ANL/CSE-14/25

Recovery of Mo from Natural Mo Targets Irradiated at MURR

Chemical Sciences and Engineering Division

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Recovery of Mo from Natural Mo Targets Irradiated at MURR

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RECOVERY OF MO FROM NATURAL MO TARGETS IRRADIATED AT MURR

1 INTRODUCTION

NorthStar Medical Technologies, LLC, is pursuing two options for production of the medical isotope Mo-99. As a long-term solution, one option is accelerator production of Mo-99 using the (γ, n) reaction on enriched Mo-100 targets. At full operational capacity, NorthStar is planning to produce 50% of the Mo-99 required for U.S. market needs. Another option being considered as a short-term solution is production of ~3000 six-day curies of Mo-99 per week by using the (n, γ) reaction on natural or Mo-98 enriched targets at the Missouri University Research Reactor (MURR). After processing of irradiated Mo targets, both options produce Mo in highly alkaline solution (~5 M KOH), which is used in the RadioGenix™ generator to separate Tc-99m from Mo-99. In both cases, valuable enriched Mo-100 or Mo-98 needs to be recycled for production of new Mo targets. In this report, we present data on recovery of Mo from natural Mo targets irradiated at MURR.

2 EXPERIMENTAL

Precipitation of Mo was performed by acidifying a solution of K_2MoO_4 in 5 M KOH (~0.2 g-Mo/mL) with glacial acetic acid (AcA) [Tkac et al. 2013] according to the following reaction:

$$K_2MoO_4 + 2\ KOH + 4\ CH_3COOH \leftrightarrows MoO_3\downarrow + 4\ CH_3COOK + 3\ H_2O$$

About 1.5 L of glacial AcA was used to precipitate Mo from ~300 mL of solution containing K_2MoO_4 in 5 M KOH. The precipitation was performed in 2 L bottles. Freshly formed precipitate was mixed for 10 min at 7000 rpm (if not noted otherwise) using a Silverson L5M-A overhead mixer, and then centrifuged at ~6200 rcf (relative centrifugal force) for 10 min using a large capacity centrifuge (Sorvall RC 12BP+). The centrifuge is capable of processing six 2 L bottles. As noted above, precipitation of MoO_3 was performed by acidifying a solution of K_2MoO_4 in 5 M KOH (~0.2 g-Mo/mL) with glacial AcA. Since AcA is a weak acid (K_a=1.8 × 10^{-5}, where the pH of concentrated acid is ~1.8), it is likely that the white precipitate that forms over time after initially precipitating MoO_3 consists of various mixed K-Mo species, and that dimeric and/or polymeric Mo species ($Mo_7O_{24}^{6-}$) are predominant in the pH=4-5 solutions with high Mo concentrations [Tkac, 2008]. The yellow color of the liquid is most likely due to the presence of soluble polymeric Mo species (Figure 1).

The benefit of using AcA is that the neutralization reaction is very mild. However, after precipitation of MoO_3 with AcA followed by centrifugation, over time a fine white precipitate that contains Mo begins to form. Not collecting this secondary precipitate will result in lower yields of Mo. The Mo precipitate is then repeatedly washed with concentrated nitric acid. Similar to the case with the acetic acid precipitation, fine Mo precipitate forms in the solution over time after the first HNO_3 wash. These fine species containing Mo cannot be collected even at 6200 rcf (Figure 2). This could be due to the presence of soluble dimeric species that temporarily form through the protonation of molybdenum trioxide but then precipitate as Mo oxide:

$$MoO_3 + H^+ = HMoO_3^+$$

$$2HMoO_3^+ = H_2Mo_2O_6^{2+}$$

$$H_3Mo_2O_6^{3+} = H_2Mo_2O_6^{2+} + H^+$$

Formation of such Mo species is common mostly in strongly acidic, non-complexing media, such as perchloric acid [Esbelin et al. 2001], but may form in the presence of nitric acid as well. To eliminate Mo losses, samples after the precipitation step with acetic acid and the first HNO_3 wash were allowed to sit for at least 24 hours.

After the final nitric-acid wash, the MoO_3 precipitate is first dried in the bottle at 80°C for a few hours and then transported into a beaker and dried at 160°C to remove the remaining HNO_3 and dehydrate the MoO_3 precipitate. The dried MoO_3 was ground into powder (Figure 3).

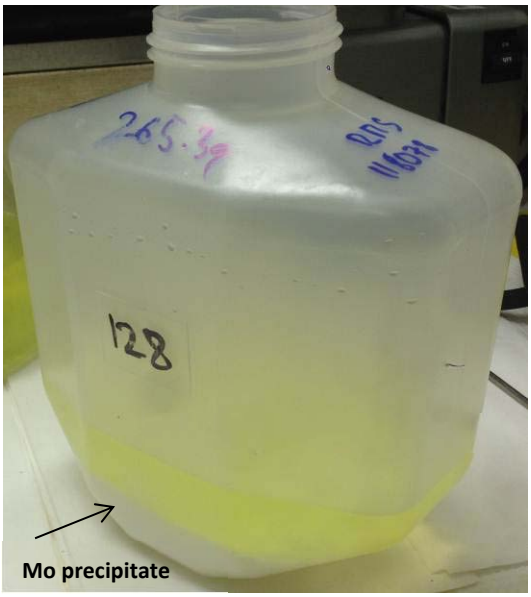

FIGURE 1 White Mo Precipitate (on the bottom of the bottle) that Forms after Adding Glacial Acetic Acid to Mo in 5 M KOH. Yellow Color of Acetic Acid/ Potassium Acetate is Most Likely Due to the Presence of Soluble Polymeric Mo Species, such as $Mo_7O_{24}^{6-}$

FIGURE 2 Formation of Fine Mo Precipitate on the Bottle Walls Next Day after Centrifugation

FIGURE 3 Recovered MoO_3 Product Obtained after Drying and Grinding

Aliquots of the MoO_3 powder were re-dissolved in NH_4OH, acidified to 5 M HCl and submitted for inductively coupled plasma/mass spectrometry (ICP-MS). Low potassium concentrations in the presence of high Mo concentrations can be determined after extracting Mo into 30% tributyl phosphate (TBP) from ~5 M HCl according to a procedure developed earlier [Tkac, 2014].

3 RESULTS AND DISCUSSION

Argonne received three bottles (Batch 1, 2, and 3) containing ~1.5 L of K_2MoO_4 in 5 M KOH from irradiated Mo targets at MURR. After dissolution of the irradiated Mo disks in hydrogen peroxide, the Mo-peroxo species were converted to K_2MoO_4 by addition of KOH. The desired composition of solution that is loaded into the RadioGenix™ generator was ~0.2 g-Mo/mL in 5 M KOH.

A small aliquot of each solution was submitted for ICP-MS analysis to determine the starting Mo and K concentrations and the concentration of other elements of interest to NorthStar. Typically, one batch containing 1.5 L of Mo solution in ~5 M KOH was divided into five 2 L bottles. One bottle would contain about 300 mL of starting Mo solution, which was precipitated with ~1.5 L of acetic acid. Due to limited hood space, two or three bottles containing ~110-160 g of Mo were processed at a time. Tables 1-3 show the concentrations of Mo and K in the dissolved irradiated Mo-targets solutions and in the recovered MoO_3 powder. Note that the concentrations of KOH and Mo vary among all three batches. Batch 3 (Table 3) has the lowest Mo concentration and highest KOH concentration. This could be very important, since the mole ratio of K/Mo changes significantly and may affect the recovery process. If a solution contains 0.2 g-Mo/mL in 5 M KOH, the K/Mo mole ratio is 4.399. All three MURR samples had higher K/Mo mole ratios: Batch 1: 4.487; Batch 2: 4.534, and Batch 3: 4.715. Tables 1-3 show that the concentration of potassium in the final MoO_3 product increased with a higher initial K/Mo mole ratio in the solution. A plot of the average potassium concentration in the final combined MoO_3 product versus the starting K/Mo mole ratio is shown in Figure 4. The data in Figure 4 indicate a correlation between K/Mo mole ratio and potassium concentration in the final MoO_3 product.

Despite slightly higher starting K/Mo ratios, the potassium concentration in the final Mo product was below 100 ppm for the first two batches. Potassium concentration in the final product of Batch 2 was 92.3 ppm, which is still below 100 ppm, but the elevated concentration of potassium in the final product, compared with that of Batch 1, is most likely due to cross-contamination of the MoO_3 powder after the drying process with a spatula that was used after the acetic acid wash when the Mo precipitate still contained 15-25% of the original potassium. Moreover, the final two washes with nitric acid for samples #135 and #136 (Batch 2) were performed after the mixer was used to wash samples #137-139 (Batch 3) with acetic acid. Although the mixer was washed with water, cross-contamination from the acetic acid wash may have occurred, and some of the potassium residue from the mixer may thus have been introduced into samples #135 and #136.

Table 3 shows that the concentration of potassium in the final MoO_3 product for the Batch 3 was above 100 ppm. This could partially be attributed to this batch having the highest K/Mo ratio, which could affect the efficiency of potassium removal (Figure 4). On the other hand, samples #140 and #141 (Batch 3) were processed with a different mixing head. Figure 5 shows the general-purpose disintegrating head (left) that was used for processing samples #127-139, along with the square-hole high-shear screen (right) that was used to process samples #140 and #141. When the high-shear screen was used for mixing, the fine precipitate that forms after the acetic acid step and first nitric acid wash did not form in the solution. Use of this screen can

TABLE 1 Mo and K Concentrations in Starting and Recovered Mo Material for Irradiated (June 3, 2013) Mo Targets from MURR, Batch 1

Batch 1 Sample #	Start Mo Solution, g	Total Start Mo, g	Total Start K, g	MoO_3 Product, g	Mo Product, g	K Product, mg	Mo Yield, %	K in Mo Product, ppm
127	516	61.0	111.5	89.8	57.0	2.84	93.4	49.7
128	518.3	61.3	112	93.2	58.8	2.67	96.0	45.3
129	420.8	49.8	90.9	74.3	47.4	1.83	95.2	38.7
130	424.2	50.2	91.7	75.3	47.9	1.57	95.4	32.9
131	417.6	49.4	90.3	75.5	49.2	1.80	99.6	36.6
Whole batch	2296.9	271.7	496.4	408.1	260.3	10.7	95.8	41.2

Note: ICP-MS data reported with 10% uncertainty. Starting K/Mo mole ratio = 4.487, c(Mo)=1.93M – 0.185g/mL, c(KOH)=4.8M. A volume of 7.6 L of acetic acid was used to precipitate Mo, and 75 L of nitric acid was used in all washing steps.

TABLE 2 Mo and K Concentrations in Starting and Recovered Mo Material for Irradiated (May 27, 2013) Mo Target from MURR, Batch 2

Batch 2 Sample #	Start Mo Solution, g	Total Start Mo, g	Total Start K, g	MoO_3 Product, g	Mo Product, g	K Product, mg	Mo Yield, %	K in Mo Product, ppm
132	465.7	55.23	102.1	81.8	51.9	3.92	93.9	75.6
133	463.3	54.94	101.6	81.6	51.9	4.06	94.5	78.2
134	464.6	55.10	101.9	82.2	51.2	3.40	92.8	66.5
135	461.8	54.76	101.2	80.3	49.3	5.73	90.0	116.1
136	464	55.03	101.7	83	52.5	6.59	95.4	125.5
Whole batch	2319.4	275.1	508.5	408.8	256.8	23.7	93.4	92.3

Note: ICP-MS data reported with 10% uncertainty. Starting K/Mo mole ratio = 4.534, c(Mo)=1.93M – 0.185g/mL, c(KOH)=4.89M. A volume of 7.6 L of acetic acid was used to precipitate Mo, and 75 L of nitric acid was used in all washing steps.

TABLE 3 Mo and K Concentrations in Starting and Recovered Mo Material for Irradiated (May 20, 2013) Mo target from MURR, Batch 3

Batch 3 Sample #	Start Mo Solution, g	Mo, M	Total Start Mo, g	Total Start K, g	MoO_3 Product, g	Mo Product, g	K Product, mg	Mo Yield, %	K in Mo Product, ppm
137	466.3	1.86	52.0	101.3	85.0	48.6	6.50	93.4%	133.9
138	466.7	KOH, M	52.1	101.4	85.3	48.6	4.69	93.4%	96.5
139	465.9	5.05	52.0	101.3	85.6	48.9	9.42	93.9%	192.9
140	466.5	Mo, g/mL	52.0	101.4	84.7	49.5	3.37	95.1%	98.2
141	464.0	0.175	51.8	100.8	85	49.0	28.16	94.6%	575.0
Whole batch	2329.4		259.8	506.3	425.6	244.4	52.1	94.1%	213.3

Note: ICP-MS data reported with 10% uncertainty. Starting K/Mo mole ratio = 4.715, c(Mo)=1.86M – 0.175g/mL, c(KOH)=5.05M.
A volume of 7.4 L of acetic acid was used to precipitate Mo, and 68 L of nitric acid was used in all washing steps.

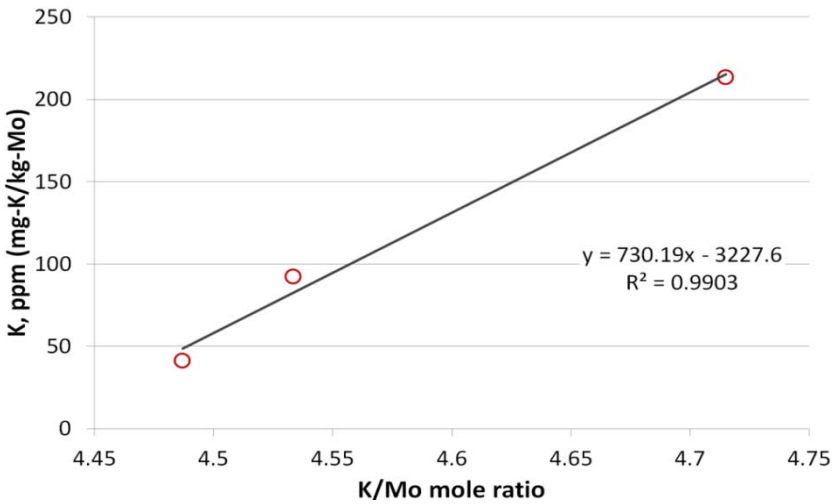

FIGURE 4 Relationship between Starting K/Mo Mole Ratios and Potassium Concentration in the Final MoO_3 Product

FIGURE 5 Photograph of General-Purpose Disintegrating Head (left) and Square-Hole High Shear Screen (right)

speed up the recovery process because there is potentially no need to wait several hours to form the fine precipitate. However, more experiments have to be performed to confirm this observation.

For the experiments when a high-shear screen was used for mixing, a splash guard that can be mounted on the bottle (Figure 6) was installed. This screen allowed mixing at a higher speed (10,000 rpm versus 7,000 rpm without it). However, mixing at 10,000 rpm with a high-shear screen produces significant heat, and the temperature of the samples increased from 21°C

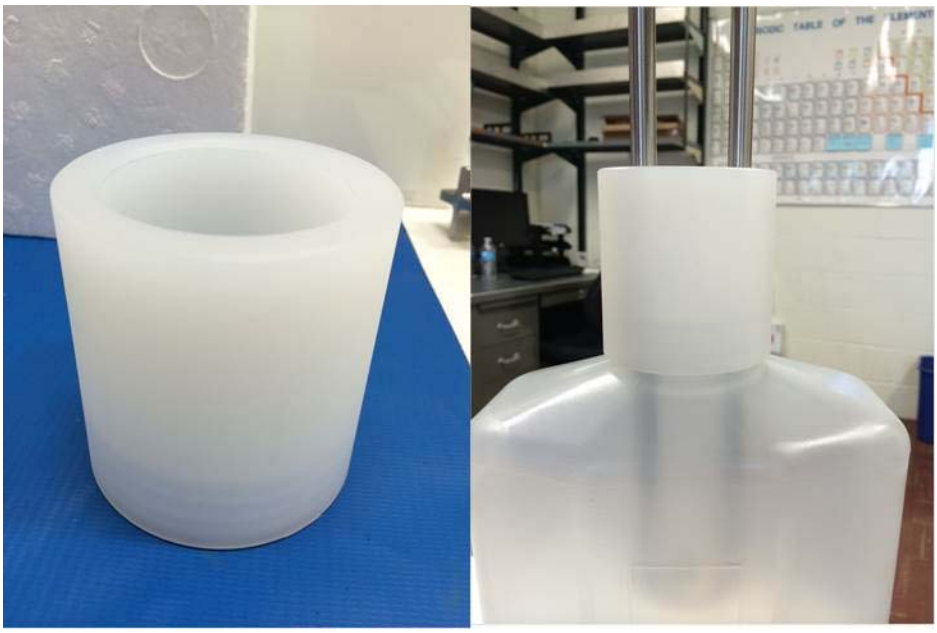

FIGURE 6 Photographs of Splash Guard that When Mounted on the Bottle Allows Mixing at Higher Speed (up to 10,000 rpm)

to 46°C after 10 minutes of mixing. This increases HNO_3 vapors and can cause corrosion problems. One of the negative effects of the high-shear screen could be lower lifting power, which makes disintegration of the centrifuged cake more difficult, and requires lifting the bottle until the rotor can mechanically disturb the Mo cake at low mixing speeds (~4,000 rpm) before mixing can continue at higher speeds (7,000-10,000 rpm). In some cases, some portion of the Mo precipitate remained stuck to the bottle even after 10 minutes of mixing, which could be one of the reasons for the elevated concentration of potassium in sample #141 (Table 3). To determine whether use of a high-shear screen is less effective in potassium removal than a general-purpose head, several runs need to be performed for the same starting solution (same K/Mo mole ratio). We plan to perform multiple large-scale runs next fiscal year.

In general, very consistent Mo recovery yields were obtained for all 15 processed samples, with average yields in the range of 93.4-95.8%. Tables 4-6 show the concentrations of various elements in the starting solutions and final purified MoO_3 product.

High concentrations of Na and Si in the starting material are most likely introduced during the processing of the irradiated targets. Typically, sodium is present in 45% KOH in the range of 0.01-0.05%, which corresponds very well with the sodium concentration found in the starting solution (~0.03% of starting potassium concentration). The presence of Si and B in the starting solution suggests that they are leached out of the borosilicate glass when KOH is added to convert the dissolved Mo into K_2MoO_4 in 5 M KOH. Most of the boron is removed during processing, but most of the sodium and silicon remain in the Mo product. Additionally, the recovery process does not remove Ti from the Mo product, and about 25 ppm of Ti remains. Some other metals, such as Fe, Co, and Ni, were also introduced into the Mo product. This can

TABLE 4 Concentrations of Various Elements in Starting Solutions and Recovered Mo Material for the Irradiated Mo target from MURR, Batch 1 (ICP-MS data reported with 10% uncertainty)

Element	Starting Solution	Conc., ppm (mg of metal per kg-Mo)				
		#127	#128	#129	#130	#131
K	1,830,000	49.7	45.3	38.7	32.9	36.6
B	NA	NA	NA	NA	NA	NA
Na	638	214	178	253	251	235
Mg	158	24.3	6.52	7.28	5.10	6.39
Al	35.6	16.8	6.68	2.86	8.69	3.69
Si	501	274	249	618	568	595
P	ND	ND	ND	ND	ND	ND
Ti	34.5	27.8	32.5	22.4	20.6	21.5
Cr	2.89	9.80	11.4	6.40	6.52	5.66
Mn	1.79	2.47	1.72	0.31	0.41	0.33
Fe	ND	73.7	49.8	ND	ND	ND
Co	ND	0.17	0.21	ND	ND	ND
Ni	3.18	7.19	6.65	0.38	0.42	0.26
Cu	ND	9.47	2.15	1.57	1.65	1.42
Zn	ND	140	8.41	3.02	3.65	9.48
Zr	NA	NA	NA	NA	NA	NA
Nb	NA	NA	NA	NA	NA	NA
Sn	2.93	1.11	1.13	0.67	0.68	0.70
Sb	1.85	1.54	1.67	0.72	0.94	1.21
Cs	ND	0.34	0.55	0.87	0.89	0.68
W	11.6	1.45	7.05	ND	ND	ND

NA – not analyzed; ND – not detected

be due to use of stainless steel (SS) mixer head and its corrosion during the washing steps with concentrated nitric acid. Another source of these metals could be due to scraping the stainless-steel spatulas that are being used during transfers of wet and dry Mo precipitate, or using a grinder with SS components. However, in general, the introduction of these metals during the recovery process is minimal, as can be seen for samples #129-141 (Table 4-6).

After ICP-MS analysis was performed on all samples, the recovered MoO_3 from all of the bottles was combined to make three separate batches (samples #127-131 from Batch 1; #132-136 from Batch 2, and #137-141 from Batch 3). It should be noted that these MoO_3 powders had different densities. Figure 7 shows the three bottles, which contain ~400-421 g of recycled MoO_3 in 2 L bottles, but the volume of MoO_3 powder differs significantly. This variation could be due to the different content of water in the samples.

TABLE 5 Concentrations of various elements in starting and recovered Mo material for the irradiated Mo target from MURR, Batch 2 (ICP-MS data reported with 10% uncertainty)

		Conc., ppm (mg of metal per kg-Mo)				
Element	Starting Solution	#132	#133	#134	#135	#136
K	1,850,000	75.6	78.2	66.5	116	126
B	80.8	3.12	3.45	2.80	ND	ND
Na	542	127	133	106	276	240
Mg	6.57	3.62	2.90	1.73	3.52	3.04
Al	31.00	5.42	8.39	1.94	4.85	2.18
Si	542	293	312	245	626	552
P	ND	ND	ND	ND	ND	ND
Ti	36.0	25.7	27.6	25.2	23.5	20.6
Cr	ND	4.55	3.15	3.13	4.88	4.21
Mn	1.45	0.47	0.34	0.36	0.40	0.33
Fe	ND	ND	ND	ND	ND	ND
Co	ND	ND	ND	ND	ND	ND
Ni	2.37	1.42	0.45	0.79	0.78	0.30
Cu	3.30	0.54	0.42	0.46	1.35	ND
Zn	ND	3.26	3.00	1.71	3.41	ND
Zr	ND	ND	ND	ND	ND	ND
Nb	3.07	6.36	5.84	5.41	6.21	5.69
Sn	ND	0.85	0.90	0.69	0.76	0.70
Sb	1.84	1.41	1.05	0.90	1.56	1.32
Cs	0.49	0.52	0.56	0.52	0.85	0.95
W	ND	ND	ND	ND	ND	ND

ND – not detected

The three bottles containing recovered MoO_3 powder were gamma counted to determine remaining activities of Sb-124 ($T_{1/2}$= 60.2 days) and Cs-134 ($T_{1/2}$= 2.062 years). Activities of Sb-124 and Cs-134 are listed in Table 7. The Sb-124 activity was determined with a HPGe (high purity germanium) detector using peaks at 722.79 keV, 645.85 keV, and 1691.04 keV. The main peak at 602.71 keV was not used because it interferes with the Cs-134 peak at 604.66 keV. The Cs-134 activity was determined using peaks at 795.86 keV and 569.29 keV. Although gamma counting results show that Sb and Cs are present in the product, their chemical concentration is very low, as it is apparent from the ICP-MS data (Tables 4-6) that the concentrations of both elements are below 2 mg/kg-Mo.

TABLE 6 Concentrations of Various Elements in Starting and Recovered Mo Material for the Irradiated Mo Target from MURR, Batch 3 (ICP-MS data reported with 10% uncertainty)

		Conc., ppm (mg of metal per kg-Mo)				
Element	Starting Solution	#137	#138	#139	#140	#141
K	1,950,000	134	96.5	193	98.2	575
B	94.5	ND	ND	ND	ND	ND
Na	627	281	285	350	291	298
Mg	26.0	3.16	5.51	ND	3.11	4.54
Al	38.3	4.28	4.95	5.13	5.26	5.36
Si	653	681	697	683	709	721
P	159	ND	ND	ND	ND	ND
Ti	40.6	30.2	29.1	28.1	27.0	28.0
Cr	2.38	9.04	5.71	6.21	5.63	5.50
Mn	2.70	0.92	0.87	0.66	0.66	0.73
Fe	80.9	18.7	11.2	10.3	13.5	10.6
Co	0.18	ND	ND	ND	ND	ND
Ni	0.69	3.14	0.45	0.99	0.70	0.38
Cu	2.74	0.91	0.79	0.58	0.42	ND
Zn	11. 7	6.29	2.73	ND	2.08	ND
Zr	0.37	0.21	0.15	0.15	ND	ND
Nb	8.97	6.61	6.34	5.98	5.99	5.61
Sn	3.36	0.83	0.84	0.73	0.63	0.58
Sb	2.39	1.97	1.90	1.82	1.57	1.58
Cs	0.34	0.85	0.94	0.96	0.96	0.98
W	36.0	13.5	11.8	10.1	8.74	8.18

ND – not detected

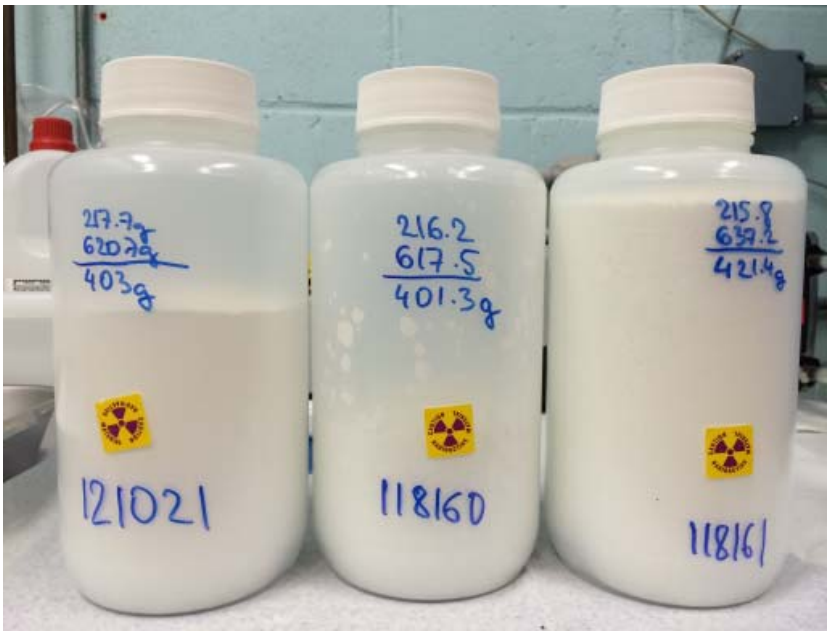

FIGURE 7 Photograph of Combined Recovered MoO_3 Powders in 2 L Bottles

TABLE 7 Activities of Sb-124 and Cs-134 in Final Recovered MoO_3 Product

#	MoO_3, g	Notes	Sb-124	Cs-134
Batch 1,		MURR Target Irrad. Date	5/20/13	5/20/13
Samples #127-131	403.0	Activity on 09-01-14	6.2 µCi	0.145 µCi
		Specific Activity	0.015385, µCi/g	0.00036, µCi/g
Batch 2,		MURR Target Irrad. Date	5/27/13	5/27/13
Samples #132-136	401.3	Activity on 09-01-14	8.644, µCi	0.261, µCi
		Specific Activity	0.02154, µCi/g	0.00065, µCi/g
Batch 3,		MURR Target Irrad. Date	6/03/13	6/03/13
Samples #137-141	421.4	Activity on 09-01-14	12.74, µCi	0.416, µCi
		Specific Activity	0.031747, µCi/g	0.001037, µCi/g

4 SUMMARY

Three batches of irradiated Mo samples from MURR, each containing ~1.5 L of K_2MoO_4 solution in ~5 M KOH, were processed to remove potassium and recover Mo as MoO_3 powder. The concentration of K in the three batches was in the range of 41-213 ppm (mg-K/kg-Mo). The reason for Batch 3 having the highest potassium concentration could be due to the starting K/Mo mole fraction being the highest, but other reasons such as using a different mixing head or lower volumes of HNO_3 could also contribute. This needs to be investigated further. Yields of recovered Mo were in the range of 93.4-95.8%.

Concentrations of other elements were also determined in the starting solutions and final products. From the elements analyzed (B, Na, Mg, Al, Si, P, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, Nb, Sn, Sb, Cs, and W), only Ti, Na, and Si had consistently elevated concentrations (>10 ppm) in the final product. The source of sodium is most likely the potassium hydroxide, as common levels of Na present in 45% KOH solution are in the range of 0.01-0.05%. The presence of both Si and B in the starting solution suggests that they are leached from the borosilicate glass when KOH is added to convert dissolved Mo into K_2MoO_4 in 5 M KOH. While boron is almost fully removed, most of the sodium and silicon remain in the Mo product.

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