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Indonesian National Atomic Energy Agency
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Argonne National Laboratory
Argonne, Illinois, U.S.A.

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ABSTRACT

The irradiation, disassembly, and processing of two full-scale low-enriched uranium (LEU) targets were recently demonstrated by personnel in the BATAN PUSPIPTEK Facilities (Serpong, Indonesia). Targets were fabricated at Argonne National Laboratory (Argonne, IL, U.S.A.) and shipped to PUSPIPTEK. The processing was done by nearly the same procedure used for the production of ^{99}Mo from high-enriched uranium (HEU) targets. The BATAN Radioisotope Production Centre produces ^{99}Mo using the Cintichem process by first dissolving the uranium in an acid cocktail; three proprietary separation steps recover the ^{99}Mo and purify it from other components of the irradiated uranium. Processing of LEU-metal targets is nearly identical to that used for HEU-oxide targets except (1) a separate dissolver is required and (2) the dissolution cocktail is nitric acid alone rather than a nitric/sulfuric acid mixture. The demonstrations went smoothly except for problems with sampling and gamma analysis to assess product purity. Foils could be removed from targets fabricated from zirconium and/or 304 stainless steel, and processing produced an equivalent yield of ^{99}Mo/^{235}U to that of the HEU target.

INTRODUCTION

The Badan Tenaga Atom Nasional (BATAN) is currently producing ^{99}Mo from neutron-irradiated HEU-UO_2 targets in the Radioisotope Production Centre (RPC) 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, and Sandia National Laboratories will begin to produce ^{99}Mo by this process in 1999. BATAN uses the process under a licensing agreement.

A collaboration is underway between BATAN and Argonne National Laboratory (ANL) under the aegis of the RERTR (Reduced Enrichment for Research and Test Reactors) program to carry out R&D and demonstration on the production of ^{99}Mo using LEU-metal foil targets. This paper gives the results of a continuation of earlier work reported at previous RERTR meetings [1-9].

EXPERIMENTAL

Six new targets were fabricated at ANL for irradiation in Indonesia; two other targets were already at PUSPIPTEK from an earlier shipment. All eight targets were irradiated in the RSG-GAS reactor for either 112 or 120 hours at 15 MW during August 1998. Two targets were processed for ^{99}Mo recovery. The others will undergo metallographic examination.

Fabrication of Targets

A schematic of the target is shown in Fig. 1. In this target, uranium foil is sandwiched between two concentric cylinders that are capped at the ends. The targets irradiated in Indonesia are considerably shorter than the conventional Cintichem target, and two targets are irradiated in tandem. One of these targets could hold up to 30 g of 130-µm uranium-metal foil. A target of the same geometry as a full-size Cintichem target could contain up to 100 g of 130-µm foil--or more if the foil were thicker. The targets from the August 1998 irradiation are described in Table 1.

The uranium was adjusted by the addition of 450-ppm iron and 1000-ppm aluminum in order to induce a fine-grain structure. The uranium foils in the new targets were given an improved beta-quench heat treatment, which is described elsewhere [10]. As discussed in earlier publications[7-9], fission-fragment barriers are necessary between the uranium foil and the target walls to prevent bonding of the foil to the walls during irradiation. To prevent fission fragments from passing completely through the fission barrier, nickel or zinc barriers must be >7-µm thick. An aluminum barrier must be >14-µm thick. Six of the targets prepared for irradiation in the RSG-GAS reactor were wrapped in thin foils of Zn, Ni, or Al. The other two fission barriers were prepared by electroplating nickel or zinc onto the uranium foil[10]. Nickel and zinc can be dissolved by nitric acid; zinc and aluminum can be dissolved by sodium hydroxide. The outer cylinder in all targets was fabricated from zirconium; the material for the inner target wall was zirconium, 304 stainless steel (SS), or aluminum.

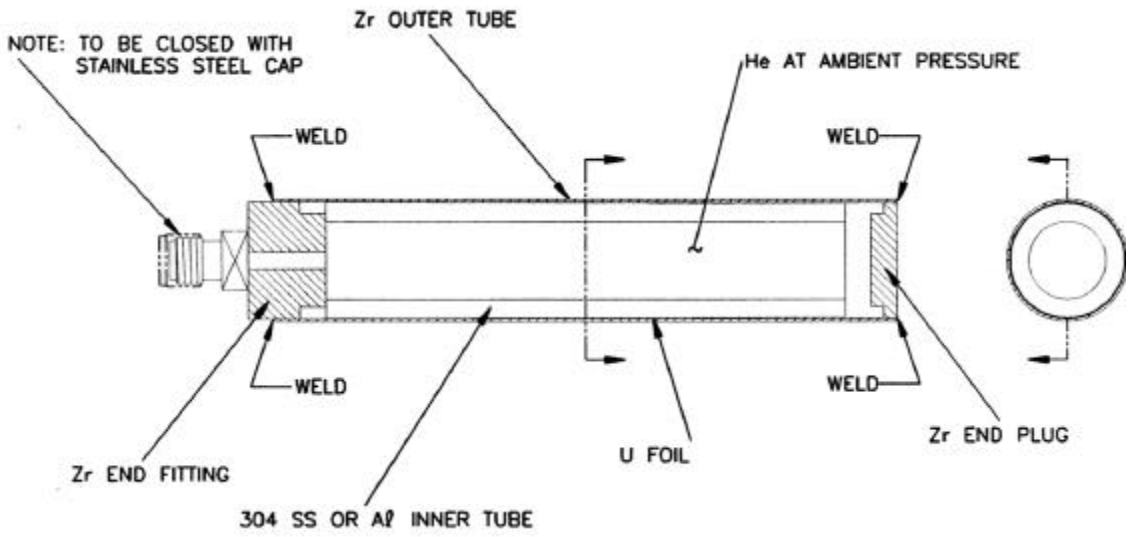


Figure 1. Schematic Diagram of Uranium Metal-Foil Target

Table 1 Characteristics of LEU Metal-Foil Targets Irradiated in the RSG-GAS Reactor during August 1998[a]

Target No.	Inner Wall	Barrier		Inner Tube Extractable?	Foil Removed?
		Material	Thickness, µm		
1	304 SS	Zn foil	15	Yes	Yes
2	304 SS	Ni foil	15	Yes	Yes
3	304 SS	Zn plate	17[b]	Yes	Yes
4	304 SS	Ni plate	11	Yes	Yes
5	304 SS	Al foil	23	Yes	No
6	Al	Al foil	23	No	-
7	Zr	Zn foil	15	Yes	Yes
8	Zr	Zn foil	15	Yes	No

a. All targets had an outer cylinder wall of zirconium.
b. By weight, calipers gave a thickness of 21 µm. The plating density of zinc on the uranium foils has always been lower than theoretical.

Disassembly of Targets

After cooling for eight hours, the targets were shipped from the reactor to the BATAN Radiometallurgy Installation (RMI) for disassembly. Disassembly consisted of cutting off both ends of the target and pushing the inner tube, and uranium foil, from the outer tube. A slight taper in the cylinders facilitates this operation. The results of disassembly are presented in Table 1. All inner tubes were extractable except for the target with an aluminum inner tube, where we believe that the combination of high temperature from decay heat and the large thermal expansion coefficient for aluminum resulted in a tight mechanical fit. A larger taper might solve this problem. The foils were easily removed from five of the seven targets that could be disassembled. The other two foils appeared to be stuck in the outer tubes. However, since all of our experience has shown that foils with fission-fragment barriers do not bond to zirconium tubes, we believe these tubes may be only mechanically stuck. Further investigation is underway at RMI. Figure 2(A) is a photograph of the Zn-foil target partly removed from the outer cylinder of the target; Fig. 2(B) shows the Ni-plated foil almost completely out of the outer cylinder.

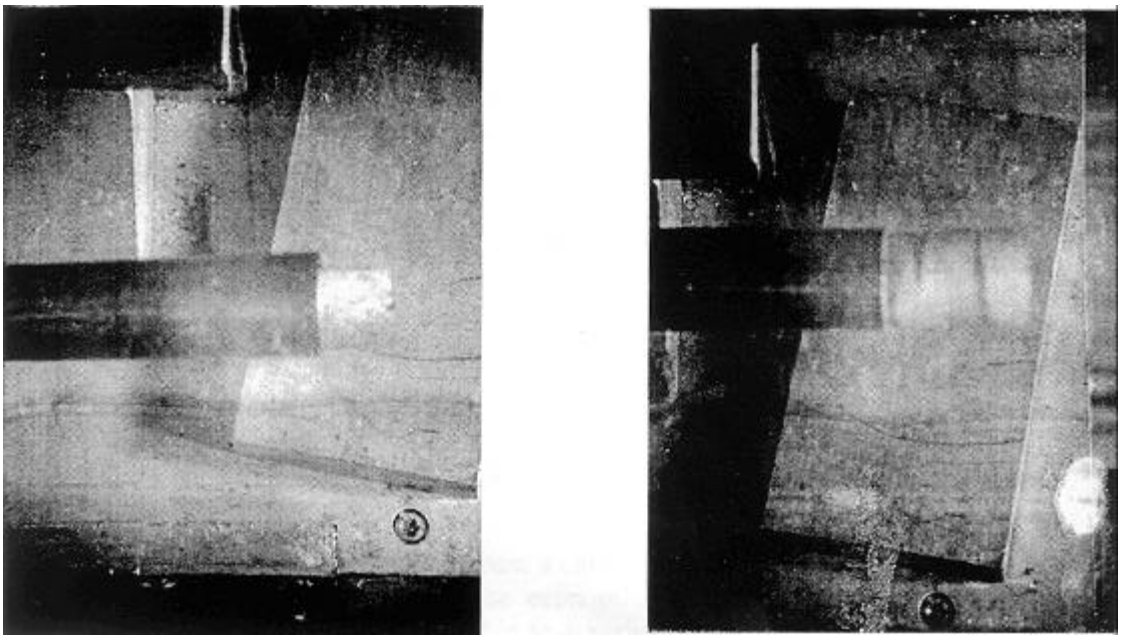
Uranium foils with nickel fission-barriers were springy and easily handled. Uranium foils with zinc fission-barriers were brittle—especially the one with the electrodeposited zinc. The electroplated foil was so brittle that handling it was difficult, and it cracked into pieces during handling. Since discovering this problem, we have been testing the variation of the conditions for zinc plating to form higher-density, bright zinc plates. We will test this new procedure in future irradiations to see if this decreases the brittleness of the zinc-plated foils.

Foil Dissolution

Following target disassembly, one target per week was shipped from RMI to the RPC for processing. Eventually, disassembly will also be performed in the RPC. The foils were dissolved in 40 mL of 6M nitric acid in a closed dissolver. Conditions were developed and tested in earlier work [1,2].

The dissolver used (see Figure 3) was an improved design over that used in the past. Changes were made to reduce the dissolver weight so that it can be handled easily by remote manipulators in a shielded-cell facility. The changes to the dissolver included (1) wing nuts in place of bar knobs, (2) smaller O-ring diameters, (3) smaller eye bolts, (4) smaller support plates for each eye-bolt hinge, (5) a pin and a retainer ring in place of the nut and bolt used as a hinge for each eye bolt, (6) reduced wall thickness for the foil-support insert in the dissolver, (7) smaller flange diameter, (8) reduced flange thickness, (9) one Viton O-ring that seals under both vacuum and pressurized operation, and (10) a protective sleeve near the mid-section of the dissolver body so that the screw on the rotation/heating jig will not scar the dissolver body. The dissolver weight was reduced from 4.3 lb (1.9 kg) to 1.8 lb (0.8 kg).

Figure 4 shows the dissolution profile (pressure buildup vs. time) for the irradiated uranium foil with the nickel-foil fission barrier. The profile for the Zn-plated uranium foil was nearly identical. The maximum pressure was predictable from earlier work based on dissolving 9.1 g of uranium and 1 g of nickel. The pressure peak is due to heating of the constituents caused by heat released in the reaction between the metals and nitric acid.



(A) (B)

Figure 2. Photographs of Irradiated Uranium Foils Partially Removed from the Outer Cylinder of the Target: (A) Foil with a Zinc-Foil Barrier and (B) Foil with Electroplated Nickel Barrier

Once the metals are dissolved, the temperature (and, consequently, the pressure) is decreased to that controlled by the heating unit—103°C. Two moles of NO gas are released for every mole of uranium metal dissolved and 2/3 mole for every mole of nickel or zinc dissolved.

The next steps in processing are to allow the dissolver to cool and then to evacuate the dissolver gases by connecting it to an evacuated cold trap (liquid nitrogen). This was the only part of the operation that did not go according to plan. For unknown reasons, the cold trap did not evacuate the dissolver completely. In the standard HEU process, a less-than-atmospheric pressure is measured in the dissolver/cold-trap system. Earlier experiments with the cold trap convinced us that it could accommodate the greater gas release of metal dissolution. In the present demonstrations, more than one contact with a cold trap was necessary to bring down the pressure. Even then, the addition of more permanganate than normal was necessary in a subsequent processing step—presumably due to more than normal amounts of NO being present in the dissolver solution following its removal from the dissolver. Work will be performed in 1999 to (1) understand the capacity and operation of the cold trap and (2) solve this problem.

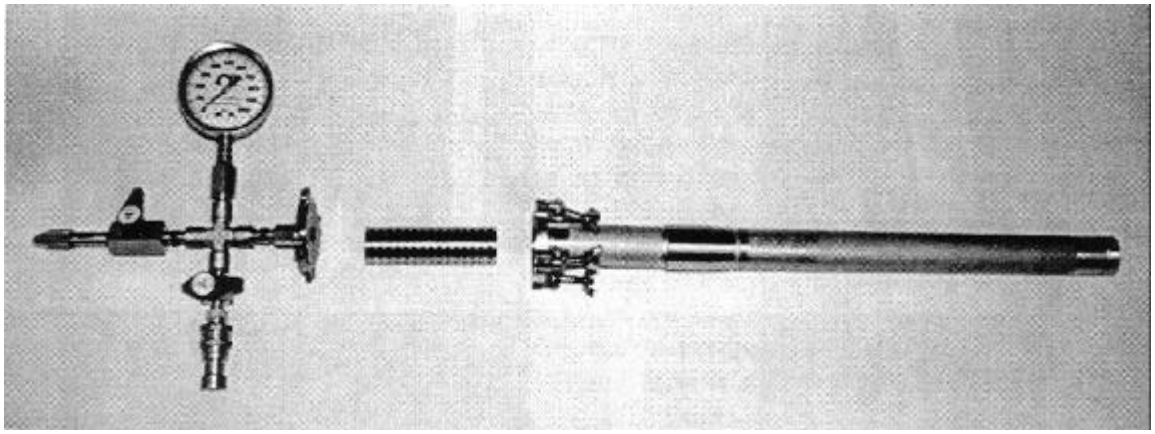


Figure 3. New, Lightweight, Closed Dissolver for Uranium Foils

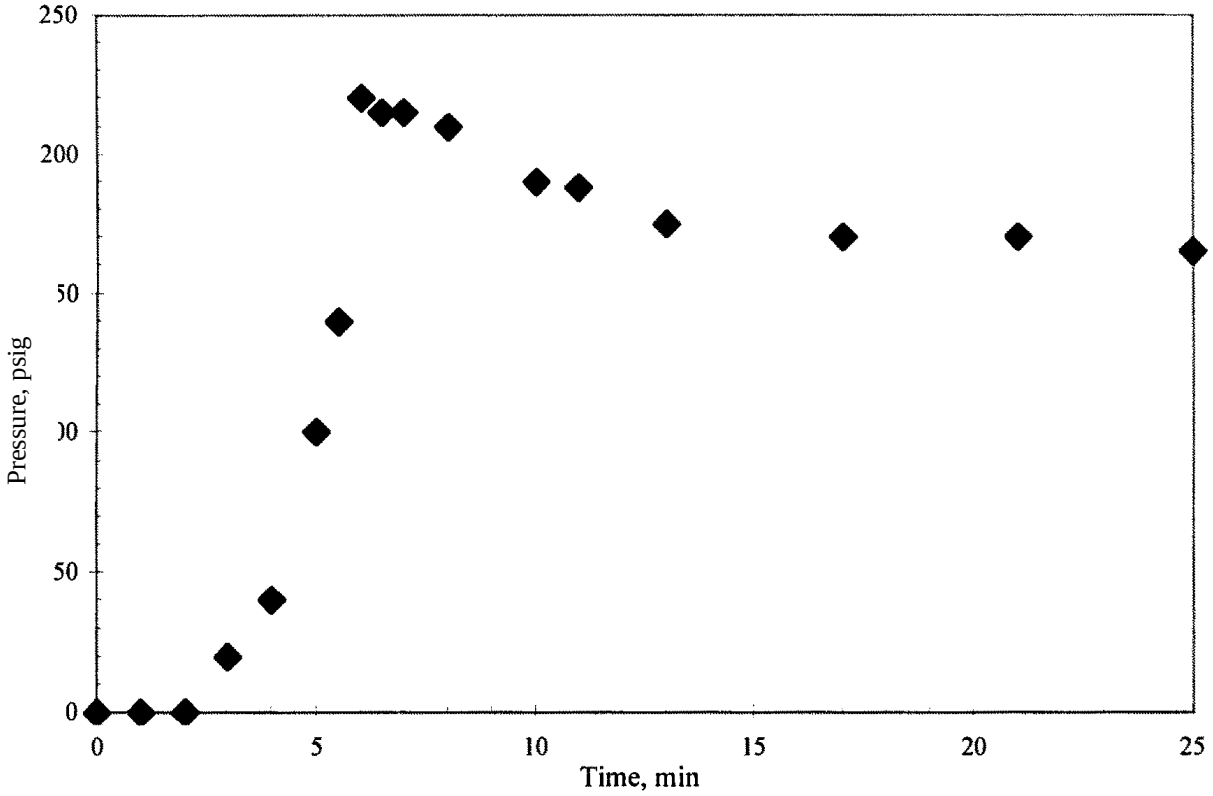


Figure 4. Pressure vs. Time for Dissolution of the Irradiated Uranium Foil with the Nickel-Foil Fission Barrier

Mo-99 Recovery and Purification

Except for requiring 20 mL rather than the typical 3-4 mL of the $KMnO_4$-reagent solution, the molybdenum recovery and purification process followed the typical behavior of HEU-target processing. With one exception, the solutions and solids were the same colors as those from processing HEU targets. (The exception was the dissolver solution for the irradiated uranium foil with a nickel barrier; this solution was noticeably green, due to the presence of nickel, rather than yellow.) All steps appeared to function as expected. Counting a fraction of the product solution in a proportional counter showed the ^{99}Mo yield to be what would be expected from a typical HEU target with the same ^{235}U content. However, problems with sampling and gamma analysis did not allow us to determine product purity. Alpha-contamination analysis has not yet been completed.

CONCLUSIONS AND FUTURE WORK

Although the demonstrations were highly successful. The following summarizes where we are and where we are going:

- The uranium-foil target has moved from a conceptual design to a reliable prototype. The next step is to provide a design that will (1) be economical to produce and (2) have the flexibility to be manufactured in geometries applicable to a variety of irradiation requirements. Future targets will likely be fabricated with zirconium or aluminum on both inner and outer walls. Doing so should aid in meeting the goals of economy and flexibility. Flexibility will also include creating an open-center target that allows both inner- and outer-wall cooling during irradiation.

- Target disassembly is still more awkward and time-consuming than necessary; improved methods for disassembly will also be designed into the next generation of targets.

- The nickel fission-fragment barrier appears to be ideal for acid dissolution. Zinc fission-fragment barriers have the advantage that the target could be used for processes that begin by either acid dissolution or alkaline digestion of irradiated uranium. However, the problem of foil brittleness needs to be addressed. For a base-side fission barrier, zinc has the advantage over aluminum in that it (1) can be electroplated onto uranium and (2) can be used for both acid- and base-side processes. However, aluminum foil is inexpensive and readily available.

- Collecting of fission and reaction gases by the standard Cintichem cold trap was the only verified problem during processing of the LEU foil targets. Studies will be completed early in 1999 to better understand the function of the cold trap and solve this problem.

- Before the next LEU demonstration, the gamma detector in the RPC will be repaired, and quality-control procedures will be updated to assure no future loss of data. The alpha-analysis method developed by A. Mutalib, during a Visiting-Scientist appointment at ANL, will be used to measure alpha contamination of the ^{99}Mo as it proceeds through the modified Cintichem process[10].

The next in a series of LEU-processing demonstrations is scheduled to be run at PUSPIPTEK in the Spring of 1999. Indonesian personnel are in the process of developing their skills in rolling and electroplating uranium foil. In the future, demonstrations will be run with targets prepared at PUSPIPTEK.

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