined for column designs using Sachtopore 40  $\mu\text{m}$  particle size sorbent, S40. Column designs were calculated for loading 500 mL LEU target solution containing ~7 mM Mo, ~450 g-U/L and the nitric-acid concentration after dissolution ~1 M. The loading time of 20 min was chosen to limit the total column process time to 1 h. Column length was limited by  $\Delta\text{P}$  constraint of 0.8 atm based on use of vacuum/gravity fluid-transfer system. Two viable column designs were determined for loading LEU target solution at 25 mL/min; 3.5 x 5 cm with superficial velocity of 2.60 cm/min and 5 x 2.5 cm with superficial velocity at 1.27 cm/min. The 3.5 x 5 cm column design was selected due to its slightly more efficient sorbent utilization and was the basis for the Mo recovery process (Table 1). The column design was tested by loading tracer Mo-99 in uranyl nitrate solution containing 7 mM Mo, 450 gU/L and 1 M  $\text{H}^+$  on the column following with recovery of Mo with 0.1 M NaOH by counter-current stripping at 3 cm/min at 25° C.  $99.4 \pm 3\%$

**Table 1: Process for recovery of Mo from LEU target solution using a 3.5 x 5 cm column and S40 sorbent.**

Step	$t_{\text{loading}}$ (min)	Volume (mL)	flow rate (mL/min)	$u_s$ (cm/min)	Solution volume (BV)
Load	20	500	25	2.6	
Wash, 1 M HNO <sub>3</sub>	8	385	48.1	5	8
Wash, H <sub>2</sub> O	3	144	48.1	5	3
Strip, 0.1 M NaOH	26.7	770	28.9	3	16

of Mo was loaded on the column and  $99.9 \pm 3\%$  was recovered in strip fractions #2 and 3 (6 BV each).

The results in Table 2 show partitioning of molybdenum, uranium, and fission products between process streams during column processing of irradiated target solution. All activities were decay corrected to the start of Mo recovery process, 5/1/2014 at 8 AM. The results show percent partitioning of irradiated solution components in process streams, normalized to 100%. The actual mass balance is also indicated in Table 2. The results of Mo partitioning indicate that 99.3% of Mo was loaded on the column. Subsequently, 28.3% of Mo was in strip fraction 2, and 70.1% of Mo was in strip fraction 3, for a total of 98.4% Mo recovery (product). Strip fraction 1 and 4 contained 1% of Mo. Mo activities in nitric-acid wash and water wash were below detection limits. Furthermore, the results in Table 2 indicate that majority of radionuclides, including uranium, iodine, alkaline (Ba, Sr), transition metals (Cd, Rh and Ru) and lanthanides (Ce, Nd) partition into the eluent and nitric acid wash streams. Only Sb (~60%) was found in the strip fraction 1. Other irradiated target components such as Ru, I, Te and Zr partitioned <1% into the Mo strip fractions 1-3. Fractions of Sb (1.6%), Te (7.1%) and Zr (1.4%) remained on the sorbent. Activities of I-131 ( $t^{1/2} = 8.02$  days) and La-140 ( $t^{1/2} = 1.68$  days), which are growing in from their respective parents Te-131m ( $t^{1/2} = 1.25$  days) and Ba-140 ( $t^{1/2} = 12.79$  days), were determined ~24 h after the column experiment. As with other irradiated solution components, I-131 and La-140 and their parent isotopes mainly partition in eluent and acid-wash process streams.

### 3. Electrochemical Dissolution

Caustic digestion of irradiated aluminum-clad HEU uranium-aluminide targets is employed for Mo-99 production [8]. However, under normal conditions, uranium metal is not digested in NaOH due to the formation of a passive uranium dioxide layer. Using alkali solutions for short-cooled irradiated target processing has several advantages over the more common acidic nitrate approach. The digestion of uranium in nitric acid produces substantial amounts of undesirable NO<sub>x</sub> gasses that demand some degree of off-gas treatment. The chemical speciation of iodine – a major fission product – exists as predominantly I<sub>2</sub> under acidic conditions [9]. In alkali solutions iodine is in the reduced anionic forms I<sup>-</sup> or I<sub>3</sub><sup>-</sup>, which are not volatile. The remainder of the fission products such as zirconium, lanthanides, barium, and cesium can be co-precipitated with uranium and calcium carbonate in basic solutions.

#### 3.1 Process and Equipment Design

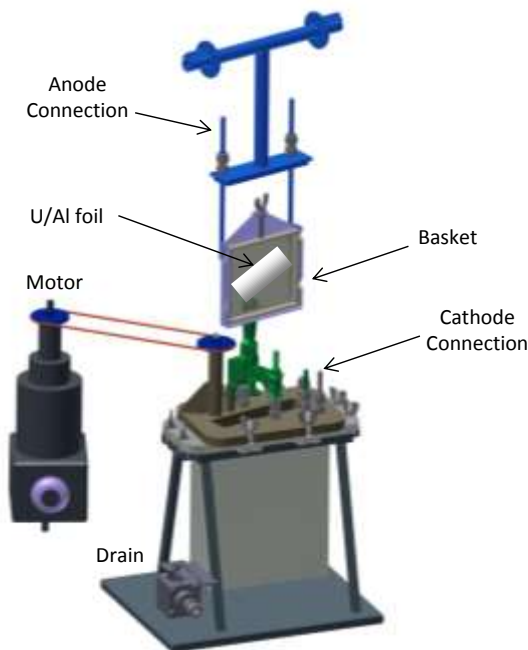
**Table 2: Percent partitioning of irradiated target solution components normalized to 100% for the nitric acid dissolution system.**

Isotope	Half-life (days)	Eluent (%)	1 M HNO <sub>3</sub> Wash (%)	H <sub>2</sub> O Wash (%)	Strip fraction 1 (%)	Strip fraction 2 (%)	Strip fraction 3 (%)	Strip fraction 4 (%)	Sorbent (%)	Mass Balance (%)
Ba-140	12.79	88.2	11.8	-	-	-	-	-	-	85.1
Ce-141	32.28	87.9	12.1	-	-	-	-	-	-	91.4
Ce-143	1.404	88.8	11.2	-	-	-	-	-	-	86.9
I-133	0.867083	89.0	10.1	0.0	0.0	0.4	0.5	-	-	87.3
I-135	0.274167	92.2	7.8	-	-	-	-	-	-	116.0
Mo-99	2.743056	0.7	-	-	0.2	28.3	70.1	0.8	-	91.3
Nd-147	11.06	82.4	17.5	-	-	0.2	-	-	-	106.3
Pm-151	1.18	88.2	11.8	-	-	-	-	-	-	91.3
Rh-105	1.473333	90.2	9.8	-	-	-	-	-	-	95.1
Ru-103	39.35	88.0	11.4	0.0	-	0.3	0.3	0.1	-	84.1
Sb-127	3.85	16.3	22.5	-	59.5	-	-	-	1.6	79.7
Sm-153	1.9285	89.2	10.7	0.0	0.0	-	-	-	-	94.4
Sr-91	0.400417	89.2	10.8	0.0	-	-	-	-	-	103.0
Te-132	3.246	79.5	11.9	0.0	0.4	0.3	0.3	0.5	7.1	12.8
U-237	6.75	86.2	13.8	-	-	-	-	-	-	69.4
Y-93	0.455417	100.0	0.0	-	-	-	-	-	-	110.0
Zr-95	64.4	98.1	-	-	0.3	0.2	0.1	-	1.4	91.8

The oxidation of uranium metal is assumed to proceed quickly through U(III), producing U(IV) oxide. Then, according to literature data, the oxidation of UO<sub>2</sub> proceeds through the formation of UO<sub>2+x</sub> species on the surface until UO<sub>3</sub>·2H<sub>2</sub>O is formed [10]. Hexavalent uranium has rather high solubility in the presence of carbonate/bicarbonate ions [11]. The dissolution occurs when these negative ions reach the positively charged surface of the uranium anode. A schematic of the basket and the dissolver is shown in Figure 3. The basket's vertical bolt pushes the outer frame with the welded screen toward the inner frame with the attached screen, thus providing the necessary contact between the anode lead and the foil. The authors have found from previous experiments that, using a basket, it takes about 2.5 h to dissolve nearly 99% of the full-size foil due to the partial reduction of the free foil surface area, caused by the mesh interference [12]. The material used for the prototype is the 304SS. Nitrogen gas was used to sweep the dissolver. Fission gases will be collected and hydrogen gas, which is generated during the electrolysis, will need to be oxidized for commercial-scale processing, but both were diluted and released in this experiment.

The first step is the dissolution of Al barrier in 1 M NaOH as NaAl(OH)<sub>4</sub>. Then the solution is drained through a ball valve on the side of the dissolver (Figure 3). After a wash step, a sodium bicarbonate solution is added in the dissolver through one of the ports on the lid. The uranium foil electrolysis is conducted at elevated temperature (85–95°C) and intense stirring. Heating is provided by an external heat gun (not shown). The approximate volume of the solution required to cover a 24 g uranium foil (10 cm × 8 cm) is about 1.2 L. However, the solution/U-metal ratio will be significantly smaller if a batch of several foils is dissolved in a vessel modified for the

full-scale process. The solubility of uranium in bicarbonate medium is 80 g-U/L (it is soluble as both the neutral and anionic complexes of uranyl carbonate) [11], thus for a 225 g batch of U foils at least 3 L of the solution is required. For the full-size dissolver, the anode basket(s) will be fabricated from nickel or nickel alloys, which are resistant to oxidation in alkaline mediums. Use of 304 SS is acceptable for several applications, so it was chosen to minimize the cost of the prototype. The cathode and the dissolver body of the production dissolver can be made from stainless steel, as there should be no significant corrosion during the processing. A 4 x 4 cm SS plate, welded to a SS rode, served as a cathode.

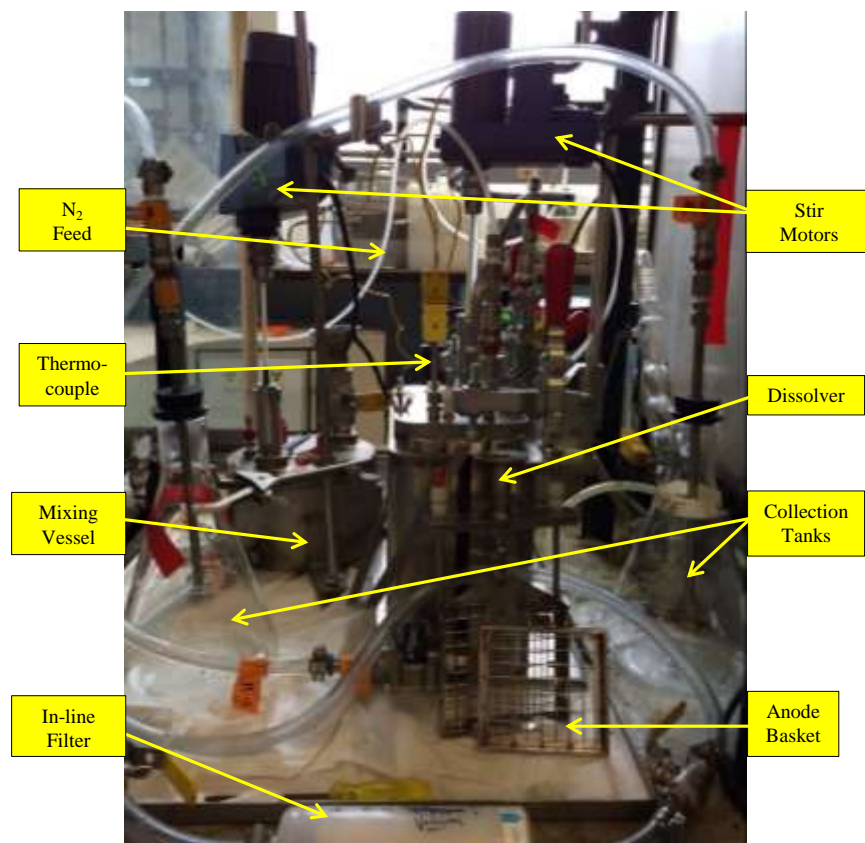


**Figure 3: An illustration of the dissolver tank and electrochemical basket.**

The *U-precipitation* step shown in Figure 1 is the critical step to remove actinides and the majority of the fission products from the product solution. Another very important role of this step is to remove the carbonate from solution as it interferes with the anion exchange step for the molybdate recovery. The mixing vessel was built with 304 SS and designed to hold 2 L of solution. The base of the vessel was built with an incline to assist the flow of the uranium-carbonate slurry and to avoid clogging. Five ports were drilled into the head of the vessel: a funnel to introduce the calcium oxide powder, a mixing port to hold the stir-shaft, and three connections for vacuum, solution entrance, and N<sub>2</sub> gas. The exit valve diameter was ½-inches to allow the slurry to move freely to the filter. An in-line 10µm Whatman filter with ½-inch barbed connections was used to capture the slurry. Figure 4 is a photograph of the entire apparatus set up in a vac-frame hood.

### 3.2 Test with Irradiated LEU-foil

A 7.6 cm × 2.5 cm LEU foil (0.32 mm thick, 15.3 g) was obtained from laboratory stocks. The foil was wrapped in aluminum and placed into an irradiation vessel that had been purged with helium. The vessel was then irradiated for ten minutes at the Argonne low energy accelerator



**Figure 4: The full process setup used for the low-burnup LEU experiment inside of a vac-frame hood.**

facility (LINAC) using a neutron converter; the thermal neutron flux was on the order of  $10^{11}$   $\text{ncm}^{-2}\text{s}^{-1}$ . The target was allowed to cool for eight hours to minimize the dose rate from short-lived fission products.

The target was removed from the irradiation vessel, secured in the dissolver basket, and placed into the dissolver. The basket was then submerged in 1.5 L of 1 M NaOH under a continuous  $\text{N}_2$  sweep with intense mixing at 70 °C. It took approximately 30 minutes to completely dissolve the aluminum and leave the uranium metal exposed and in contact with the basket. The NaOH solution was drained from the dissolver into a collection flask for analysis and the dissolver was filled with 1.2 L of 0.9M  $\text{NaHCO}_3$ . Anode and cathode leads were connected to the basket and the cathode, respectively. At a constant voltage of 9 V, the current was maintained at  $41 \pm 2$  A using a Magna Power supply. The temperature of the solution was  $92 \pm 2$  °C. Nitrogen gas was continuously flushed through the dissolver and into a condensing coil and liquid trap. After four hours, the electrolysis was stopped, and the foil was examined. Over 98% of the uranium foil was dissolved in approximately 650 mL of carbonate solution (pH 10.0). An aliquot of the solution was taken for analysis. The solution was then fed into the mixing vessel where it was contacted with 89 g of calcium oxide. The solution was mixed for approximately 15 minutes and rinsed with de-ionized water to flush precipitates from the mixing vessel. The slurry then exited the vessel through the in-line Whatman filter and weighed in a collection flask (1.16 L total, pH 13.0). The product solution was visibly clear indicating that most of the uranium had been



**Table 3: Gamma analysis results of the electrochemical dissolver process.**  
**These values have been decay-corrected to the time of irradiation.**

Isotope	$\mu\text{Ci}$			% Mo Recovered <sup>†</sup>
	Dissolver Solution*	Mo Separated Filter	Product Solution	
Mo-99	32.1	1.9	25.6	92 ± 3
U-237	16.7		0.7	
La-140	5.6		0.7	
Zr-95	1.4		0.1	
Nb-95	0.2		ND	
Nd-147	0.8		0.1	
Np-239	10.2		0.3	
Te-132	0.7		0.1	
I-133	32.3		11.1	
I-131	1.6		0.9	
Ru-103	0.3		< 0.1	
Rh-105	1.4		0.1	

\* These values may not represent the true values on account of the uncertainty in solution volume and gamma-interference.

<sup>†</sup> Calculated from the sum of the Mo Separated Filter and the Product Solution

ND = below detection limit or complications with the peak shape.

removed. An aliquot of the product solution was taken for analysis. The uranium precipitate that was captured on the filter was eluted with concentrated nitric acid. A small volume of acidic 0.5 M stable Mo-carrier was added to the eluted solution with 2% alpha-benzoin-oxime ( $\alpha\text{BO}$ ) [13]. The precipitate was filtered, dried, and then digested in sodium hydroxide and hydrogen peroxide to a final volume of 0.215 L. An aliquot of this solution (labelled *Mo Separated Filter* in Table 3) was taken for analysis.

### 3.3 Results and Discussion

The samples were gamma-counted on a high-purity germanium well-detector (calibrated with Eckert & Ziegler mixed isotope standard). The activities of each nuclide were decay-corrected to the time of irradiation. The dead time in each case was less than 5%. The Mo-99 concentrations were determined from the gamma peak at 778 keV. Table 3 lists the results of the analysis. The NaOH-aluminum solution showed only trace amounts of technetium and iodine. The activity from this solution was considered negligible in the recovery calculations. The dissolved LEU solution (listed as *Dissolver Solution* in Table 3) was heavily loaded with fission product and actinide gamma peaks thus the results may not be quantitative due to interference. There is also some uncertainty in the solution volume because of evaporation. Gamma analysis of this solution calculated approximately 32  $\mu\text{Ci}$  of Mo-99 assuming a final volume of 700 mL. The *Mo Separated Filter* contained approximately 1.9  $\mu\text{Ci}$  of Mo-99. To obtain this number, the precipitate was dissolved in acid followed by the quantitative precipitation of Mo with alpha-benzoin oxime ( $\alpha\text{BO}$ ). This loss in product can be afforded to Mo-99 being trapped within the precipitate.

Gamma analysis of the product solution showed strong signals from technetium, molybdenum,

iodine, and several xenon isotopes as a result of iodine decay. We assume the concentrations of Mo-99 in the product solution to be quantitative on account of the chemical treatment and hindered gamma-interference. The Mo-99 recovery was calculated from the sum of the Mo-99 captured on the filter (loss) and the product solution which results in over 92% recovery ( $\pm$  one-sigma notation from counting statistics). From Table 3 we can approximate that between 30-50% of the iodine was recovered based on the 133 and 131 isotopes; though it was anticipated that more I-131 should be present. The U-237 tracer (induced by photon reactions with U-238) confirms that over 95% of the uranium was removed with the precipitation step and the single in-line filter. This removal can be improved with additional filters and Mo-purification.

To demonstrate the compatibility of this technology with backend Mo purification procedures, an aliquot of the product solution was mixed with AG-MP-1(BioRad) anion exchange resin. Gamma analysis of the product solution afterwards showed that technetium, molybdenum, and iodine were strongly retained on the resin.

#### 4. Conclusion

Two front-end processes of dissolving LEU-foil for Mo-99 recovery were demonstrated with irradiated foils at Argonne. Both technologies are compatible with Mo-purification procedures. A full-scale prototype of the ambient pressure, nitric acid LEU-foil dissolver system (capable of dissolving 250 grams of irradiated LEU) was designed and tested. Tests results show that the cooling system of the dissolver (reflux condenser) is sufficient to remove all heat produced by the exothermic dissolution reaction and continuous decay heat of the irradiated foils. The uranyl-nitrate solution (895 g,  $\sim$ 350 gU/L) was loaded onto 3.5 x 5 cm S40 column at 25 mL/min at 60° C. The majority of Mo, 98.4% was recovered in strip fractions 2 and 3 (product). Partitioning of actinides, lanthanides and fission products between different process streams was determined. The second process to separate Mo-99 from irradiated uranium targets electrochemically dissolved LEU foil in an alkali carbonate media. The actinides and the majority of the fission products were precipitated with calcium oxide which left technetium, molybdenum, and iodine in solution. This technology demonstrated over 92% Mo-99 recovery with appreciable separation from actinides and fission products.

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