ANL/CSE-14/32

Scalability of the LEU-Modified Cintichem Process

Chemical Sciences and Engineering Division

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Scalability of the LEU-Modified Cintichem Process

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CONTENTS

1 INTRODUCTION 1

2 EXPERIMENTAL 4

2.1 Van de Graaff Beam Profiling 4
2.2 Temperature Profiling of Van de Graaff Sample 5
2.3 Oxalic-Acid Dosimetry 7
2.4 Molybdenum-ABO Experiments 8

3 RESULTS AND DISCUSSION 11

4 CONCLUSION 14

5 REFERENCES 15

FIGURES

1 Structure of α-Benzoin Oxime 1

2 Rig for Mo-ABO Dose Experiment 4

3 Sample Held in Aluminum Block with Compressed Air Line Rear of Sample Relative to the Beam 5

4 Rig for Mo-ABO Dose with Enclosure and Insulated Dewar for Chilled Compressed Air 6

5 Plot of VDG Beam Profile at 10 in. and 15 in. from Beam Shutter Window with 20 μA Current. 6

6 Temperature Profile of Sample without Cooling and Cooled with the Compressed Air 7

7 Oxalic Acid Dosimetry Calibration Curve Obtained at 15 in. from the Window at 30-μA Current 8

8 Image of Sample ~40 min into Irradiation 9

9 Image of Sample Prior to Irradiation 9

10 Sample of Mo-ABO Precipitate after Receiving a Dose of 3.4 Grad 11

FIGURES (Cont.)

11 Mo Recovery Results for Mo-ABO Irradiations in Presence of 0.1 M HNO_3 12

12 Photographs of HNO3 Wash Solution and NaOH Product Solution after Processing of Irradiated Mo-ABO Sample 13

TABLES

1 Equations for Calculating Dose 3

2 Distribution of Mo-99 in 0.1M HNO3 Wash, in NaOH Product Solution, and on Filter 11

SCALABILITY OF THE LEU-MODIFIED CINTICHEM PROCESS

1 INTRODUCTION

As part of the National Nuclear Security Admiration's (NNSA) Global Threat Reduction Initiate (GTRI), Argonne National Laboratory and SHINE Medical Technologies are developing technologies for the domestic production of Mo-99 for nuclear medicine while minimizing the civilian use of highly enriched uramium-235. SHINE is planning to produce Mo-99 by fission of low enriched uranium-235 (LEU) in a subcritical aqueous solution using accelerator-based neutron generation [1].

The Cintichem process has been modified for the purification of Mo-99 from LEU targets (the LEU-Modified Cintichem process—LMC), and LMC has been chosen by SHINE to process their irradiated solutions [2, 3]. Cintichem rarely processed more than 1000 Ci of Mo-99 in a single batch. In this process, Mo(VI) is precipitated by α-benzoin oxime (ABO, Figure 1), a standard analytical method for molybdenum quantification [4, 5, 6-11].

BO is the key reagent of the Cintichem and LMC process, allowing for selective precipitation of molybdenum from acidic solutions (~1 M HNO_3), where molybdenum is present as molybdenyl cation (MoO_2^{2+}):

$$MoO_2^{2+} + 2C_6H_5CH(OH)C(NOH)C_6H_5 \rightarrow MoO_2(C_6H_5CH(O)C(NOH)C_6H_5)_2 + 2H^+ \quad \text{Eq. 1}$$

A concern is that ABO will break down under high dose conditions, and the recovery of Mo-99 will decrease. Previous experiments performed at Argonne National Laboratory determined the adsorbed dose that ABO-precipitated molybdenum could tolerate without causing losses of Mo. These experiments indicated that the Mo-ABO precipitate would not begin to lose

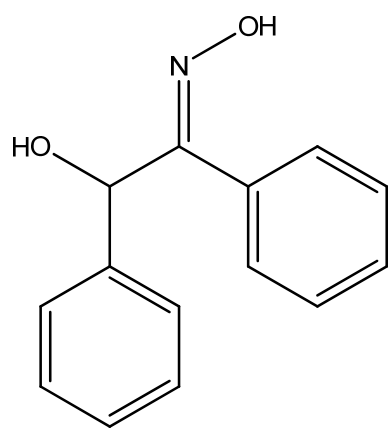

FIGURE 1 Structure of α-Benzoin Oxime (ABO)

Mo-99 until over 28,000 Ci when dry or 7,000 Ci when in contact with nitric acid. The experiments were performed by direct irradiation of small samples in test tubes with an electron beam. Samples were placed directly next to the Van de Graaff (VDG) beam exit window and irradiated for various time lengths at set beam parameters. The narrow beam and beam "wandering" introduced errors in perceived doses, so these experiments have been repeated using a wider beam. A wider beam will allow for a more uniform irradiation zone and thus uniform dose even if beam "wandering" occurs.

Computational studies have also been conducted at Argonne to determine the absorbed dose in ABO during the actual processing time of this step in the LMC with 1 kCi of Mo-99. Simulations with the Monte Carlo N-Particle Transport Code (MCNPX) [12] were conducted under several conditions and provided a link between Mo-99 activity and dose in ABO. For a more conservative scenario when the Mo-ABO precipitate is present as a monolayer, Makarashvili et al. determined that the precipitate containing 1 kCi of Mo-99 receives a dose of 148.3 Mrad [12].

Experimentally determining the dose received by the sample requires a dosimeter of identical geometry. Draganic reported that oxalic acid can be used as a dosimeter in aqueous solution and is the only dosimeter available for reactors [13]. Other aqueous chemical dosimeters exist, but oxalic acid holds advantages over them, such as 1) higher dose limits, 2) no activation of dosimeter, and 3) insensitivity to impurities and absence of photosensitivity. The dose is determined by comparing the concentration of acid prior to and after irradiation as the acid decomposes with irradiation [13]:

$$(COOH)_2 + OH \rightleftharpoons CO_2 + H_2O + COOH \qquad \text{Eq. 2}$$

$$O_2 + H \rightleftharpoons HO_2 \qquad \text{Eq. 3}$$

$$COOH + HO_2 \rightleftharpoons H_2O_2 + CO_2 \qquad \text{Eq. 4}$$

$$HO_2 + HO_2 \rightleftharpoons H_2O_2 + O_2 \qquad \text{Eq. 5}$$

$$(COOH)_2 + H \rightleftharpoons H_2O + CO_2 + CHO \qquad \text{Eq. 6}$$

Irradiated oxalic acid solution is titrated with NaOH to determine the amount of oxalic acid remaining. Once the concentration of acid is determined, the dose can be calculated from the following [13]:

$$\mathrm{Log}(D) = a \log(C) + b \qquad \text{Eq. 7}$$

where D = absorbed dose in eV/mL; C = number of oxalic acid molecules decomposed in 1 mL; and a and b are constants depending on the initial concentration of oxalic acid and the number of moles of oxalic acid decomposed, found in Table 1.

TABLE 1 Equations for Calculating Dose

Initial $(COOH)_2$ Concentration (M)	Range of log (C)	Dose Equation
25	$18.380 < \log(C) < 18.676$	$\log(D) = 0.999 \log(C) + 1.344$
	$18.676 < \log(C) < 19.057$	$\log(D) = 1.512 \log(C) - 8.244$
50	$18.380 < \log(C) < 18.954$	$\log(D) = 0.999 \log(C) + 1.344$
	$18.954 < \log(C) < 19.373$	$\log(D) = 1.426 \log(C) - 6.774$
100	$18.380 < \log(C) < 19.230$	$\log(D) = 0.999 \log(C) + 1.344$
	$19.230 < \log(C) < 19.663$	$\log(D) = 1.295 \log(C) - 4.359$
600	$18.380 < \log(C) < 19.978$	$\log(D) = 0.999 \log(C) + 1.344$
	$19.978 < \log(C) < 20.505$	$\log(D) = 1.400 \log(C) - 6.668$

2 EXPERIMENTAL

To achieve a uniform dose across the sample and eliminate beam "wandering," we placed the sample 15 in. from the beam window. Laser levels were used to position the apparatus built for this experiment (Figure 2). The sample was centered in the middle of the beam path and cooled with a stream of compressed air (Figure 3). Under radiological conditions (the use of tracer Mo-99), the rig needed to be enclosed and exhausted through a HEPA filter (Figure 4). The beam current was read on the beam shutter before irradiation of the sample and correlated with the current on the sample holder. During irradiation, the current was monitored on the aluminum sample holder, allowing for real-time beam current readout.

2.1 VAN DE GRAAFF BEAM PROFILING

Beam profiles were performed at 10 in. and 15 in. from the window using a faraday cup. The faraday cup was centered on the beam and then moved from the center in half-inch increments to obtain a profile. The results are summarized in Figure 5. It can be seen that a one-inch radial uniform beam can be achieved 15 in. from the beam shutter window.

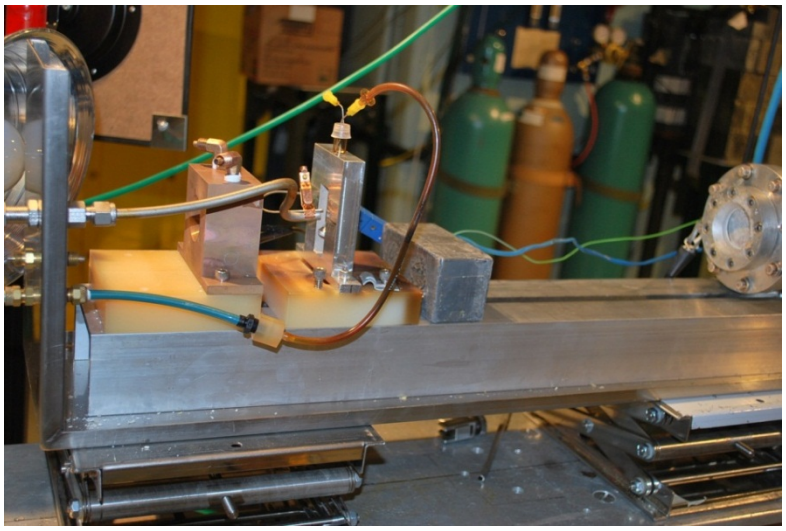

FIGURE 2 Rig for Mo-ABO Dose Experiment. The sample is located in the aluminum block with the beam window to the left. The sample holder was placed on a rail system allowing samples to be irradiated at different distances. Compressed air was passed through an ice bath to cool the samples. A copper beam stop is located behind the sample.

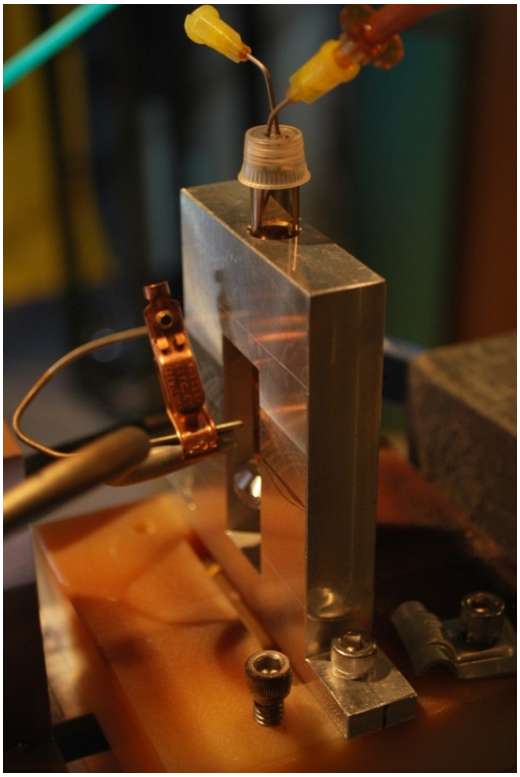

FIGURE 3 Sample Held in Aluminum Block with Compressed Air Line Rear of Sample Relative to the Beam. Current was monitored on the sample holder using alligator clips. A sweep gas was used to dissipate hydrogen accumulation from radiolysis. The cooling air supply tube can be seen on the left side of the image. Attached to the top of the air supply tube is a thermocouple wire.

2.2 TEMPERATURE PROFILING OF VAN DE GRAAFF SAMPLE

The temperature of the sample will increase with irradiation to the point at which the solution will boil or a solid will burn. Therefore, the sample temperature was monitored with a thermocouple to obtain a temperature profile. Molybdenum was precipitated with ABO, isolated, covered with HNO_3, and used to obtain a temperature profile prior to the actual sample irradiation. A thermocouple was immersed in the Mo-ABO sample in HNO_3 (0.1 M, 200 μL). To prevent boiling of the solution, the sample needed to be cooled. A copper coil was attached to the compressed air lines and submerged in a salted ice bath prior to passing the stream of air through the coil and over the sample. An insulated Dewar can be seen in Figure 3. The temperature of the sample with cooling in this manner reached ~43±2 °C after ~30 min and did not change significantly over time. For longer irradiations, the salted ice bath was replaced as necessary. During irradiation, the temperature of the sample was externally monitored by a thermocouple

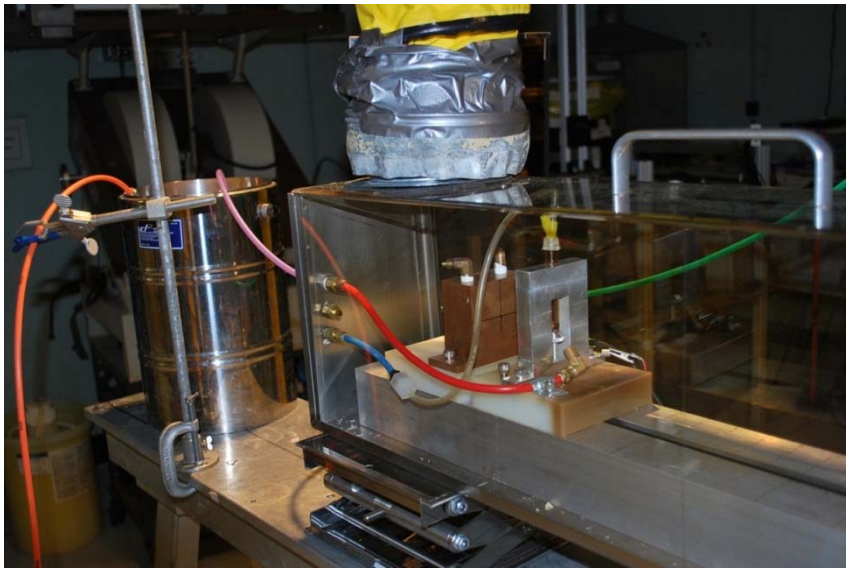

FIGURE 4 Rig for Mo-ABO Dose with Enclosure and Insulated Dewar for Chilled Compressed Air. Not shown is the exhaust tube to HEPA filter system.

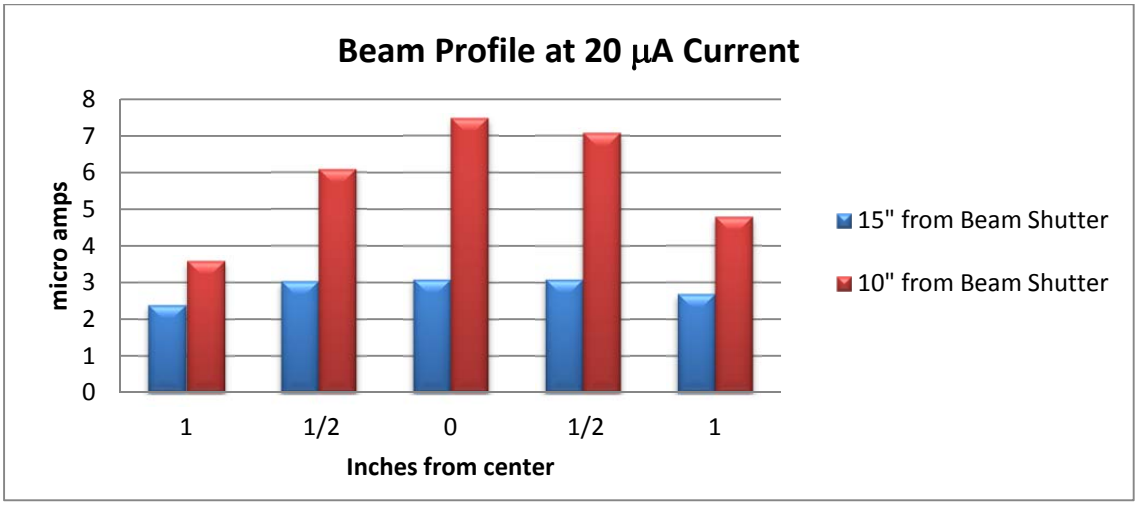

FIGURE 5 Plot of VDG Beam Profile at 10 in. and 15 in. from Beam Shutter Window with 20 μA Current

placed in contact with the exterior of the irradiated vial. The thermocouple can be seen attached to the top of the cooling air supply tube in Figure 3. The results of temperature profile are summarized in Figure 6. Note that the temperature readings from the external thermocouple touching the sample (triangles) are lower than those from the thermocouple immersed in the sample (circles) because it was in the direct stream of the cooling air.

2.3 OXALIC-ACID DOSIMETRY

Doses were determined by oxalic-acid dosimetry. A solution of oxalic acid (~0.6 M, 1 mL) was irradiated with a 30 μA beam at 15 in. from the window for various time lengths and cooled with a constant stream of compressed air (chilled by a salted ice bath). The clear glass test tube browned over time. Irradiated solutions of oxalic acid (0.9 mL) were titrated with standardized NaOH to determine the final concentration of oxalic acid. Plotting the results yielded a linear dose curve (Figure 7). It is important to realize that the dose is extremely dependent on the position of the sample. Even slight variations in the sample's position will dramatically alter the dose received. Therefore, dose calibrations must be obtained with the rig in place right before experimental work begins.

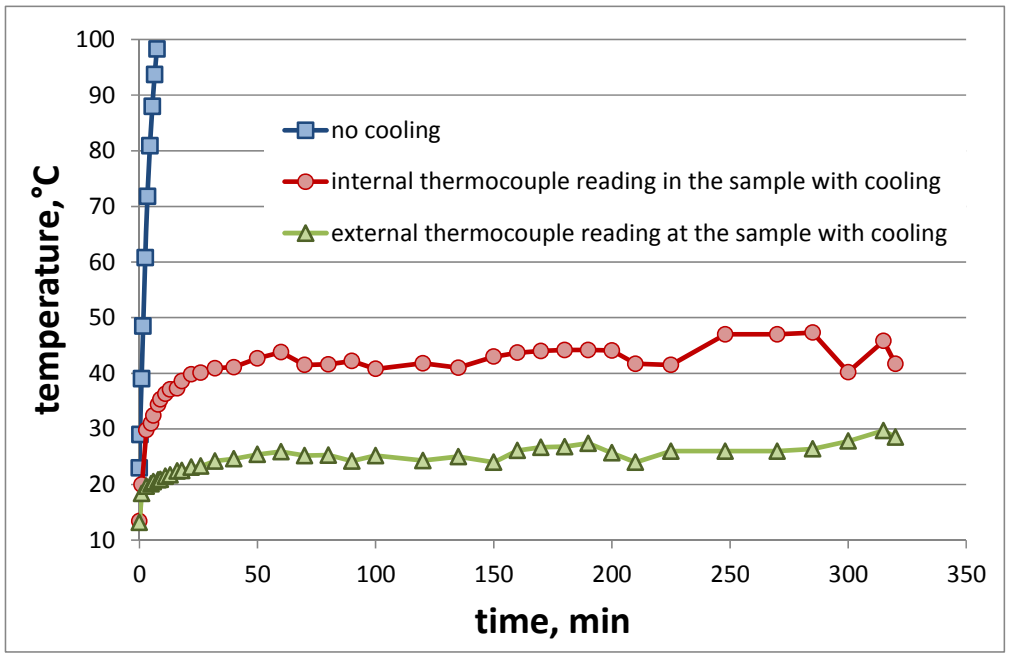

FIGURE 6 Temperature Profile of Sample without Cooling (squares) and Cooled with the Compressed Air

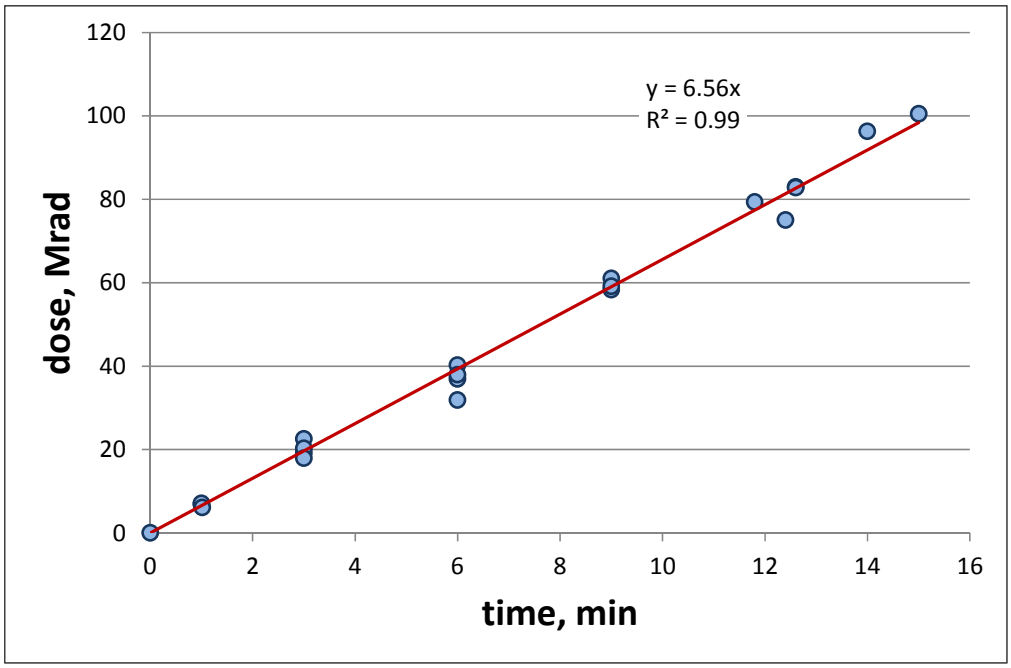

FIGURE 7 Oxalic Acid Dosimetry Calibration Curve Obtained at 15 in. from the Window at 30-μA Current

2.4 MOLYBDENUM-ABO EXPERIMENTS

A molybdenum carrier solution (10 mg-Mo/mL) was prepared by dissolving MoO_3 in NaOH (1 M) and neutralizing it with HNO_3 (8 M). Prior to precipitation with ABO, the Mo carrier solution was spiked with a known amount of Mo-99. The ABO (2%) was prepared by dissolution in hot NaOH (0.4 M). A Mo carrier (11.0 μL) spiked with Mo-99 was diluted with HNO_3 (~1.43 M, ~1.47 mL) and oxidized with $KMnO_4$ (2.5 % $KMnO_4$, 68 μL). The resultant solution was mixed and then precipitated with the ABO stock solution (453 mL). The mixture was centrifuged, and the supernatant was removed. The Mo-ABO solid was covered with HNO_3 (0.1 M, 200 μL) and irradiated in a glass vial. Figure 8 shows the bubble formation that occurred during the irradiation. This phenomenon is also being studied at Argonne [14]. Figure 9 shows a representative sample before irradiation.

After irradiation, the samples were filtered with a 0.22-μm polyvinylidene fluoride (PVDF) membrane filter (Millipore). The vessel and filter were washed with HNO_3 (0.1 M, 2.0 mL). The wash was kept for gamma counting. The Mo-ABO precipitate was dissolved from the filter using a hot NaOH/H_2O_2 solution (0.4 M NaOH, 1 % H_2O_2, 1.5 mL). To ensure complete dissolution of the ABO-Mo precipitate, the filter was washed with an additional 1.0 mL of NaOH/H_2O_2 (0.2 M NaOH, 1 % H_2O_2). Another NaOH wash (0.2 M, 0.5 mL) was used to rinse any remaining residue left on the filter. All NaOH fractions were collected, combined, and kept for gamma counting. A high purity germanium (HPGe) gamma detector was used to determine the amount of Mo-99 (739.4 keV emission) in the HNO_3 wash, NaOH product, and any remaining on the filter.

FIGURE 8 Image of Sample ~40 min into Irradiation. Obtained by a remote camera placed within the irradiation cell and through the Mo-ABO rig lid.

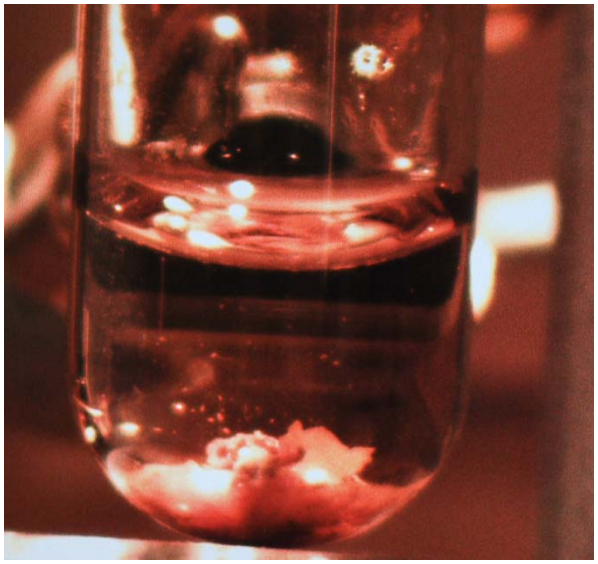

FIGURE 9 Image of Sample Prior to Irradiation. Obtained by a remote camera placed within the irradiation cell. The lid to the Mo-ABO rig was removed to obtain this image.

Under these scaled-down conditions, it was difficult to achieve quantitative dissolution of the Mo-ABO complex, and some Mo was left on the filter. In the Cintichem process, the Mo-ABO complex is dissolved in a $NaOH/H_2O_2$ mixture, which was then heated (via heat gun) and shaken using glass beads to achieve quantitative dissolution. Moreover, the dissolved Mo in the NaOH solution was removed from the glass filter in a vacuum, which led to a very high Mo recovery. These conditions are difficult to replicate in small-scale experiments.

3 RESULTS AND DISCUSSION

Freshly prepared samples of Mo-ABO precipitate in the presence of HNO_3 were irradiated and received a dose in the range of 0.4-3.4 Grad. After irradiation, a significant change in color was observed for the precipitate (Figure 10). The data in Table 2 and Figure 11 show the distribution of Mo-99 in the HNO_3 wash, NaOH product solution, and on the filter.

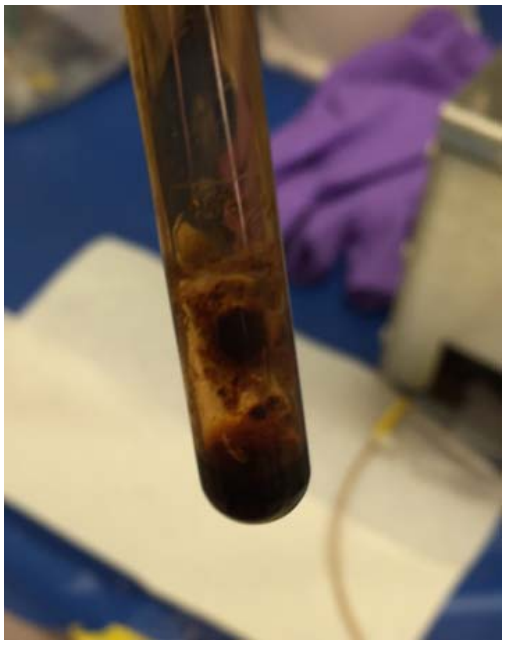

FIGURE 10 Sample of Mo-ABO Precipitate after Receiving a Dose of 3.4 Grad

TABLE 2 Distribution of Mo-99 in 0.1M HNO_3 Wash, in NaOH Product Solution, and on Filter

			Mo-99 Distribution (%)		
Sample #	Dose, Grad	kCi of Mo-99	HNO_3	Filter	NaOH
1	0.395	2.66	4.27	9.31	86.43
2	0.816	5.50	4.72	10.20	85.08
3	1.23	8.29	4.77	3.62	91.62
4	1.85	12.5	5.88	2.43	91.70
5	2.17	14.7	9.69	2.34	87.96
6	2.52	17.0	20.27	1.94	77.79
7	3.39	22.9	12.20	3.29	84.51

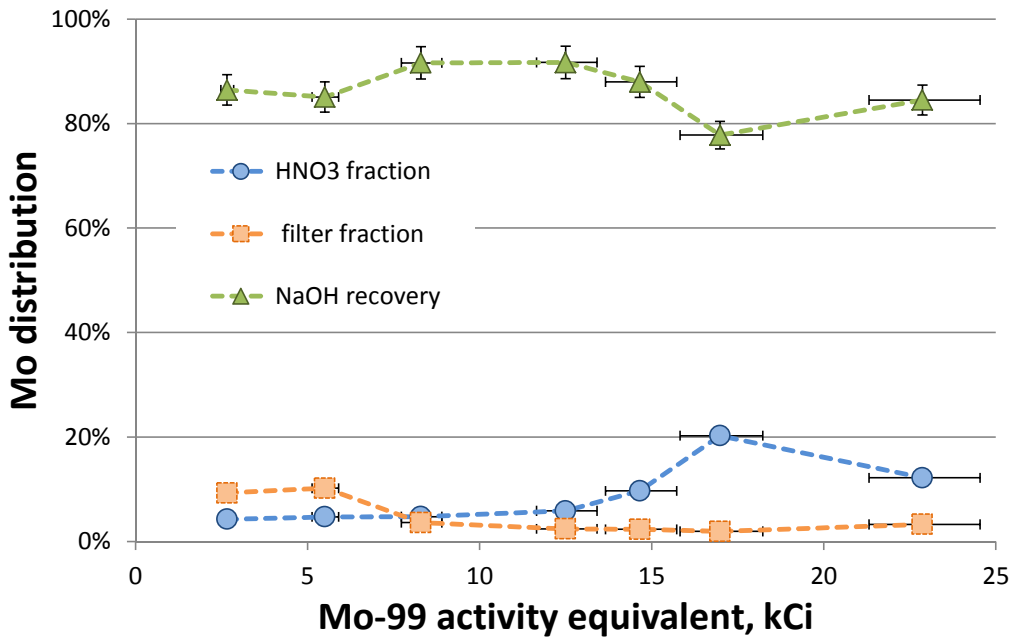

FIGURE 11 Mo Recovery Results for Mo-ABO Irradiations in Presence of 0.1 M HNO_3

As mentioned earlier, due to several limitations, it was difficult to achieve very good recovery of Mo under these scaled-down conditions, and even after centrifugation at 6,500 rpm, a small portion of solution remained on the filter. We found that adding the $NaOH/H_2O_2$ solution in several aliquots and increasing the centrifugation time from 2.5 min to 5 min increased Mo recovery from the filter.

All samples showed signs of degradation after irradiation; colorless HNO_3 turned a light yellow color, and the NaOH solutions were dark brown (Figure 12). Despite the discoloration of the HNO_3 wash, only a small portion of Mo was detected in this fraction. Larger fractions of Mo were observed in these washes at doses starting at ~1.85 Grad (12.5 kCi Mo-99 equivalent). From the data in Table 2 and Figure 11, it is evident that at doses >1.85 Grad, the decomposition of the Mo-ABO complex leads to the formation of Mo species that are soluble in 0.1 M HNO_3, which causes a noticeable decrease in Mo recovery. Previously obtained data [12] using a narrower beam showed a noticeable degradation of the Mo-ABO complex and increased Mo content in the HNO_3 wash at the dose equivalent to ~7.5 kCi of Mo-99. We were not able to achieve dose rates to determine the "breaking point" of the Mo-ABO solid but have shown that >90% of Mo was retained in the solid up to 1.9 Grad (12.5 kCi Mo-99 equivalence) and >80% of Mo was retained in the solid up to ~3.4 Grad (22.9 kCi Mo-99 equivalence). Higher doses are needed to determine the "breaking point" of Mo-ABO.

An advantage of irradiating at a longer distance is that a more uniform beam (Figure 5) can be achieved; however, the dose rates are significantly lower than when irradiating samples closer to the beam exit window. This condition leads to much longer irradiation times. For example, to achieve a dose of 3.4 Grad with a beam current of 30 μA, the sample was irradiated

FIGURE 12 Photographs of HNO_3 Wash Solution and NaOH Product Solution after Processing of Irradiated Mo-ABO Sample (1.2 Grad)

for 517 min. To increase the dose, the distance must be decreased, or the current or the time must be increased. A uniform beam is desirable; therefore, the distance cannot be changed. Increasing the time of an experiment is not a desirable feature, so the current must be increased. This condition leads to a new challenge because the sample must be more adequately cooled. The current cooling system passes compressed air through coiled tubing submerged in an ice bath onto the back side of the sample/target (opposite of beam) by a single outlet. An improvement would be to cool the sample from two angles, preferably where the beam strikes the target. The target holder is currently being redesigned to integrate these changes. These experiments will be continued at higher current to achieve higher doses after the new target holder has been redesigned and tested.

4 CONCLUSION

Irradiation of the Mo-alpha-benzoin oxime (Mo-ABO) precipitate spiked with Mo-99 was performed at a Van de Graaff generator using a wide electron beam at 15 in. from the window. Samples of the Mo-ABO precipitate in a glass vial were irradiated in the presence of HNO_3 and cooled by a jet of cold compressed air. The temperature of the sample during irradiation was ~43 °C. Dose equivalents up to 22.9 kCi of Mo-99 were applied. After irradiation, the Mo-ABO precipitate was filtered, washed with HNO_3, dissolved in a hot $NaOH/H_2O_2$ mixture, and rinsed with NaOH. All washes, dissolutions, and rinses were gamma counted to determine Mo-99 content. The experimental data demonstrate good radiation stability of the Mo-ABO complex up to ~12.5 kCi dose equivalents of Mo-99. At higher doses, the decomposition of the Mo-ABO complex leads to the formation of Mo species that are soluble in HNO_3 (10-20%) and causes a noticeable decrease in Mo recovery. The irradiations of the Mo-ABO precipitate will continue at higher doses after modifications are made to the sample holder to allow for better cooling and higher currents (up to 50 μA).

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