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2 of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ medical isotope without use of enriched

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11 Recycle of enriched Mo targets for economic

12 production of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ medical isotope

13 without use of enriched uranium

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17 Abstract

18 A new recycle process for recovery of enriched ^{98}Mo or ^{100}Mo used for production of
19 $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ medical isotope was developed. In this process, Mo is precipitated from spent
20 NorthStar Mo/Tc generator solution containing $\sim 200\text{g/L}$ Mo as K_2MoO_4 in 5M KOH
21 using acetic acid and then washed with nitric acid. High purification factors from
22 potassium were achieved, and typical Mo recovery yields were $\sim 95\%$. The recycle
23 process was performed with up to 260g of Mo per batch and can be easily implemented
24 for processing of up to 400g of Mo.

25 **Keywords**

26 ⁹⁹Mo, enriched molybdenum, accelerator, production, purification, recycle

27 Introduction

28 Lately, a lot of attention has been paid to a potential shortage of the world's supply of
 29 $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ [1-7]. Currently, most of the ^{99}Mo is produced by the fissioning of ^{235}U in
 30 high-enriched uranium targets (HEU, generally 93% ^{235}U) [1] using aging reactors. A

31 major supplier of ^{99}Mo is Nordion, Inc., (Ottawa, Canada) providing ~40 percent of the
32 world's supply. However, the Chalk River Laboratories' NRU reactor, which produces
33 ^{99}Mo for Nordion, plans to cease production in 2016. [2, 8-9]. The majority of the US
34 supply of ^{99}Mo is provided by Nordion and Mallinckrodt [1]. The US currently
35 consumes about 50% of the world's production of ^{99}Mo . The National Nuclear Security
36 Administration's Material Management and Minimization office works with U.S.
37 commercial entities and the national laboratories to accelerate the establishment of a
38 reliable supply of the ^{99}Mo by supporting a diverse set of technologies to produce ^{99}Mo
39 without the use of highly enriched uranium. Recently, several possible technologies were
40 proposed for the production of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ or directly $^{99\text{m}}\text{Tc}$ without use of uranium
41 targets [10-11]: i) by linear accelerators using fast neutrons $^{100}\text{Mo}(\text{n},2\text{n})^{99}\text{Mo}$ reaction
42 [12], or photonuclear reaction with photon source from bremsstrahlung $^{100}\text{Mo}(\gamma,\text{n})^{99}\text{Mo}$
43 [13-19], ii) direct production of $^{99\text{m}}\text{Tc}$ in cyclotrons using $^{100}\text{Mo}(\text{p}, 2\text{n})^{99\text{m}}\text{Tc}$ [8, 20-23],
44 or iii) neutron capture on ^{98}Mo [24-27]. These technologies offer a lower-cost alternative
45 to reactor production using fissioning of ^{235}U , but generally with a lower yield of ^{99}Mo or
46 $^{99\text{m}}\text{Tc}$. To produce significant activities of ^{99}Mo (several TBq of ^{99}Mo , 1000Ci=37TBq),
47 enriched ^{100}Mo or ^{98}Mo needs to be used for all these non-uranium alternatives (enriched
48 ^{100}Mo could be available for ~ \$1000/g for kg quantities). Use of Mo targets requires a
49 major change in the generator technology to accommodate for the high concentration of
50 Mo. The main advantage of all these proposed technologies is very limited waste stream,
51 because only limited or no further purification/separation is needed.

52 Argonne National Laboratory, in cooperation with Los Alamos and Oak Ridge National
53 Laboratories and NorthStar Medical Technologies, LLC, is developing technology for the
54 production of ^{99}Mo . As a short-term solution, NorthStar is planning to produce ^{99}Mo via
55 $^{98}\text{Mo}(\text{n}, \gamma)^{99}\text{Mo}$ reaction at the University of Missouri Research Reactor, and as a long-
56 term solution using photonuclear reaction $^{100}\text{Mo}(\gamma, \text{n})^{99}\text{Mo}$. For economic production of
57 ^{99}Mo , both technologies require enriched Mo material (^{98}Mo or ^{100}Mo). To accommodate
58 for low specific activity of Mo-99, separation of $^{99\text{m}}\text{Tc}$ from the ^{99}Mo (and the rest of Mo
59 present in the target) is performed using the RadioGenix[®] generator from NorthStar. The
60 RadioGenix[®] uses ABEC resin, consisting of monomethylated polyethylene glycol

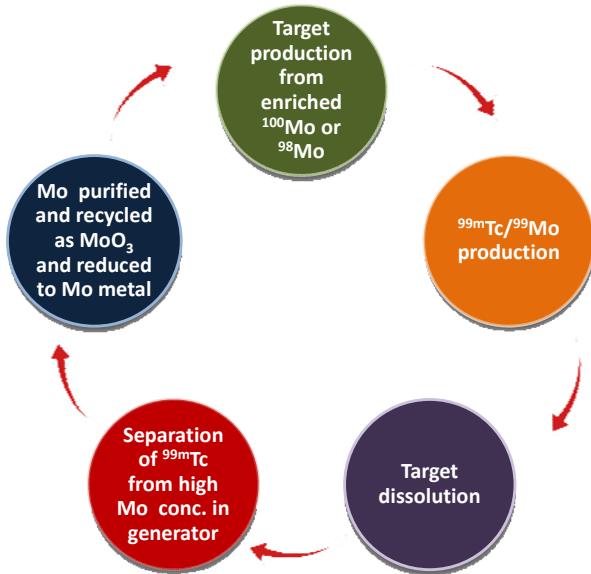
61 (PEG)-5000 covalently bound to a polystyrene support [28]. The ABEC resin is highly
62 selective for Tc from highly alkaline solutions while Mo passes through the column and
63 is recovered for another milking. This allows efficient separation of Tc from highly
64 concentrated Mo solution. Irradiated Mo-metal targets are dissolved in H₂O₂, and Mo is
65 converted to K₂MoO₄ by addition of KOH. The advantage of using KOH over NaOH is
66 higher Mo solubility in KOH [17]. Used generator solution needs to be treated to recover
67 valuable enriched Mo (¹⁰⁰Mo or ⁹⁸Mo) for future production of Mo targets. One of the
68 possible ways to recycle Mo is the conversion of K₂MoO₄ in 5 M KOH solution into
69 MoO₃ powder that can be further reduced to Mo metal [29]. Recovery of Mo during the
70 reduction of Mo from MoO₃ to MoO₂ and Mo metal is nearly quantitative [29].

71 The most challenging step is purification of Mo from potassium. The starting ¹⁰⁰Mo
72 enriched material usually contains \leq 100 mg-K/kg-Mo. However, the spent generator
73 solution contains about 1.8 kg-K/kg-Mo. One requirement is that the impurities in the
74 recycled material need to be at the same or below the concentration present in the starting
75 material to facilitate acceptance for use of recycled ¹⁰⁰Mo by the FDA (U.S. Food and
76 Drug Administration). Therefore, the amount of potassium in purified MoO₃ powder
77 should be below 100 mg-K/kg-Mo. Recently, Bénard et al [8] reported about 85%
78 recovery yield for recycled enriched ¹⁰⁰Mo material, although no process was discussed
79 on how the recycle was performed. Gagnon et al. [30] reported similar recovery yields
80 ~87% from metal to metal for ammonium molybdate system. Authors discuss that
81 ammonium molybdate is better starting material for Mo reduction than MoO₃, because it
82 shows increased density for sintered Mo. It should be noted that increased density is not
83 the only parameter that drives the target production. Target density under certain
84 sintering condition can severely affect the dissolution kinetics of the target in hydrogen
85 peroxide [31-32].

86 Here we report a simple and very effective method for recycle of enriched Mo targets that
87 achieves very high purification factors from potassium and provide high Mo yields
88 (~95%). Cyclotron production of ^{99m}Tc, and accelerator production of ⁹⁹Mo using ¹⁰⁰Mo
89 enriched targets with a recycle process that can handle several hundred grams of Mo per
90 batch can be a very attractive and cost-effective alternative to fission made molybdenum.

91 The general scheme for the molybdenum cycle from the target production through the
92 recycle of the spent Mo solution is illustrated in Figure 1.

93



94

95 **Figure 1** Diagram of molybdenum cycle from the disk production through the recycle
96 process

97 Experimental

98 ^{99}Mo was obtained from Lantheus as a 1Ci $^{99\text{m}}\text{Tc}$ generator (TechneLite). A starting
99 solution of K_2MoO_4 (0.2 g-Mo/mL) in 5 M KOH solution was prepared by dissolving
100 MoO_3 in KOH. Concentrated nitric acid (70% vol HNO_3), glacial acetic acid and
101 ammonium hydroxide were trace-metal grade-purity. HNO_3 and HCl used for ICP-MS
102 analysis were Optima grade. All other chemical reagents used in this work were of
103 analytical-reagent-grade purity and were used without further purification. All aqueous
104 solutions were prepared with deionized water with a resistivity $\geq 18 \text{ M}\Omega\text{-cm}$.

105 *Small-scale experiments*

106 For small-scale experiments, precipitation of Mo was performed by adding acid: acetic
107 acid (AcOH), nitric acid (HNO₃) and sulfuric acid (H₂SO₄) or ethanol (EtOH) to a
108 solution of K₂MoO₄ in 5 M KOH (0.2 g-Mo/mL). The Mo precipitate was then
109 repeatedly washed with concentrated AcOH and/or HNO₃ acid using a vortex mixer. The
110 wash solution was separated from the Mo precipitate using a Beckman Allegra X-30
111 centrifuge at 3400 rcf (relative centrifugal force) for 5-10 minutes. After a final wash, the
112 precipitate was re-dissolved in NH₄OH, acidified by HCl, and analyzed for K content
113 using inductively coupled plasma mass spectrometry (ICP-MS), PerkinElmer SCIEX
114 ELAN DRC II. Recovery of Mo for experiments with ⁹⁹Mo was determined
115 radiometrically using a high purity germanium detector (Ortec) using a peak at 739.5 keV
116 and using ICP-MS for experiments performed without ⁹⁹Mo tracer.

117 *Large-scale processing*

118 Large-scale experiments were performed in 2-L bottles using ~300 mL of K₂MoO₄
119 solution (~0.2g-Mo/mL) in 5 M KOH. About 1.5 L of glacial AcOH was used to
120 precipitate Mo from a 300 mL of solution. Freshly formed precipitate was mixed for 10
121 min at 7000-10000 rpm using a Silverson L5M-A overhead mixer, and then centrifuged
122 at ~6200 rcf for 10 min using a large capacity centrifuge (Sorvall RC 12BP+). The
123 centrifuge is capable of processing six 2 L bottles. After the final nitric-acid wash, the
124 MoO₃ precipitate was first dried in the bottle at 80°C for a few hours and then transferred
125 into a beaker and dried at 160°C to remove the remaining HNO₃ and dehydrate the MoO₃
126 precipitate. The dried MoO₃ was ground into powder. Aliquots of the MoO₃ powder were
127 re-dissolved in NH₄OH and acidified by HCl, and the concentration of the metals of
128 interest was determined using ICP-MS.

129 **Results**

130 *Small-scale experiment*

131 *Precipitation of Mo*

132 Several reagents were tested to precipitate Mo from highly alkaline solution. Table 1
133 shows the effectiveness of potassium removal by different reagents as well as Mo losses
134 in precipitation step. For these experiments, 1 mL of K_2MoO_4 in 5 M KOH (0.2 g-
135 Mo/mL) was combined with 5 mL of each reagent listed in Table 1.

136 **Table 1.** Mo Losses and Removal of Potassium for Different Reagents Used to Precipitate Mo.

Reagent Used to Precipitate Mo ^a	Mo Lost	K Removed
Glacial AcOH	0.2–2%	75–85%
EtOH	~0–0.2%	30–45%
4:1 v/v EtOH:AcOH	0–0.4%	30–40%
15.9 M HNO_3	5–20%	80–90%

^a AcOH = acetic acid; EtOH = ethanol

137 Besides reagents listed in Table 1, we also investigated H_2SO_4 . HNO_3 and H_2SO_4 are
138 strong acids; therefore, adding these acids to a highly alkaline solution causes a highly
139 exothermic neutralization reaction. If H_2SO_4 is carefully added to K_2MoO_4 in 5 M KOH
140 in 1:1 v/v ratio, a white precipitate containing Mo forms. However, when more H_2SO_4 is
141 added, both phases collapse and form a gelatinous suspension, which prevents
142 purification of Mo from potassium. A combination of sulfuric and acetic acid was also
143 tested, but a significant portion of Mo was found to be soluble in the mixtures. Careful
144 addition of HNO_3 forms a yellow precipitate containing Mo and a clear liquid phase.
145 When ethanol is added to a highly alkaline Mo solution, a white precipitate containing
146 K_2MoO_4 is formed. Since AcOH is a weak acid ($K_a=1.8\times 10^{-5}$) it is likely that the white
147 precipitate that forms after precipitating Mo consists of various mixed K-Mo species, and
148 possibly dimeric or polymeric Mo species that are predominant in the pH=2–4 region and
149 high Mo concentrations [33]. The benefit of using AcOH is that the neutralization
150 reaction is mild. After precipitation of molybdenum with AcOH followed by
151 centrifugation (6200 rcf), over time a fine white precipitate that contains Mo can form.
152 This is most likely due to the formation of soluble polymeric Mo species that
153 predominate at pH~4 [34]. These species probably decompose over time to the hydrated
154 MoO_3 and form a fine precipitate. Therefore, it is important to allow sufficient time for
155 this conversion and collect the secondary precipitate in order to keep Mo recovery as high
156 as possible. It should be noted that in order to achieve ~100 mg-K/kg-Mo in the final Mo

157 product, 99.995% of starting potassium has to be removed during Mo recovery process.
158 To achieve this purity level, multiple washes of the Mo precipitate are needed.

159 *Washing step*

160 Based on the data presented in Table 1, nitric and acetic acid were studied in more detail.
161 Experimental data on the effectiveness of AcOH and HNO₃ as washing agents are
162 presented in Table 2.

163 **Table 2.** Distribution of K and Mo in AcOH and HNO₃ fractions. Balance of Mo and K is affected by
164 some carryover of washing solution into Mo precipitate. Starting solution: 5 mL of K₂MoO₄ in 5 M KOH
165 (0.2 g-Mo/mL)

step	volume, mL	reagent	K, %	Mo, %	mg of K/kg of Mo
precipitation	25	17.4 M AcOH	80.0%	0.5%	
wash #1	25	17.4 M AcOH	8.8%	0.1%	
wash #2	25	17.4 M AcOH	2.3%	0.2%	
wash #3	25	15.9 M HNO ₃	13.7%	0.8%	
wash #4	25	15.9 M HNO ₃	3.3%	0.4%	
		product	1.05%	95.15%	20219
precipitation	25	17.4 M AcOH	78.1%	0.5%	
wash #1	25	17.4 M AcOH	7.4%	0.1%	
wash #2	25	15.9 M HNO ₃	15.6%	1.2%	
wash #3	25	15.9 M HNO ₃	3.9%	0.6%	
wash #4	25	15.9 M HNO ₃	0.9%	0.3%	
		product	0.50%	96.86%	9403
precipitation	25	17.4 M AcOH	81.5%	0.6%	
wash #1	25	15.9 M HNO ₃	23.4%	1.3%	
wash #2	25	15.9 M HNO ₃	3.0%	0.3%	
wash #3	25	15.9 M HNO ₃	0.6%	0.2%	
wash #4	25	15.9 M HNO ₃	0.2%	0.2%	
		product	0.14%	96.48%	2703.1
precipitation	25	15.9 M HNO ₃	84.9%	5.4%	
wash #1	25	15.9 M HNO ₃	11.2%	0.5%	
wash #2	25	15.9 M HNO ₃	1.4%	0.3%	
wash #3	25	15.9 M HNO ₃	0.2%	0.3%	
wash #4	25	15.9 M HNO ₃	0.04%	0.4%	
		product	0.017%	91.96%	347.3

166

167 If acetic acid is used for precipitation, about 80% of the potassium is removed with <1%
168 Mo losses. However, when HNO₃ is used to precipitate Mo, the Mo losses in first

169 precipitation step are significant (Tables 1-2). Results clearly show that HNO_3 is more
170 effective in removing potassium during the wash step and the sooner the AcOH is
171 replaced by HNO_3 to wash the Mo precipitate, the more effective the removal of
172 potassium is.

173 Similar to the case with the AcOH precipitation, a fine Mo precipitate forms in the
174 solution over time (after several hours) after the first HNO_3 wash. This could be due to
175 the presence of soluble dimeric species that temporarily form through the protonation of
176 molybdenum trioxide [35]. Formation of such Mo species is common mostly in strongly
177 acidic, non-complexing media, such as perchloric acid [35], but may form in the presence
178 of HNO_3 as well. To eliminate Mo losses, samples after the precipitation step with acetic
179 acid and the first HNO_3 wash were allowed to sit for at least 24 hours. This significantly
180 improved the recovery yields of Mo.

181 Data in Table 3 show the effectiveness of potassium removal when HNO_3 is used after
182 precipitation of Mo using acetic acid. Small-scale experiments showed that the best
183 potassium removal and Mo recoveries are obtained when acetic acid is used to precipitate
184 Mo followed by washing the Mo precipitate with concentrated HNO_3 .

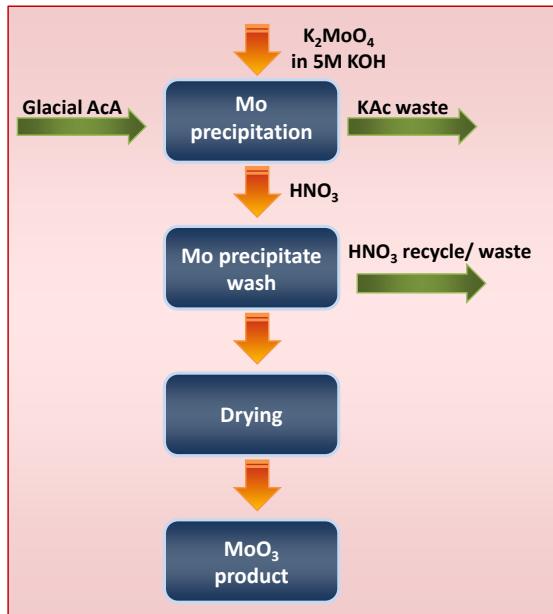
185 **Table 3.** Effect of number of HNO_3 washes and mixing time on removal of potassium after
186 precipitation of Mo using glacial acetic acid. Minimum of 24 hours were allowed after precipitation step
187 and first HNO_3 washing step to recover Mo from the fine precipitate that forms after these steps.

precipitation	Mixing time, min	# HNO_3 washes	Product content		
			K removed, %	Mo, %	mg of K/kg of Mo
AcOH	4	6	99.956%	98.11%	777.3
AcOH	4	8	99.985%	99.51%	261.2
AcOH	4	10	99.992%	99.37%	142.7
AcOH	4	12	99.996%	96.74%	71.6
AcOH	10	12	99.999%	~100%	14.7
AcOH	10	14	99.999%	~100%	10.8

188
189

190 *Large-scale processing*

191 Based on the results from small-scale experiments, large-scale experiments were
 192 performed by precipitating Mo with glacial acetic acid followed by washing with 70%
 193 HNO_3 . The major steps of the process are outlined in Figure 2.



194

195 **Figure 2.** Flow diagram of Mo recovery process using acetic and HNO_3

196 **Table 4.** Mo and K Concentrations in the Starting Solution and Recovered Mo Material for Large-Scale
 197 Experiment^a. Mo from ~300 mL of K_2MoO_4 solution was precipitated using ~1.5 L of AcOH.

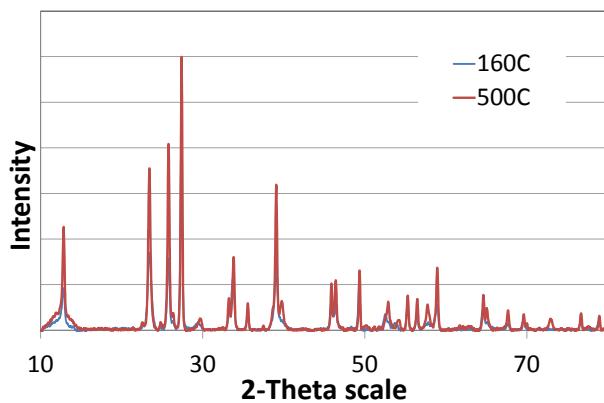
Sample	Start Mo Solution, g	Total Start Mo, g	Total Start K, g	MoO ₃ Product, g	Mo Product, g	K Product, mg	Mo Yield, %	mg of K/kg of Mo
Batch 1A	469.2	60.06	108.9	86.6	59.45	12.2	99.0	205
Batch 1B	468.3	59.94	108.7	85.4	56.51	5.7	94.3	102
Whole batch 1	937.5	120	217.6	172	115.95	17.9	96.6	154.4

198 ^a ICP-MS data reported with 10% uncertainty. K/Mo mole ratio: 4.44. c(Mo)=2.08M – 0.20g/mL,
 199 c(KOH)=5.07M. A volume of 3 L of acetic acid was used to precipitate Mo, and 24 L of HNO_3 was used in
 200 all washing steps per whole batch.

201

202 Table 4 shows the concentrations of K and Mo in the starting solution and in the
 203 recovered Mo material after the precipitation step with acetic acid and 8 washes with
 204 HNO_3 . The large-scale experiment showed some promising results with a final potassium
 205 concentration of ~154 mg/kg-Mo after only 8 washes with HNO_3 . In small-scale

206 experiments, more than 10 washes were required to achieve this level of purity. This
207 could be due to better mixing with an overhead mixer compared to vortex mixer.
208 Moreover, very good Mo recovery was observed, with total yield of 96.6%. This could be
209 attributed to the fact that precipitation step was performed by adding K_2MoO_4 solution
210 into the acetic acid while mixing, which prevented the formation of the fine secondary
211 precipitate that usually forms over time after centrifugation and positively affected the
212 Mo recovery. Recovered MoO_3 material after drying at 160°C was analyzed using X-ray
213 diffraction (XRD) and peaks were characteristic for MoO_3 . MoO_3 was also dried at
214 500°C with no significant changes in XRD spectrum (Figure 3).



215

216 **Figure 3.** XRD spectra of final MoO_3 product heated at 160°C and 500°C.

217 *Recycle of HNO_3*

218 Since significant quantities of HNO_3 are required to wash the MoO_3 precipitate to
219 achieve low K levels in the final Mo product, we also investigated an option to recycle
220 the HNO_3 used in the washing steps to remove potassium from the Mo precipitate. In this
221 process, ~80% of K is present in the acetic acid fraction that is used to precipitate Mo.
222 The first HNO_3 wash usually contains 15-20% of the starting K concentration.
223 Concentration of potassium in later washes decreases rapidly (Table 5). HNO_3 from the
224 2nd wash can be re-used for the next batch in the 1st wash and so on. Also, if little to no K
225 is present, e.g. if the first two HNO_3 washes are disposed, the rest of the HNO_3 fractions
226 combined usually contain 100-200 mg-K/L and can be recycled using rotary evaporation.

227 **Table 5.** Concentration of potassium in HNO_3 washes determined using ICP-MS. ICP-MS data reported
 228 with 10% uncertainty
 229

HNO_3 wash	K, ppm
1	11100
2	1960
3	500
4	220
5	15
6	4.7
7	0.7
8	0.3

230 Combined HNO_3 washes containing \sim 165 ppm of potassium were distilled by rotary
 231 evaporation. The potassium concentration in the recycled HNO_3 was very low (\sim 0.1
 232 ppm).

233 *Processing of irradiated samples*

234 Three \sim 1.5L batches of K_2MoO_4 solution in \sim 5 M KOH obtained after the dissolution of
 235 natural Mo targets irradiated at University of Missouri Research Reactor (MURR) were
 236 also processed. Main side reaction products Zr and Nb that are formed during irradiation
 237 were removed from the dissolved irradiated target using ferric co-precipitation. Solutions
 238 were allowed to decay and then shipped to Argonne. As received, the composition of the
 239 samples was determined using ICP-MS. Table 6 show the concentrations of Mo and K in
 240 the dissolved irradiated Mo-targets solutions and in the recovered MoO_3 powder.

241 **Table 6.** Mo and K Concentrations in Starting and Recovered Mo Material for Irradiated
 242 Mo target from MURR, total volume of Mo in \sim 5M KOH processed \sim 4.5L

batch #	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
1	2329.4	259.8	506.3	425.6	244.4	52.1	94.1%	213.3
2	2319.4	275.1	508.5	408.8	256.8	23.7	93.4	92.3
3	2296.9	271.7	496.4	408.1	260.3	10.7	95.8	41.2
						Average	94.4	115.6

243 Note: ICP-MS data reported with 10% uncertainty. Starting composition for batches 1-3: K/Mo mole ratio:
 244 4.7; 4.53; 4.49, $c(\text{Mo})=1.86\text{M}$; 1.93M ; 1.93M , $c(\text{KOH})=5.05\text{M}$; 4.89M ; 4.8M . A volume of 7.4 -7.6L of
 245 acetic acid per batch was used to precipitate Mo, and 68-75 L of HNO_3 was used in all washing steps per
 246 batch.
 247

248 In general, very consistent Mo recovery yields were obtained for all three MURR batches
249 (5 samples were processed per batch) with an average Mo yield in the range of 94.4%.

250 Table 7 shows the concentrations of various elements in the starting solutions, and final
251 purified MoO₃ product.

252 **Table 7.** Concentrations of Various Elements in Starting and Recovered Mo Material for the Irradiated Mo
253 Target from MURR, batch 1-3 (ICP-MS data reported with 10% uncertainty)

	starting concentration, ppm			final product, ppm		
	batch 1	batch 2	batch 3	batch 1	batch 2	batch 3
K	1,950,000	1,850,000	1,830,000	219	92.5	40.6
B	94.5	80.8	NA	ND	1.87	ND
Na	627	542	638	301	176	226
Mg	26	6.57	158	3.26	2.96	9.92
Al	38.3	31	35.6	5.00	4.56	7.74
Si	653	542	501	698	406	461
P	159	ND	ND	ND	ND	ND
Ti	40.6	36	34.5	28.5	24.5	25.0
Cr	2.38	ND	2.89	6.42	3.98	7.96
Mn	2.7	1.45	1.79	0.768	0.380	1.05
Fe	80.9	ND	ND	12.9	ND	24.7
Co	0.18	ND	ND	ND	ND	ND
Ni	0.69	2.37	3.18	1.13	0.748	2.98
Cu	2.74	3.30	ND	0.540	0.554	3.25
Zn	11.7	ND	ND	2.22	2.28	6.14
Zr	0.37	ND	NA	0.102	ND	ND
Nb	8.97	3.07	NA	6.11	5.9	ND
Sn	3.36	ND	2.93	0.722	0.780	0.858
Sb	2.39	1.84	1.85	1.77	1.25	1.22
Cs	0.34	0.49	ND	0.938	0.680	0.666
W	36	ND	11.6	10.5	ND	1.70

254

255 Discussion

256 Although low Mo losses were observed when ethanol or its mixture with AcOH was used
257 to precipitate Mo, multiple washes are needed to completely remove KOH, and in order
258 to purify molybdenum from K, acid has to be used to precipitate Mo as an oxide. Due to

259 the oxidation of ethanol by HNO_3 , and possible formation of explosive ethyl nitrate,
260 washing with AcOH must follow any step involving ethanol before adding HNO_3 .

261 The average concentration of potassium in the final MoO_3 product for the MURR
262 samples was slightly above 100 ppm. The elevated concentration of K in the Mo product
263 in MURR batch 1 could be due to the highest starting K/Mo ratio, which could affect the
264 efficiency of potassium removal. If more pure product is required, additional HNO_3
265 washes can be added when the starting K/Mo mole ratio is rather higher. It should be
266 noted that when irradiated Mo targets are dissolved in peroxide and converted to base, Si
267 and B are usually leached out from the glass. Also KOH can introduce some Na into
268 dissolved target solution. In general, most of the contaminants are being removed during
269 the process. Since Si is not soluble under acidic conditions, its removal is limited;
270 however, silicon does not cause any significant issue regarding the production of side
271 reaction products. An elevated concentration of sodium on the other hand, causes
272 production of short lived ^{24}Na ($T_{1/2}=15\text{h}$) and long-lived ^{22}Na ($T_{1/2}=2.6\text{years}$), which
273 increases the total gamma impurities in the product. Also when Ti is present in the target
274 material, several Sc isotopes (mostly ^{47}Sc , ^{48}Sc and ^{44}Sc) may form through (γ , p)
275 reactions, and therefore it is desirable for the starting concentration of Ti to be low.
276 Another metal that can cause significant problems when present in starting material is W,
277 which is a common Mo impurity. An elevated concentration of W can cause noticeable
278 activities of ^{187}W , ^{185}W and ^{181}W in the Mo product. It should be noted that the
279 experiments reported here were performed with natural Mo, and elevated concentrations
280 of several elements were present in starting material, compared to the actual enriched
281 ^{100}Mo or ^{98}Mo targets that have significantly lower concentration of troublesome
282 elements e.g. the concentration of Ti and W in enriched ^{100}Mo material is $\leq 10\text{ ppm}$, and
283 the total gamma activity of side reaction products from Ti and W is not problematic.
284 Nevertheless, it is worth noting that the concentration of majority of the elements in the
285 purified product was below the concentration in starting enriched Mo-100 material
286 (besides much higher starting concentration), and significant amount of troublesome
287 elements such as W, Ti and Na are being removed during the recycle process.

288

289 **CONCLUSIONS**

290 We developed a simple process that can effectively recover enriched ^{100}Mo or ^{98}Mo
291 materials by precipitating Mo using glacial acetic acid and subsequent washing with
292 concentrated HNO_3 . Typical recovery yields for Mo are $\sim 95\%$, which is very important
293 for the economic production of medical isotope $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ using enriched ^{98}Mo or ^{100}Mo
294 targets. The recovery process is capable of achieving a very high purification factor for
295 potassium. The process can be easily implemented for processing of up to 400 g of Mo
296 per batch.

297

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304

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