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PROGRESS IN CHEMICAL TREATMENT OF LEU TARGETS BY THE MODIFIED CINTICHEM PROCESS*

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PROGRESS IN CHEMICAL TREATMENT OF LEU TARGETS BY THE MODIFIED CINTICHEM PROCESS

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ABSTRACT

Presented here are recent experimental results on tests of a modified Cintichem process for producing ^{99}Mo from low enriched uranium (LEU). Studies were focused in three areas: (1) testing the effects on ^{99}Mo recovery and purity of dissolving LEU foil in nitric acid alone, rather than in the sulfuric/nitric acid mixture currently used, (2) measuring decontamination factors for radionuclide impurities in each purification step, and (3) testing the effects on processing of adding barrier materials to the LEU metal-foil target. The experimental results show that switching from dissolving the target in the sulfuric/nitric mixture to using nitric acid alone should cause no significant difference in ^{99}Mo product yield or purity. Further, the results show that overall decontamination factors for gamma emitters in the LEU-target processing are high enough to meet the purity requirements for the ^{99}Mo product. The results also show that the selected barrier materials, Cu, Fe, and Ni, do not interfere with ^{99}Mo recovery and can be removed during chemical processing of the LEU target.

INTRODUCTION

The Cintichem process for producing ^{99}Mo 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, effects of modifying the dissolver solution due to this conversion must be studied, and necessary modifications to processing must be developed. In the Cintichem process, the UO_2 in the target is dissolved in a mixture of sulfuric and nitric acid. After the target is dissolved, the solution is prepared (by the addition of several reagents) for molybdenum precipitation with α-benzoin oxime. Following precipitation, the precipitate is collected, washed, and redissolved. The redissolved molybdenum solution is then passed through two additional purification steps. It is our objective in switching 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. It is also our goal to make improvements to the process that will alleviate any economic detriment to conversion to LEU.

Argonne National Laboratory and the University of Illinois at Urban/Champaign are collaborating with the National Atomic Energy Agency (BATAN) of Indonesia to develop and demonstrate the use of LEU targets in the Cintichem process. This work is a follow-up to work on this project reported last year [3-6]. In the next few months, we plan to perform the demonstration of processing a fully irradiated LEU metal foil at the PUSPIPTEK Radioisotope Production Center in Serpong, Indonesia. During 1996, we focused on three technical areas: (1) testing the effects on

^{99}Mo recovery and purity of dissolving the LEU foil in nitric acid alone rather than in the sulfuric/nitric acid mixture currently used, (2) measuring decontamination factors for radionuclide impurities in each purification step, and (3) testing the effects on processing of adding barrier materials to the LEU metal-foil target. The results of these studies are reported below.

NITRIC ACID ALONE AS DISSOLVER SOLUTION

The primary consideration for converting the dissolver solution to nitric acid alone is facilitating waste treatment and disposal. Sulfate in the acidic waste solution from the ^{99}Mo recovery step complicates uranium recovery, waste volume reduction, and waste disposal [7]. Therefore, removal of sulfuric acid from the dissolver solution is likely to significantly reduce total processing costs. A series of experiments was performed to measure the molybdenum recovery efficiency and radioisotope decontamination over a range of nitric acid concentrations. Results were compared to earlier data for the mixed-acid dissolver solution.

Precipitation of Mo(VI) by α-benzoin oxime (α-BO) is a standard analytical method for molybdenum. The standard procedure requires molybdenum in 1M sulfuric acid [8-10]. Molybdenum precipitation is quantitative, and the precipitate contains very little impurities. In our previous tests, we found that molybdenum can be also precipitated quantitatively with α-benzoin oxime from a nitric acid solution [3,4]. However, to prove the feasibility of using the nitric acid alone as a dissolver solution, we had to verify that radionuclide decontamination of the ^{99}Mo product is not degraded by this modification. Four irradiated-LEU-tracer and several ^{99}Mo-tracer experiments were also run for determining the effects of the nitric acid concentration of the dissolver solution.

Table 1 compares the results of LEU tracer experiments using nitric-acid-alone dissolver solutions and the results for sulfuric/nitric mixtures reported last year [4]. The compositions of the simulated nitric-acid dissolver solutions were (1) 0.7M nitric acid and 1.7M uranium, (2) 5M nitric acid with 0.7M uranium, (3) 0.8M nitric acid and 0.8M uranium, and (4) 0.7M nitric acid and 1.8M uranium. To each solution was added a small volume of an irradiated solution of ~10 mg/mL LEU in 0.3M HNO_3 and 0.2M $H2SO_4$. Within the nitric acid data, no trend was evident for differences in radioisotope decontamination with solution composition. Where different values were measured for the four nitric acid experiments, a range was reported. When detection limits were all that could be measured, the lowest detection limit was reported. As can be seen from the comparison in Table 1, there is no significant effect of eliminating sulfuric acid for either molybdenum recovery or radioisotope decontamination. As reported in the past [3-5], most of the decontamination is done in the precipitation of molybdenum with α-BO, and the following purifications are polishing steps. We must note that our tracer tests only indicate chemical behavior; verification of this behavior will require full-scale demonstrations using fully irradiated uranium-foil targets.

Other experiments were performed to measure molybdenum recovery as a function of uranium and nitric acid concentrations (see Table 2). The experiments included solutions prepared from dissolving depleted uranium foil in nitric acid (#1 and 2), UO_2 in nitric acid (#3), uranyl nitrate dissolved in nitric acid (#4 and 5) or sulfuric acid (#6), and nitric acid solution with no uranium (#7). The results validate the results reported last year [3] that molybdenum recovery efficiency is high under a variety of conditions. Again, we see no loss of ^{99}Mo yield by dissolving targets in nitric acid alone.

Table 1. Comparison of Impurity Levels after Each Purification Step of the Cintichem Processing of Simulated LEU Targets Dissolved in Either Nitric Acid Alone or the Standard Sulfuric/Nitric-Acid Cocktail Solution

Nuclide	Impurity Levels, μCi/mCi-^{99}Mo					
	α-BO Precipitation		Purification 1		Purification 2	
	Nitric Alone[a]	Mixture	Nitric Alone	Mixture	Nitric Alone	Mixture
Ba-140	0.03	0.07	<0.38	<0.17	<0.42	<0.13
Ce-141	<0.12	<0.04	<0.11	<0.03	<0.12	<0.02
Ce-143	<0.3	0.41	<0.15	<0.21	<0.17	<0.20
I-131	1.6-2.4	2.35	<3.25	<0.58	<3.60	<0.32
Te/I-132	0.22-0.44	0.56	<0.55	<0.08	<0.62	<0.09
I-133	6.7-15.6	27.99	1.16	1.64	0.92	1.25
I-135	4.0-8.5	18.88	0.59	0.71	0.42	0.48
La-140	<0.04	0.43	<0.01	0.18	<0.01	<0.15
Nb-95	0.47-1.3	0.80	0.23	<0.18	<0.04	<0.02
Zr/Nb-97	5.5-274	24.79	14.57	34.65	<0.08	5.53
Nd-147	<0.6	0.10	<0.20	<0.16	<0.25	<0.09
Np-239	<0.74	<0.74	<0.33	<0.49	<0.38	<0.47
Pm-151	<0.54	<0.54	<0.26	<0.38	<0.29	<0.37
Rh-105	<0.55	<0.55	<0.26	<0.38	<0.28	<0.38
Ru-103	0.43-1.02	0.50	0.05	<0.04	0.03	<0.02
Sb-127	<0.10	<0.24	<0.15	<0.25	<0.17	<0.26
Sr-91	<0.36	0.32	<0.20	<0.34	<0.26	<0.33
Sr-92	<0.04	n.m.[b]	<0.02	n.m[b]	<0.02	n.m.[b]
Y-93	<3.70	<2.27	<0.88	<1.26	<1.10	<1.19
Zr-95	0.2-5.9	0.66	<0.08	0.59	<0.09	0.52
	^{99}Mo Recovery, %					
Mo-99	94.8-98.0	98.3	95.2	96.5	93.2	91.3

[a]Range of results of four separate tests with different nitric acid and uranium concentrations. When two or more of the results were below detection limits (< values), the lowest detection limit is presented

[b]n.m.: not measured in the experiment.

Table 2. Molybdenum Recovery by α-Benzoin Oxime Precipitations from Various Acidic Uranyl Nitrate Solutions

Solution No.[a]	Concentration, M				Mo Recovery, %
	H^+	NO_3^-	SO_4^{2-}	UO_2^{2+}	
1	0.75	2.35	0	0.8	100±3
2	5.0	6.5	0	0.75	93±3
3	0.4	4.0	0	1.8	100±3
4	0.75	4.35	0	1.8	94±3
5	0.75	2.75	0	1.0	100±3
6	1.5	2.0	0.75	1.0	100±3
7	0.75	0.75	0	0	100±3

[a]See text for description of how solutions were prepared.

DECONTAMINATION OF THE ^{99}MO FROM OTHER RADIOISOTOPES

The allowed radiochemical impurity levels in ^{99}Mo product are very low, ranging from 0.1 to 10^{-7} μCi/mCi-^{99}Mo. Therefore, each purification step must work effectively. The gamma-emitting isotopes that need to be analyzed in the ^{99}Mo product are tabulated in Table 3. By using ORIGEN2, we calculated the activities of these radioisotopes in an 18-gram LEU target at 24 hours after discharge from the Indonesian RGS-GAS reactor, following a 120-hour irradiation at full power (second column of Table 3). Columns 3 through 5 contain decontamination factors we measured in our tracer experiments for each processing step. The Cintichem process uses three purification steps: the α-BO precipitation and two polishing steps (purifications 1 and 2). The predicted impurity levels in units of μCi/mCi-^{99}Mo in the irradiated LEU target are listed in the last column. The calculations show that, except for ^{103}Ru, radioisotopic decontamination levels can be met easily. Because ^{103}Ru contamination is not a concern in the current Cintichem product from HEU targets and because substitution of LEU will not affect the fission yield, this result for ^{103}Ru may indicate a limitation of tracer experiments more than a problem with LEU substitution.

Table 3. Calculated Impurity Levels of a Fully Irradiated LEU Target and the ^{99}Mo Product[a]

Nuclide	Calculated Target Activity, Ci	Measured Decontamination Factors[b]			Calculated Product Impurity Level, μCi/mCi-^{99}Mo
		Precipitation	Purification 1	Purification 2	
Ba-140	292	>516	>162	>165	<3.6E-05
Ce-141	121	>1116	328	419	<1.3E-06
Ce-143	685	>3354	313	641	<1.7E-06
I-131	186	51	28	41	5.3E-03
I-133	628	91	35	51	6.3E-03
I-135	104	121	38	43	8.8E-04
La-140	224	>2409	>104	>149	<1.0E-05
Mo-99	697	1.04	1.05	1.08	-
Nb-95	4.7	4	>13	>9.5	<1.7E-02
Nb-97	480	11	56	1410	9.2E-04
Nd-147	119	208	>62	>59	<2.6E-04
Np-239	1610	>1770	>247	>333	<1.9E-05
Pm-151	45	103	>16	>21	<2.1E-03
Rh-105	102	>276	>34	>46	<4.0E-04
Ru-103	54	113	1.3	3.7	1.7E-01
Sb-127	13.6	>41	1.3	>10	<4.3E-02
Sr-89	65.7	-	-	-	<2.3E-07[c]
Sr-90	0.39	-	-	-	<1.4E-09[c]
Sr-91	209	>3452	235	>586	<7.4E-07
Sr-92	2.65	>2101	>71	>63	<4.7E-07
Te/I-132	464	>5083	327	657	<7.1E-07
Y-93	258	>1294	511	822	<8.0E-07
Zr-95	70	13	27	>49	<6.8E-03
Zr-97	447	17	23	>41	<4.6E-02

[a]Basis is an 18-g LEU target, 24 hours after discharge from the RGS-GAS reactor, following a 120-hour irradiation at full power.
[b]Ratio of activity in the molybdenum solution before and after treatment.
[c]Predicted from Sr-91 behavior.

EFFECTS OF BARRIER MATERIALS ON PROCESSING

Development of LEU metal-foil targets has led to the use of thin (10 μm) metal barriers between the uranium foil and the target walls [11]. Three metals (Cu, Fe, and Ni) were selected as primary candidates for the barrier material based on the basis of their physical, chemical, and nuclear properties. Physical characteristics are important to target fabrication and are discussed in reference 10. Important chemical characteristics are foil dissolution (reported in reference 12) and the effect of barrier materials on the recovery and purity of ^{99}Mo (discussed below). The nuclear properties of interest are the radioisotopes generated in the barrier during target irradiation and their activity levels, which must be removed from molybdenum during processing, as discussed below.

Table 4 shows ORIGEN2 calculations for the radioisotopes generated in Fe, Ni, and Cu barriers during LEU target irradiations in the RGS-GAS reactor. The target contains an 18-g uranium-metal foil with a 10-μm metal barrier on each side of the foil. The results of these calculations show that only a copper barrier would generate enough radioactivity to be of concern. For ^{64}Cu to be less than 0.1 μCi/mCi-^{99}Mo in the molybdenum product, its overall decontamination factor must be >3,100.

Neither the barrier materials nor their neutron-activation products are reported to interfere with the precipitation of molybdenum by α-BO [8-10]. Experiments were run to verify the noninterference of these metal ions by using solutions prepared to simulate dissolving the barrier-clad uranium foil in nitric acid. In the same experiments, we measured the amount of each barrier metal that carried with the molybdenum precipitate. Table 5 shows the results of these experiments. The molybdenum recovery was high for all experiments, as were the measured decontamination factors. It is likely that the differences in the decontamination factors are more an indication of how well the precipitate was washed in each experiment rather than chemical differences in the barrier-metal ions.

Table 4. Radioisotopes Generated from Barrier Metals during Irradiation of an LEU Metal Targets[a]

Metal	Isotope	Half-Life	Activity, mCi
Fe	Fe-55	2.73 y	37
	Fe-59	44.5 d	26
	Mn-54	312 d	1.3
	Mn-56	2.56 h	1.0
Ni	Ni-65	2.52 h	53
	Co-58	70.92 d	97
Cu	Cu-64	12.7 h	153,000
	Cu-67	61.9 h	0.6

[a]Barrier material (10 μm) on both sides of an 18-g metal foil irradiated in the RGS-GAS reactor at full power for 120 hours.

Table 5. Effects of Barrier Materials on α-BO Precipitation: Molybdenum Yield and Barrier-Metal Decontamination Factors[a]

	Cu	Fe	Ni
Molybdenum recovery (%)	99 ± 3	96 ± 3	96 ± 3
Decontamination factors	1680	258	660

[a]Solution contained 0.75M HNO_3, 1.5M $UO_2(NO_3)_2$, and the concentration of Cu, Ni, or Fe corresponding to a 10-μm barrier on either side of the uranium foil.

The decontamination factors measured for iron and nickel are more than high enough to meet impurity requirements for the molybdenum product. However, the removal of ^{64}Cu may require additional decontamination. For this reason, we tested the removal of copper by the two polishing steps; these tests showed that the overall decontamination factor for the two polishing steps should be >10,000. A combination of all three steps should therefore effectively reduce ^{64}Cu contamination to well below concern.

CONCLUSION

Testing and development activities are continuing at Argonne National Laboratory and the University of Illinois at Urbana/Champaign to support modification of the Cintichem process for use with LEU targets and to assist BATAN researchers at the PUSPIPTEK Radioisotope Production Center, who are preparing to demonstrate this process on a fully irradiated LEU target. Our collaboration with BATAN is vital to developing and validating this process. Results this year have added to the database showing that substitution of LEU in the Cintichem process will be successful and advanced our progress toward the full-scale demonstration to be done by BATAN.

Our experimental results predict that replacing the current dissolution cocktail, which contains both nitric and sulfuric acids, with nitric acid alone will not compromise the effectiveness of the Cintichem process. In our tracer experiments with this substitution, molybdenum recovery and purity were not degraded. Removal of sulfuric acid from the dissolver solution will decrease waste treatment and disposal costs and increase the stability of the disposed waste form. On the basis of measured decontamination factors from our tracer experiments, molybdenum produced from processing fully irradiated LEU targets is predicted to meet radiochemical purity limits. Its yield will be equivalent to that currently produced from HEU. Likewise, addition of barrier materials will not affect the process. A full-scale demonstration of process will take place in the near future at PUSPIPTEK.

FUTURE WORK

Future activities will be aimed at supporting the full-scale demonstrations to be performed in Indonesia.

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Progress in Chemical Treatment of LEU Targets by the Modified Cintichem Process

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OUTLINE

Background

- Goals for LEU Conversion
- Cintichem Process
- Collaborative Program with BATAN

Progress

- Effects of Barrier Materials on Target Processing
- Effects on Process of Dissolving LEU-Metal Foil with HNO_3 Alone
- Predicted Decontamination Factors for Target Processing

Conclusions

Future Work

LEU Conversion Goals

To develop modified <u>targets and separations</u> for converting current ^{99}Mo processes from HEU to LEU

To assist current and future producers to convert

To make conversion to LEU as painless as possible

- Attain same ^{99}Mo yield for LEU target and process
- Attain same product purity
- Minimize changes in target geometry
- Minimize modification to target processing
- Minimize economic costs of conversion

To address key technical issues of LEU conversion

To utilize international cooperation--working with ^{99}Mo producers

Overall Process

Target fabrication

Irradiation

Post irradiation disassembly

Target dissolution

Separation and purification of ^{99}Mo

Waste treatment and disposal

Reference HEU and LEU Targets

	HEU	**LEU**
^{235}U Enrichment, %	93	19.75
^{235}U, g	15	18.5
Total U, g	16.1	93.7
^{99}Mo yield, Ci	532	545
Total Mo, mg	9.8	10.0
^{239}Pu, μCi	30	720
$^{234, 235, 238}$U, μCi	1280	840
Total α, μCi	1310	1560

High Priority Processing Needs for Conversion to LEU

Dissolution of irradiated uranium

- Different target means modified dissolver solution
- Different dissolver design
- Dissolved uranium solution must be compatible with ^{99}Mo recovery and purification steps that follow

Initial ^{99}Mo recovery step is key to processing

- Once uranium is removed, HEU and LEU processing should be same

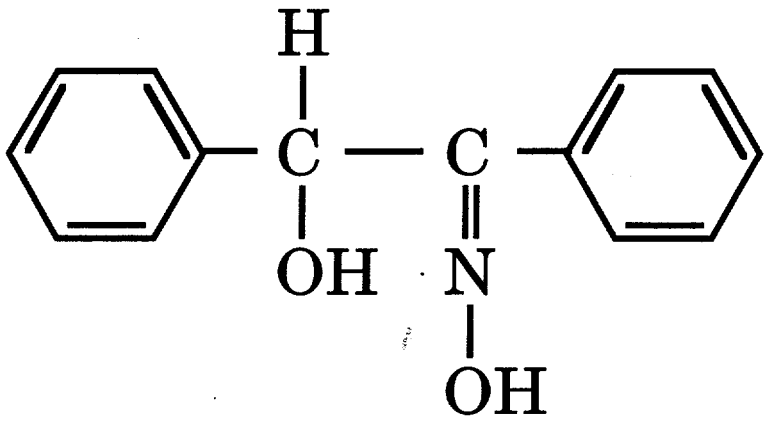
To meet product alpha contamination limits in LEU calls for

- Greater Pu reduction
- Slightly less U reduction

Cintichem Process

Current HEU Process

- Dissolution of irradiated UO_2 by a cocktail of nitric and sulfuric acids
- Recovery and initial purification of ^{99}Mo by precipitation by α-benzoin oxime



- Two additional purification steps

Modified LEU Process

- Dissolution of irradiated uranium metal foil by nitric/sulfuric acids or nitric acid alone
- ^{99}Mo recovery and purification steps need not be modified

Collaboration with BATAN

Through a BATAN/ANL agreement, PUSPIPTEK researchers are performing

- Target irradiations
- Process demonstrations

Thus far at the PUSPIPTEK Radioisotope Production Center has done demonstrations

- At full-scale
- But using tracer amounts of activity.

We are planning a full-scale demonstration on a fully irradiated LEU target

Effects of Barrier Materials on Processing

Thin (10 μm) barriers are needed between the U foil and the target walls

- To stop fission-recoil atoms
- Whose passage bonds the foil to the target walls

Nickel, copper, and iron were chosen based on their:

- Mechanical properties
- Chemical properties
- Nuclear properties

Effects of Barrier Materials on Processing

Mechanical Properties

- Ni, Cu, and Fe can all be rolled into foils
- Ni and Cu can be electroless plated

Nuclear Properties

- Ni and Fe barriers will produce negligible amounts of radioactivity during target irradiation
- A Cu barrier will produce significant gamma activity
  - Substantial decontamination is needed to meet ^{99}Mo radiochemical-purity requirements
  - Tracer tests show decontamination should be achievable

Chemical Properties

- All quickly dissolve in nitric acid with or with out sulfuric acid
- Their presence should not diminish yield or radiochemical purity of the ^{99}Mo-product

Effects of Barrier Materials on Processing Nuclear Properties

Activation-product Decay for a 2-sided, 10-μm barrier after 12-h decay

Metal	Isotope	Half-Life	Activity (mCi)
Fe	Fe-55	2.73 y	37
	Fe-59	44.5 d	26
	Mn-54	312 d	1.3
	Mn-56	2.56 h	1.0
Ni	Ni-65	2.52 h	53
	Co-58	70.92 d	97
Cu	Cu-64	12.7 h	153,000
	Cu-67	61.9 h	0.6

Decontamination Factor to meet < 0.1μCi/mCi-^{99}Mo

Barrier Metal	Element	DF Required
Fe	Fe	>1.1
	Mn	none
Ni	Ni	>1.1
	Co	>1.9
Cu	Cu	>3,100

Effects of Barrier Materials on Processing Chemical Properties

Effects of Barrier Materials on α-BO Precipitation: Mo Yield and Barrier Decontamination Factors[a]

	Cu	Fe	Ni
Mo Recovery (%)	99 ± 3	96 ± 3	96 ± 3
Decontamination Factors	1680	258	660

[a] Solution contained 0.75M HNO_3, 1.5M $UO_2(NO_3)_2$, and the conc. of Cu, Ni, or Fe corresponding to a two-sided 10-μm barrier on uranium foil.

Mo recovery is not diminished by presence of barrier metals

Contamination of ^{99}Mo is only of concern for Cu barrier

Overall decontamination factor for Cu in two polishing steps was measured to be >10,000

Therefore, contamination of ^{99}Mo product by ^{64}Cu should not be a problem

Dissolving LEU Foil by Nitric Acid Alone

Currently, HEU oxide is dissolved by a cocktail of nitric and sulfuric acid

Removal of sulfuric acid from the dissolver solution is likely to cut waste treatment and disposal costs. Sulfuric Acid

- Increases complexity of waste treatment
- Increases volume of stabilized waste
- Decreases stability of disposed waste form

We studied this alternative as a means to make conversion to LEU more attractive

Tracer studies show no loss in Cintichem process effectiveness with this substitution

Dissolving LEU Foil by Nitric Acid Alone

Comparison of impurity levels and Mo recovery after each Cintichem purification step shows (1) no effect of removing sulfuric acid and (2) no trend in impurity levels from varying concentrations of nitric acid or uranium.

Nuclide	Impurity Levels, μCi/mCi ^{99}Mo					
	a-BO Precipitation		Purification 1		Purification 2	
	Nitric	Mixture	Nitric	Mixture	Nitric	Mixture
Ba-140	0.03	0.07	<0.38	<0.17	<0.42	<0.13
Ce-141	<0.12	<0.04	<0.11	<0.03	<0.12	<0.02
Ce-143	<0.3	0.41	<0.15	<0.21	<0.17	<0.20
I-131	1.6-2.4	2.35	<3.25	<0.58	<3.60	<0.32
Te/I-132	0.22-0.44	0.56	<0.55	<0.08	<0.62	<0.09
I-133	6.7-15.6	27.99	1.16	1.64	0.92	1.25
I-135	4.0-8.5	18.88	0.59	0.71	0.42	0.48
La-140	<0.04	0.43	<0.01	0.18	<0.01	<0.15
Nb-95	0.47-1.3	0.80	0.23	<0.18	<0.04	<0.02
Zr/Nb-97	5.5-274	24.79	14.57	34.65	<0.08	5.53
Nd-147	<0.6	0.10	<0.20	<0.16	<0.25	<0.09
Np-239	<0.74	<0.74	<0.33	<0.49	<0.38	<0.47
Pm-151	<0.54	<0.54	<0.26	<0.38	<0.29	<0.37
Rh-105	<0.55	<0.55	<0.26	<0.38	<0.28	<0.38
Ru-103	0.43-1.02	0.50	0.05	<0.04	0.03	<0.02
Sb-127	<0.10	<0.24	<0.15	<0.25	<0.17	<0.26
Sr-91	<0.36	0.32	<0.20	<0.34	<0.26	<0.33
Sr-92	<0.04	n.m.[b]	<0.02	n.m[b]	<0.02	n.m.[b]
Y-93	<3.70	<2.27	<0.88	<1.26	<1.10	<1.19
Zr-95	0.2-5.9	0.66	<0.08	0.59	<0.09	0.52
	^{99}Mo Recovery, %					
Mo-99	94.8-98.0	98.3	95.2	96.5	93.2	91.3

Dissolving LEU Foil by Nitric Acid Alone

Molybdenum recovery by α-benzoin oxime precipitation is unaffected by the composition of acidic uranyl nitrate solutions

Concentration, M				Mo Recovery,
H^+	NO_3^-	SO_4^{2-}	UO_2^{2+}	%
0.75	2.35	0	0.8	100±3
5.0	6.5	0	0.75	93±3
0.4	4.0	0	1.8	100±3
0.75	4.35	0	1.8	94±3
0.75	2.75	0	1.0	100±3
1.5	2.0	0.75	1.0	100±3
0.75	0.75	0	0	100±3

Removal of sulfuric acid from the dissolver solution looks like a good idea

Full-scale process demonstrations with fully irradiated LEU targets are necessary to validate these results

Predicted Radiochemical Decontamination of ^{99}Mo Based on UIUC Tracer Experiments

Irradiated-LEU-tracer experiments have been performed separately for each purifications step

- to measure decontamination factors for each radioisotope
- to predict overall decontamination of each radioisotope

Overall decontamination factors are calculated by multiplying decontamination factors for each step

$$DF\text{-}overall_{Sr\text{-}92} = (DF\text{-}\alpha BO_{Sr\text{-}92})\,(DF\text{-}P1_{Sr\text{-}92})\,(DF\text{-}P2_{Sr\text{-}92})$$

$$DF\text{-}overall_{Sr\text{-}92} = (>2100)\,(>71)\,(>63) = >9.4\times10^{6}$$

Predicted Radiochemical Decontamination of ^{99}Mo Based on UIUC Tracer Experiments

Nuclide	Predicted Activity (Ci)	DF Values			Impurity Level μCi/mCi Mo-99
		α-BO ppt.	Purify 1	Purify 2	
Ba-140	292	>516	>162	>165	<3.6E-05
Ce-141	121	>1120	328	419	<1.3E-06
Ce-143	685	>3350	313	641	<1.7E-06
I-131	186	51	28	41	5.3E-03
I-133	628	91	36	51	6.3E-03
I-135	104	121	38	43	8.8E-04
La-140	224	>2410	>104	>149	<1.0E-05
Mo-99	697	1.04	1.05	1.08	-
Nb-95	4.7	4	>13	>9.5	<1.6E-02
Nb/Zr-97	480	11	56	1410	9.2E-04
Nd-147	119	208	>62	>59	<2.6E-04
Np-239	1610	>1770	>247	>333	<1.9E-05
Pm-151	45	103	>16	>22	<2.1E-03
Rh-105	102	>276	>34	>46	<4.0E-04
Ru-103	**54**	**113**	**1.3**	**3.7**	**0.17**
Sb-127	13.6	>41	1.3	>10.0	<4.3E-02
Sr-89*	**65.7**	**-**	**-**	**-**	**<2.3E-07**
Sr-90*	**0.39**	**-**	**-**	**-**	**<1.4E-09**
Sr-91	209	>3450	235	>586	<7.4E-07
Sr-92	2.65	>2100	>71	>63	<4.7E-07
Te/I-132	464	>5083	327	657	<7.1E-07
Y-93	258	>1294	511	822	<8.0E-07
Zr-95	70	13	27	>49	<6.8E-03
Zr-97	447	17	23	>41	<4.6E-02

* Predicted from Sr-91 behavior

Predicted Radiochemical Decontamination of ^{99}Mo Based on UIUC Tracer Experiments

Tracer experiments indicate no purity problems for the ^{99}Mo product

^{103}Ru only possible problem. However,

- ^{103}Ru not a problem in the current HEU processing
- Fission yield is the same for HEU or LEU
- Therefore, may be an artifact of tracer experiments

Full-scale demonstration on fully irradiated target is necessary to verify these tracer results

Conclusions

On the basis of our tracer experiments, ^{99}Mo yield and purity for Cintichem processing won't be degraded by

- Substitution of LEU for HEU
- Addition of Ni, Cu, or Fe barriers in LEU targets
- Removal of sulfuric acid from the dissolver solution

Full-scale demonstration on a fully irradiated LEU target is necessary to verify these findings

Future Work

A demonstration should be performed in Indonesia in the next few months.