Facts and Myths Concerning ^{99}Mo Production with HEU and LEU Targets

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ABSTRACT

Currently, most of the world's supply of ^{99}Mo is produced from the fissioning of ^{235}U in targets of high-enriched uranium (HEU). Conversion of these targets to low-enriched uranium (LEU) would ease worldwide concern over the use and transport of this weapons-grade material. The four major producers (MDS Nordion, Mallinckrodt, IRE, and NTP) have not converted to LEU; they have stated that they have concerns over the economic penalties that may be associated with conversion to demonstrated LEU technologies. They have also stated concerns over the potential purity of the Mo-99 produced from LEU. This paper discusses (1) the main concerns that potential suppliers may have regarding the conversion of existing facilities for Mo-99 production to use of LEU and (2) the technical and, to a limited extent, economic bases associated with conversion to LEU. These discussions attempt to separate the facts from the myths

1. Introduction

Technetium-99m (^{99m}Tc), the daughter (decay) product of molybdenum-99 (^{99}Mo), is the most commonly used medical radioisotope in the world. Annually, it is used for approximately 20-25 million medical diagnostic procedures, comprising some 80% of all nuclear medicine procedures [1]. Other important medical isotopes such as ^{131}I (iodine-131) and ^{133}Xe (xenon-133) can be recovered during ^{99}Mo processing. These radioactive isotopes are being used in therapeutic and diagnostic applications, and have promising applications in the area of radio-immunotherapy.

^{99m}Tc is recovered from ^{99}Mo generators by clinics and treated chemically using "kits" supplied by radiopharmaceutical suppliers that allow the ^{99m}Tc tagged protein to bind to specific organs. The patients are then gamma scanned to look at the organ, blood flow through the organ, and other effects. ^{99m}Tc decays with a half-life of six-and-a-half hours.

Today, 95-99 % of all ^{99}Mo is produced in research, test, or isotope production reactors by irradiation of highly enriched uranium (HEU) targets that are subsequently processed primarily to recover molybdenum ^{99}Mo. There are four major producers of ^{99}Mo, all of them utilizing HEU targets and dedicated processing facilities. These producers are:

- MDS Nordion (Canada)
- Mallinckrodt (Netherlands)
- IRE (the Institut National des Radioéléments, Belgium)
- NTP Radioisotopes (Pty) Ltd (South Africa)

At this time, the remaining 1-5 % of global ^{99}Mo production is derived from the irradiation of low-enriched uranium (LEU) targets. The Australian National Science and Technology Organization (ANSTO) has always used LEU, and the Argentine Comisión Nacional de Energía

Atómica (CNEA) converted in September 2003. Additionally, very small quantities of ^{99}Mo are produced from the irradiation of natural molybdenum (by neutron activation of ^{98}Mo). To yield equivalent amounts of ^{99}Mo, an LEU target must contain approximately five times the uranium of an HEU target. Consequently, substituting LEU for HEU requires changes in both target design and chemical processing. Three major challenges are evident for the substitution of LEU for HEU: (1) modifying the targets and purification processes as little as possible, (2) assuring continued high yield and purity of the ^{99}Mo product, and (3) limiting economic disadvantages. Argentina and Indonesia were committed to conversion and have been successful. The larger producers have not committed and see only problems in doing so. Discussed in this paper are facts and myths associated with conversion. We hope that discussing them openly will clarify the situation and lead to honest dialog about the real challenges in converting, so they may be addressed, and conversion for all ^{99}Mo producers will move forward.

2. Facts

Factual statements governing conversion of ^{99}Mo production from HEU to LEU targets include:

- **LEU targets require approximately five times more uranium than 93% ^{235}U targets.** In other words, the amount of ^{235}U in an LEU target must be the same or slightly greater than in an HEU target to produce an equal number of fissions.
- **Approximately 25-times more ^{239}Pu is produced in a 19.7 %LEU target than in an equivalent a 93% ^{235}U target.** ^{239}Pu is a decay product of ^{239}Np ($t_{1/2}$ = 2.355 day), which is formed from neutron absorption by ^{238}U.
- **Most of the world's ^{99}Mo is produced using HEU.** The big four (MDS-Nordion, Mallinckrodt, IRE, and NPT) all use HEU.
- **Worldwide concerns about the transport of weapons-grade material will eventually end its use.** This is a fact for both reactor fuels and targets and is the driver for the Global Threat Reduction Initiative (GTRI) and the RERTR program.

3. Myths

- **No serious producers use LEU.** Both ANSTO and CNEA use and plan to continue to use LEU. ANSTO has been a regional supplier of ^{99}Mo for many years using 2.2% enriched ^{235}U targets containing UO_2 pellets. It has recently decided to use the CNEA LEU target and process as a means to increase its production. CNEA converted to LEU targets three years ago and has successfully met its market requirements during and following conversion [2].

- **More targets will be required to be irradiated to meet the same Mo-99 production rate.** Because of the high density of uranium metal, the annular LEU-foil target can contain much greater quantities of uranium and, therefore, produce equivalent fission yields. ^{235}U loadings will be set by ability of the reactor coolant to remove heat from the target, not by geometric considerations.

- **The LEU target is not developed.** The LEU-foil target has been successfully irradiated, disassembled, and processed in Indonesia, Argentina, and Australia. A commercial fuel supplier (CERCA) has fabricated LEU-foil annular targets for ANSTO [3]. Argonne National Laboratory has fabricated a mock IRE target that will produce an equivalent amount of ^{99}Mo as their current HEU target.

  CNEA has irradiated and processed its LEU dispersion target commercially for three years.

- **Longer processing time will be required to meet production.** The answer to this myth varies within the broad spectrum of processing and the LEU target. For alkaline-digestion processes (CNEA, IRE, Mallinckrodt, and NPT) using the CNEA high-density dispersion-plate LEU target should have no effect on processing time. Use of the Argonne LEU-foil target could actually decrease processing time by as much as four hours due to the significantly reduced volume of the dissolver solution. In the case of the LEU foil target, only the uranium foil and the thin aluminum fission-recoil barrier are digested. For dispersion targets, the entire target is dissolved, requiring volumes five to ten times greater than that required for the LEU foil. This lower volume and the lower concentration of hydroxide in solution allow columns to be downsized considerably, lowering total processing time for the CNEA process by about four hours [4, 5].

  For acid dissolution processes (Cintichem, MDS Nordion), uranium metal dissolves significantly faster than uranium oxides. Further, when dissolving LEU foils, dissolution volumes are set by solubility of uranium in nitric acid rather than by target geometry (as is the case for the Cintichem and the new MDS Nordion targets). As seen in figure 1, the solubility of uranium in 1 M HNO_3 is quite high, >800 g/L under typical processing conditions.

  In the case of the Cintichem process, dissolution time is cut from two hours for the HEU target to less than one hour for the LEU foil. Further, because the volume used for the HEU target is set only by geometry, Argonne typically requires only half the dissolver-solution volume, further reducing processing time. Overall, processing time for the LEU-modified Cintichem process is decreased by almost two hours [6].

- **Significantly more radioactive waste is generated using LEU targets.** It is certainly a fact that five times more uranium will be processed and eventually become waste. However, uranium is not the only waste generated from processing. Use of the LEU-foil target in alkaline-digestion processes will reduce liquid waste volumes by at least five times and greatly limit the amount of aluminum hydroxide and column material that must be disposed. In the Cintichem process, lower dissolution volume may generate slightly less waste.

- **LEU targets and processing generate a ^{99}Mo product with lower purity.**

  Alpha contamination. Irradiating an LEU target does produce ~25-times more ^{239}Np as does an equivalent HEU target. The ^{239}Np decays with a half-life of 2.355 day to ^{239}Pu ($t_{1/2}$ = 24,119 year). The characteristic gamma ray emitted during ^{239}Np decay can be

easily followed during Mo-purification. In the LEU-modified Cintichem process, we see purification from ^{239}Np of greater than 10^9. Based on its much longer half-life, this is equivalent to contamination by ^{239}Pu in the Mo product of $<10^{-15}$ Ci/Ci of ^{99}Mo. This degree of decontamination is likely similar for all processes.

However, for both HEU and LEU targets, the bulk of the alpha decay in an irradiated target is from uranium isotopes (^{235}U, ^{238}U, and especially ^{234}U); see table 1. The data in figure 2 show that when uranium is enriched in ^{235}U, it is even more enriched in the relatively short-lived ^{234}U (2.45×10^5 year, compared with 7.04×10^8 year for ^{235}U and 4.47×10^9 for ^{238}U). For uranium enriched to 93%, the ratio of $^{234}U/^{235}U$ is increased by about 40%. For LEU, the ratio of $^{234}U/^{235}U$ is much more variable than for HEU material but always lower than HEU and, on average, lower than natural uranium. Due to alpha decay from uranium isotopes, the irradiated LEU targets will only contain ~20% more alpha activity than in an irradiated HEU target, not 25 times more. This small difference will have no effect on the radioisotopic purity of the ^{99}Mo product.

Overall product purity. In general, once uranium is removed in the first processing step, the following HEU and LEU process steps are essentially identical. In demonstrations of the LEU-modified Cintichem process in Indonesia, BATAN researchers have consistently found that the purity and yield using LEU-foil targets have been equal to or better than those produced by HEU targets, CNEA has demonstrative evidence that its LEU-product purity has been more consistent and higher than that it achieved with processing HEU targets [7]. The Indonesian and Argentine data are apples-to-apples comparisons, where the same process was used to treat both kinds of targets.

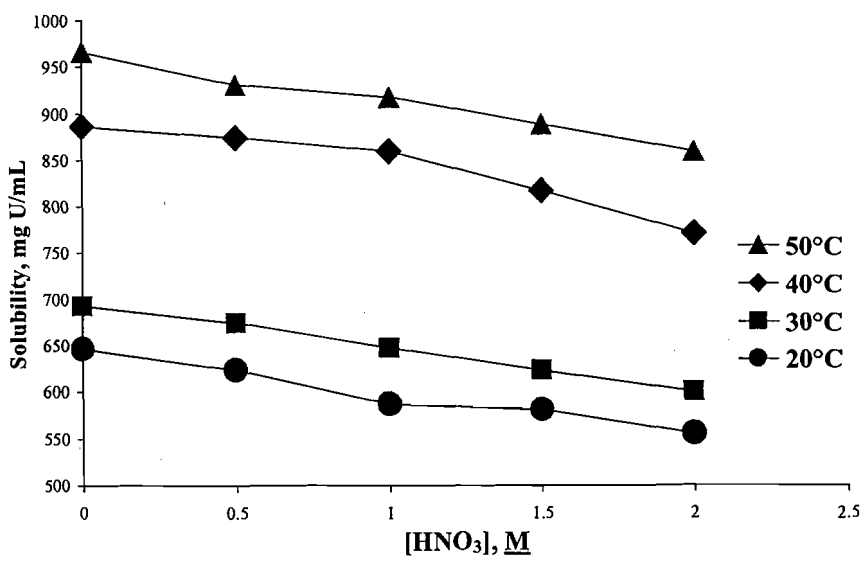

Fig. 1. Solubility of uranium as uranyl nitrate in nitric acid solutions between 20 and 50°C.

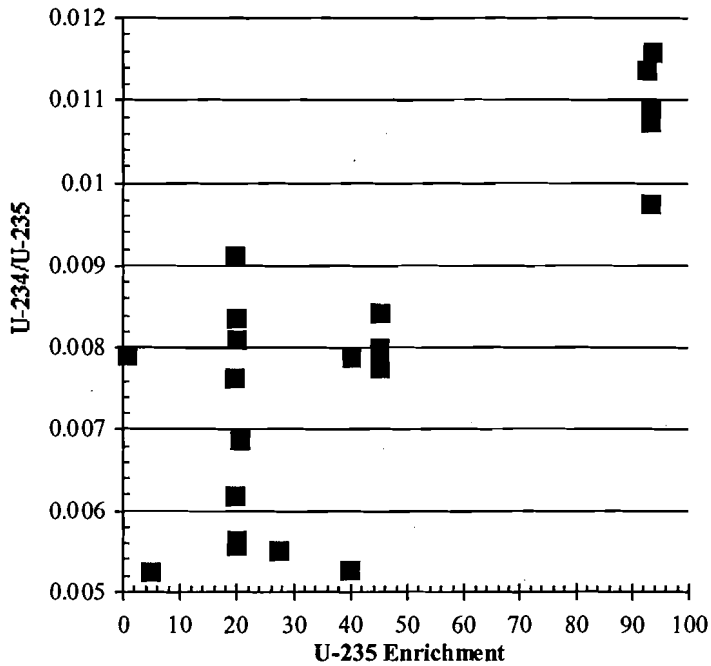

Fig. 2. Ratio of $^{234}U/^{235}U$ vs. ^{235}U content for various U.S. uranium exports.

Table 1. Reference HEU vs. LEU target: Comparison of calculated values for an HEU target in the Cintichem HEU-fueled reactor vs. an LEU target in an LEU-fueled Cintichem reactor

Reference Target	**HEU** (93% -^{235}U)	**LEU** (19.8%-^{235}U)
^{99}Mo yield, Ci	530	540
Total U (^{235}U), g	16 (15)	94 (18)
^{239}Pu, μCi	30	720
$^{234,235,238}U$, μCi	1280	840
Total α, μCi	1310	1560

4. Summary and Conclusions

Conversion from HEU to LEU targets is technically feasible for **all** current processes. LEU-foil annular targets and CNEA LEU dispersion targets have been successfully irradiated and processed. Producers in Argentina and Indonesia, who set out to convert, have been successful.

In these two instances, conversion has been demonstrated to not affect product purity, product yield, or operating costs. However, CNEA and BATAN have had to invest in converting.

Large-scale technology for conversion to LEU targets can never be completely demonstrated without the cooperation of the commercial producers. Each producer must weigh its commercial interests vs. international nuclear-nonproliferation and security interests when making the

decision to commit to full-scale technology demonstration and production of LEU-generated ^{99}Mo.

5. Acknowledgment

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6. References

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