ANL/CSE-13/32

Conceptual Process and Facility Design to Produce Low-Enriched Uranium Targets for the Production of ^{99}Mo Medical Radioisotopes

Chemical Science and Engineering Division

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Conceptual Process and Facility Design to Produce Low-Enriched Uranium Targets for the Production of ^{99}Mo Medical Radioisotopes

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CONCEPTUAL PROCESS AND FACILITY DESIGN TO PRODUCE LOW-ENRICHED URANIUM TARGETS FOR THE PRODUCTION OF ^{99}Mo MEDICAL RADIOISOTOPES

ABSTRACT

The Global Threat Reduction—Conversion Program, which is part of the National Nuclear Security Agency, aims to minimize the use of high-enriched uranium (HEU) worldwide by substituting low-enriched uranium (LEU) in processes that currently use HEU. A significant amount of HEU is processed to make medical isotopes, especially ^{99}Mo. Argonne National Laboratory has developed a process using LEU foil and an annular target design to produce ^{99}Mo without the need for HEU. A high-level study was conducted to find the unit operations and considerations that would be required in a facility designed to meet the next decade of U.S. demand for ^{99}Mo using this LEU target. This study should give a commercial enterprise interested in pursuing production of targets enough information to (1) understand what is required in the facility and (2) perform a high-level cost estimate to build and operate the facility.

1 INTRODUCTION

The most important clinical radionuclide currently is ^{99m}Tc (Manning 1995). This radionuclide is the product of the decay of Molybdenum-99 (^{99}Mo). Most of the ^{99}Mo used today is derived from the fissioning of ^{235}U; approximately 95% of the current ^{99}Mo supply came from four large-scale producers, all of which use high-enriched uranium (HEU: ≥20% enriched in ^{235}U):

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

Technetium-99m (^{99m}Tc) possesses two key characteristics that make it very useful in medical imaging. It emits an energetic gamma ray that is easily detectable through considerable amounts of tissue, and it has a short half-life of ~6 hours, which minimizes the radiation dose to the patient. Because of its 2.7-day half-life, ^{99}Mo must be continuously produced. The United States has not had a domestic source for ^{99}Mo since the Cintichem Tuxedo, New York, reactor was shut down in 1989 (NRC 1998). MDS Nordion produces most domestically used ^{99}Mo in the reactor at Chalk River, Canada (Brown 2005).

A process has been developed by Argonne National Laboratory (Argonne) to produce ^{99}Mo from low-enriched uranium (LEU: <20% enriched in ^{235}U) (Vandegrift et al. 1999). The annular irradiation target consists of a uranium ≥125-µm (≥0.005-in.) LEU foil that is clad with aluminum alloy (Figure 1). During assembly, a fission-recoil barrier is wrapped around the LEU foil before it is inserted between the cladding tubes. Its purpose is to allow the LEU foil to be removed from the cladding after irradiation by cutting the two ends of the target, making a longitudinal cut through the outer cylinder (where a U-free gap is left as the foil is wrapped along the inner tube), and prying the outer tube away from the inner tube. The fission-recoil barrier is 15-µm nickel foil for acid dissolution and 40-µm aluminum foil for acid digestion of the irradiated LEU foil. The thickness of the foil and the dimensions of the annular target are set by irradiation conditions. The outer diameter of the target and the irradiation rig design are set by allowing equal coolant flow on both the outer and inner walls of the target during irradiation. Its length will likely be set by the vertical flux profile to take advantage of the “sweet spot” in the flux, to achieve maximum production of ^{99}Mo.

Argonne has designed and fabricated targets for irradiation in Indonesia and Argentina. Target dimensions were set by their individual irradiation positions and ^{99}Mo requirements. The Indonesian target was designed to have the same outer dimension as their current HEU target and to contain up to 20 g of LEU (~4 g of ^{235}U), which is their current limit for insertion in the irradiation position. Table 1 gives the dimensions of the finished target, and Appendix A is an engineering drawing of the target. The Argentine target was designed so that up to four targets

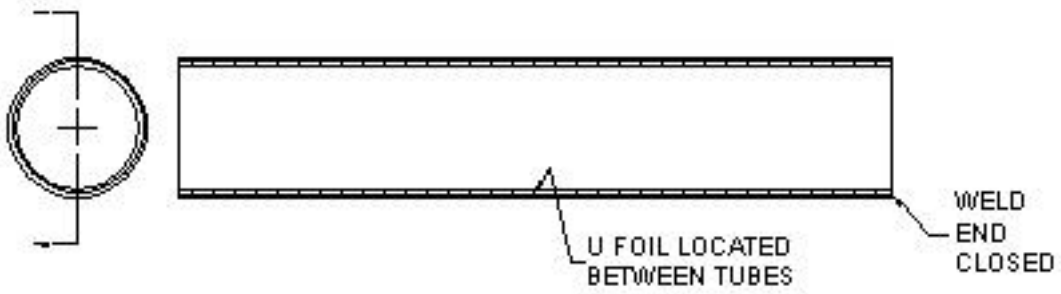

FIGURE 1 Schematic of the Argonne Annular Foil Target

TABLE 1 Dimensions of the Targets Developed for Use in Indonesia and Argentina

				Wall Thickness, mm	
Target	Length, mm	Outer Diameter, mm	Inner Diameter, mm	Inner	Outer
Argentina	155	36	33.7	2.3	2
Indonesia	150	30	28	2.3	2

could be placed in a square irradiation fixture that holds up to 12 of their current LEU dispersion-plate targets; each target will contain 30 g of LEU, 6 g of ^{235}U.

The University of Missouri Research Reactor (MURR) is studying the use of annular LEU-foil targets and the LEU-Modified Cintichem process developed at Argonne to produce about 50% of the current U.S. requirements (~3000 6-day Ci per week[1]).

If domestic production of ^{99}Mo using the LEU process developed at Argonne is to become a reality, a facility must be built to produce the LEU foil and to manufacture the targets. This report presents a high-level, preliminary study of the requirements (processes and equipment) related to producing rolled foil and targets for industrial-scale production of ^{99}Mo using the Argonne annular LEU-foil target.

There are multiple processes that can be used to produce foils. Four assumptions are made regarding the process:

- The incoming material will be supplied as LEU metal;
- The fabrication method must be a proven technology;

1 Producers generally sell ^{99}Mo by the 6-day Ci. This means they assure that, 6 days after delivery, *X* 6-day Ci will decay to no less than *X* Ci. Given the 66-hour half-life, the producers must actually send about four times the amount of Ci that will be available 6 days later.

- Recycling will be an important part of the process; and

- The plant will produce a final product of targets ready to be irradiated.

All processes begin with incoming material being inspected, weighed, melted, and cast. The defect-free section of the ingot is then protected from oxidation, heated, and reduced in thickness on a rolling mill. After rolling, the material is cleaned, sheared, and cold-rolled to final thickness. From this material, individual foils are sheared to the specified dimensions. The foils are then heat treated and inspected for meeting the requestor's specifications. While the foils are awaiting further processing, they are stored in a protective atmosphere. A flowchart of this general foil-making process is given in Figure 2. Figure 3 shows a flow chart of the target-making process.

The LEU target has been described above. The cladding of the LEU foil begins with wrapping the LEU foil in a 15-µm thick nickel foil, which acts as a fission-recoil barrier during irradiation to keep the LEU foil from bonding to the aluminum walls of the target. Cleaned concentric tubes are then assembled with the LEU foil sandwiched in between. The inner tube is expanded, forming a seal except for the ends. After facing, the ends are welded, the welds are sized, and the target is inspected for leaks and dimensions. The targets that pass inspection are then stored under vacuum until they are shipped. More details about the process are given in Guelis et al. (2003).

Most of the equipment used in producing LEU foils for targets is duplicated in producing U-Mo foils for monolithic fuel fabrication, as is a large portion of the storage facilities, security systems, and equipment required for quality control of the finished targets. Therefore, a section of this report is dedicated to looking at what capabilities could be added to a facility producing U-Mo monolithic fuel elements to also produce LEU-foil annular targets for ^{99}Mo production.

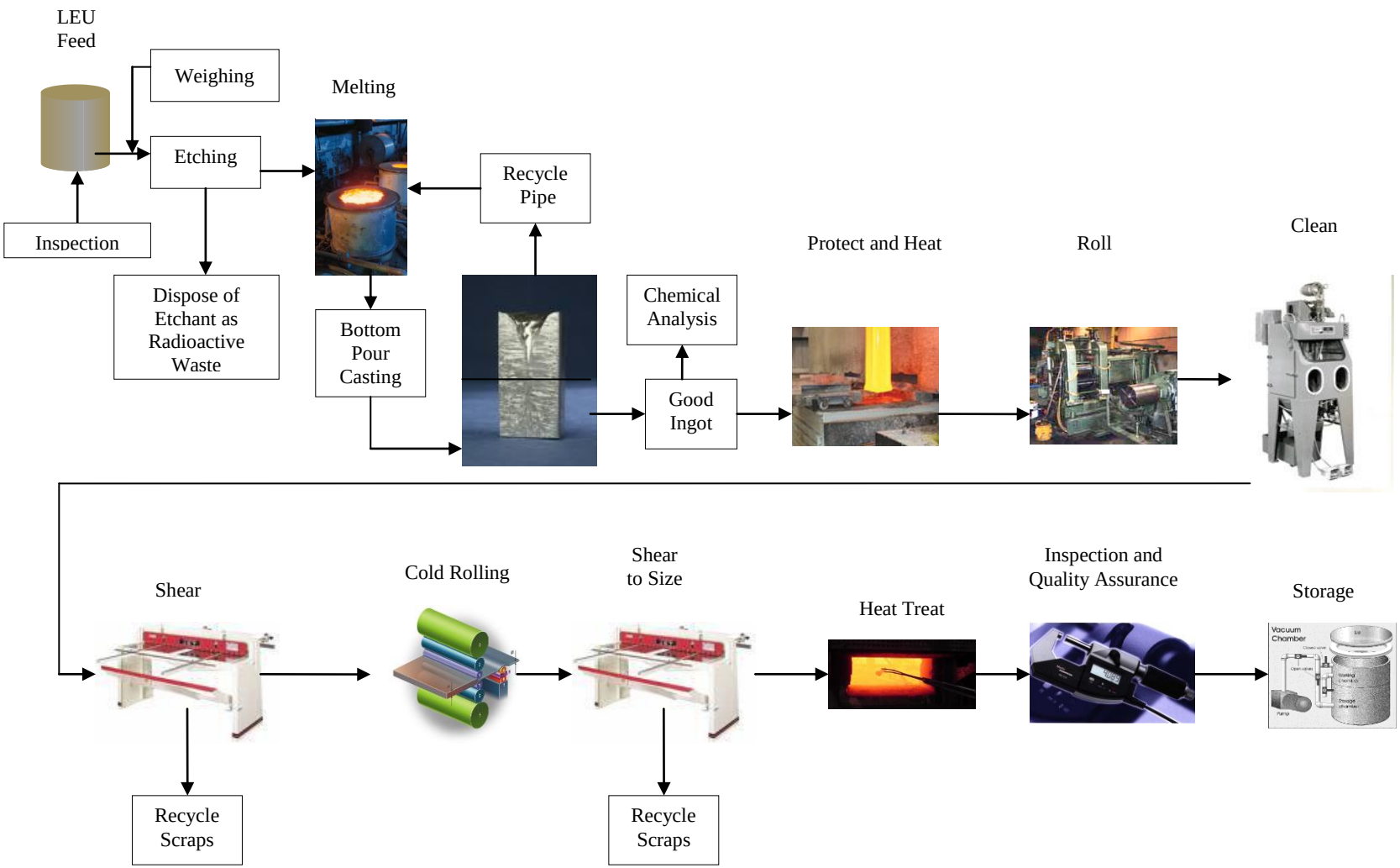

FIGURE 2 Flowchart of Foil-Making Process

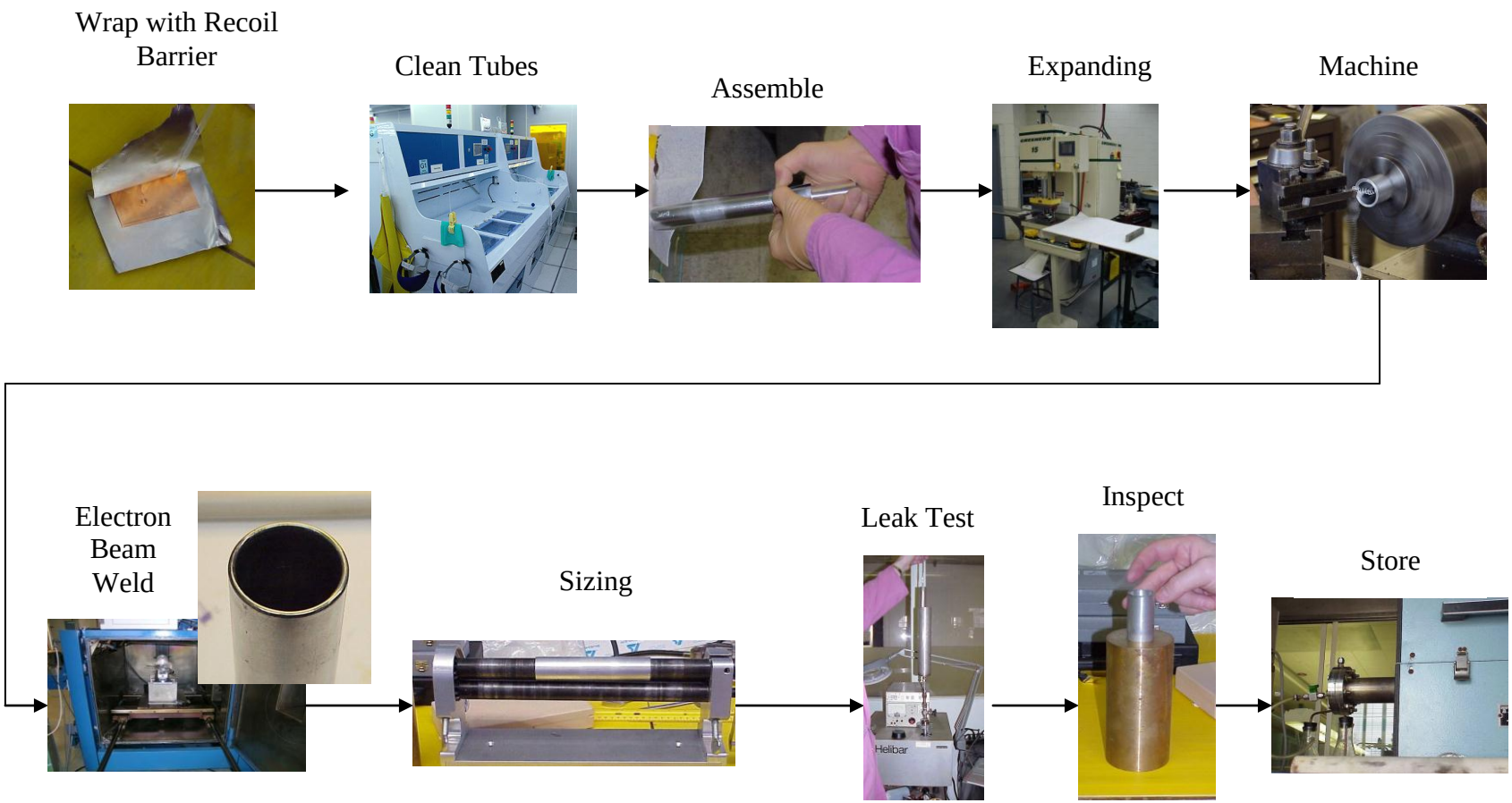

FIGURE 3 Flowchart of Target Assembly

2 LEU-TARGET REQUIREMENTS

2.1 ESTIMATED ANNUAL U.S. DEMAND FOR ^{99}Mo

A personal communication Charles Allen (2008) from the MURR staff contained the following statement: "Based on the results of preliminary neutronics calculations (ORIGEN 2.2), approximately forty to fifty LEU-foil targets (20-gram foil, 19.8 % enriched) would need to be irradiated and processed in order to produce 50% of the U.S. weekly ^{99}Mo demand." MURR has not yet finalized the size of the targets or the mass of LEU per target.

At a flux of ~1×10^{14} neutrons/cm^2-sec, ~150 Ci of ^{99}Mo (out of reactor) are produced per gram of ^{235}U, which translates into ~30 Ci/g-LEU. Figure 4 illustrates the number of out-of-reactor Ci of ^{99}Mo that must be produced each week to supply 6000 6-day Ci/week at the end of processing—between 40,000 and 50,000, depending on the chemical yield and decay during processing. Typically, the chemical yields for the LEU-Modified Cintichem process are ≥85% (compared to the 75% assumed in this illustration), and the processing time is ~5 hours, compared to up to 14 hours for other processes. Therefore, an assumption of 45,000 Ci/week is justified. Based on 30 Ci/g-LEU, producing 45,000 Ci/week of ^{99}Mo would require the irradiation of ~1.5 kg of LEU/week. This is equivalent to 75 20-g targets, 50 30-g targets, 38 40-g targets, or 30 50-g targets, because MURR is irradiating on the reflector area, where the flux is #8×10^{13}, assuming 110 20-g targets is a reasonable assumption.

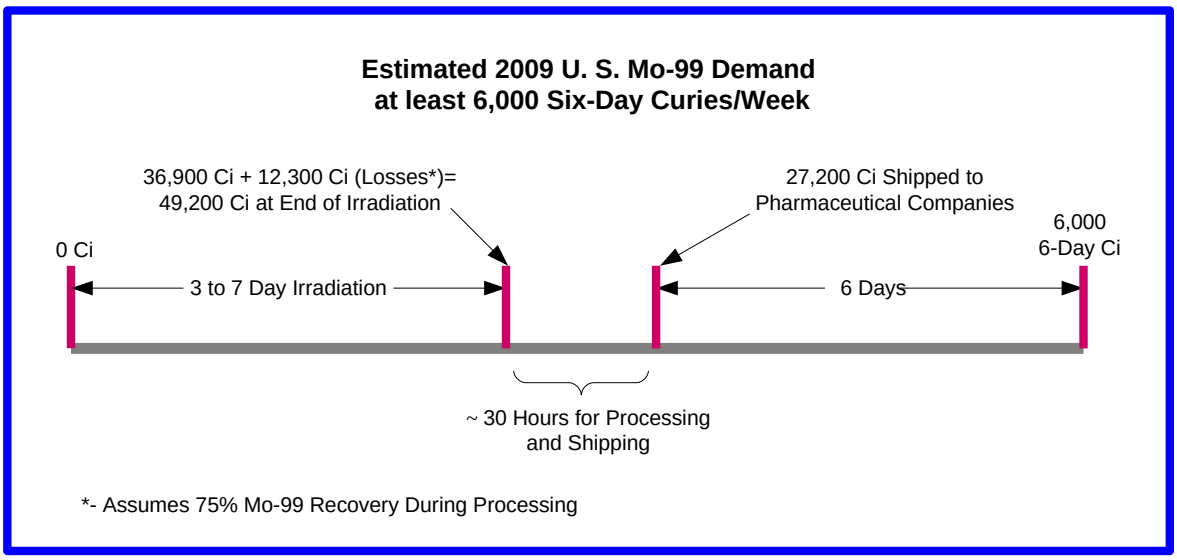

FIGURE 4 Illustration of Out-of-Reactor Ci/week of ^{99}Mo Required to Meet a Demand of 6000 6-day Ci/Week

According to a recent report, "Mo-99 use is expected to grow between 5 and 10% per year over the next decade" (Grant & Gray 2007). Because of the nature of the workshop, this can be expected to refer to the worldwide demand; therefore, the U.S. demand would be at the low end of this estimate.

According to Bio-Tech Systems, Inc. (2008), U.S. sales of diagnostic radiopharmaceuticals reached $1.93 billion in 2007 and are expected to rise to $4.06 billion by 2014. Radiopharmaceutical sales grew 9.0% in 2007, recovering from a temporary slowdown the previous year. However, growth should resume in the range of 11% 13% per year as new products are introduced for cardiology and oncology. Continued growth of PET imaging and sales of FDG will also contribute favorably. Sales of nuclear cardiology products will continue to bolster the radiopharmaceutical market, with high utilization of nuclear perfusion studies coupled with advanced pharmacologic stress agents and introduction of new products for imaging myocardial infarction and congestive heart failure. Therefore, nuclear cardiology sales of $1.29 billion in 2007 will increase to $2.10 billion by 2014.

FDG sales will increase from $398 million in 2007 to $1,358 million by 2014. FDG distribution will continue to improve, allowing more widespread use of PET in community hospitals. With reimbursement stabilizing and procedure volume increasing, PET should become more profitable for providers as well as suppliers of FDG. In addition, new PET radiopharmaceuticals in the pipeline for specialized applications should add to these sales. Market growth should also benefit from higher prices for many of the new products. More biopharmaceutical products for SPECT imaging will incorporate combined molecular imaging and therapy platforms with broad applicability to many different types of cancer, expanding the range of nuclear procedures. Availability of SPECT-CT will also add an important dimension to these new agents, aiding in image interpretation. This technical influx will help all segments of nuclear medicine, expanding the platform for molecular imaging. One effect is that clinicians will have more options as alternatives to higher risk and more costly invasive procedures. This will stimulate more research and investment, adding strength and stability to newer venture companies as well as those more established in the field.[2]

The growth of ^{99}Mo demand is not specifically cited in this reference, but it does point to a healthy growth in radiopharmaceuticals.

Projecting a 6% growth in the U.S. ^{99}Mo demand in the future would lead to doubling of the U.S. demand in 12 years; it would triple in 19 years. Thus, it is not unreasonable to take future growth requirements into account and look at a full capacity of the proposed facility to be 220 20-g targets/week.

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2 PET = Positron emission tomography. FDG = fluorodeoxyglucose. SPECT = Single Photon Emission Computed Tomography. SPECT-CT = combined Single Photon Emission Computed Tomography with High-Resolution Computed Tomography (CT) equipment in a single system.

2.1.1 Estimated LEU Targets Required

The number of targets produced per year would be 220 targets/week × 52 weeks/year = 11,440 targets/year. Using a conservative 90% acceptance rate, 12,700 targets/year will be required. It should be noted that the number of targets required will not have a large effect on the equipment needs or the footprint of the facility. Because of the relatively low mass of LEU to be fabricated into foils, the melting and rolling equipment is underutilized. One induction melter and two rolling machines (one for hot rolling and one for cold rolling) are all that are necessary for extensive expansion of production. At some point, an additional e-beam welder and lathe(s) for sizing target tubes may be required for expansion. More will be said about throughput limits when specific operations are discussed in the meat of the report.

2.1.2 Estimated LEU Metal Required

This study assumes that the uranium will not require needed down-blending and will be delivered as LEU metal. Assuming each target weighs 20 g (with approximate dimensions of 8.9 × 9.4 × 0.0125 cm, or 3.5 × 3.7 × 0.005 in.), the weight of LEU required would be 20 g × 12,700 targets/year = 254 kg/year. Using a conservative 70% (20% shearing and cleaning losses, 5% reject losses for foils and 5% reject losses for targets) acceptance rate, 18,200 foils will need to be produced each year, or approximately 350 foils/week. The combined weight for one week's worth of these foils is 7 kg. Using a very conservative 50% acceptance rate (80–85% would be a more typical acceptance rate) for the losses due to the pipe, and melting, a charge of 14 kg will be used for the melting furnace. To have extra melting capacity, a furnace with a capacity of 50 kg of uranium will be specified. From the charge weight, a reasonable ingot size to cast can be determined. If 8.9 cm (3.5 in.) is the standard foil width and 2.5 cm (1 in.) is the minimum height required to break down the cast structure, the ingot length is calculated to be 33 cm (13 in.). After cutting off the pipe, the ingot will be 16.5 × 8.9 × 2.5 cm (6.5 × 3.5 × 1 in.). Given these ultraconservative rejection rates, it is clear that one melter will suffice for any expansion scenario envisioned.

2.2 FOIL PRODUCTION METHODS

2.2.1 Direct Casting

Direct casting of uranium foils has been demonstrated by the Korean Atomic Energy Research Institute (KAERI) (Kim et al. 2004). The method is the least labor-intensive production method, has the lowest amount of recycle material, and has the minimum cost per foil (if the foils do not require any further processing after casting). The large investment in the direct caster equipment is offset by the savings of not needing to purchase rolling mills, heating furnaces, and heat treatment equipment. However, the current single-roll system produces foils that have extensive roughness on the side not in contact with the roller. The roughness and methods to reduce the amount of surface variance have been studied (Wiencek et al. 2008). It has not been proven that the direct cast foils can be used as cast, with or without a heat treatment. KAERI has

an experimental program in place to improve their foils (Kim et al. 2008). This may eliminate the problems with thickness seen in the single-roll method, but it is not a proven technology and may not be successful. Because of the uncertainty associated with the direct cast foil method, it was not considered a feasible method for this report.

2.2.2 Heat Treat and Cold Roll

Casting a thin slab and cold rolling it to foil with intermediate annealing heat treatments was proposed as another method to fabricate uranium foil (Clark et al. 2003). This method was also found to have several limitations. Casting a defect- and crack-free thin slab is not a trivial task. An as-cast structure usually has large grains, which lead to poor foil quality (cracks) and high reject rates. This is mostly due to the lower ductility of the as-cast slab and the cold-rolling temperatures. The method was not considered because of the low-quality foil produced and the significant cost increase due to the higher reject rate.

2.2.3 Hot and Cold Roll

The most common method used to produce uranium foils is the hot- and cold-roll method. Commercial fabricators have been in business making foil by this method since the 1980s. It is a well-established and proven method. The foils produced are of high quality, with very uniform thicknesses, and contain no pinhole defects. The equipment necessary for this method is readily available and is not prohibitively expensive. Because of these reasons, this method was chosen to use for the production of the foils.

3 CONCEPTUAL LEU FOIL FABRICATION PROCESS

3.1 RAW MATERIALS—ORDER, RECEIVE, INSPECT, AND STORE

3.1.1 Uranium

Since only pure LEU uranium will be used, there will be no down-blending in the facility and no (modified) uranium alloying. The most likely source for this material will be the Y-12 plant in Oak Ridge, Tennessee. A chemical analysis for the trace elements and isotopic analysis will be specified for each batch of incoming material. A year's supply of uranium will be kept on hand (250 kg) to ensure a constant supply of ^{99}Mo will be available. All uranium not in process will be stored in the same area. To preclude oxidation, the uranium must not be allowed long-term contact with air. The most economical means for storage would be vacuum-sealed containers or vacuum chambers. The use of inert gas would add more cost for storage with very little gain. Increasing the amount of uranium stored will have minimal effect on the equipment required or the amount of floor space.

All material weights will have to be verified in a high-efficiency particulate air (HEPA)–filtered hood in a separate loose-contamination area. All nonradioactive materials will be stored and inspected in a different area. A loose-contamination area will also be set up for cutting and machining operations. This is referred to as the "hot shop."

3.1.2 3003 Aluminum Tubing

A year's supply of 3003 aluminum alloy tubing will be kept on hand. This equates to about 6500 feet (12,700 tubes/year × 6 in.) of tubing. Domestic commercial tubing vendors (Precision Tube Company 2008) routinely fabricate batches of this size. A chemical analysis will be specified for each batch of tubing.

3.1.3 Material Tracking System for Accountability and Safeguards

A system for identifying and tracking all enriched uranium must be established. A computer-based system with barcode labels will be used to identify and track every piece of processed uranium and account for every 0.5 g of the original batch. Security will have to meet the requirements for a U.S. Nuclear Regulatory Commission (NRC) Category II special nuclear material facility (NRC 2013) based on an inventory of 254 kg of LEU. The cost to obtain a license from the NRC to operate such a facility must be added to the facility costs. If the amount of ^{235}U is increased, the facility will still be Category II as long as all the uranium is >20% enriched.

3.2 CRITICALITY CONSIDERATIONS

Processing and storage of this material will require a nuclear criticality safety evaluation. The single parameter limit for a solid metal infinitely reflected ^{235}U sphere is 20.1 kg (NRC 1978). The factor by which ^{235}U subcritical mass limits may be increased for <20% enrichment is 15 (NRC 1978). The resulting amount of 301.5 kg is well below the anticipated facility inventory. However, as the material is divided, much smaller amounts of uranium are required for a criticality. When solutions are considered, the minimum critical mass is 4.5 kg for <20 ^{235}U (NRC 1978). Controls and limits will need to be placed on the amount of reflective and moderator materials allowed in the storage area. Material in storage will also have to be sealed to prevent water intrusion and spaced into grids for additional protection. This amount of uranium can be processed safely with the proper documentation and the proper fissionable materials control procedures. If the amount of material is increased, extra space may be needed to keep the material in a subcritical array.

3.3 CLEANING, MELTING, CASTING, AND RECYCLING

The subsections below describe options for performing cleaning, melting, casting, and recycling of uranium during processing. In all these operations, surface oxide formation must be minimized. Uranium metal oxidizes rapidly when exposed to normal atmospheric conditions. The oxidation rate is even greater for liquid uranium. The presence of oxides in the starting material could lead to low ductility and unacceptable rejection rates. Controlled atmospheres must be used to minimize oxidation. In addition, LEU costs approximately $10K/kg and maximizing recycling appears to be a good business decision.

3.3.1 Cleaning/Etching/Weighing

The uranium must be cleaned by chemical etching of excess oxide before melting to ensure a clean, solid ingot. Typically a 50% nitric/water acid solution is used as the etchant. The etching area will require ventilation with HEPA filter exhaust and chemical vapor filters. The solution is not heated, so a simple commercial etching tank could be used. Weights will be measured in the loose contamination weighing hood referred to in Section 3.1.1 to account for the weight losses. Due to criticality concerns, the solution will be changed before the total loss reaches 350 g of ^{235}U and the material will be recycled or disposed of as waste depending on economic considerations. Due to the presence of uranium in solution, this operation will be the largest criticality concern in the facility. This area could easily meet the demands of cleaning uranium for one 50-kg melt/48-hour period.

3.3.2 Melting Method

There are many ways to safely melt uranium. Only commonly used methods were reviewed. As mentioned in Section 2.1.2 the furnace will have a 50-kg uranium capacity. This equates to a volume of about 2700 cm^3 (165 $in.^3$), or a crucible about 13 cm × 13 cm × 17 cm

(5 in. × 5 in. × 7 in.). As mentioned in the previous section, a total capacity of 6000 kg/year is possible with a 50-kg furnace (10 melts/month × 12 months × 50 kg).

3.3.2.1 Arc-Melting

Arc-melting (Beall 1968) uses a non-consumable electrode to directly heat a melt. It has many advantages and can produce very clean ingots; however, most of applications are for very small (<1 kg) ingots. This process may be the best choice for the minimization of contamination during melting since a water-cooled copper hearth is used, and the ingot could be cast directly in the hearth with minimal loss of material. It was not chosen for this facility because it is not a commercially proven production method for 15–50 kg melts. It should be noted that arc-melting should not be confused with electric arc melting, which is used commercially for melting steels. This method uses an arc between carbon electrodes, which would lead to carbon pickup during melting.

3.3.2.2 Microwave Melting

Microwave furnaces for the melting and holding of metal melts are energy efficient and environmentally green. They also have the advantage of using new types of uncoated crucibles. This would minimize the impurities in the cast ingot. However, this method is still in the developmental stages. It will be many more years before this will be a commercial process.

3.3.2.3 Electron Beam Melting

Electron-beam melting has been applied to the fabrication of refractory metals for many years. It is a proven process, but it was not considered due to the high cost of the equipment, its lack of robustness, and costly maintenance.

3.3.2.4 Induction Melting

Induction melting is a well-established method for melting metals. It can be done under an inert atmosphere or a vacuum to prevent oxidation. The equipment is robust and relatively inexpensive. Melting under vacuum has two advantages: (1) it will refine the melt due to the floating of oxides to the top of the melt and (2) it is the cheapest atmosphere to use for melting. A bottom-pour, coated graphite or oxide crucible will be used to contain the melt. A 100-kW power supply, furnace, vacuum chamber, and controls will be required. The melting area will require ventilation with HEPA filter exhaust.

3.3.3 Casting

The subsections below describe options for casting the uranium ingot.

3.3.3.1 Melt Size and Criticality Considerations

As mentioned in Section 2.1.2, a typical melt size will be 14 kg. The criticality limits of Section 3.2 are for a solid metal sphere and not molten metal. A much more detailed analysis will be required for the melting process at the facility. If a water leak should occur during melting, and water contacts the molten metal, an explosion most likely will be a greater safety concern than a criticality. The explosion most likely will disperse the material before any homogeneous mixing required for criticality can occur.

3.3.3.2 Graphite Molds

Graphite molds are not recommended for the process. Graphite molds must be coated to prevent carbon pickup. They are also prone to erosion and cracking. Graphite will also have to be machined since the mold shape required is not standard.

3.3.3.3 Copper Molds

The use of a permanent water-cooled copper mold has many advantages and is recommended for the process. These molds have long lifetimes and produce an ingot with uniform dimensions and good surface finish. Both of these characteristics are very important for producing foil of uniform thickness and free of pinholes. Ingot removal is also simplified if a split mold is used.

3.3.4 Pipe Removal (Recycle)

The casting operation will generate a pipe on the top of the ingot. This void space must be removed because if it is rolled it will initiate cracks in the ingot. A conventional band saw with fluid coverage will be required. The chips should be recycled and criticality controls will be needed. This equipment is located in the hot shop processing area. The hot shop area will require ventilation with HEPA filter exhausts.

3.3.5 Chemical Analysis

At this stage, a chemical analysis will be required for each ingot to be rolled that contains recycled uranium. Virgin material will not need a second chemical analysis. Since the material is enriched uranium, it would be most economical if transportation of the material were kept to a minimum. An in-house chemical analysis area would be set up. It would contain two spectrometers to do isotopic analysis and trace element analysis.

3.4 INGOT PREPARATION FOR HOT ROLLING

The subsections below describe considerations and options for ingot preparation.

3.4.1 Surface Preparation

Through the use of the copper mold, surface preparation should be kept to a minimum. Any laps or seams should be dressed by grinding before rolling. This grinding equipment will also be located in the hot shop area. This area will easily handle one ingot/48-hour period or up to 6000 kg/year.

3.4.1.1 Storage and Criticality Considerations

If multiple ingots are produced and stored for future use, a system of spaced, sealed containers must be used. These ingots would be stored under vacuum in the uranium storage area, which will be designed to have minimal water intrusion. Ingots will be stored in the uranium sealed-container storage area referred to in Section 3.1.1.

3.4.2 Ingot Protection from Oxidation

As mentioned previously, uranium metal must be protected from oxidation. The speed of the oxidation reaction increases rapidly with increasing temperature. Two methods to prevent oxidation are used: (1) mechanical methods, where the uranium is covered with a protective layer; and (2) atmosphere control, where the uranium is processed in an inert atmosphere. The second method is very costly and will not be considered. Most uranium metal processing uses the first method, and variants of it are discussed below.

3.4.2.1 Steel Canning

Canning the uranium with an oxidation resistant metal (typically iron alloys) is a common procedure where loose contamination must be kept to a minimum. Each can must be machined or stamped to dimensions, assembled, and welded. These steps add significantly to the cost of the processing. This method is not recommended because of the higher cost.

3.4.2.2 Protective Oil and Salt Baths

The rolling processing temperature for uranium can vary from 100 to 800°C. The most efficient temperature to use is somewhere in the range of 620 to 650°C. The required temperature range eliminates the use of oil, which would not be stable. Only molten salts can operate at these temperatures. The choice of salt is up to the operator’s preference. A list of commonly used salts is given in reference (Jackson 1989). Great care must be taken to keep all ingots dry before

insertion into the bath. Water and hot salt react violently. Lead baths were not considered because of toxicity issues.

3.5 HOT ROLLING TO SHEET

Hot rolling to sheet will be performed on a standard 2-high or 4-high rolling mill with a 50-horsepower motor. It is assumed that the mill will be located in a loose-contamination area and will be ventilated through HEPA filters on both sides. The rolling-mill area will contain the protective salt baths and the rolling mills.

3.5.1 Mill Requirements

The ingot must be reduced by hot rolling from 2.5 cm (1 in.) to approximately 0.5 mm (0.020 in.). We have found that a 9-in.-diameter roll is well suited for this size of ingot. A roll width of 6 in. is sufficient for the foil size to be rolled. It is also highly recommended that a 4-high mill for the hot rolling be considered to optimize foil flatness. As long as the ingot is cast to a maximum thickness of 1 in. (2.54 cm) and a width of 3.5 in., the length could be increased to accommodate a larger melt size, or two 25-kg ingots could be rolled in succession. This mill could easily process 50 kg/48 hours.

3.5.2 Reduction Schedule

Appendix B gives a suggested hot rolling schedule. The cold ingot should be heated for a minimum of 30 minutes before the first pass. There are 33 passes, and a 10-minute reheat will be required between passes. The reheating between passes will take approximately 6 hours. The piece will have to be cut twice when the length approaches the salt bath depth (122 cm, or 48 in.). This will add to the time required to complete a pass, so a full 8-hour day will be needed for the hot rolling. One ingot will yield four 133 × 8.9 × 0.078 cm (52 × 3.5 × 0.031 in.) pieces.

3.6 SHEET PREPARATION FOR COLD ROLLING

3.6.1 Cleaning Residue Salts

A wet (water) grit blaster will be used to remove the residual salts. To account for the weight losses, the sheets will have to be weighed before and after treatment. When the total loss reaches 350 g of ^{235}U, the wet-blasting unit will be cleaned and the material will be recycled or disposed of as waste depending on economic considerations. Due to the presence of finely divided uranium in water, this operation will be the second largest criticality concern in the facility. This equipment will be located in the hot shop area.

3.6.2 Shearing to Size and Recycle

After hot rolling, the sheets will be sheared into 65 cm (26 in.) lengths. The edges will be squared up and any cracks or defective sections will be removed. As mentioned in Section 2.1.2, 20% is allowed for cleaning and shearing losses. The sheared pieces will be recycled. To give a conservative estimate of the production schedule, these losses are not included, and the full amount of material is assumed to be used for the cold rolling. The shearing equipment will be located in the hot shop area.

3.6.3 Storage

If the sheets are to be stored, they will require vacuum chambers. Criticality issues must also be addressed for the size of a stored batch. Once again, amount of moderators and reflectors will have to be controlled. The sheets will be stored in the uranium sealed-container storage area.

3.7 COLD ROLLING

The equipment chosen for the cold rolling must match the design requirements of the foil. The typical foil used in a target will be in the range of 125 to 140 µm, with a variance of ±10% (Wiencek et al. 1994). The mill recommended for the cold rolling will be based on the most economical method of achieving 25 µm or better flatness.

3.7.1 Mill Requirements

3.7.1.1 Standard 2-High Mill

Using a standard 2-high mill for the cold rolling will most likely result in foils with a larger-than 25-µm range in thickness. The reason for this is that the rolls bend as the piece passes through the mill. However, there are 2-high mills that can correct for bending. Even though they can produce foils that have a much smaller range in thickness than is required for the annular Argonne ^{99}Mo target design, they are not recommended because they are complex and expensive.

3.7.1.2 Specialty Mill

It would seem advantageous to use a Sendzimir-type mill (U.S. Patent 3815401) because of its ability to roll very flat foils to 125 µm or less. For this process, the 0.078-cm-thick feedstock output of the hot rolled material would be well sized to use as input. However, these machines are typically used for making high-volume foil. The extra cost and complexity may not be justified given the “small” volume of foil required. Even 6000 kg/year would be considered a small volume.

3.7.1.3 4-High Mill

The recommended cold-rolling mill is a standard 4-high. This type of mill minimizes roll bending by adding a larger roll above the work roll. It is a simple, robust, inexpensive (compared to the mills described above) piece of equipment. We have found that a 5-cm (2-in.) diameter work roll is a good match for producing 125-µm foils. The cold rolling mill will be located in the hot- and cold-rolling area mentioned in Section 3.5. This mill will easily be capable of reducing 50 kg of hot rolled input sheet per 48-hour period. The first hot rolling may need to be staggered so that both the hot and cold rolling mills will have material to operate at the same time.

3.7.2 Rolling Schedule

Appendix C gives a suggested cold-rolling schedule. The starting sheet will be eight pieces, each about 65 cm (26 in.) long. There are 36 passes, and since cold working raises the temperature of the piece, a 5-minute cooling period will be required between passes. The total cooling time between passes will be approximately 3 hours. The piece will have to be cut once when the length approaches a reasonable length to handle (200 cm, or 80 in.). Because of these operations and the fact that rolling multiple pieces will add to the time required to complete a pass, a full 8-hour day will be needed for the cold rolling. The final foil will consist of 16 pieces that are 210 cm (84 in.) long. Each piece will yield about 22 foils, for a total of 350 foils (or 7000 g). Taking into account the losses for shearing and rejects, which was estimated to be 25%, approximately 260 good foils will be produced per ingot.

3.8 HEAT TREATMENT AND INSPECTION

Cold working of uranium tends to produce a textured preferential grain-orientation with the (100) plane parallel to the rolling plane (Kaufmann 1962). It has also been shown that highly textured pure uranium will exhibit severe antistrophic growth when irradiated (Kaufmann 1962). Growth of this type might cause the foil to bond to the cladding, which would make removal of the foil after irradiation difficult. In addition, the target might warp under irradiation, making its removal from the irradiation position complicated. In the worst case, the anisotropic growth could cause the target cladding to fail during irradiation. A heat treatment has been developed to produce small, randomly oriented grains (no texture) in uranium foil (Conner et al. 1998). The foil is heated to 720°C in a vacuum-sealed metal bag, forming the beta uranium phase of uranium metal, and then cooled by dropping the sealed pouch into water. The vacuum is needed to prevent oxidation of the foil and to allow for good contact between the pouch and the foil, so the heat can be rapidly removed. The rapid cooling through the beta to alpha crystal structure results in the formation of small and randomly-oriented grains.

3.8.1 Sealing Foils for Heat Treatment Using Tungsten Inert Gas (TIG) Welding

As mentioned in the previous section, Argonne has developed a method of heat treating foils by vacuum sealing them in stainless steel pouches (U.S. Patent 7350280). This method was

developed because of the loss of an electron beam welding facility. Previously, the samples were sealed by electron-beam welding, which also results in a vacuum. It is not recommended that the production facility use this Tungsten Inert Gas (TIG) welding method to seal the pouches.

3.8.2 E-Beam Welding

Electron-beam (e-beam) welding produces excellent welds with an internal vacuum; however, for production a batch process will have to be designed since setting up the samples to be welded and pumping down the chamber requires a minimum of 1–2 hours. If batches of 50–60 foils (10–12 foils, each sealed in five pouches) could be sealed per run, the entire run of 260 good foils could be sealed in 3–5 days. Another possible option is to cut the as-cold-rolled foils into 20-in. lengths, heat treat the uncut foil, and then shear to final size. If capacity is to be increased, it is recommended that an additional e-beam welder is added for every 200 kg/year to be produced.

3.8.3 Heat Treatment and Shearing Open Pouches

The heat treatment will require a furnace large enough to hold five pouches. A vented quench tank and a shear will also be needed. The sheared pouches will be opened in a HEPA filtered hood. This furnace will easily heat treat 6000 kg/year.

3.8.4 Radiographic Inspection of Foils

A diffractometer will be needed to confirm a successful heat treatment or the lack of texture will need to be confirmed by an outside vendor. An economic study of both solutions will have to be done to determine which method to use. One-hundred-percent sampling is not recommended. A much lesser sampling system should be chosen after confirmation that the chosen heat treating procedure is effective.

3.9 FOIL INSPECTION AND QUALITY ASSURANCE

The final foil should be inspected to minimize the number of targets rejected due to foil defects. Because of this, a separate dedicated area will be set up. Since the foils are not clad, the inspection area will have to be a loose-contamination area. A fume hood will need to be set up with inspection equipment. Although uranium oxidizes in air, the rate is such that brief exposure to air causes only a thin oxide layer (which is not protective) to form. Thus, exposure to air should be minimized. The foils will be weighed and dimensions measured.

3.10 STORAGE

After inspection, the foils will need to be stored under vacuum until needed. This will be part of the storage area mentioned in Section 3.4.1.1.

Once again, the uranium must be stored in a manner such that a criticality will not occur. Distance between batches of foil and moderator/reflector minimization will be required. This area may have to be expanded if >250 kg/year of foil are required.

4 CONCEPTUAL LEU TARGET FABRICATION PROCESS

4.1 MACHINING TUBES

Before target assembly, the aluminum tubes must be machined. Machining is performed on a lathe, where the inner tube surface is machined to a smooth defined dimension and an insert is cut into the outer surface of the inner tube to hold the nickel-wrapped uranium foil. A longitudinal scribe mark should also be made on the outer tube to aid in target assembly and disassembly; it is used to show where the gap between the two ends of the foil is located.

A separate clean (nonradioactive) machining area would have to be included if the tubes are to be machined in-house. This area could also be used for other nonradioactive material machining. However, it likely would be better to have this machining performed by an outside vendor. A separate cost analysis is required to determine which method would be more economical.

4.2 MEASUREMENT AND WEIGHING

The inner and outer tubes will need to be inspected prior to expanding. A simple go/no-go gauge type system would allow for 100% inspection. The mass of aluminum will be determined by subtracting the weight of the foil (measured in Section 3.9) from the weight of the final target. This inspection area will be included in the clean inspection area mentioned in Section 3.1.1

4.3 CLEANING

A commercial cleaning bath unit will be used to clean the inner and outer tubes. Cleaning is also performed on the drawn and machined target before the target ends are welded closed. Ventilation will be required with the proper chemical filters. It may be advantageous to investigate biodegradable chemicals for the cleaning. The tube cleaning will be a separate area, and no uranium will be allowed in this area. Additional cleaning baths may be required if >250 kg/year of targets are needed.

4.4 ASSEMBLY

A separate dust-free area is required for assembly. It will be difficult to mechanize this process, and it most likely will have to be done by hand inside a HEPA filtered fume hood. The fission-recoil-barrier foils could be precut to size and jigs could be made for uniformly folding the fission-recoil-barrier foil over the uranium foil. The uranium-nickel/aluminum foil sandwich is wrapped around the inner tube inside the groove cut in the machining. The inner tube is then slipped inside the outer tube while keeping the foil inside the groove. Additional assembly hoods may be required if >250 kg/year of targets are needed.

4.5 DRAWING

It should be noted that although the operation used to mechanically cold work the inner tube/foil/outer tube assembly may involve the use of a draw bench, it is more accurately described as expansion. In this process, the target is inserted into the draw die, and by pulling or pushing a plug or expandable multiple rollers through the inside of a tube, the inner tube is expanded plastically and the outer tube elastically to achieve good contact between the target and the U/Ni foils.

Although the process could be completed on a commercial hydraulic draw bench, these machines are more suited to expand longer (3 m or 10 ft.) tubes. Due to the short lengths required (typically 15–20 cm, 6–8 in.) for the targets, a vertically mounted expansion plug on a hydraulic press would be a better choice. This would allow for setting up of fixtures, which should hold tighter tolerances and increase the production rate. Additional presses or multiple targets may need to be pressed in one cycle if >250 kg/year of targets are needed.

4.6 RADIOGRAPHIC INSPECTION

Radiographic inspection is needed after the expansion to confirm the location of the foil. During expansion, the foil could twist about the center axis. This would cause the uranium to enter the area that is used to cut open the target after irradiation. A low-power unit (150 Kv, 3 ma) will have sufficient power for determining the foil location in the targets. It is recommended that a digital imaging system be used to eliminate the need for film and the chemicals required to develop the film. All images could be archived electronically. This inspection area will be included in the radiographic inspection area mentioned in Section 3.8.4.

4.7 FINISHING TARGET ENDS

After expanding, the ends of the tubes are usually uneven. Machining the target ends is required to reach the correct length by facing off equal amounts of material from both ends to create flat parallel surfaces. At Argonne, a ~0.5 mm × 45° (~0.020 in. × 45°) chamfer is cut on the outer diameter and inner diameter (OD and ID) surfaces after facing. Additional lathes may be required if >250 kg/year of targets are needed.

For production, other methods such as shearing or sawing could be considered.

4.8 IDENTIFICATION

Each target will have a unique identification number. This number will be added to the barcoded uranium tracking information mentioned in Section 3.1.3. It will be up to the final end user whether to have this identification stamped into the target or individually identify targets after receipt from the target fabrication facility. Most likely, the identification will be stamped or pressed into the tube. This will provide positive identification even after irradiation.

4.9 DRAWN TARGET CLEANING AND STORAGE

The targets must be thoroughly cleaned and then stored in a vacuum before welding. Storage in vacuum is essential to minimize the oxidation of the aluminum in the targets ends, which will impede welding. A bright-dip cleaning is used at Argonne to dissolve the aluminum oxide at the target ends before welding.

4.10 E-BEAM WELDING

Since the e-beam welder mentioned in Section 3.8.2 is a major equipment cost, it would make economical sense for it to also be used to the welding of the target ends. Due to the sealed nature of the target, making the final weld is difficult because the internal pressure increase from heating causes porosity in the weld area. Since e-beam welding is done in a vacuum, it eliminates the problem. This welder becomes a limiting factor and additional welders would be required if >250 kg/year of targets are needed.

4.11 SIZING

Welding and identification can cause changes to the outside and inside diameter of the target. Sizing returns the tube to the specified outer diameter and inner diameter. Mechanized equipment like this is commercially available. The sizing area will be included in the target finishing area (Section 4.7).

4.12 LEAK TESTING AND FINAL INSPECTION

The process of leak checking will also need to be changed to a batch operation. If a leak is found in a batch, individual targets in that batch will need to be inspected. Dimensional inspections will also need to be done. The leak testing and final inspection area will be included in the target finishing area.

4.13 STORAGE

4.13.1 Storage Media

The final targets will be stored in individual water tight plastic pouches. These will keep the outer surfaces from being damaged.

4.13.2 Target Storage Criticality Considerations

An analysis will be required to prevent criticality during the storage of the final targets. Credit for preventing a criticality will be taken for the watertight pouches.

4.14 FINAL PACKAGING AND SHIPPING

U.S. Department of Transportation (DOT) requirements will have to be determined for the design of the final packaging container. The DOT requirements will dictate the size and type of container that will be needed to ship the targets to the end user's reactor.

5 QUALITY CONTROL

Many aspects of quality control have been described in the previous sections. Each target will have a computer database record of the entire fabrication history. It will include the mass of the uranium, its composition, the mass of the target, its dimensions, and other data collected during its manufacture. Traceable standards will be used for calibration of all the equipment used to generate the measured data. In addition, random destructive sampling will be done to confirm the uniformity of the fuel/barrier/cladding interface.

6 OFFICE, LOCKER ROOMS, AND RESTROOM SPACE

Space will need to be provided for the managers, secretaries, engineers, and other facility operation personnel. Locker rooms must also be provided to allow for separate clothing to be worn in contamination areas.

7 OTHER EXPENSES

Other major expenses will be associated with a facility of this type. The three most important are acquiring a NRC license to operate with LEU, writing and getting approved an environmental impact statement, and the security/radiation protection requirements imposed by the NRC license.

8 LEU-FOIL AND LEU-TARGET FABRICATION INCORPORATED IN A U-Mo MONOLITHIC-FUEL FABRICATION PLANT

A similar process and facility has been conceived for the fabrication of U-Mo monolithic fuel plates (Wachs et al. 2008); however, the scope of the fuel-fabrication endeavor is much larger. It was estimated that 17,000 foils (15,000 fuel plates) will be required each year. At a typical 100-g LEU/U-10 Mo fuel-plate, this equals 1700 kg/year, or roughly seven times more LEU required than the ^{99}Mo process. Economically, it makes sense to combine these two facilities.

Because the fuel plates require alloyed U-10 Mo foils and the target-fabrication process uses pure uranium, it may be prudent to have two separate lines that do not use any common processing equipment. In this case, the savings would come from combining the safeguards and NRC licensing expenses for the fuel plate fabrication facility with the target-fabrication facility. These expenses will be quite large and will only be minimally increased with the addition of the target requirements. All other expenses should be the same if the facilities were built separately.

9 CONCLUSIONS

A high-level study was conducted to find the unit operations and considerations required in a facility designed to meet U.S. demand for LEU-foil targets for the production of ^{99}Mo over the next decade. The option of incorporating this facility into the U-10 Mo monolithic fuel fabrication facility was also addressed. This study should give a commercial enterprise interested in pursuing production of targets enough information to (1) understand what is required in the facility and (2) perform a high-level cost estimate to build and operate the facility.

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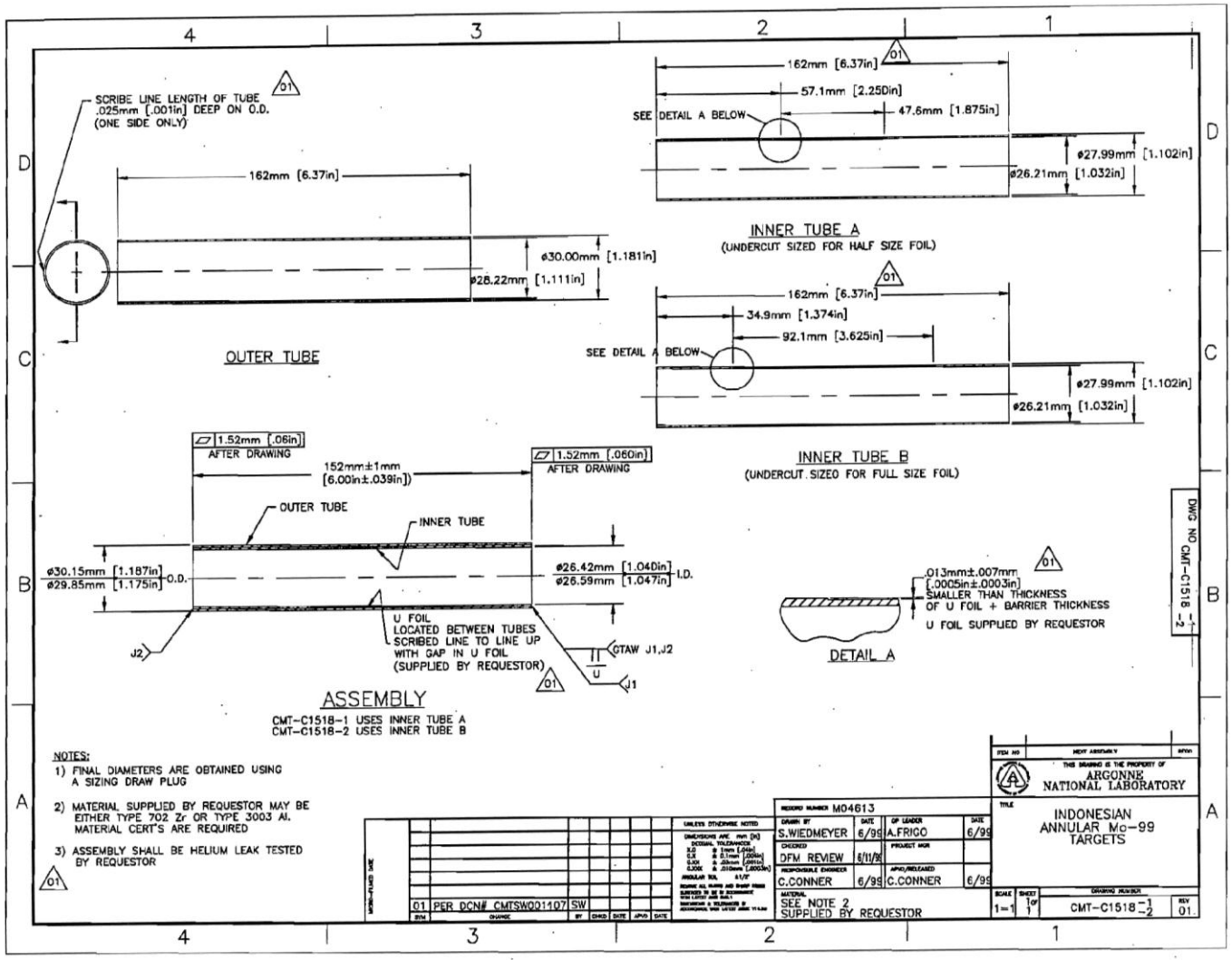
APPENDIX A:

INDONESIA TARGET



APPENDIX B:

HOT ROLLING SCHEDULE

Pass Number	Reduction in Thickness	Thickness (cm)	Thickness (in.)	Length (cm.)	Length (in.)
0	-	2.540	1.000	16.400	6.530
1	10%	2.286	0.900	18.222	7.256
2	10%	2.057	0.810	20.247	8.062
3	10%	1.852	0.729	22.497	8.957
4	10%	1.666	0.656	24.996	9.953
5	10%	1.500	0.590	27.774	11.059
6	10%	1.350	0.531	30.859	12.287
7	10%	1.215	0.478	34.288	13.653
8	10%	1.093	0.430	38.098	15.170
9	10%	0.984	0.387	42.331	16.855
10	10%	0.886	0.349	47.035	18.728
11	10%	0.797	0.314	52.261	20.809
12	10%	0.717	0.282	58.068	23.121
13	10%	0.646	0.254	64.519	25.690
14	10%	0.581	0.229	71.688	28.544
15	10%	0.523	0.206	79.654	31.716
16	10%	0.471	0.185	88.504	35.240
17	10%	0.424	0.167	98.338	39.155
18	10%	0.381	0.150	109.264	Cut 43.506
19	10%	0.343	0.135	60.702	24.170
20	10%	0.309	0.122	67.447	26.855
21	10%	0.278	0.109	74.941	29.839
22	10%	0.250	0.098	83.268	33.155
23	10%	0.225	0.089	92.520	36.839
24	10%	0.203	0.080	102.800	40.932
25	10%	0.182	0.072	114.222	Cut 45.480
26	10%	0.164	0.065	63.457	25.267
27	10%	0.148	0.058	70.507	28.074
28	10%	0.133	0.052	78.342	31.193
29	10%	0.120	0.047	87.046	34.659
30	10%	0.108	0.042	96.718	38.510
31	10%	0.097	0.038	107.464	42.789
32	10%	0.087	0.034	119.405	47.544
33	10%	0.078	0.031	132.672	Cut 52.826

APPENDIX C:

COLD ROLLING SCHEDULE

Pass Number	Reduction in Thickness	Thickness (μm)	Thickness (in.)	Length (cm)	Length (in.)
0	-	784.9	0.0310	66.34	26.41
1	5%	745.7	0.0295	69.83	27.80
2	5%	708.4	0.0280	73.50	29.26
3	5%	673.0	0.0266	77.37	30.80
4	5%	639.3	0.0252	81.44	32.42
5	5%	607.4	0.0240	85.73	34.13
6	5%	577.0	0.0228	90.24	35.93
7	5%	548.2	0.0216	94.99	37.81
8	5%	520.7	0.0206	99.99	39.80
9	5%	494.7	0.0195	105.25	41.90
10	5%	470.0	0.0186	110.79	44.10
11	5%	446.5	0.0176	116.62	46.42
12	5%	424.2	0.0168	122.76	48.86
13	5%	402.9	0.0159	129.22	51.43
14	5%	382.8	0.0151	136.03	54.14
15	5%	363.7	0.0144	143.18	56.99
16	5%	345.5	0.0136	150.72	59.98
17	5%	328.2	0.0130	158.65	63.14
18	5%	311.8	0.0123	167.00	66.46
19	5%	296.2	0.0117	175.79	69.96
20	5%	281.4	0.0111	185.05	73.64
21	5%	267.3	0.0106	194.78	Cut 77.51
22	5%	254.0	0.0100	205.04	40.90
23	5%	241.3	0.0095	215.83	43.06
24	5%	229.2	0.0091	227.19	45.32
25	5%	217.7	0.0086	239.14	47.70
26	5%	206.8	0.0082	251.73	50.21
27	5%	196.5	0.0078	264.98	52.86
28	5%	186.7	0.0074	278.93	55.64
29	5%	177.3	0.0070	293.61	58.56
30	5%	168.5	0.0067	309.06	61.64
31	5%	160.1	0.0063	325.33	64.88
32	5%	152.1	0.0060	342.45	68.30
33	5%	144.4	0.0057	360.47	71.89
34	5%	137.2	0.0054	379.44	75.67
35	5%	130.4	0.0051	399.41	79.65
36	5%	123.8	0.0049	420.44	83.84

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