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PROCESSING OF LEU TARGETS FOR ^{99}Mo PRODUCTION -- TESTING AND MODIFICATION OF THE CINTICHEM PROCESS

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ABSTRACT

Recent experimental results on testing and modification of the Cintichem process to allow substitution of low enriched uranium (LEU) for high enriched uranium (HEU) targets are presented in this report. The main focus is on ^{99}Mo recovery and purification by its precipitation with α-benzoin oxime. Parameters that were studied include concentrations of nitric and sulfuric acids, partial neutralization of the acids, molybdenum and uranium concentrations, and the ratio of α-benzoin oxime to molybdenum. Decontamination factors for uranium, neptunium, and various fission products were measured. Experiments with tracer levels of irradiated LEU were conducted for testing the ^{99}Mo recovery and purification during each step of the Cintichem process. Improving the process with additional processing steps was also attempted. The results indicate that the conversion of molybdenum chemical processing from HEU to LEU targets is possible.

INTRODUCTION

The Cintichem process for ^{99}Mo production currently uses high enriched uranium (HEU, ~93% ^{235}U) as irradiated UO_2 deposited on the inside of a cylindrical target [1, 2]. In order to convert the process to low enriched uranium (LEU, < 20% ^{235}U) as a uranium metal-foil target, the effects of modifying the dissolver solution must be studied, and necessary modifications must be made.

In the Cintichem process, the UO_2 in the target is dissolved in a mixture of H_2SO_4 and HNO_3. After the target is dissolved, the solution is prepared for an α-benzoin oxime precipitation by the addition of several reagents. Following this precipitation step, the precipitate is collected, washed, and redissolved. The redissolved molybdenum solution is then passed through two additional purification steps.

It was our objective in switching from HEU to LEU to maintain the process for molybdenum recovery and separation from uranium and its fission and absorption products as close as possible to the current Cintichem process. Except for the potential of higher concentrations of uranium and ^{239}Np/Pu following the precipitation, it is likely that no other major differences will exist between the LEU and HEU processes. The three main goals in this study were to

(1) measure the effectiveness of the current process for LEU targets, (2) measure the robustness of the precipitation step to variations in the composition of the dissolver solution, and (3) develop modifications of the process to accommodate LEU substitution.

MOLYBDENUM RECOVERY

Molybdenum recovery efficiency is defined as the ratio of ^{99}Mo recovered in the final product to that produced from uranium fission. The Cintichem process involves a series of steps for separating in the ^{99}Mo from uranium, transuranic isotopes, and other fission products. Our attention was paid to the precipitation part, since its efficiency depends on many variables, as we will discuss in the following subsections. Molybdenum recovery efficiency was measured by two techniques:

1. Adding a known amount of natural molybdenum to solution, performing the precipitation, and measuring the amount of molybdenum in the precipitate by neutron activation analysis (NAA).

2. Activating molybdenum with neutron irradiation, adding the radioactive ^{99}Mo tracer, performing the precipitation, and measuring the ^{99}Mo in the filtrate solution.

The effects of varying the composition of the dissolver solution on the precipitation are discussed below. The test procedure followed the proprietary methods developed by Cintichem. The overall conclusion from this study is that the precipitation of Mo(VI) by α-benzoin oxime is quite robust, and the yield of ^{99}Mo from LEU is about the same as from HEU.

Molybdenum Concentration

In a series of seven tests, molybdenum recovery in the precipitate was measured as a function of molybdenum concentration. In tests 1-5, the molybdenum concentration was varied from 0.028 to 1.11 mg/mL as the mass ratio of α-benzoin oxime to molybdenum (αBZ/Mo) was kept at 10 mg to 1 mg (10/1). The filtration was performed using a glass filtration unit, which included a Millipore membrane filter (MF, mixed esters of cellulose) having a diameter of 47 mm and a pore size of 0.22 µm. The molybdenum recovery was determined by measuring the molybdenum in the precipitate by NAA. Tests 6 and 7 were run using 100 mL of 0.94$\underline{M}$ H_2SO_4 as a matrix solution and adding 1 mL and 0.2 mL of 10 mg/mL Mo standard solutions, respectively. A Mitex (PTFE) filter with 5-µm pore size was used for a first filtration, since the MF filter could not handle the large amount of precipitate. To prevent loss of precipitated molybdenum to the filtrate, a second filtration was conducted using an MF filter. The feed solution in test 6 was chilled in ice water to compare the effects without this procedure in test 7.

Results of these tests are shown in Table 1. As can be seen, molybdenum recoveries are above 90% for all solutions except for test 5, which has the lowest molybdenum and α-benzoin oxime concentrations. More α-benzoin oxime was added in tests 6 and 7 than tests 1-5. As seen in Table 1, higher initial concentrations of α-benzoin oxime can increase recoveries for low molybdenum concentrations. As seen in comparing tests 6 and 7, chilling had no effect on molybdenum recovery.

Table 1. Molybdenum Recovery as Function of Initial Molybdenum Concentration

Test	Initial Mo Conc. (mg/mL)	Mass Ratio αBZ/Mo	% Mo Recovery
1	1.11	10	93
2	0.22	10	100
3	0.11	10	95
4	0.055	10	100
5	0.028	10	89
6	0.1	20	99.5
7	0.02	50	99.6

Mass Ratio of α-Benzoin Oxime to Molybdenum

The mass ratio of α-benzoin oxime to molybdenum affects the molybdenum recovery during the precipitation. From the stoichiometry of the chemical reaction:

$$MoO_2^{++} + 2\,C_6H_5CH(OH)C{:}NOHC_6H_5 \leftrightarrow MoO_2[C_6H_5CH(O)C{:}NOHC_6H_5]_2 + 2H^+$$

the stoichiometric mass ratio (αBZ/Mo) is 4.7. To verify the actual amount of α-benzoin oxime needed, two tests of molybdenum recovery efficiency as a function of the αBZ/Mo ratio were conducted, one at a low molybdenum concentration (10 μg/mL) and the other at a high molybdenum concentration (200 μg/mL). The test procedure is quite similar to the previous one. The results shown in Table 2 indicate that an αBZ/Mo ratio of 10 or higher is necessary for quantitative molybdenum recovery.

Table 2. Molybdenum Recovery vs. Mass Ratio of α-Benzoin Oxime to Molybdenum

Mass Ratio	% Mo Recovery	
αBZ/Mo	[Mo] = 10 μg/mL	[Mo] = 200 μg/mL
5:1	9 ± 1	33 ± 1
10:1	37 ± 1	98 ± 2
20:1	97 ± 1	100 ± 2
50:1	100 ± 2	100 ± 2
100:1	100 ± 2	100 ± 2

Concentration of Acid in Dissolver Solution

The dissolution rate of uranium metal can be increased by increasing the concentrations of either nitric or sulfuric acid or both [3]. The effect of higher acid concentrations on the molybdenum recovery during the precipitation step was, therefore, measured as a function of nitric and sulfuric acid concentrations ranging from 0.1 to 8M. The results in Table 3 show that molybdenum recovery is satisfactory when the hydrogen ion concentration is in the range of 0.5M to 4M.

Table 3. Molybdenum Recovery Efficiency as Function of Solution Acidity

[HNO_3], M	% Mo Recovery	[H_2SO_4], M	% Mo Recovery
0.1	90	0.1	100
0.5	100	0.5	100
1.0	100	1.0	98
2.0	100	2.0	95
4.0	100	4.0	93
6.0	88	6.0	98
8.0	-	8.0	8.7

Partial Neutralization of Dissolver Solution

If higher acid concentrations are needed to dissolve the uranium metal foil, one option is to partially neutralize the acid in the dissolver solution before the precipitation is done. After the dissolution, part of the acid can be neutralized by a concentrated base solution, such as the 8M NaOH used in this work. The results in Table 4 show that molybdenum recovery can remain high if the acid solution is partially neutralized by base.

Table 4. Effects of Partial Neutralization of Acidic Dissolver Solutions on Molybdenum Recovery by Precipitation Using α-Benzoin Oxime

Solution	Approximate [H^+], M	% Mo Recovery
10 mL 8M HNO_3 + 7.77 mL 8M NaOH	1	100 ± 2
10 mL 8M HNO_3 + 6.00 mL 8M NaOH	2	100 ± 2
10 mL 8M HNO_3 + 3.33 mL 8M NaOH	4	92 ± 2
10 mL 8M HNO_3 + 1.43 mL 8M NaOH	6	100 ± 2
10 mL 8M H_2SO_4 + 14 mL 8M NaOH	2[a]	94 ± 2
10 mL 8M H_2SO_4 + 10 mL 8M NaOH	4[a]	100 ± 2
10 mL 8M H_2SO_4 + 5 mL 8M NaOH	8[a]	97 ± 2
10 mL 8M H_2SO_4 + 2 mL 8M NaOH	12[a]	99 ± 2

[a] Based on 2 moles of acid per mole of H_2SO_4.

Uranium Concentration

All tests discussed above were conducted in an acid solution without the presence of uranium. The effect of uranium concentration is shown in Table 5. The uranium solutions were prepared by dissolving uranyl nitrate in sulfuric acid solutions. Four different uranium concentrations were prepared at two sulfuric acid concentrations. Molybdenum recovery was measured for (1) ^{99}Mo in the precipitate and (2) ^{99}Mo which passed through a filter. The ^{99}Mo tracer was produced by using molybdenum standard solution with a concentration of 10 mg/mL [National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 3134]. The solution was irradiated in the "Lazy Susan" facility with a thermal neutron flux of 3.4×10^{12} n/cm^2 at the University of Illinois TRIGA reactor. The reactor power was 1.5 MW, and irradiation time was 2 hours. About 1 mg of the radioactive ^{99}Mo tracer was added to each solution. A solution of 2% α-benzoin oxime in 0.4M NaOH was added to form the precipitate. After about 10 min, the precipitate was collected on the filter by using Nalgene disposable filterware (Nalgene Analytical Filter Unit, Cat. No. 130-4045). The filter material is a Triton-free cellulose nitrate membrane with a pore size of 0.45 µm. The filtration was done by using a hand-operated vacuum pump. Both the precipitate on the filter and the filtrate were analyzed for molybdenum by using a high purity germanium (HPGe) gamma-ray detector. Molybdenum recovery efficiencies ranged from 98-100% for uranium and sulfuric acid concentrations anticipated during processing (Table 5). Recovery efficiencies were measured by counting the precipitate and the filtrate. Since most of the ^{99}Mo precipitated, the activity and background were much higher for those samples. In these experiments, molybdenum recovery efficiency could be measured more precisely by counting the filtrate.

Table 5. Molybdenum Recovery Efficiencies as Function of Uranium Concentrations and Measurement Method

[$UO_2(NO_3)_2$], M	[H_2SO_4], M	%Mo Recovery Precipitate	%Mo Recovery Filtrate
0.5	1.0	100 ± 2	99.5 ± 0.1
1.0	1.0	100 ± 2	99.3 ± 0.1
1.5	1.0	100 ± 2	99.1 ± 0.1
2.0	1.0	98 ± 2	98.4 ± 0.1
0.5	2.0	-	99.1 ± 0.1
1.0	2.0	-	99.3 ± 0.1
1.5	2.0	-	99.0 ± 0.1

ALPHA DECONTAMINATION

Alpha contamination of ^{99}Mo is an important concern associated with the switch to LEU. Compared with the HEU, the initial ^{99}Mo in the LEU target has an alpha activity about three times higher because of the greater amount of ^{238}U and 50 times higher because of the additional ^{239}Pu. To achieve an alpha activity less than 10^{-7} µCi/mCi of ^{99}Mo, the decontamination factors (DF) must be ~200 for uranium and ~8000 for ^{239}Np (which decays to ^{239}Pu). The decontamination factors from uranium and neptunium are discussed below.

Uranium

Under conditions expected for Cintichem processing of an LEU target, the precipitation of the molybdenum/α-benzoin oxime salt produced a uranium decontamination factor of $\sim 3 \times 10^4$. This value would be sufficient to reach the alpha contamination limit of 10^{-7} µCi/mCi of ^{99}Mo.

Neptunium and Plutonium

Neptunium-239 is the major activation product from irradiation of ^{238}U and has a β-decay half-life of 2.35 days to Pu-239, which α-decays with a half-life of 2.4 x 10^4 years. The neptunium decontamination factor was determined by using a radioactive Np-239 tracer, produced from thermal irradiation of natural uranium.

The simulated dissolver solution was prepared by dissolving various concentrations of uranyl nitrate in 2M sulfuric acid solution. The volume of uranium solution used for each case is 10 mL. In each case, 0.1 mg of molybdenum was mixed with 2 mL of 2% α-benzoin oxime in 0.4M NaOH solution. The ^{239}Np on the filter was measured by using its 277 keV peak. As shown in Table 6, the decontamination factors, (the ratio of the ^{239}Np added to that on the filter) for all cases are about 10^4, which is lower than the 10^5 reported in previous work [4]. This DF value is higher than the required value of ~8000.

Table 6. Decontamination of ^{99}Mo from Neptunium by α-Benzoin Oxime Precipitation

$[UO_2(NO_3)_2]$, (M)	239-Np DF
0.5	0.9×10^4
1.0	1.3×10^4
1.0[a]	1.1×10^4
1.5	1.1×10^4

[a] Six times more ^{239}Np tracer solution was added.

We did not determine the decontamination factors for separating molybdenum from plutonium. However, in previous work [4], the DF for Pu-239 was measured to be 10^3 for a single precipitation, and 5 x 10^6 for two precipitations followed by a silver-coated activated-charcoal column treatment.

ADDITIONAL ACTINIDE DECONTAMINATION

We sought to increase the purity of ^{99}Mo in a modified Cintichem process by improving actinide and fission product decontamination early in the process. Traditional methods for molybdenum separation from acidic solutions, alumina columns, and hydroxide precipitation [5] were investigated and adjusted for the expected process solutions produced by the acidic dissolution of LEU foil.

Alumina columns are often used to remove ^{99}Mo from solution. The effects of uranium and hydrogen ion concentration on molybdenum adsorption were studied at ANL in 1987 [6]. It was found that the molybdenum partitioning coefficient, $K_d(Mo)$, surpassed 50 mL/g for 0.1M H^+ but fell quickly to less than 5 mL/g for 1.0M H^+ when the feed solution contained 7 x 10^{-4}M Mo. The effect of uranium concentration on $K_d(Mo)$ was similar, dropping from 25 mL/g at 0.01M U to 3 mL/g at 0.95M U with constant 1.4 x 10^{-3}M Mo and 0.5M H^+. To perform an effective column separation, the K_d value should be greater than 10 mL/g. Since the expected dissolver solution will have 2-9M H^+ and 0.73M UO_2^{2+}, addition of an alumina column after the initial dissolution of LEU foil targets was rejected because of the poor partitioning coefficients for molybdenum under the high concentrations of acid and UO_2^{2+} anticipated.

As an alternative to the alumina column, we studied hydroxide precipitation of actinides, lanthanides, and ruthenium. Simulated acidic dissolver solutions were prepared and spiked with radiotracers ^{99}Mo, ^{239}Np, and $^{152/154}$Eu. The simulants were neutralized and made alkaline by adding concentrated sodium hydroxide. Actinides and lanthanides precipitated as expected. Precipitation of uranium from dissolver simulant solutions with concentrated sodium hydroxide carried ~50% of the molybdenum from solution. Rinsing with 0.2M NaOH washed an additional 20% of the ^{99}Mo into solution, but 30% was trapped in the precipitate. Reducing the uranium concentration in the simulant by factors of 10 and 100 yielded 78% and 94% recovery of ^{99}Mo. Figure 1 shows the improvement in ^{99}Mo recovery as uranium concentration decreases from 0.73M to 0.0073M.

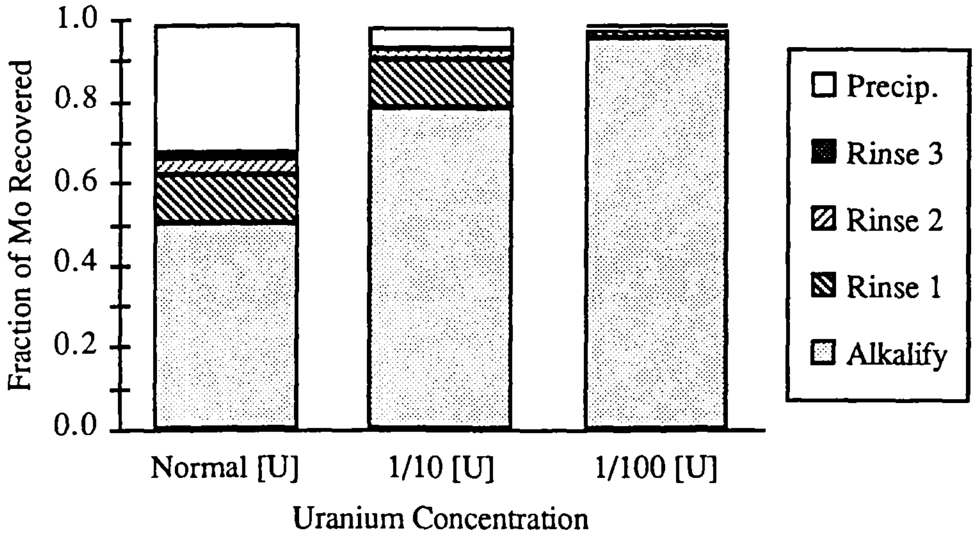

Figure 1. Increase in Molybdenum Recovery with Decreases in Uranium Concentration. Normal, 1/10, and 1/100 uranium concentrations are 0.73, 0.073, and 0.0073M U, respectively. Rinses were done with 0.2M NaOH.

Reducing the concentration of uranium significantly reduced the volume of precipitate and coprecipitation of molybdenum. The fraction of molybdenum retained in solution after increasing the pH increased from 40-50% to 79% and 95% for reductions in uranium concentration by factors of 1/10 and 1/100, respectively. Rinses easily washed the remaining ^{99}Mo from the precipitate of reduced uranium simulants, suggesting that ^{99}Mo was entrapped in the bulky uranyl hydroxide precipitate. The coprecipitation of molybdenum with uranyl hydroxide makes hydroxide precipitation unacceptable as a pretreatment process for removing α-activity.

At this point, we have not developed a suitable pretreatment step to the α-benzoin oxime precipitation. However, the results presented in the next section suggest that this step may not be necessary.

TRACER EXPERIMENT

An LEU-tracer experiment was performed to simulate the entire Cintichem process. In this experiment, a spike of irradiated LEU was added to a simulated dissolver solution, and samples were taken and analyzed for each step of the Cintichem process. The objectives of this experiment were to (1) prepare for an LEU process demonstration being planned for Indonesia, (2) validate the ^{99}Mo separation and purification process for LEU, (2) identify suitable gamma rays for determining the activity of radionuclides in the irradiated LEU, (3) compare measured fission yields and those calculated by using the ORIGEN2 code [7], and (4) determine the molybdenum yield and decontamination in each step of the process. A schematic diagram of the experimental procedures is presented in Figure 2, showing sampling points for each step.

Gamma Analysis of Irradiated LEU Isotopes

A 204-mg piece of LEU foil was dissolved in 3 mL of 3.0$\underline{M}$ HNO_3 and 2$\underline{M}$ H_2SO_4. A part of that solution was irradiated for one hour, at a reactor power of 1500 kW, which corresponds to 3.4×10^{12} n/(cm^2 s) thermal neutron flux. Following a 17-hour decay, a 0.1-mL sample of the original solution was saved for counting. The LEU tracer sample (S-1) was used to define the specific activity levels of the simulated LEU dissolver solution. After Sample S-1 was counted several times, a decay constant was fit for each gamma-ray peak. Only peaks where the measured decay constants are very close to the listed decay constants for an isotope were considered to be interference free. Thus, these peaks were used to measure the activity of corresponding isotopes. These activities of 24 isotopes were measured and compared with results calculated from the ORIGEN2 code under the test condition. The comparisons are listed in Table 7. As can been seen, measured and calculated results agree very well, except for the volatile isotopes iodine and xenon. Some isotope pairs, for example, Ba-140 and La-140, were theoretically fitted with experimental results, and agreement was also good.

Simulated Cintichem Processing

The experiment simulating the Cintichem process in a glovebox was begun. A simulated uranium dissolver solution was prepared, with concentrations of 0.75$\underline{M}$ $UO_2(NO_3)_2$ and 2$\underline{M}$ H_2SO_4. The volumes of all solutions were 1/10 of those used in the Cintichem process. Solution additions specified by the proprietary Cintichem process were made in preparation for the molybdenum precipitation, and the filtration was conducted with a disposable filter unit. The precipitate was collected on the filter, while the filtrate was saved. The precipitate was washed several times using dilute sulfuric acid. The filter with precipitate was removed from the filter unit and saved for further purification. A 1-mL sample was taken from the filtrate; this sample is designated S-2 in Figure 2.

A second, equivalent experiment was performed with another spiked simulated dissolver solution. The precipitate was formed and filtered as above. This filter sample was saved for counting activity in the precipitate; it is designated S-2(ppt) in Figure 2.

The next step was the dissolution of the precipitate. A sample was taken from the redissolved precipitate solution (designated as S-3 in Figure 2), and the filter residue [designated as S-3(res)] was saved to count any residual radioactivity. It is important to note that 100% mass balance cannot be achieved because activity could also be on the glassware used in the precipitate dissolution.

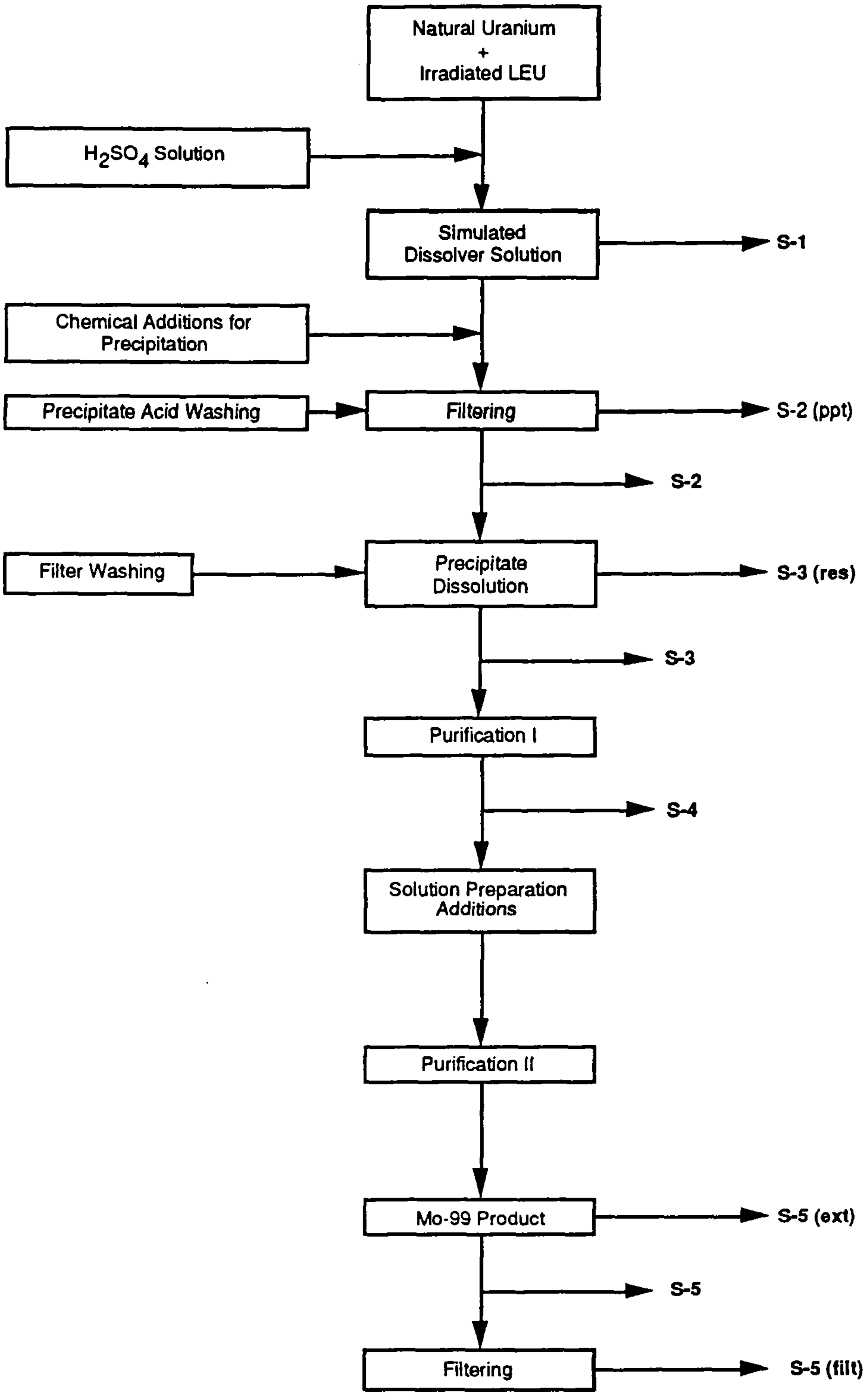

Figure 2. Schematic Diagram of the Procedures for LEU Tracer Experiment

The redissolved solution was then passed through the first molybdenum purification step. Following this procedure, a 1-mL sample was taken from the product solution (designated as S-4).

Before the second purification step, solution additions specified by the Cintichem process were made. The product of that step was sampled for counting (designated as S-5). The product solution was then passed through a 0.45-μm filter and resampled; this sample is designated as S-5(filt). The Cintichem "Quality Control Manual" contains a solvent extraction procedure that removes molybdenum from the product solution to more easily measure impurities in the product. We also performed this process on our final product. This sample is designated S-5(ext) in Figure 2.

Gamma-Analysis Results

The results of the simulated Cintichem process using LEU are presented in Table 8. The results are presented as the ratio of each impurity's activity in solution to the activity of ^{99}Mo (μCi-impurity/mCi-^{99}Mo). Because the isotopes have different half-lives (see Table 7), this ratio is decay-time dependent. Therefore, all ratios were corrected to one day after the end of irradiation. On the bottom of Table 8, the ^{99}Mo activity in each processing-step sample is also listed. The ^{99}Mo activities in the samples do not balance very well due to cumulative errors of the measurements. However, the overall ^{99}Mo yield appears be more than 90%.

Comparing the activity ratios found in the dissolver solution (S-1) to those in the filtrate (S-2) and the dissolved precipitate (S-3) shows the effectiveness of the molybdenum precipitation by α-benzoin oxime. Decontamination factors from most isotopes were ≥1000. The only isotopes not removed efficiently were iodine isotopes, where decontamination factors were in the range of 40. The S-2 precipitate shows that a fair fraction of the iodine isotopes precipitated was in the precipitate. The residue left with the filter [S-3(res)] indicates that a significant fraction of the iodine did not dissolve with the molybdenum.

The first purification step (S-4) lowered the activity of most isotopes below our detection limit. Iodine isotopes were lowered by another factor of 20. This step had no effect on the level of ^{97}Nb/^{97}Zr and ^{95}Zr contamination. The second purification step (S-5) had little effect on molybdenum purity. Filtering the sample did appear to lower the ^{97}Nb/^{97}Zr contamination. The product solution [S-5(ext)], where the ^{99}Mo had been extracted, gives a better indication of the impurity level of the final product. Reducing the ^{99}Mo by a factor of 130 considerably lowered the overall activity of the sample and allowed lower detection limits for all samples.

CONCLUSIONS AND FUTURE ACTIVITIES

This effort has met its goal of helping us to understand the function and performance of the steps in the Cintichem process. This knowledge helped to prepare for the mock demonstrations at PUSPIPTEK [8, 9] in Indonesia and provides confidence that the switch to an LEU target can be accomplished with no or minor modification to the Cintichem process. Our attempts to modify the Cintichem process by an early precipitation of uranium hydroxide were unsuccessful. However, our results with the standard Cintichem process have shown that such steps may not be necessary. The ^{99}Mo separation from uranium and its fission and activation products by α-benzoin oxime precipitation provides excellent molybdenum yield and decontamination. Standard purification processes are likely to reduce impurity levels to the required level.

Table 8. Gamma-Analysis Results of the Simulated Cintichem Demonstration on LEU Targets

Nuclide	Impurity Levels in Each Step During Processing (μCi/mCi Mo-99)[a]								
	Dissolver Soln.	Filtrate	Precipitate	Dissolved Ppt.	Ppt. Dis. Residue	Product of First Purification	Product of Second Purification	Filtered Product	Mo-Extracted Product
	S-1	S-2	S-2(ppt)	S-3	S-3(res)	S-4	S-5	S-5(filt)	S-5(ext)
Te-132	1,080	127,000	0.16	<0.13	<0.007	<0.09	<0.09	<0.09	0.005
Xe-135	345	71,900	14.1	7.86	2.40	0.19	0.06	0.06	0.018
Y-93	2,760	297,000	9.89	<2.27	0.505	<1.26	<1.19	<1.34	<0.124
Np-239	2,510	299,000	0.90	<0.74	0.033	<0.49	<0.47	<0.48	<0.048
Ce-143	2,070	235,000	0.46	0.41	0.038	<0.21	<0.2	<0.2	<0.019
Rh-105	222	32,500	0.82	<0.55	0.019	<0.38	<0.38	<0.37	0.018
Pm-151	115	11,400	0.89	<0.54	<0.0423	<0.38	<0.37	<0.37	<0.033
Zr-97	2,640	273,000	9.83	<5.99	1.56	<4.12	<3.95	<4.09	<0.340
I-133	1,060	241,000	191	28.0	27.3	1.64	1.25	1.25	0.479
Sr-91	2,860	289,000	0.65	0.32	0.066	<0.34	<0.33	<0.38	<0.017
Nb-97/Zr-97	2,860	311,000	11.6	24.8	0.913	34.7	5.53	1.18	0.755
I-132/Te-132	554	70,600	0.09	0.56	0.248	<0.08	<0.09	<0.08	0.022
Sb-127	20	2,380	0.24	<0.24	<0.016	<0.25	<0.26	<0.25	<0.018
Te-131M	117	14,400	0.68	0.48	<0.049	<0.41	<0.41	<0.42	<0.055
I-135	870	163,000	127	18.9	19.2	0.71	0.48	0.52	0.398
Nd-147	81	11,400	0.11	0.10	0.001	<0.16	<0.09	<0.11	<0.005
Ce-141	100	14,700	<0.07	<0.04	<0.003	<0.03	<0.02	<0.02	<0.003
Ru-103	48	6,700	0.61	0.50	0.037	<0.04	<0.02	<0.03	0.005
Ba-140	289	38,300	0.07	0.07	0.197	<0.17	<0.13	<0.16	<0.009
I-131	87	22,700	15.9	2.35	2.56	<0.58	<0.32	<0.4	0.051
Zr-95	60	8,490	0.70	0.66	0.025	0.59	0.52	0.50	<0.003
Nb-95	11	1,260	0.85	0.80	0.033	<0.18	<0.02	<0.01	<0.002
La-140	426	23,700	0.68	0.43	2.61	0.18	<0.15	<0.1	<0.070
	Mo-99 activity in each Processing Step (μCi)								
Mo-99	86.7	0.5	86.3	85.2	0.3	83.7	79.2	79.0	0.6

[a] Data are corrected to one day after the end of irradiation.

Table 7. Identified γ-Emitting Isotopes in Irradiated LEU. Comparison of the Activities from some Radionuclides between Measured and Calculated Results.[a]

Nuclide	Gamma (keV)	Half-life Published	Half-life Measured	Activity (Ci) Measured[b]	Activity (Ci) Calculated	Ratio[b] (M/C)	Notes
Ba-140	537	12.75 d	13.2 d	1.67E-05	1.60E-05	1.05	
Ce-141	145	32.5 d	34.2 d	5.94E-06	5.52E-06	1.08	
Ce-143	293	1.38 d	1.39 d	1.18E-04	1.13E-04	1.05	
I-131	637	8.04 d	8.15 d	5.76E-06	1.03E-05	0.56	volatile
I-132/Te-132	523	3.26 d	3.18 d	4.17E-05	3.93E-05	1.06	
I-133	530	20.8 h	21.3 h	7.81E-05	1.78E-04	0.44	volatile, no Mo-99 correction
I-135	1260	6.57 h	6.67 h	8.63E-05	2.08E-04	0.41	volatile
La-140	1596	1.678 d	-	3.65E-06	3.22E-06	1.1	fitted well from Ba-140 decay
Mo-99	739	2.75 d	2.95 d	5.98E-05	6.48E-05	0.92	
Nb-95	766	34.97 d	-	3.36E-08	3.36E-08	1.0	fitted well from Zr-95 decay
Nb-97/Zr-97	658	16.8 h	16.6 h	1.86E-04	1.69E-04	1.10	
Nd-147	91	10.98 d	11.6 d	7.81E-06	6.67E-06	1.17	
Np-239	277	2.35 d	2.39 d	1.33E-04	2.31E-05	5.76	from epithermal and fast neutrons
Pm-151	340	28.4 h	32.6 h	6.53E-06	8.85E-06	0.74	interference?
Rh-105	319	35.4 h	32.6 h	1.59E-05	1.71E-05	0.93	interf. with Nd-147
Ru-103	497	39.27 d	42.0 d	2.74E-06	2.66E-06	1.03	
Sb-127	685	3.84 d	3.58 d	1.19E-06	1.04E-06	1.14	interference?
Sr-91	556	9.5 h	9.50 h	2.11E-04	2.01E-04	1.05	
Sr-92	1384	2.71 h	2.57 h	7.26E-05	7.34E-05	0.99	short T1/2, not able to see
Te-131M	794	1.35 d	1.23 d	6.60E-06	7.34E-06	0.90	
Te-132	228	3.26 d	3.22 d	4.29E-05	3.90E-05	1.10	corrected for Np-239
Xe-135	250	9.1 h	-	1.23E-05	2.47E-04	0.05	estimated from I-135
Y-93	267	10.2 h	11.0 h	1.77E-04	2.22E-04	0.80	interference with Y-91 (58.5 d)
Zr-95	757	64.02 d	-	3.24E-06	3.35E-06	0.97	
Zr-97	355	16.8 h	16.8 h	1.60E-04	1.68E-04	0.95	

[a] The values are corrected at 12 hours after the end of irradiation.

[b] The low ratios of iodine and xenon isotopes result from volatility.

Future activities will be aimed at supporting the demonstrations being performed in Indonesia. As questions arise on means to improve processing to achieve the switch to LEU, we will perform tracer experiments to better understand the chemistry driving the separation.

REFERENCES

[1] H. Arino, H. H. Kramer, J. J. McGovern, and A. K. Thornton, "Production of High Purity Fission Product Molybdenum-99," U.S. Patent 3,799,883 (1974).

[2] H. Arino, F. J. Cosolito, K. D. George, and A. K. Thornton, "Preparation of a Primary Target for the Production of Fission Products in a Nuclear Reactor," U.S. Patent 3,940,318 (1976).

[3] B. Srinivasan and G. F. Vandegrift, "Dissolution of Uranium Metal Foil in Nitric and Sulfuric Acid Mixtures," presented at the 1994 RERTR meeting in Williamsbury, VA, September 1994.

[4] G. F. Vandegrift, D. J. Chaiko R. R. Heinrich, E. T. Kucera, K. J. Jensen, D. S. Poa, R. Varma, and D. R. Vissers, "Preliminary Investigations for Technology Assessment of ^{99}Mo Production from LEU Targets," proceedings paper for 1987 International Meeting on Reduced Enrichment for Research and Test Reactors, *Fission Molybdenum for Medical Use*, IAEA-TECDOC-515, International Atomic Energy Agency, Vienna (1989).

[5] W. T. Elwell and D. F. Wood, *Analytical Chemistry of Molybdenum and Tungsten*, Pergamon Press, New York (1971).

[6] G. F. Vandegrift, J. D. Kwok, S. L. Marshall, D. R. Vissers, and J. E. Matos, "Continuing Investigations for Technology Assessment of Mo-99 Production from LEU Targets," proceedings paper for 1987 International Meeting on Reduced Enrichment for Research and Test Reactors, *Fission Molybdenum for Medical Use*, IAEA-TECDOC-515, International Atomic Energy Agency, Vienna (1989).

[7] A. G. Croff, Nucl. Tech. 62(11983) 335.

[8] J. L. Snelgrove, G. L. Hofman, T. C. Wiencek, C. T. Wu, G. F. Vandegrift, S. Aase, B. A. Buchholz, D. J. Dong, R. A. Leonard, B. Srinivasan, and D. Wu, "Development and Processing of LEU Targets for ^{99}Mo Production--Overview of the ANL Program," these proceedings.

[9] Z. Aliluddin, A. Mutalib, A. Sukkmaria, Kardarisman, G. F. Vandegrift, D. Wu, B. Srinivasan, and J. L. Snelgrove, "Processing of LEU Targets for ^{99}Mo Production--Demonstration of a Modified Cintichem Process," these proceedings.