OVERVIEW OF 2007 ANL PROGRESS FOR CONVERSION OF HEU-BASED MO-99 PRODUCTION AS PART OF THE U.S. GLOBAL THREAT REDUCTION--CONVERSION PROGRAM

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ABSTRACT

ANL effort is divided into five areas: (1) cooperation with Argentina to demonstrate the use of LEU-foil targets in alkaline-based processes, (2) cooperation with Indonesia in converting their HEU-based Cintichem process to LEU-foil targets, (3) technical assistance to two potential U.S. domestic suppliers (MURR and BWTX), (4) responding to the National Academies Study, and (5) participation in the IAEA CRP for Indigenous Mo-99 production. This paper presents highlights of these activities. A short description of how the dose emitted by spent HEU target material compared to spent fuel is also included.

1. Introduction

The Reduced Enrichment for Research and Test Reactors (RERTR) program was begun in 1978 to develop technical means to convert reactor fuels from HEU to LEU. By 1986, RERTR's success in developing uranium silicide fuel and codes and analyses for proving the efficacy of converting many reactors made the hitherto minor amount of HEU exported for ^{99}Mo production begin to look important. It had become approximately 20% of exported HEU. With the ^{99}Mo market continuing to grow by ~10%/yr and continuing success in reactor conversion, that fraction will continue to grow. Other concerns with spent HEU used in ^{99}Mo production are (1) only a few percent of the ^{235}U is burned up during irradiation, making it very desirable material and (2) because of the low burn up, the material, it becomes essentially contact-handled waste after ~3 years. Therefore, a new task to develop means to convert HEU targets for the production of ^{99}Mo was begun.

Currently, 95% of all ^{99}Mo is produced by irradiation of HEU targets that are subsequently processed primarily to recover the molybdenum. The four major producers of ^{99}Mo, all of them utilizing HEU targets and dedicated processing facilities, are:

- MDS Nordion (Canada)
- Tyco Healthcare, Mallinckrodt (Netherlands)
- IRE (Belgium)
- NTP Radioisotopes (Pty) Ltd (South Africa)

These four act as both competitors and partners, depending on the circumstances. For example, IRE supplies much of MDS Nordion's ^{99}Mo to the European market, and IRE and NTP Radioisotopes have a formal cooperative agreement in place. A small producer, the Indonesian National Atomic Energy Agency (Badan Tenaga Atom Nasional, BATAN) is currently using HEU targets but will be converting to LEU in the near future. The remaining 5% of global ^{99}Mo production is primarily derived from the irradiation of LEU targets. The Australian National Science and Technology Organization (ANSTO) have always used LEU, and CNEA (Comisión Nacional de Energia Atómica, Argentina) converted to LEU in September 2002. Additionally, very small amounts of ^{99}Mo are being made from the irradiation of natural molybdenum (by neutron activation of ^{98}Mo); China, India, Brazil, and Kazakhstan use this technique.

The Global Threat Reduction, Conversion program, formerly known as the Reduced Enrichment for Research and Test Reactors (RERTR) program has made significant progress in developing technologies that will allow conversion of HEU targets to LEU. Conversion to LEU usually necessitates changes in the target, in dissolution/digestion, and the Mo-recovery step. Once the uranium is removed, the remaining purification steps will be identical for HEU and LEU targets.

Our ^{99}Mo conversion effort is divided into five areas: (1) cooperation with Argentina to demonstrate the use of LEU-foil targets in alkaline-based processes, (2) cooperation with Indonesia in converting their HEU-based Cintichem process to LEU-foil targets, (3) technical assistance to two potential U.S. domestic suppliers (MURR and BWTX), (4) responding to the National Academies Study, and (5) participation in the IAEA CRP for Indigenous ^{99}Mo production. A short summary of our progress in each of these areas follows. Technical highlights are discussed in an accompanying publication [1]. A short discussion of the dose rate from spent-target material is also included.

2. Cooperation with Argentina

The Comisión Nacional de Energia Atómica, Argentina (CNEA) has been a strong partner of the RERTR program for many years. It converted its HEU processing to LEU in 2002 by developing a high-density LEU dispersion plate that can be used in its current process with minor changes [2]. This target will allow them to continue production at their current rate but allows limited opportunity for increasing production. For the long term, development of the process using LEU-foil targets is in their plans. This will not only allow a tripling of their ^{99}Mo production in the same irradiation hole, but also will lower the liquid waste from processing by 6-10 times and processing time by at least 4 hours. Major development challenges for the CNEA conversion using LEU-foil targets were (1) designing a prototype production vessel for digesting irradiated LEU foils in alkaline solutions, (2) developing means to improve digestion efficiency, and (3) modifying ion-exchange processes used in the CNEA recovery and purification of ^{99}Mo to deal with the lower liquid volumes generated from LEU-foil digestion. A demonstration of irradiating and processing an LEU foil target was performed in December 2006.

An LEU target (24.35 g of LEU-total; 4.87 g of ^{235}U), fabricated at Argonne had previously been shipped to Argentina. The target was in a low-flux irradiation position at 5 MW power; therefore, the fission yield was much lower than usual. Due to scheduling problems, it also sat for 5 days after being removed from the reactor. The target yielded 53.87 Ci of ^{99}Mo, and 22.62

Ci of ^{131}I, as measured in the dissolved target. For comparison, a similarly sized HEU target, irradiated in the CINTICHEM reactor typically yielded 600 Ci of ^{99}Mo and 200 Ci of ^{131}I. The irradiated target was transferred into the isotope production hot cell facility, and the target was disassembled. An apparatus designed and built at Argonne was used to cut the two welded ends off of the target and to cut the outer tube of the target. The outer tube was then separated from the inner tube and the target package, consisting of the LEU foil and the Al fission-barrier, was removed. The entire disassembly operation was easily accomplished in about 1.5 hours. It should be noted that this was the first time the operators had carried out the operation; therefore, the disassembly time would be expected to decrease significantly with experience. For example, after practice, it took about 20 minutes in the ANL mockup facility.

The target package was placed in a digestion vessel, sealed, and heated to 280ºC and 100 bar. The oxidant $KMnO_4$ and only 400 mL of 1 M NaOH were required to digest the target. The digestion was complete in one hour; the entire heating/cooling cycle took about three hours. The residual pressure was 20 bar. This compares favorably with the current CNEA process that requires about 4000 mL of base and 4 hours to digest a target. Once the digestion vessel had cooled, the slurry was vacuum drawn out of the vessel and through two filters. The filtration was quick and effective. The subsequent ion-exchange loading, washing, and stripping operations were also done without any problems. The flow rate through the column was significantly higher than planned; the 500 mL of feed passed through the column in about 12 minutes. .

The gamma counting results showed that we recovered 92% of the ^{99}Mo in the product stream. Less than 10% of the total ^{131}I was recovered in the wash and product streams. Evidently, most of the iodine was reduced and, therefore, was sorbed onto the column, and not removed in the stripping step. The chemistry of iodine under these chemical and radiological conditions is being investigated in laboratory experiments at ANL [1].

3. Cooperation with Indonesia

The Indonesian National Atomic Energy Agency, *Badan Tenaga Atom Nasional* (BATAN), has been cooperating with ANL for many years and is expected to convert its Cintichem targets and processing to LEU-foil targets and the LEU-modified Cintichem process during 2008 [3, 4]. As well as developing its own process, BATAN has irradiated and demonstrated digestion of targets for alkaline-side processing.

During 2006, BATAN personnel were trained in rolling foils using depleted uranium metal. They have successfully rolled depleted uranium foils and await shipment of LEU metal from ANL. We are in the process of sending them a kg of LEU metal. Another task that needs be completed for conversion is solidification of the HEU waste currently stored. A visit is being planned for January 2008 to (1) develop means for solidification of waste using calcination and (2) optimize conditions for foil rolling using LEU metal.

4. Technical Support to BWXT and MURR

BWXT (BWX Technologies, Inc.) and MURR (University of Missouri Research Reactor Center) have provided assurances of interest in written form to NNSA to produce Mo-99 domestically. These documents are commercially sensitive and unavailable for public access.

BWXT is proposing the use of LEU-fueled solution reactors for producing ^{99}Mo; MURR is proposing use of the LEU-modified Cintichem processing of LEU-foil targets (technology developed at ANL as part of the RERTR program). In both cases, once the decision is made to proceed, at least three years will be required to begin production. Current planning by these two entities leads to each supplying 30-50% of the US market.

Argonne's interactions with BWXT has thus far been limited to (1) providing them with a white paper that summarizes R&D by Argonne and others for recovering molybdenum from uranyl nitrate and sulfate solutions at pH 1, and (2) defining a workscope for optimizing the recovery of molybdenum from these solutions.

Argonne has had a significant interaction with MURR on their implementing the LEU-modified Cintichem process for recovering and purifying molybdenum from LEU-foil targets. We have assisted MURR and the University of Missouri at Columbia Engineering College to develop the technology to the MURR environment. We have designed, fabricated, and are currently testing a small-footprint dissolver for nitric-acid dissolution of LEU foils; once testing is complete, it will be sent to MURR for testing with irradiated foils [1].

5. Responding to the National Academies Study

According to their web site [5], the "National Academies will conduct a study and provide findings and recommendations to the Department of Energy on the production of medical isotopes without highly enriched uranium. This project is sponsored by the U.S. Department of Energy, National Nuclear Security Administration. As mandated by Congress in Section 630 of the Energy Policy Act of 2005 [See Section 630(A) in Attachment 1], the 24 –month study will determine the following:

1. The feasibility of procuring supplies of medical isotopes from commercial sources that do not use highly enriched uranium, using the definition of feasibility defined in Section 630 of the Energy Policy Act of 2005.

2. The current and projected demand and availability of medical isotopes in regular current domestic use.

3. The progress that is being made by the Department of Energy and others to eliminate all use of highly enriched uranium in reactor fuel, reactor targets, and medical isotope production facilities.

4. The potential cost differential in medical isotope production in the reactors and target processing facilities if the products were derived from production systems that do not involve fuels and targets with highly enriched uranium."

The start date for the project is 9/25/2006. A report will be issued at the completion of the project.

Argonne has presented to the committee at an open meeting and will be meeting with them during their visit to MURR in mid October 2007. Six white papers were also written for use by

the Committee: (1) Primer on Mo-99 production, (2) The progress that is being made by the Department of Energy and others to eliminate all use of highly enriched uranium in reactor targets and medical isotope production facilities, (3) RERTR/GTRI Mo-99 technology-development history, (4) The current and projected demand and availability of medical isotopes in regular current domestic use, (5) The potential cost differential in medical isotope production in the reactors and target processing facilities if the products were derived from production systems that do not involve fuels and targets with highly enriched uranium, and (6) The feasibility of procuring supplies of medical isotopes from commercial sources that do not use highly enriched uranium.

6. Participation in the IAEA CRP for Indigenous Mo-99 production

Argonne's role in the IAEA CRP on Developing Techniques for Small-Scale, Indigenous Production of Mo-99 Using Low-Enriched Uranium is to provide the technology for the LEU-foil annular target and –Modified-Cintichem process to the participants. During 2007, we have communicated with participants in one-on-one and in meetings to meet that aim. A low foot-print dissolver was designed and is now being tested. It will be shipped to MURR later this year for demonstration using an irradiated foil.

7. Dose Associated with Spent HEU from Mo-99 Production

Because of its relatively low burn up, the ^{235}U content of the spent HEU is still above 90%. Further, the amount of long-lived fission products in the spent target material is minimal, meaning that it can be contact handled after a relatively short period of time after processing. To look at this effect, simple calculations were done to quantitate the dose per g of HEU irradiated for 5 days at a flux of $1x10^{14}$ neutrons/cm^2-sec [6]. These calculations were done for two scenarios—(1) acid dissolution, where all actinides and fission products but Mo, I, Kr, and Xe were present and (2) for alkaline digestion, where the base-soluble Cs is also removed. Three distances from the point source were looked at (1) 5 cm (approximate distance to the outside of a one-gallon can, (2) 10 cm (approximate distance to the outside of a five-gallon can, and (3) one meter (typical working distance for laboratory operations). No shielding was accounted for. Once specific information is known about the waste-storage container, the amount and form of the HEU stored per container, etc., a more detailed analysis can be performed.

For both waste types, the dose drops by nearly five orders of magnitude between the day the target is processed and three-year storage. For acid-dissolution waste, the dose rate after 3-years storage is 1.5 mrem/hour per g of HEU at 100 cm, <u>with no shielding</u>. For alkaline-digested HEU, the dose rate is 0.5 mrem/hr per g. Shielding would considerably lower that dose rate.

Figure 1 shows the drop in dose rate (mrem/hr) per g of HEU waste from alkaline processes vs. time after end of irradiation for the three distances. Considering that 5-8 million mrem are required to cause immediate disorientation and coma in seconds or minutes [7], the received dose for removal of large quantities of this material would not be consequential to a dedicated terrorist. Converting this material to a weapon would not require elaborate shielding and could be performed in a garage with minimal dose to the processors.

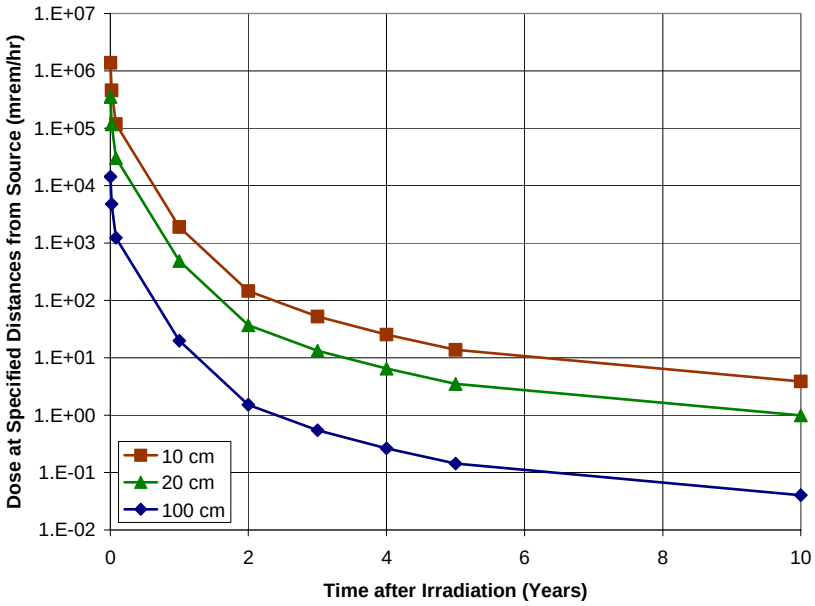

Figure 1. Dose rate/g at three distances from point source vs. time for HEU waste derived from alkaline digestion of targets (Cs, Mo, I, Xe, Kr removed) from 1-day to 10-year storage.

Based on a report published by Ponds and Matos [8], the dose rate at one meter/g-uranium for a spent HEU MTR fuel element burned to 60% would be ~40 mrem/h. Clearly, because of its greater ^{235}U fraction (~92%) and its lower dose rate, spent HEU target material is a far greater security and safeguards concern than spent research reactor fuel.

8. Future Plans

We will continue to work in the five areas over the next year. It is planned that we will have all in place to allow the conversion of the Indonesian production in 2008.

9. Acknowledgements

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