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PRODUCTION OF MO-99 FROM LEU TARGETS ACID-SIDE PROCESSING

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DISCLAIMER

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ABSTRACT

During 2000, additional targets of the new annular design containing low-enriched uranium (LEU) foils were irradiated in the Indonesian RSG-GAS reactor. This new design significantly decreases the fabrication target cost. This irradiation allowed us to compare the irradiation performance of several batches of LEU foil. We also processed one of the irradiated foils to recover ^{99}Mo using a slightly modified Cintichem process. Finally, we measured some important physical properties of uranyl nitrate solutions (i.e., density and solubility), which will be useful in future efforts to further increase the amount of uranium that can be processed by the Cintichem process.

INTRODUCTION

To reduce nuclear-proliferation concerns, the U.S. Reduced Enrichment for Research and Test Reactors (RERTR) Program is working to reduce the use of high-enriched uranium (HEU) by substituting low-enriched uranium (LEU) fuel and targets. Low-enriched uranium contains <20% ^{235}U. Currently, most of the world's supply of ^{99}Mo is produced by fissioning the ^{235}U in HEU targets, generally 93% ^{235}U. Targets for the production of ^{99}Mo are generally either (1) miniature Al-clad fuel plates or pins containing U-Al alloy or UAl_X dispersion fuel or (2) a thin film of UO_2 coated on the inside of a stainless steel tube. After irradiation, the ^{99}Mo is separated from the uranium and fission products [1].

To yield equivalent amounts of ^{99}Mo, an LEU target must contain approximately five times as much uranium as an HEU target. Consequently, substituting LEU for HEU will require changes in both target design and chemical processing. Three major challenges have been identified with substituting LEU for HEU: (1) modify the targets

and purification processes as little as possible, (2) assure continued high yield and purity of the ^{99}Mo product, and (3) limit economic disadvantages.

A denser form of uranium is required in order to keep the target geometry the same when changing from HEU to LEU targets. Targets containing LEU in the form of a metal foil (~125-150 μm thick) are being developed. A new annular target was developed last year, and several targets were irradiated [1]. This year, five annular targets containing six foils and two tapered style [2] targets containing two foils were fabricated and irradiated. Performance of the targets is discussed below.

Also, chemical processing of one of the irradiated foils was completed using a slightly modified Cintichem process [3] to recover ^{99}Mo from the irradiated LEU foil. The foil was processed in the same hot cell and equipment used for recovering ^{99}Mo from the HEU target. The same equipment was used except for the dissolver. Also, the same processing steps were used except that sulfuric acid was eliminated from all process solutions. Samples of process solutions were collected during processing and analyzed by gamma spectrometry to measure ^{99}Mo yield and purity from step to step. Results are discussed below. Also discussed are several experiments to determine the effects of further increasing the uranium concentration in the Cintichem process.

TARGET IRRADIATIONS

The uranium foils irradiated in the targets were adjusted uranium (specified to contain approximately 450 ppm iron, 1000 ppm aluminum) that had been heat-treated to produce a fine, random grain structure [4]. Various fission-recoil barriers were added to the uranium foils [4,5]. Table 1 shows the details of the fabricated targets. The foils irradiated during August 1999 in the annular targets were more brittle after irradiation than expected [1]. This test matrix allowed us to compare the performance of several batches of foil in both the annular and the tapered style targets. It also allowed us to study thicker nickel-plate and aluminum-foil fission recoil barriers.

The annular targets were loaded onto the re-usable irradiation rigs described last year [1]. Each irradiation rig can accommodate up to two annular targets. The tapered targets were assembled into a tandem assembly [2] and irradiated separately. All the targets were irradiated in the Indonesian RSG-GAS reactor at a reactor power of 15 MW (~2 x 10^{14} n/cm^2/s) for approximately 120 hours. The overall irradiation performance of the targets was good. There was no evidence of heat-transfer problems during irradiation. All the annular targets were easily removed from their rigs after irradiation, indicating that no significant mechanical distortions had occurred during irradiation. The tapered targets also had no signs of mechanical distortions or problems after irradiation.

Table 1. Targets Irradiated 6/00 in the Indonesian RSG-GAS Reactor

Target No.	Target Style	Barrier	Notes
00-1[a]	Tapered	15-μm Electroplated Ni	-Tubes were Type 702 Zr. -Foil from ingot 99-3 (excess from 8/99 irradiation).
00-2[b]	Annular	15-μm Electroplated Ni	-Tubes were Type 3003 Al. -Contact surfaces were anodized.[c] -Foil from ingot 98-1 (excess from 3/99 irradiation).
00-3[a]	Tapered	15-μm Electroplated Ni	-Tubes were Type 702 Zr. -Foil from ingot 99-2 (new for 6/00 irradiation).
00-4[b]	Annular	15-μm Electroplated Ni	-Tubes were Type 3003 Al. -Contact surfaces were anodized.[c] -Foil from ingot 99-2 (new for 6/00 irradiation)
00-5	Annular	30-μm Electroplated Ni	-Tubes were Type 3003 Al -Contact surfaces were anodized.[c] -Foil from ingot 99-2 (new for 6/00 irradiation).
00-6	Annular	15-μm Electroplated Zn	-Tubes were Type 3003 Al. -Contact surfaces were anodized.[c] -Foil from ingot 99-2 (new for 6/00 irradiation).
00-8	Annular	25-μm Al Foil	-Tubes were Type 3003 Al. -Contact surfaces were anodized.[c] -Foil from ingot 99-2 (new for 6/00 irradiation).
00-9	Annular	25-μm Al Foil	-Tubes were Type 3003 Al. -Contact surfaces were not anodized. -Foil from ingot 99-2 (new for 6/00 irradiation).

[a]Two targets were assembled into a tandem assembly [2].
[b]Two foils assembled into one set of target tubes.
[c]Black sulfuric acid anodization following MIL A 8625 F Type II Class 2 specifications. Only the inner surface of the outer tube and the outer face of the inner tube were anodized. The inner and outer tubes were masked at either end so that a clean, oxide-free surface remained for welding.

After irradiation, the targets were transported to a target disassembly hot cell at the Radiometallurgy Installation. Disassembly of the annular targets was accomplished by cutting off the ends of the target and then making a longitudinal cut in the outer tube. After cutting the outer tube, our procedure called for the outer tube to be pried off the inner tube and the uranium foil to be recovered. Disassembly of the tapered targets was accomplished by cutting off the ends of the target and then pushing the inner tube out. Results of the disassembly are shown in Table 2.

Table 2. Performance Results of Targets Irradiated 6/00 in Indonesia

Target No.	Foil Condition	Notes
00-1	Excellent	No bonding of the foil to the target was observed. The foil was recovered in one piece and remained ductile for several days after removal from the target (see Fig. 1).
00-2	Very Good	No bonding of the foil to the target was observed. The foil was easily recovered in one piece. However, after sitting unencapsulated in the hot cell overnight it became slightly brittle and broke into a few large pieces.
00-3	Excellent	No bonding of the foil to the target was observed. The foil was recovered in one piece and remained ductile for several days after removal from the target (see Fig. 1).
00-4	Very Good	No bonding of the foil to the target was observed. The foil was easily recovered in one piece (see Fig. 2). However, after sitting unencapsulated in the hot cell overnight it became slightly brittle and broke into a few large pieces.
00-5	Very Good	No bonding of the foil to the target was observed. The foil was easily recovered in one piece. However, after sitting unencapsulated in the hot cell overnight, it became slightly brittle and broke into a few large pieces.
00-6	Poor	No bonding of the foil to the target was observed. However, the foil was very brittle and broke into many pieces during disassembly.
00-8	Poor	No bonding of the foil to the target was observed. However, the foil was very brittle and broke into many pieces during disassembly.
00-9	Poor	A small amount of bonding of the foil to the target was observed. Also, the foil was very brittle and broke into many pieces during disassembly.

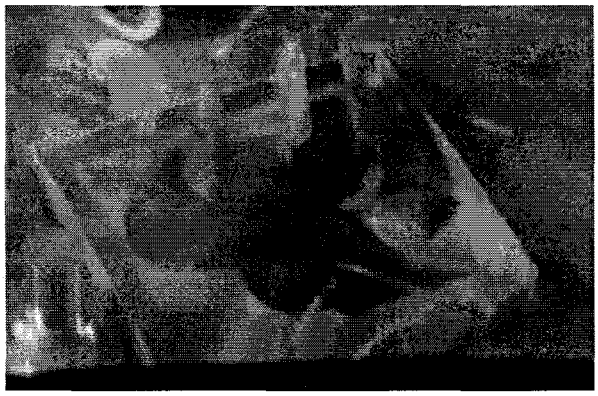

Figure 1. Foils from Targets 00-1 and 003

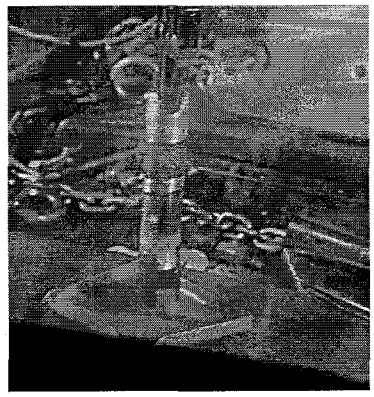

Figure 2. Foil from Target 00-4

In general, the irradiation test was very successful. Foils irradiated in the annular target with the 15-μm electroplated nickel fission-recoil barriers did not bond and were not brittle when initially removed from the target. Also, foils irradiated in the tapered target with the 15-μm electroplated nickel fission-recoil barriers did not bond and were not brittle, even after sitting a couple of days unencapsulated in the hot cell. Since the foils are to be processed immediately after being unencapsulated, foils from either target would be acceptable. To increase mechanical strength after irradiation and possibly decrease the fabrication effort, we are considering irradiating targets with 15-μm nickel foil fission-recoil barriers.

The targets irradiated with aluminum or zinc fission-recoil barriers are being evaluated for potential use in alkaline recovery processes. The foil with the zinc barrier did not bond to the target. However, it was very brittle after irradiation and broke into many pieces. Although a system could be designed to collect those pieces prior to processing, the resulting equipment and procedures would be more complex than currently envisioned. Therefore, we are considering ways to improve zinc fission-recoil barrier performance.

The foils irradiated with 25-μm aluminum foil barriers were also very brittle. The foil irradiated in the tube without anodizing was lightly bonded to the target tubes. However, the foil irradiated in the target with anodizing was not bonded to the target tubes and was easily recovered. To improve the performance of targets with aluminum fission-recoil barriers we are considering increasing the thickness of the barrier. When using the annular-style target with aluminum fission recoil barriers, anodization of the tubes is necessary to prevent bonding, and increasing the thickness of the aluminum barrier may enhance the performance.

CHEMICAL PROCESSING

Chemical processing of one of the irradiated LEU foils was completed using the slightly modified Cintichem process described earlier [3]. In general, the separation process consists of dissolving the irradiated LEU foil in a reusable dissolver, precipitating the molybdenum with α-benzoin oxime (ABO), washing the precipitate, dissolving the precipitate, then passing the resultant solution through two purification columns. Samples of solutions were collected during processing and analyzed by gamma spectrometry to measure ^{99}Mo yield and purity from step to step. The yield for ^{99}Mo following the dissolution of the ABO precipitate was >95% for the LEU target, which is excellent. Following the dissolution of the ABO precipitate, the solution volumes and compositions are identical for both the HEU and LEU Cintichem processes. Therefore, no differences between the HEU and LEU process are evident.

Radiochemical purity was also measured at each of the process steps for the LEU foil that was processed. Figures 3-6 show the purity of the ^{99}Mo after each of the processing steps. Note that the impurity levels in the final product sample (that following 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 (samples taken between purification steps), the higher ^{99}Mo and ^{99m}Tc activities make determination of impurity activities difficult.

The purity of the ^{99}Mo from radioiodine is shown in Fig. 3. The purity from ^{131}I, the isotope of primary concern, exceeded purity specification by 100 times. Figures 4 and 5 show the purity of the ^{99}Mo from transition metal and rare-earth radioisotopes, respectively. Figure 6 shows the ^{99}Mo purity from a variety of other radionuclides. The use of LEU has always raised concerns about the increased production of ^{239}Pu (due to the increased amount of ^{238}U) and ability to meet alpha purity specifications. However, note that in Fig. 6 the purity of the ^{99}Mo product from ^{239}Np (the parent of ^{239}Pu) exceeded 0.01 μCi/mCi ^{99}Mo. Based on their relative half-lives, this level of ^{239}Np would generate a product containing $<3\times10^{-9}$ μCi/mCi ^{99}Mo of ^{239}Pu. This exceeds the purity specification for alpha emitters by >300 times.

As seen in Figures 3-6 the purity of the ^{99}Mo product exceeded the product purity specification of 0.1 μCi/mCi ^{99}Mo. Thus the ^{99}Mo recovered from LEU metal foil using the Cintichem process appears to be a viable alternative to HEU. However, we still need to verify how well the ^{99}Mo product is loaded onto the ^{99m}Tc generators. We plan to complete this activity next year.

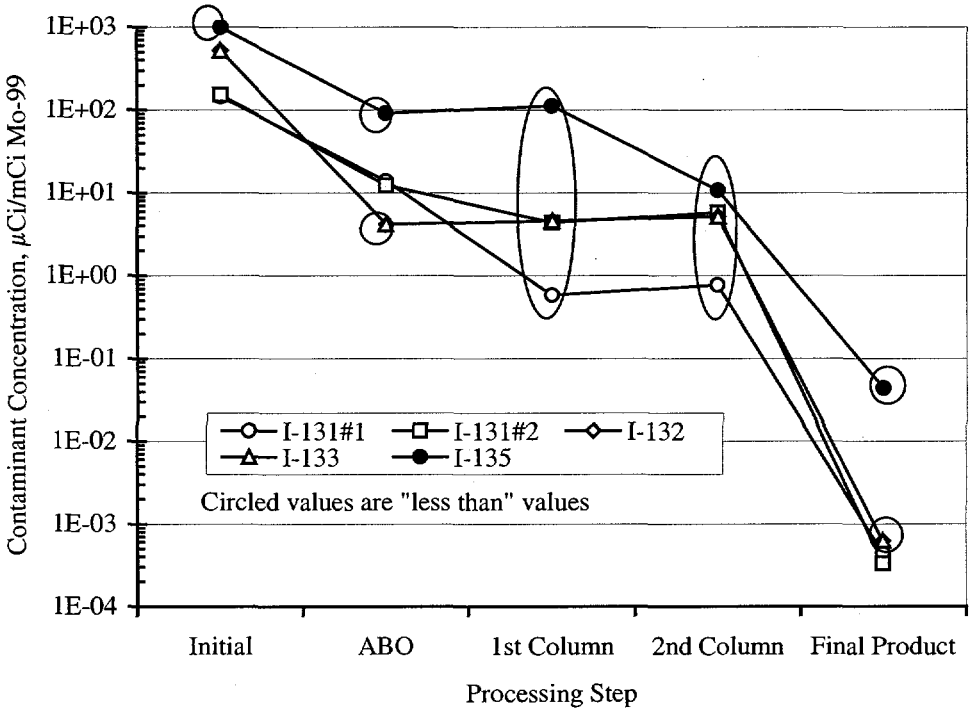

Figure 3. Purity of ^{99}Mo from Radioiodine

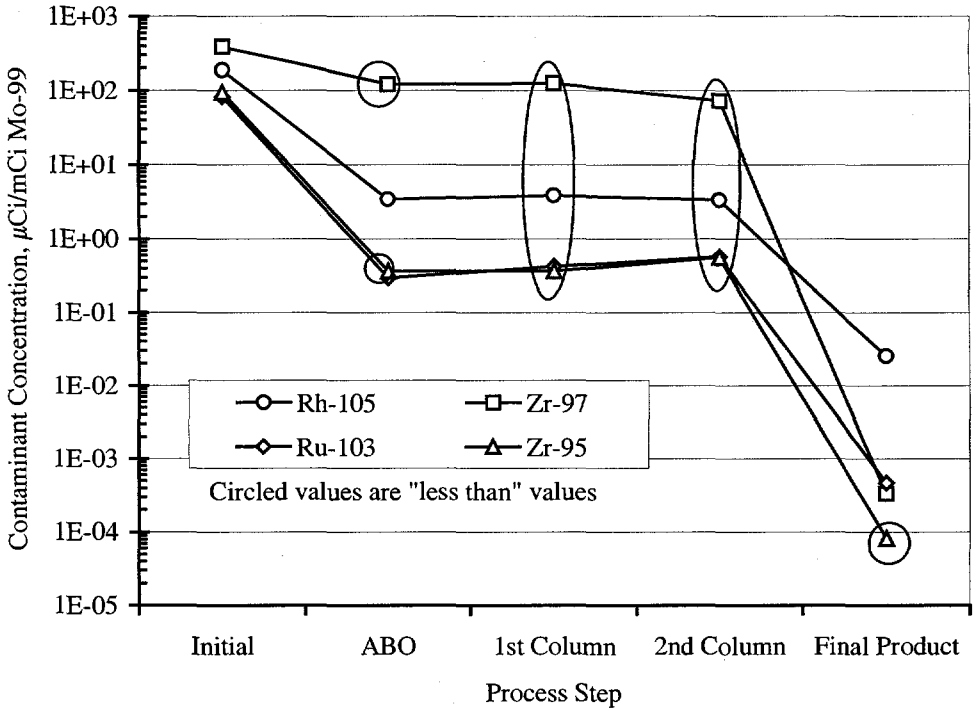

Figure 4. Purity of ^{99}Mo from Transition Metals

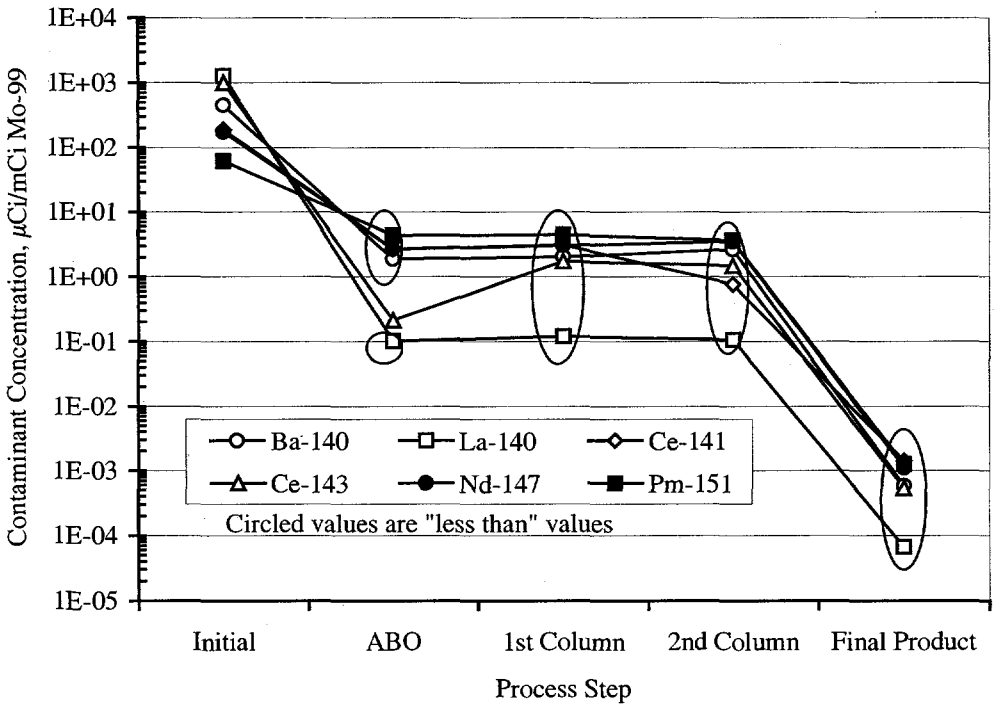

Figure 5. Purity of ^{99}Mo from Rare Earth Isotopes

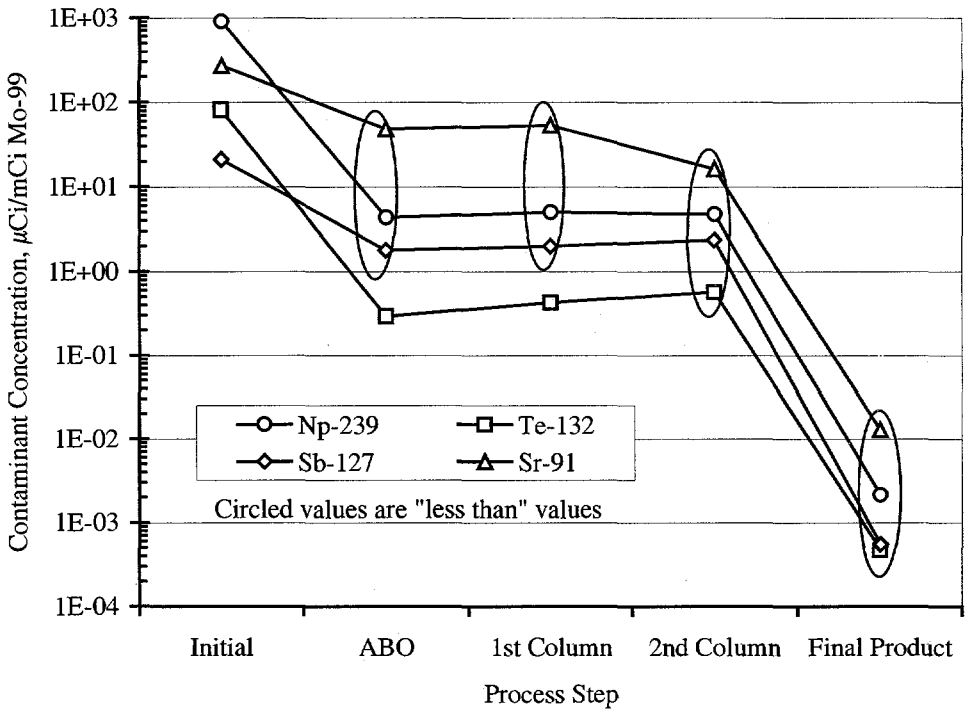

Figure 6. Purity of ^{99}Mo from Various Other Radioisotopes

The use of LEU requires about five times more uranium to be processed to yield equivalent amounts of ^{99}Mo. In many cases the same equipment and the same procedures are used for LEU as for HEU; thus, the solubility of uranium as a function of nitric acid concentration becomes important. Since the published data on the solubility are inadequate, the solubility of uranyl nitrate was measured at temperatures ranging from 20°-50°C and in nitric acid concentrations of 0-2 M. The densities of each saturated solution were also measured.

As seen in Figs. 7 and 8 the solubilities increase with increasing temperature and decrease with increasing nitric acid concentrations. These data will be useful for further efforts in predicting how much uranium can be dissolved and processed in the Cintichem process.

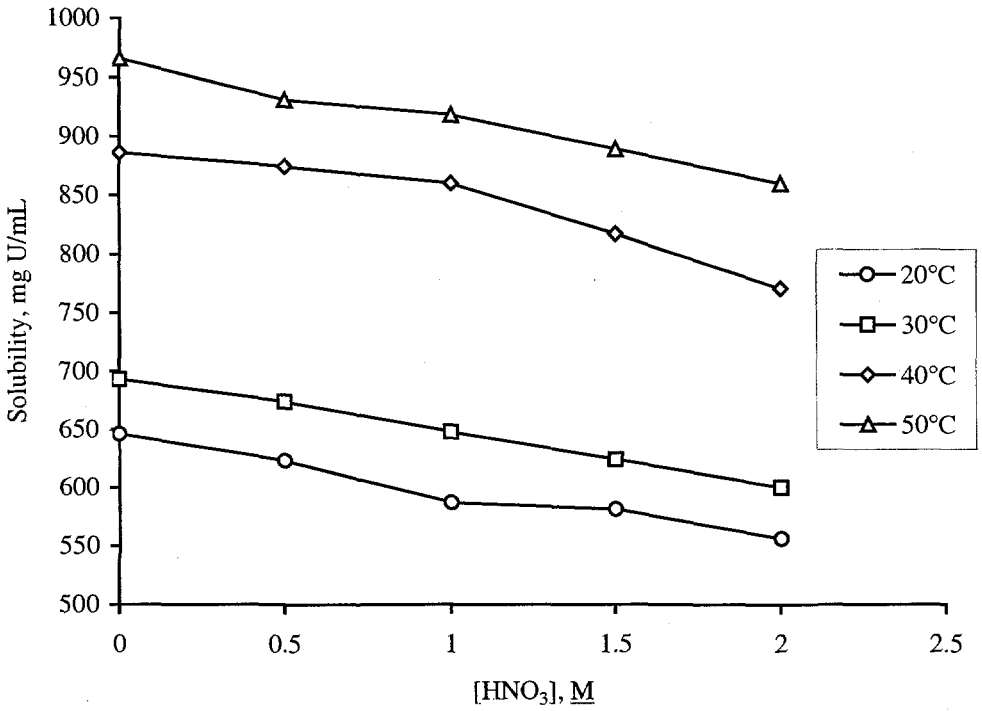

Figure. 7. Solubility of Uranyl Nitrate Solutions

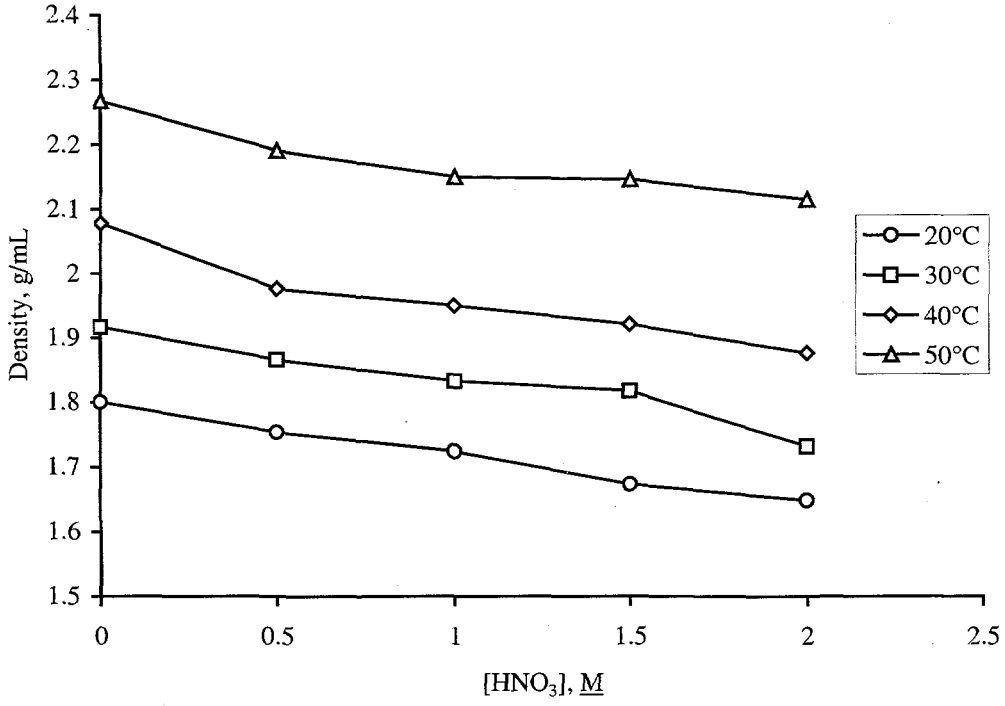

Figure. 8. Density of Saturated Uranyl Nitrate Solutions

CONCLUSIONS

The substitution of LEU for HEU is viable when using the Cintichem process. The annular targets using nickel-plated barriers perform well and can be used for processes using acid dissolution. The ^{99}Mo product recovered from an LEU foil target achieved the specified purity of the ^{99}Mo from gamma-emitting impurities. At least one additional demonstration in Indonesia during 2001 will provide additional proof of these conclusions.

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