Modifications to LEU-Modified Cintichem Process to Allow Processing of Fission-Produced ^{99}Mo from Up to 750 g of Irradiated Uranium Targets

Chemical and Fuel Cycle Technologies Division

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by

Peter Tkac and George F. Vandegrift

Chemical and Fuel Cycle Technologies Division

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CONTENTS

1. Introduction.....	1
2. The LEU-Modified Cintichem Process.....	1
3. Potential Radiation Damage to the Mo-ABO Complex.....	3
4. Potential LMC Modifications for Eden	4
4.1 Iodine Carrier	4
4.2 Silver Nitrate.....	4
4.3 Ru and Rh Carriers.....	5
4.4 Mo Carrier and ABO.....	5
4.5 Acidification Before Mo-ABO Precipitation	5
4.6 Ag-Charcoal and Combination Column	6
5. Conclusion	7
6. References.....	8

FIGURES

Figure 1. α -benzoin oxime (ABO).....	2
Figure 2. Scheme of LEU-modified Cintichem process for purification of ^{99}Mo from fission products (Ag/C = silver coated activated charcoal column, HZO = hydrated zirconium oxide).	3

1. INTRODUCTION

Eden Radioisotopes, LLC is developing a TRIGA*-sized, 2 MW all-target reactor system licensed from Sandia National Laboratories [1]. This all-target reactor uses up to 60 Argonne-designed annular LEU-foil targets to produce fission-produced ^{99}Mo . The Eden low-enriched uranium (LEU) target contains ~ 137 g of uranium and will be irradiated at up to 30 kW, with a target burnup determined by the number of targets processed each week. The annular target utilizes metallic uranium foils 400 mm long by 72 mm wide by 250 ± 25 μm -thick covered by a thin (10 μm on each face) nickel fission barrier. The foil is placed between two 35 mil aluminum tubes that form the outer and inner walls of the cladding cylinder, which is hydroformed to provide high thermal conductivity within the target. Eden expects to process between three and five targets per process batch, to maintain the process batch ^{99}Mo inventory at or below 5000 curies to maintain high ^{99}Mo recovery, within a few hours of discharge from the reactor. Eden expects to meet full U.S. demand by processing 30 targets per week, with each target operated for two weeks at 30 kW. To meet less than full U.S. demand, both the target power and the number of targets processed weekly will be adjusted. With increased ^{99}Mo production and given the size of U targets processed in a single batch (up to $\sim 750\text{g-U}$), it is important to identify whether any modifications are needed to the LEU-modified Cintichem process.

Eden's proposed separation scheme starts with acidic dissolution of irradiated U-metal targets using nitric acid. Dissolved U containing ^{99}Mo and other fission products is then loaded on a titania (titanium dioxide) column to separate ^{99}Mo from the bulk uranium. Uranium and most fission products in the feed solution are not adsorbed on the column and remain in the eluate. After washing steps, molybdenum and other fission products are stripped from the TiO_2 column using a sodium-hydroxide solution. The strip-eluate solution is then acidified and submitted for a final purification using the LEU-modified Cintichem (LMC) process. The uranium is then precipitated by base to drive the formation of insoluble uranyl-hydroxide (most of the Tc is removed), redissolved in nitric acid, and submitted for solvent extraction with tri-*n*-butyl phosphate (TBP; UREX = uranium extraction) to purify and recover the uranium. After acetohydroxamic acid is used to scrub the co-extracted Np/Pu, U is stripped from the TBP using dilute nitric acid.

To recycle the uranium into new targets, this solution must be converted to uranium metal. The conversion of the UREX strip solution to uranium metal is a multistep process. The two primary steps for conversion are (1) conversion of the solution to UO_2 and (2) conversion of UO_2 to U metal [2].

2. THE LEU-MODIFIED CINTICHEM PROCESS

The Cintichem process, developed in 1974 at the Cintichem facility in Tuxedo, N.Y., is a small-scale (starting solution is ~ 50 mL) method for purification of fission-produced molybdenum using specific glassware designed for remote operation in a hot cell. It was developed to separate useful isotopes like ^{99}Mo from mixed fission products produced by irradiation of 93% ^{235}U HEU (UO_2) in a 5-megawatt open-

* General Atomics' TRIGA® (Training, Research, Isotopes, General Atomics)

pool reactor [3]. From 1980 to 1989, Cintichem Inc.[†] was the only U.S. producer of ⁹⁹Mo, with a production rate of ~2000 6-day Ci/week (1Ci=3.7×10¹⁰Bq).

The Cintichem process was later modified by Argonne National Laboratory to enable production of ⁹⁹Mo by LEU foil target, and sulfuric acid was replaced by nitric acid. The modified process is called the LEU-modified Cintichem process or LMC process. Several full-scale demonstrations of the LMC process were performed at the Indonesian Badan Tenaga Atom Nasional (BATAN) facility in Indonesia between 1998 and 2010 [4]. Several papers on the LMC process have been published [5, 6, 7, 8], indicating continuous interest in this small-scale purification process.

The key reagent of the Cintichem process, α-benzoin oxime (ABO), shown in Figure 1, allows selective precipitation of Mo from acidic media (~1M HNO₃) where molybdenum is primarily present as molybdenyl cation (MoO₂²⁺) in the following reaction:

Figure 1. α-benzoin oxime (ABO).

The LMC process (Figure 2), in which Mo is purified from actinides and other fission products, has several steps, most of which are for the removal of radioiodine. During the dissolution of uranium foil in nitric acid, most of the iodine present as I₂ is volatilized and collected in an iodine trap, although other iodine species such as I⁻, IO₃⁻ and IO₄⁻ may be present. The fission-produced ⁹⁹Mo can be separated from uranium by loading the uranium solution obtained after dissolution of irradiated targets on a titania-based column. While uranium and some other fission products pass through, Mo is retained on the column [9, 10] and then recovered using ~1 M NaOH, which needs to be converted to ~1 M HNO₃ in order to further purify Mo from other fission products by the LMC process. After the Mo-strip solution is converted to ~1 M HNO₃, iodide is precipitated as AgI by adding NaI and AgNO₃ (Figure 2). The remaining silver is precipitated by adding HCl. Precipitation of iodide ions by the addition of silver nitrate is considered quantitative; the solubility product of AgI is 8.3×10⁻¹⁷ M [11]. The I₂, which does not co-precipitate with silver ions, can be removed by reaction with silver metal to form the insoluble AgI. The solubility of silver iodate is also limited but is significantly larger (3.1×10⁻⁸ M) [11] than that of AgI. It is likely that AgIO₄ is even more soluble than AgIO₃. Vandegrift et al. [12] have shown that radioactive iodine present

[†] The research facility was later sold to Hoffman-LaRoche.

as periodate can be removed by co-precipitation with AgI and AgCl if sufficient time is allowed for isotopic exchange.

The next step in the LMC process is the addition of an Mo carrier, to aid in its precipitation, and Ru and Rh hold-back reagents, to prevent their co-precipitation with Mo. To keep Mo in a hexavalent oxidation state, potassium permanganate is added. After the addition of ABO, a white flocculate precipitate is formed and is collected on a fritted-glass filter containing glass beads. The Mo-ABO precipitate is then washed multiple times with 0.1 M HNO_3 . The Mo-ABO precipitate is then dissolved in a heated $\text{NaOH}/\text{H}_2\text{O}_2$ solution. The glass beads are present to aid in washing the precipitate and to allow for better dissolution. The solution containing Mo and re-dissolved ABO is passed through a silver-coated charcoal column (Ag/C). Molybdenum passes through, while organic residuals are retained on the charcoal bed. If I_2 is present, it reacts with the silver metal on the Ag/C column to form insoluble AgI. Before the solution is loaded on a combination column, which consists of equal volume beds of activated charcoal, hydrous ZrO (HZO, which acts as a cation exchanger), and Ag/C, precipitation of trace iodine is repeated by adding NaI and AgNO_3 . The AgI precipitate is collected on the combination column, and Mo is eluted with 0.2M NaOH .

Figure 2. Scheme of LEU-modified Cintichem process for purification of ^{99}Mo from fission products (Ag/C = silver coated activated charcoal column, HZO = hydrated zirconium oxide).

3. POTENTIAL RADIATION DAMAGE TO THE Mo-ABO COMPLEX

Typically, Cintichem and BATAN processed targets containing less than 1000 Ci of ^{99}Mo and did so without losses in Mo recovery, due to a radiolytic instability of the Mo-ABO complex. Since Eden is

expecting to produce several kCi of ^{99}Mo and process it using a single LMC run, it is important to understand the activity limit (kCi of ^{99}Mo) that could cause radiation damage to the Mo-ABO complex and negatively affect Mo recovery. With increased ^{99}Mo production, due to the size of U targets processed in a single batch (up to $\sim 750\text{g-U}$), it is important to identify whether any modifications are needed to the LEU-modified Cintichem process. To cover 50% of U.S. demand, Eden plans to process 21 targets per week, producing $\sim 1,300$ 6-day Ci/week. By processing five targets in a single batch, Eden is expected to process $\sim 2.7\text{kCi}$ of ^{99}Mo activity—more than what was normally processed from 1980 to 1989 in the Cintichem facility, the only former U.S. producer of fission-produced ^{99}Mo with weekly production of ~ 2000 6-day Ci/week [13, 14, 15, 16, 5]. With higher activities of ^{99}Mo processed in a single batch, a concern is that α -benzoin oxime (ABO), the key reagent in the process, could break down under high radiation conditions. However, previous experiments on radiation stability of ABO performed at Argonne indicate that ^{99}Mo yields will not be decreased when up to $\sim 7\text{kCi}$ of ^{99}Mo is processed in a single batch [17, 18, 19].

4. POTENTIAL LMC MODIFICATIONS FOR EDEN

With the increased amount of irradiated uranium processed in a single batch, it is important to see whether any modifications are needed in the LMC process to offset the higher concentration of certain elements (assuming 30kW for 21d irradiation per target). By processing up to five U targets in a single batch, it is expected that final volume of acidic HNO_3 solution produced after dissolution of U targets would be around 2 L. This solution would be passed through a TiO_2 column to remove bulk U, and the Mo solution would then be eluted in a basic NaOH solution ($<50\text{ mL}$). After acidification to $\sim 1\text{ M HNO}_3$, this solution could then be directly used by LMC to purify ^{99}Mo . For this exercise, it is assumed that fission-produced Mo, I, Ru, and Rh are not retained by the TiO_2 column. Because natural Mo, I, Ru, and Rh compounds are added as carriers in the LMC process, this is an important assumption for the discussion below.

4.1 Iodine Carrier

The first step in the LMC process is iodine precipitation. In this step, 4 mg of iodine are added as 4 mL NaI (4mg/mL) to facilitate the precipitation of radioiodine. This is equivalent to $\sim 0.0315\text{ mmol}$ of iodine. However, the expected amount of iodine produced in five U targets is $\sim 45\text{ mg}$ (0.425 mmol). Although it is likely that the bulk of the iodine will be volatilized during the nitric acid dissolution of the foils, it is currently not clear what fraction will remain in solution, and iodine is constantly being formed by the decay of radioactive tellurium isotopes. In the standard LMC process, 50 mg of Ag (0.463 mmol) are added as 0.5 mL of 10% AgNO_3 in 0.1M HNO_3 to precipitate added the 4mg of iodine carrier. Due to the much greater quantity of Ag than I in the standard LMC process ($\sim 15\text{x}$), to effectively remove iodine produced in five irradiated targets, the amount of Ag needs to be increased to a maximum of 560 mg of Ag (5.2 mmol of Ag), or $\sim 5.6\text{ mL}$ of 10% AgNO_3 solution. The effectiveness of this iodine removal should be explored experimentally.

4.2 Silver Nitrate

Because large amounts of Ag are needed to effectively remove iodine, the next step in the standard LMC process is to remove Ag by adding HCl (1 mL of 1M HCl or 1 mmol of HCl). This needs to be increased to 11.3 mmol of HCl or 1.13 mL of 10 M HCl.

4.3 Ru and Rh Carriers

Then, before the selective precipitation of Mo with ABO, Ru and Rh carriers are added to prevent their coprecipitation with Mo. In the standard LMC process, 12 mg of Rh (0.117 mmol of Rh) is added as 1.5 mL of 8 mg-Rh/mL, and 10 mg of Ru (0.099 mmol of Ru) is added as 2 mL of 5 mg-Ru/mL. The amount of Ru produced in five irradiated targets—204.5mg (2 mmol of Ru)—greatly exceeds the amount that is added as a carrier in the standard LMC process. The amount of Rh produced is 9.1 mg (~0.088 mmol), which is close to the amount currently added as a carrier. It is expected that addition of Ru and Rh carriers should not affect Mo decontamination, but experiments should be performed to validate this claim.

4.4 Mo Carrier and ABO

The addition of a Mo carrier for precipitation of ^{99}Mo with ABO can affect the specific activity of the final product. This depends on curie production of ^{99}Mo and how much carrier is added. In the standard LMC process, 5 mg of Mo (0.052 mmol of Mo) is added as 0.5 mL of 10mg-Mo/mL solution. It is expected that up to ~260 mg of Mo (2.7 mmol of Mo) can be produced in five irradiated targets; therefore no addition of Mo carrier would be needed. However, due to the higher content of Mo, the amount of ABO needs to be increased to ensure the complete precipitation of Mo. In the standard LMC process, 20 mL of fresh 2% ABO (1.76 mmol of ABO) in 0.4M NaOH is added, which produces an ABO:Mo mol ratio of 33.8. To keep the same ABO:Mo ratio as in the standard LMC process, ~91.5 mmol of ABO would be required. Due to the limited solubility of ABO in 0.4M NaOH, its concentration needs to remain the same, and practically speaking, only the volume of solution can be increased. This, however, leads to the unreasonable volume of ~1 L of 2% ABO. Previous studies that looked at the minimum ABO:Mo ratio for effective precipitation of Mo suggest that much smaller amounts of ABO could be sufficient [20]. It was demonstrated that an ABO:Mo ratio of 3.6 or higher led to complete Mo precipitation. Therefore, it is expected that 4x ABO:Mo mole ratio should provide satisfactory Mo precipitation yields. Based on these results, about 2.5 g of ABO would be needed, which could be added as ~120 mL of 2% ABO in 0.4 M NaOH.

It should be noted, however, that increased amounts of KMnO_4 can have a negative impact. The addition of KMnO_4 in the LMC process is performed as a colorimetric titration, where the addition is stopped after a deep pink color develops. This ensures that Mo is in the appropriate oxidation state for complexation with ABO. At low KMnO_4 concentrations, the Mo-ABO complexation formed a white flocculent precipitate that was easy to transfer and that dissolved readily in hot NaOH/ H_2O_2 solutions. When increased amounts of 2.5% KMnO_4 solution were used (20x), significant losses of Mo were observed. First, the precipitation of Mo with ABO was affected, and 24% of Mo was found in the effluent stream [20]. Secondly, the precipitate morphology changed, forming dense, viscid clumps that resisted dissolution and adsorbed easily to glass and plastic surfaces. These precipitate characteristics led to downstream losses during the mixing and dissolution steps. About 12% of the Mo was left undissolved on the filter after dissolution, with other losses accruing on surfaces and mixing equipment, leading to a total Mo loss of 36% [20].

4.5 Acidification Before Mo-ABO Precipitation

As indicated previously, it is important that Mo is present as molybdenyl species to allow for complexation with ABO (Eq. 1). This can only be achieved under acidic conditions, so an appropriate amount of HNO_3 is needed. In the standard LMC process, 11 mL of 4 M HNO_3 is added after the iodine precipitation step to wash the AgI precipitate; however, with increased amounts of ABO in 0.4 M NaOH added to offset higher the Mo content in Eden's process, the amount of HNO_3 needs to be appropriately

increased to keep the solution ~ 1 M HNO_3 . Total H^+/OH^- concentrations in the standard LMC process are shown in Table 1. In Table 1, the reagents are listed horizontally as they are added. The first column is the reagent added followed by its volume and hydrogen ion or hydroxide concentrations. The next column is the total volume of the resulting solution, followed by the resulting hydrogen ion concentration. After the addition of the ABO solution, the hydrogen ion concentration is calculated to be 0.971 M.

Table 1. Concentration of H^+ before precipitation of Mo-ABO versus reagents added in the standard LMC process.

Reagent	V ml	$[\text{H}^+]$ M	$[\text{OH}^-]$ M	V(total) mL	$[\text{H}^+](\text{total})$ M
1M HNO_3	60	1	0	60	1.000
NaI	4	0	0	64	0.938
AgNO_3	0.5	0.1	0	64.5	0.931
HNO_3	11	4	0	75.5	1.378
HCl	1	1	0	76.5	1.373
Mo carrier	0.5	0	0	77	1.364
KMnO_4	3	0	0	80	1.313
ABO	20	0	0.4	100	0.971

If no additional HNO_3 is added, and the volume of ABO is increased to 120 mL to offset the higher Mo content, the concentration of HNO_3 before Mo precipitation would be 0.282 M, which is not sufficient. Therefore, it is important to adjust the acidity of the solution before the Mo-ABO precipitation step. This could be done by addition of 12 mL of 16 M HNO_3 after the iodine precipitation step, or, more conveniently, adjust the HNO_3 concentration after the acidification of the titania strip solution to ~ 3.4 M (instead of ~ 1 M). Based on this exercise, modified volumes of reagents and the adjusted starting HNO_3 concentration are shown in Table 2. The final hydrogen ion concentration in this case is 0.98 M.

Table 2. Concentration of H^+ before precipitation of Mo-ABO versus reagents added for the LMC process with higher volume of ABO

Reagent	V ml	$[\text{H}^+]$ M	$[\text{OH}^-]$ M	V(total) mL	$[\text{H}^+](\text{total})$ M
1M HNO_3	60	3.4	0	60	3.400
NaI	4	0	0	64	3.188
AgNO_3	6.75	0.1	0	70.75	2.893
HNO_3	11	4	0	81.75	3.042
HCl	1.35	1	0	83.1	3.009
Mo carrier	0	0	0	83.1	3.009
KMnO_4	3	0	0	86.1	2.904
ABO	120	0	0.4	206.1	0.980

4.6 Ag-Charcoal and Combination Column

The final purification steps performed in the LMC process are performed by a combination of the Ag-charcoal column, which removes residual organic content after the dissolution of the Mo-ABO complex, and the combination column, composed of Ag-charcoal, hydrous zirconia and activated charcoal, which acts as a final step for the removal of any impurities left in Mo product. Due to the increased amount of ABO needed for the Eden-modified LMC process, it is possible that a larger column will be needed for

effective purification. The column size needed for effective removal of organic content needs to be determined experimentally.

5. CONCLUSION

Overall, the LMC process is applicable to purifying the ^{99}Mo produced by the Eden system, but because of the amount of fission products expected in five irradiated U targets (750g of U total, at 30kW and 21d irradiation) several modifications are needed:

- Due to the higher iodine content expected, the amount of AgNO_3 and HCl needs to be increased.
- Due to an expected higher Mo content, the amount of ABO needed to effectively remove Mo by precipitation rises from 20 mL of 2% ABO to about 120 mL.
- This in turn leads to the need to increase the amount of HNO_3 added to ensure acidic conditions before the Mo-ABO precipitation step.
- It is also possible that the appropriate size of the Ag-charcoal and combination columns would need to change; this should be determined experimentally.

Because of the complex redox chemistry in these solutions, the changes suggested above must be tested with irradiated uranium to assure high yields and purity of the ^{99}Mo product.

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Argonne National Laboratory
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