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DEMONSTRATION OF ^{99}MO PRODUCTION USING LEU METAL-FOIL TARGETS IN THE CINTICHEM PROCESS—1999*

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ABSTRACT

In March and September 1999, demonstrations of the irradiation, disassembly, and processing of LEU metal foil targets were performed in the Indonesian BATAN PUSPIPTEK Facilities. These demonstrations showed that (1) irradiation and disassembly can be performed so that the uranium foil can be easily removed from the target body, and (2) with only minor changes to the current process, the LEU foil can produce yield and purity of the ^{99}Mo product at least as great as that obtained with the HEU target. Further, because of these modifications, two hours are cut from the processing time, and the liquid waste volume is reduced. Results of these demonstrations will be presented along with conclusions and plans for future work.

INTRODUCTION

The Indonesian National Nuclear Energy Agency (BATAN) is currently producing ^{99}Mo from neutron-irradiated HEU-UO_2 targets in the Radioisotope and Radiopharmaceutical Production Centre at PUSPIPTEK, Serpong, Indonesia. The chemical procedure that is used to recover and purify the ^{99}Mo is the Cintichem process. Cintichem (a subsidiary of Medi-Physics Inc./Hofmann La-Roche) employed the process until 1989 at their reactor facilities in Tuxedo, New York. Now, the proprietary rights for the process rest with the United States Department of Energy (DOE). Sandia National Laboratories in Albuquerque, New Mexico, had planned to begin production of ^{99}Mo by this process in 1999, but a funding shortfall has put the schedule on hold. BATAN uses the process under a licensing agreement.

A collaboration is continuing between BATAN and Argonne National Laboratory (ANL) under the aegis of the RERTR (Reduced Enrichment for Research and Test Reactors) program to develop means for ^{99}Mo production using LEU-metal foil targets. Earlier work has been reported at previous RERTR meetings [1-10]. This paper provides a progress report on the process-demonstration phase of our collaboration. A second paper at this conference reports our progress in the design, fabrication, and irradiation of a new annular target [11]. By running a

series of demonstrations using LEU-foil targets with a slightly modified Cintichem process, we are able to (1) gain experience in the processing with LEU and (2) show that purity and yield of ^{99}Mo are not compromised by conversion. Moreover, as a result of the new, easily fabricated target, significantly shorter processing times due to dissolving uranium metal rather than uranium oxide, and other process improvements, conversion to LEU now may make economic sense.

EXPERIMENTAL

Two series of targets were fabricated at ANL for irradiation in Indonesia during March and August/September 1999. The earlier targets were the tapered design discussed in previous years [8-10], and the results of irradiation are summarized in another paper at these proceedings [12]. Targets of the new design were irradiated in August/September [11]. All targets designated for processing were irradiated in the RSG-GAS reactor for ~120 hours at 15 MW reactor power. Most of the targets were irradiated to evaluate irradiation and disassembly performance, but three targets were processed for ^{99}Mo recovery.

The demonstrations were performed in the same hot cell as HEU processing is normally done. Except for the dissolver, all equipment was identical for HEU and LEU processing. Except for (1) the dissolution step and (2) elimination of sulfuric acid from all process solutions, all processing steps and reagents are identical. Samples of process solutions were collected during processing and analyzed by gamma spectrometry to measure ^{99}Mo yield and purity from step to step.

LEU-FOIL PROCESSING

Foil Extraction and Dissolution

The irradiation targets are essentially two concentric tubes with the uranium foil sandwiched between them. The uranium foil has a Zn- or Ni-electroplated fission-recoil barrier (10- to 15-μm thickness) that prevents its bonding to the target walls during irradiation. Foils are extracted from the targets by cutting off both ends of the concentric tubes and separating the inner and outer tubes. If all goes well, the foil should be extracted from the target in 15 min. In addition to its primary purpose of preventing bonding of the uranium foil to the target tubes, the fission-recoil barrier holds fission gases inside the uranium until it is dissolved.

Once the foil is placed inside the dissolver (Fig. 1), the dissolver is evacuated to test for leaks, and then 40 mL of nitric acid is added through a septum. The concentration of nitric acid is set by the mass of uranium and fission barrier, the stoichiometries of the dissolution reactions, and the requirement to have the hydrogen concentration be ~1.5 M following dissolution. Typically, for the 8-9 g uranium foils we have used in our demonstrations, the initial concentration of nitric acid is 6 M. Following addition of nitric acid, the dissolver is placed in a heating rotation rig. Dissolution is completed in ≤10 min as observed by the pressure inside the dissolver first rising then falling as the exothermic reaction concludes. Because of three modifications to the Cintichem process due to dissolving LEU metal rather than a uranium-oxide coating, two hours are cut from the processing time. These modifications are (1) the need to trap

fission gases from the target before dissolution is eliminated, (2) dissolution of a uranium-metal foil is much faster than dissolution of the UO_2 coating from a target tube, and (3) dissolution volumes are considerably less. Details of dissolution and the dissolver were presented in earlier meetings [1, 2, 4, 8, 10]. Following dissolution, the gas in the dissolver (mostly NO and fission gases) is evacuated using a liquid-nitrogen-cooled sorption pump. Then, the solution is transferred from the dissolver, and a rinse solution is added and then combined with the dissolver solution. All yields are based on assuming this solution contains 100% of the molybdenum.

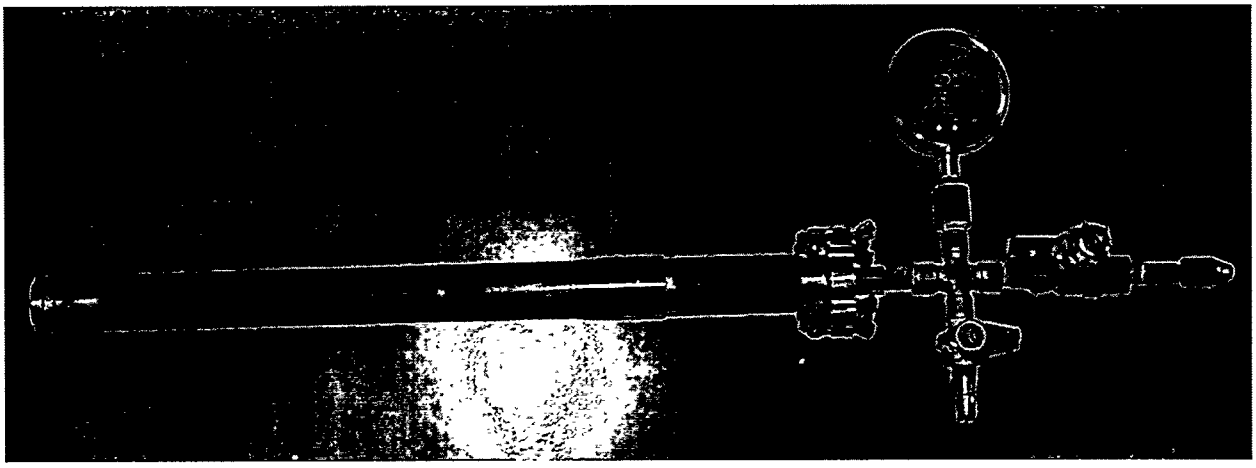

Figure 1. Photograph of ANL Dissolver

Initial Molybdenum Recovery Step

Molybdenum is recovered from the uranium solution by precipitation with alpha-benzoin oxime (ABO). This precipitation is highly selective and is a standard method used in quantitative analysis for molybdenum [13,14]. Earlier work reported at these conferences has shown that, given proper washing and moving quickly to avoid radiation damage to the ABO, high yield and decontamination are possible for this step. It is likely that the lower feed volume and, therefore, higher concentrations of constituents (due to a lower dissolver-solution volume) facilitate this separation. Earlier studies have shown that increased uranium concentrations do not compromise this separation.

Further ^{99}Mo Purification Steps

Following the precipitation, the following steps (Mo-ABO dissolution and two column separations) are identical for HEU and LEU targets. Gamma measurements of yield and decontamination for each step show them equivalent for HEU and LEU target processing.

RESULTS

The overall ^{99}Mo yield for LEU-foil processing has consistently been ~79% overall.[1] The yield for the HEU runs that we have monitored has been ~65%. The difference, as stated above, appears to be in the initial Mo-ABO precipitation step. Other factors may also be

[1] These yields were calculated from the total ^{99}Mo measured in the dissolver solutions.

important. For example, the LEU process has a better control of the acid concentration following dissolution. Based on earlier studies that showed that the Mo-ABO precipitation was quantitative between 0.5 and 2 M HNO_3, we calculate the amount of nitric acid added to the dissolver to produce 1.5 M nitric acid following foil dissolution. Another factor, although hard to quantitate, is the greater care that may be taken in an R&D activity vs. production.

Figure 2 shows the loss of ^{99}Mo throughout the process. The overall loss was ~21%. Because the volumes of the process solutions are estimated to ±5%, there are significant errors in calculating yield at each step. Volume estimates are on the basis of process knowledge and the total volumes added to the process step.

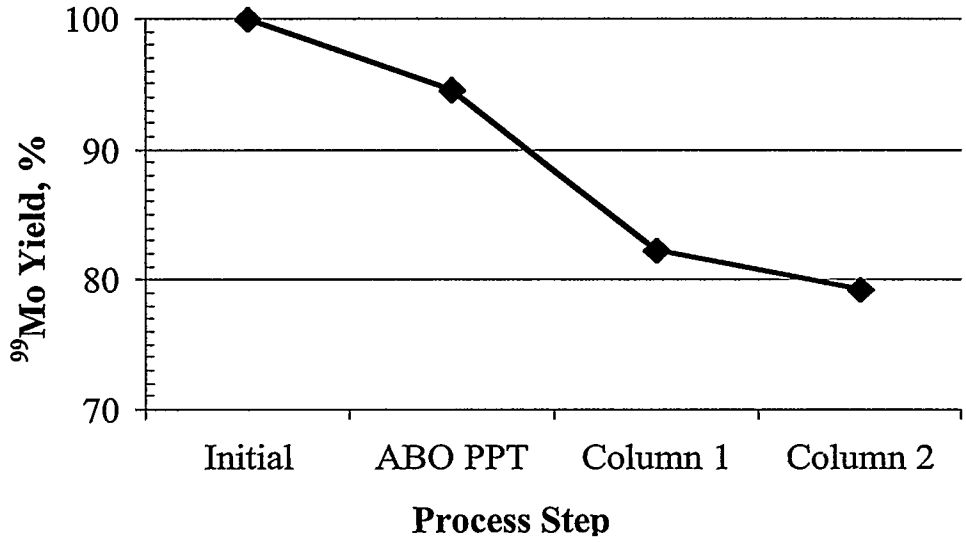

Figure 2. Typical Yield of ^{99}Mo vs. Processing Step in LEU-Modified Cintichem Process

Figure 3 shows the decontamination of the ^{99}Mo from isotopes of iodine. Figure 4 shows the decontamination from a variety of other isotopes. Note that the impurity levels in the product sample (that effluent of column 2) are well known. An extraction process quantitatively separates radioiodine from the bulk of the ^{99}Mo, making accurate analysis possible. Likewise, another extraction process removes the bulk of the molybdenum from the other isotopes to measure their contamination levels. On the other hand, in the intermediate samples, the orders-of-magnitude higher ^{99}Mo and ^{99m}Tc activities (and the many possible interference peaks they possess) make determination of impurity activities tenuous and inaccurate. Table 1 shows potential interferences from ^{99}Mo and ^{99m}Tc in determining impurity activities. (The gamma spectrum contains 42 peaks for ^{99}Mo and five for ^{99m}Tc, as well as four potential summation peaks from these isotopes [15].) When the activity levels of ^{99}Mo and/or ^{99m}Tc are extremely high, the summation peak at 281 keV can interfere with the 277.6 keV ^{239}Np peak. In addition, a ^{99}Mo summation peak at 321.6 keV can interfere with the 318.9 keV ^{105}Rh peak. Even in the final product analysis, extraction of 99.9% of the molybdenum may not be sufficient for accurate ^{140}Ba or ^{105}Rh analyses. Table 1 shows the extreme potential for interferences at high ^{99}Mo levels. These difficulties aside, based on these results, the ^{99}Mo from this demonstration would have been an acceptable feed for a technetium generator.

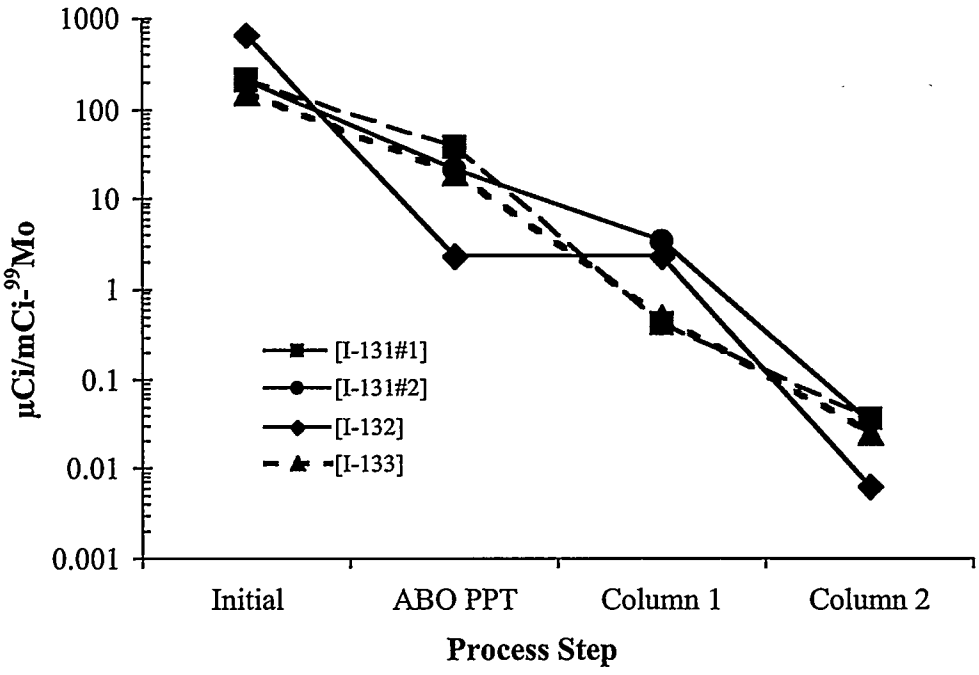

Figure 3. Decontamination of ^{99}Mo from Radioiodine in the LEU-Modified Cintichem Process

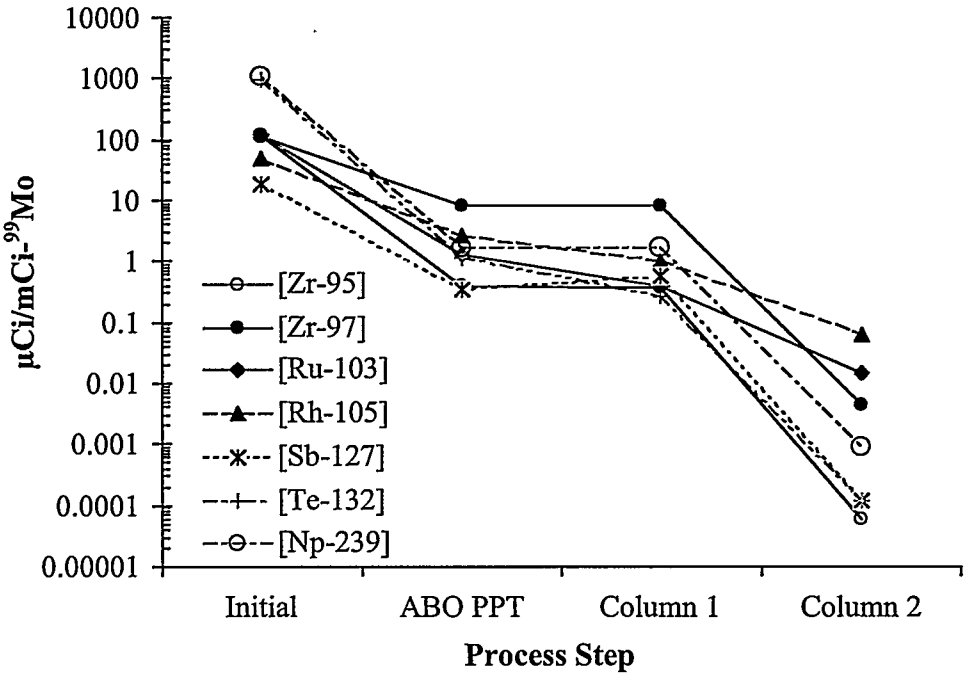

Figure 4. Decontamination of ^{99}Mo from ^{239}Np and Various Fission Products by the LEU-Modified Cintichem Process

Table 1. Extent of Interference from ^{99}Mo and ^{99m}Tc Peaks on Radioisotopic Impurities Measured by Gamma Counting

	Impurity		**^{99}Mo**					**^{99m}Tc**				
Isotope	Peak Energy, keV	Intensity, %	Peak Energy, keV	Intensity, %	Δ Energy, keV	Intensity Ratio	Potential for Interference	Peak Energy, keV	Intensity, %	Δ Energy, keV	Intensity Ratio	Potential Interference
Nd-147	91.1	2.79E+01	-	-	-	-	none	89.6	2.60E-03	1.5	1.1E+04	yes
Ce-141	145.4	4.84E+01	140.511	4.60E+00	4.9	1.1E+01	yes	140.511	8.91E+01	4.9	5.4E-01	high
	145.4	4.84E+01	-	-	-	-	none	142.63	1.90E-02	2.8	2.5E+03	yes
Te-132	227.9	8.82E+01	-	-	-	-	none	232.8	8.80E-06	-4.9	1.0E+07	none
Rh-105	318.9	1.92E+01	319.8	6.30E-03	-0.9	3.0E+03	extreme	322.4	9.90E-05	-3.5	1.9E+05	none
	318.9	1.92E+01	321	6.80E-03	-2.1	2.8E+03	high	-	-	-	-	none
Zr-97	355.4	2.38E+00	352.9	2.50E-03	2.5	9.5E+02	high	-	-	-	-	none
I-131	364.5	8.12E+01	366.5	1.16E+00	-2.0	7.0E+01	extreme	-	-	-	-	none
I-133	529.9	8.63E+01	528.788	5.42E-02	1.1	1.6E+03	yes	-	-	-	-	none
Ba-140	537.3	2.44E-01	537.79	4.40E-02	-0.5	5.5E+00	extreme	-	-	-	-	none
Zr-95	756.7	5.49E+01	761.774	1.10E-03	-5.1	5.0E+04	very low	-	-	-	-	none
Nb-95	765.8	1.00E+02	761.774	1.10E-03	4.0	9.1E+04	low	-	-	-	-	none

CONCLUSIONS AND FUTURE WORK

The substitution of LEU for HEU is viable. Several conclusions come from the BATAN process demonstration activities. Our method for LEU substitution:

- Provides the specified purity of the ^{99}Mo from gamma-emitting impurities.
- Provides a higher yield of ^{99}Mo due to a decrease of 1.5 to 2.0 hours in the processing time and potentially higher processing efficiency.
- Cuts waste-treatment and disposal costs by less liquid waste per target and elimination of sulfuric acid.

Future demonstrations will provide additional proof of these conclusions and provide alpha-decontamination data. We will assess additional process modifications to improve yield. Thus far, target disassembly has been performed in a hot cell located in the BATAN Radio-metallurgical Installation; ultimately, targets will be disassembled in a hot cell in the Radioisotope and Radiopharmaceutical Production Centre, where the ^{99}Mo is processed. Demonstrations will continue during 2000. At that point, a decision will be reached on the need for further activity.

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