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STATUS AND PROGRESS OF FOIL AND TARGET FABRICATION ACTIVITIES FOR THE PRODUCTION OF ^{99}MO FROM LEU

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ABSTRACT

Argonne National Laboratory has produced most of the hot-rolled uranium foils used in experiments to produce ^{99}Mo from LEU-foil targets. We are working with the Indonesian BATAN and several other members of the IAEA coordinated research project (CRP) on "Small-Scale Indigenous Production of ^{99}Mo Using LEU Targets or Neutron Activation" to develop their own capability to roll foils. This paper details the foil fabrication and other activities at Argonne, in particular, (1) characterization of direct-cast foils supplied by KAERI as an alternative to Argonne's hot rolled foils and (2) a high-level, preliminary study of the requirements (equipment and footprint,) related to producing rolled foil and targets for industrial-scale production of ^{99}Mo using the Argonne annular LEU-foil target. Preliminary findings do not eliminate the feasibility of using the direct-cast foils for demonstration purposes.

1. Introduction

Because of its potential use in improvised nuclear devises, transport of fresh high enriched uranium (HEU) and storage and disposal of spent HEU from Mo production present a global threat.[1,2] Conversion to low enriched uranium (LEU) with subsequent removal of HEU from commerce would make the world a safer place.[3]

Currently, 95% of all ^{99}Mo is produced by irradiation of HEU targets that are subsequently processed primarily to recover the molybdenum.[4]

The four major producers of ^{99}Mo, all of them utilizing HEU targets and dedicated processing facilities, are:

- MDS Nordion/AECL (Canada)
- Mallinckrodt (Netherlands)
- IRE (Belgium)
- NTP Radioisotopes (Pty) Ltd (South Africa)

A smaller 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 very near future. The remaining 5% of global ^{99}Mo production is primarily derived from the irradiation of LEU targets. ANSTO Radiopharmaceuticals and Industrials (ARI, Australia) has 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).[4] The specific activity of fission-product ^{99}Mo is 10,000 times higher than absorption ^{99}Mo, allowing the use of the present-day ^{99m}Tc generator, where one Ci of ^{99m}Tc can be eluted into 5 mL of saline solution.
Production of fission-product ^{99}Mo can be characterized in five steps: (1) Foil production (2) target fabrication, (3) target irradiation, (4) dissolution or digestion of target and/or uranium fuel, (5) recovery and purification of molybdenum from all other target components. A significant fraction of this paper describes the microstructure of direct-cast foils provided by KAERI and compares these foils to Argonne rolled foils. The cost, availability, quality and irradiation behavior of LEU foils will be key to any large scale ^{99}Mo production program using the annular targets designed at Argonne.

Other fabrication activities at Argonne related to ^{99}Mo development are also briefly described.

2. Results and Discussion

The primary tool for allowing conversion is the Argonne-developed LEU-foil annular target (Figure 1). In this target, uranium foil is drawn between two Al tubes. Prior to drawing, an Al or Ni fission-recoil barrier is wrapped around the U foil to prevent bonding of the U foil to the target walls during irradiation, so foil can be removed from the target prior to dissolution/digestion and ^{99}Mo recovery. Because of the high density of uranium metal, the annular LEU-foil target can contain much greater quantities of uranium compared to aluminide, silicide or oxide targets and, therefore, can produce equivalent yields in the same irradiation position. An example of how this can be accomplished is to look at the possible conversion of the current IRE HEU target.[5] The IRE target is in the form of an HEU-aluminide dispersion plate that has been curved and then welded into an annular target. The current target contains 3.7 g of ^{235}U at 93% enriched uranium. An LEU-foil target with a standard 135-µm thick foil of the same dimensions of 19.9% enriched uranium would contain 4.1 g of ^{235}U. If the LEU foil were made as thick as the current fuel meat (510 µm), the target would contain 16.6 g of ^{235}U.

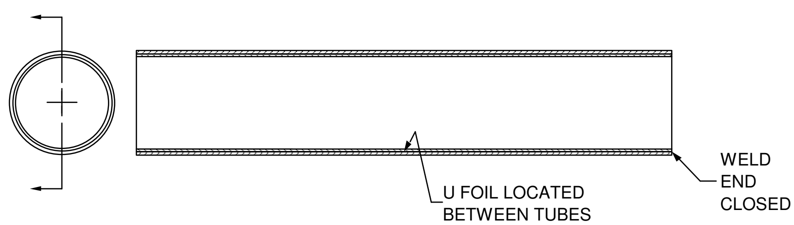

Figure 1. Schematic of the Argonne annular foil target[6]

A key step in the large scale production of annular targets is the availability of LEU foils with good irradiation behaviors.

Examination of Direct-Cast Foils

The Korean Atomic Energy Agency (KAERI) has developed a method to fabricate thin uranium foils economically using a direct casting method.[7] They can produce a continuous polycrystalline LEU foil with an average thickness range of 100 to 150 µm and a width of about 5 cm, exceeding 5 m in length per batch. The foil by this production method is not as uniform as that produced by rolling, and the side that is not in contact with the cooling roller is rougher than rolled foils.(Figure 2). A typical transverse cross section had a minimum value of 65 µ (0.0026 in.) and a maximum of 205 microns (0.0082 in.)

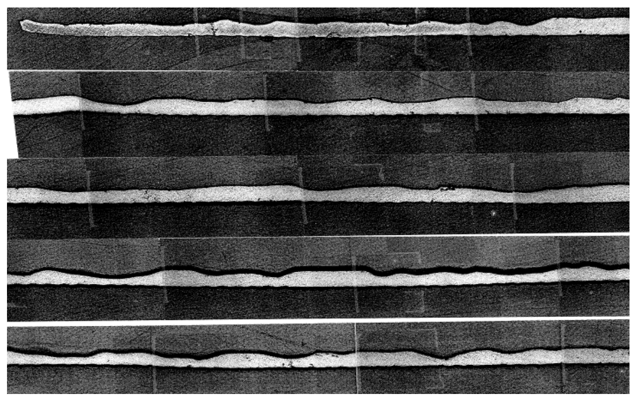

Figure 2. Optical 12X cross-sectional micrographs of as-received KAERI LEU foils (bright areas). The measured foil thickness ranges from 65 to 205 µm.

The reason for this roughness is twofold. Since the outer foil surface is not constrained, surface tension will cause peaks and valleys to form while the uranium is molten. Once formed, these grains will seek to continue to grow parallel to the casting (heat removal) direction. Figure 3 is a low magnification photograph of the free surface of a foil showing the elongated grains and the "hill and valley" surface.

Casting Direction ➔

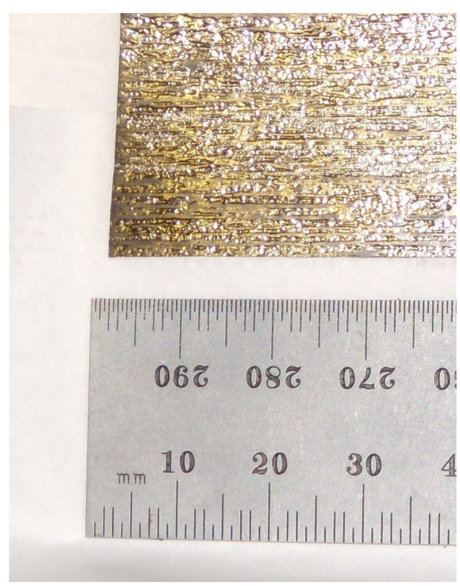

Figure 3. Typical unconstrained (free) side surface of direct-cast uranium foil

This could affect (1) target fabrication, where the U foil, the barrier foil, or the target walls may be damaged during drawing, and (2) irradiation behavior, where gaps between the target walls and the U metal could affect cooling of the targets. Argonne initiated a R&D effort to study the roughness of these foils, potential means to smooth them by rolling techniques, and any effects that the observed uniformity had on target fabrication.

Cold Rolling
To reduce the surface roughness, it was decided that cold rolling would be the quickest and simplest method. Samples for cold rolling were prepared by shearing a typical foil along the casting direction and perpendicular to the casting direction (Figure 4).

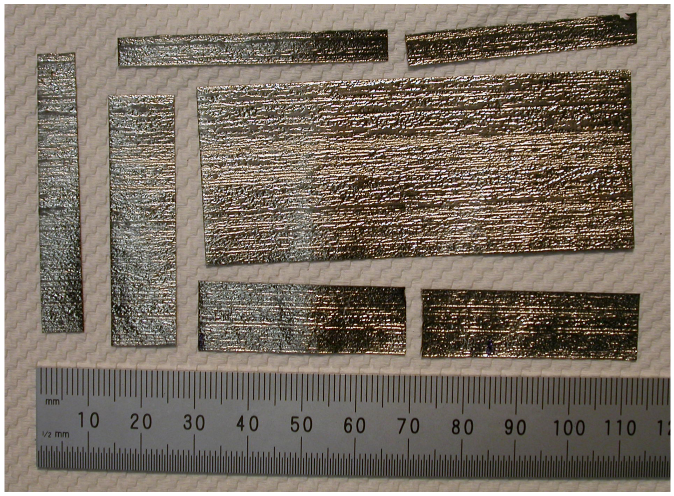

Figure 4. Optical photograph of sectioned direct-cast LEU foil used for experiments

Two test samples were cold rolled 25% reduction in thickness parallel and perpendicular to the casting direction and then cold rolled an additional 25% (44% total cold reduction). Rolling of the small (1 cm x 2 cm) pieces of the foil perpendicular and parallel to the casting direction went very well with the foils staying quite flat. No holes were seen in the foils. The samples after cold rolling are shown in Figure 5.

Samples at 150 µ and 110 µ and rolled both parallel and perpendicular (four samples total) were mounted and polished to examine the cross sections after rolling and measure thicknesses. Figures 6 and 7 show cross sections cut perpendicular to the casting direction for both rolling directions. Table 1 gives the results of the measurements. A 59% reduction in thickness variation was measured after 44% cold reduction for the sample rolled parallel to the casting direction. This is a major improvement but is still approximately 40% higher than the 40-µ range for a typical hot and cold rolled foil.[8]

Table 1. Difference between maximum and minimum thickness µ (in.) for KAERI direct-cast natural uranium Foil

	As-Received Range of Thickness µm (in)	After First Cold Roll Range of Thickness µm (in)	After Second Cold Roll Range of Thickness µm (in)
Rolled Parallel to Casting Direction	137 (0.0054)	71 (0.0028)	56 (0.0022)
Rolled Perpendicular to Casting Direction	137 (0.0054)	99 (0.0039)	74 (0.0029)

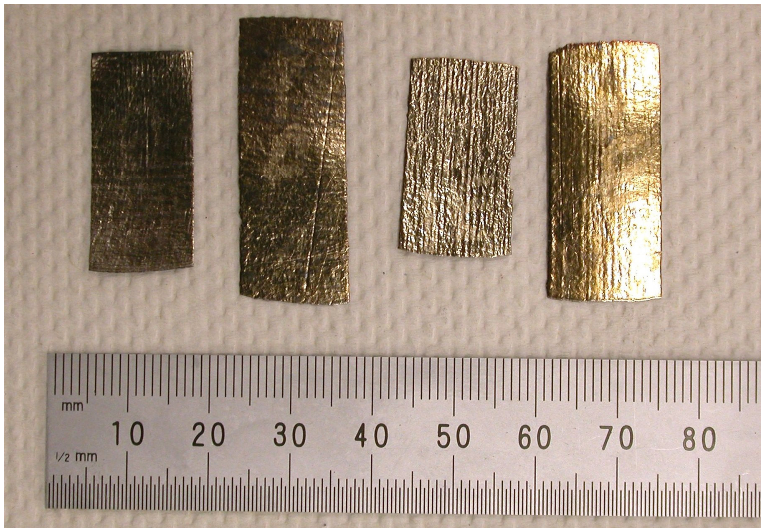

Figure 5. Cold rolled direct cast foils (from left to right), 25% and 44% reduction perpendicular to the casting direction, and 25% and 44% reduction parallel to the casting direction

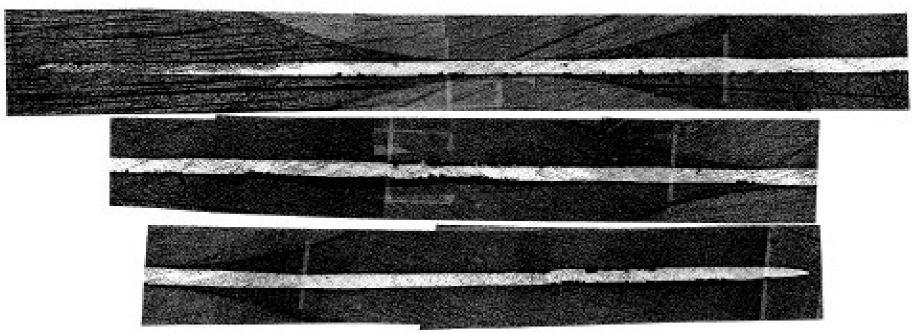

Figure 6. Cross section cut perpendicular to the casting direction for sample cold rolled 44% parallel to the casting direction

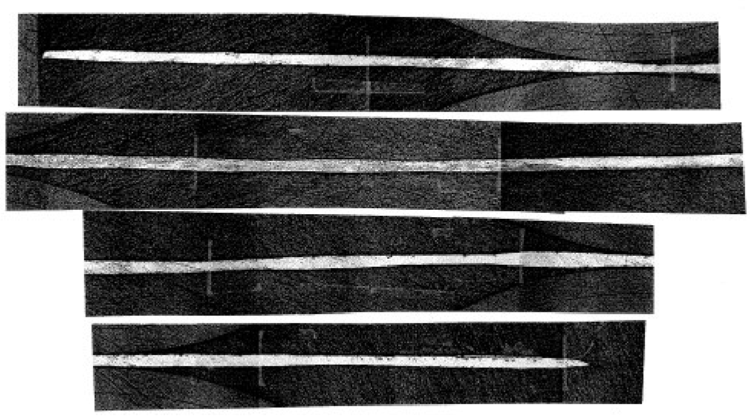

Figure 7. Cross section cut perpendicular to the casting direction for sample cold rolled 44% perpendicular to the casting direction

Full Size Direct-Cast Foil Rolling

Once an optimized method for cold rolling was established, a full-size (5.5 x 10 cm) direct-cast KAERI foil was easily cold rolled from 200µm to 125µm. After cold rolling, the thickness was measured using a 3 x 5 point array and a flat end micrometer. This information is shown graphically in Figure 8. The readings varied from a maximum of 155 µm to a minimum of 110 µm. The resulting 45 µm range is similar to the range typically seen for standard hot and cold rolled foils. However, it is very important to note that two different methods were used to measure the foil. The as-received values are from a previous foil which was sectioned for the rolling method optimization tests and measured metalographically. Since we planned to use the full-size foil for target assembly tests, it was not sectioned. Thickness measurements were made using a flat end micrometer. A point end micrometer was not used because previously it was found that it left impressions on the foil after each measurement. Intuitively, it seems impossible that the minimum thickness for the cold rolled foils could be less than the as-received foils and indeed it is. Optical examination revealed multiple pin holes in the foil which were too small to measure with a flat end micrometer. Using back lighting, the pin holes are clearly seen as shown in Figure 9. The holes were most likely caused by the surface roughness of the non-roll contact side. The thin areas between the thick areas did not have sufficient material to elongate as much as the thick areas and were stretched to failure. If we use zero as the true minimum thickness, then the max/min range would be 155 µm which is very similar to the as-received value of 137 µm. Based on these data and observations, cold rolling does not improve the flatness of the foil; it just lowers the average thickness.

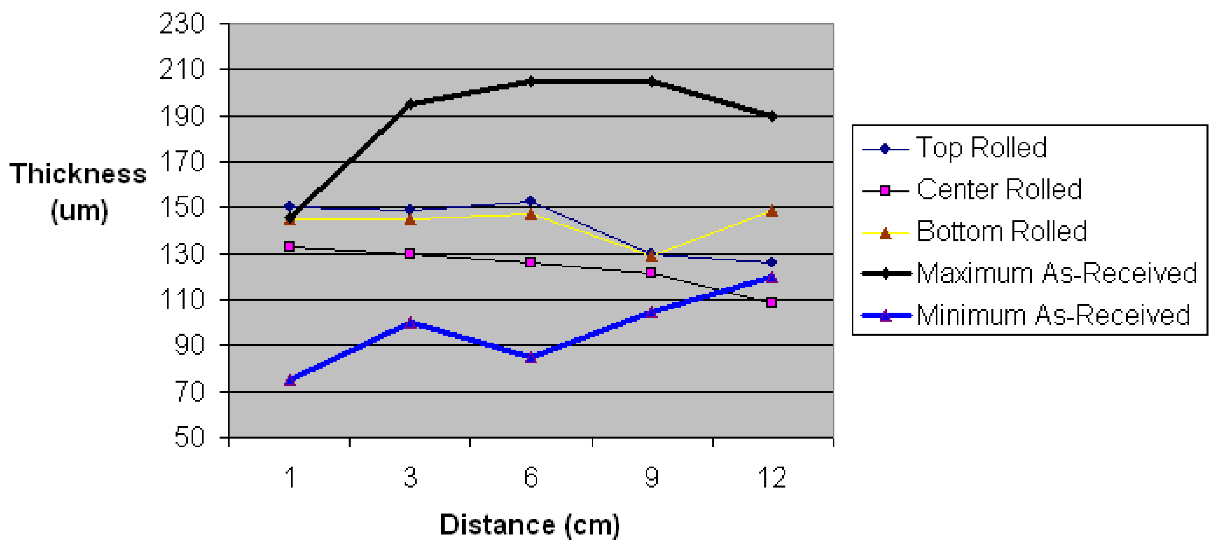

Figure 8. Range of thickness readings for as-received direct-cast LEU foil and for as-cold rolled direct-cast LEU foil

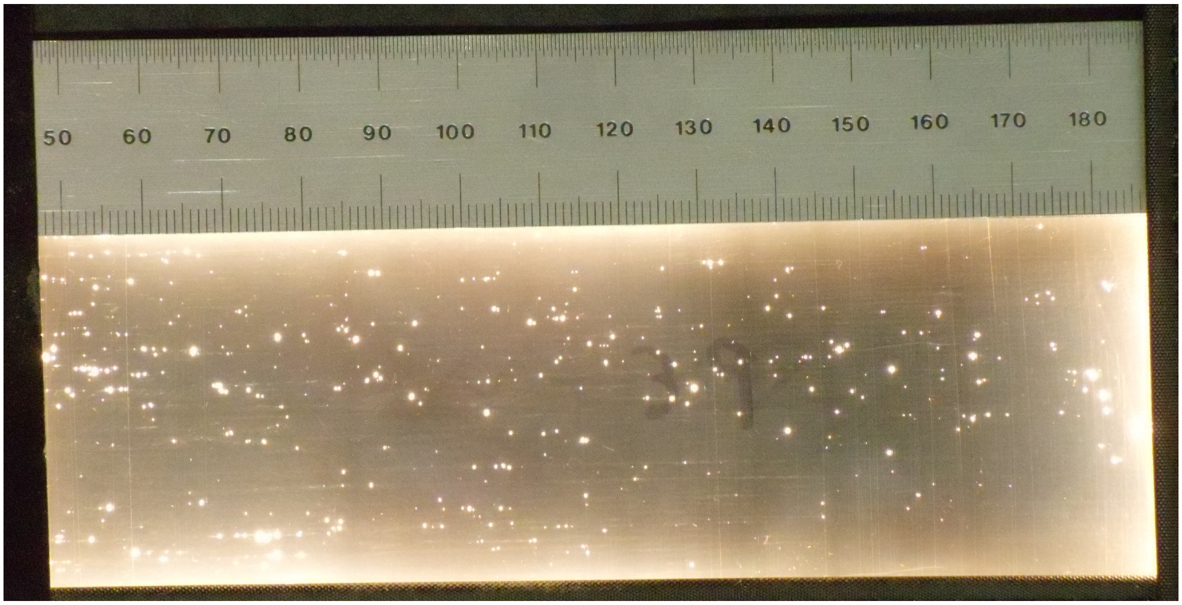

Figure 9. Direct-cast LEU foil after cold rolling showing pin holes

X-ray Diffraction Results

Cold working of uranium tends to produce a textured (aligned grains) preferential grain orientation with the (100) plane parallel to the rolling plane.[9] It has also been shown that highly textured pure uranium will exhibit severe antistrophic growth when irradiated**.**[9] 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, or no texture, in uranium foil.[10] 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 foil in water. The rapid cooling results in the formation of small and randomly oriented grains. The extent of grain orientation in a foil sample is evaluated by comparing its XRD pattern to that of a standard, randomly oriented foil. Table 2 lists the major uranium peak intensities for uranium metal with random grain orientation.

Table 2. Major uranium peak intensities for texture free uranium[11]

Crystal Plane	(021)	(110)	(111)	(002)	(112)	(131)
Intensity	100%	73%	54%	51%	41%	32%

X-ray diffraction measurements were taken on both sides of the as-received foil, the as rolled foil rolled parallel and perpendicular to the casting direction and the heat treated foils (also both rolling directions). These results were compared to the data shown in Table 2, and the data are plotted in Figure 10. A ratio (hlk/021) matching the standard (1, 0.73, 0.54, 0.51, 0.41 and 0.32

respectively) indicates that the extent of grain orientation in the sample is the same as in the standard, i.e., highly random grain orientation. High ratios indicate a highly oriented grain structure.

As would be expected, the cooling-wheel side surface (WS in Figure 10) which has the fastest cooling rate has the least amount of texture. The free side of the foil (FS in Figure 10) has higher grain orientation than the cooling-wheel side, which can be explained by its longer cooling time. Overall, the amount of texture is quite small, and foils produced by this method most likely will not require any heat treatment before irradiation.

The as-cold-rolled foils (AR in Figure 10) exhibit a very large amount of texturing after rolling. The directionality of the foil results in more deformation being given to the material when rolled in the casting direction. This causes the sample which was rolled parallel to the casting direction to have less grain orientation than the sample cold rolled perpendicular to the casting direction after similar hear treatments.

The extent of grain orientation in the foils that were heat–treated and cold-rolled parallel to the casting direction (HT Par) is slightly greater than in the as-received foil. Preliminary results suggest that the amount of texture is small and should not affect the irradiation of targets made from direct-cast foil processed this way.

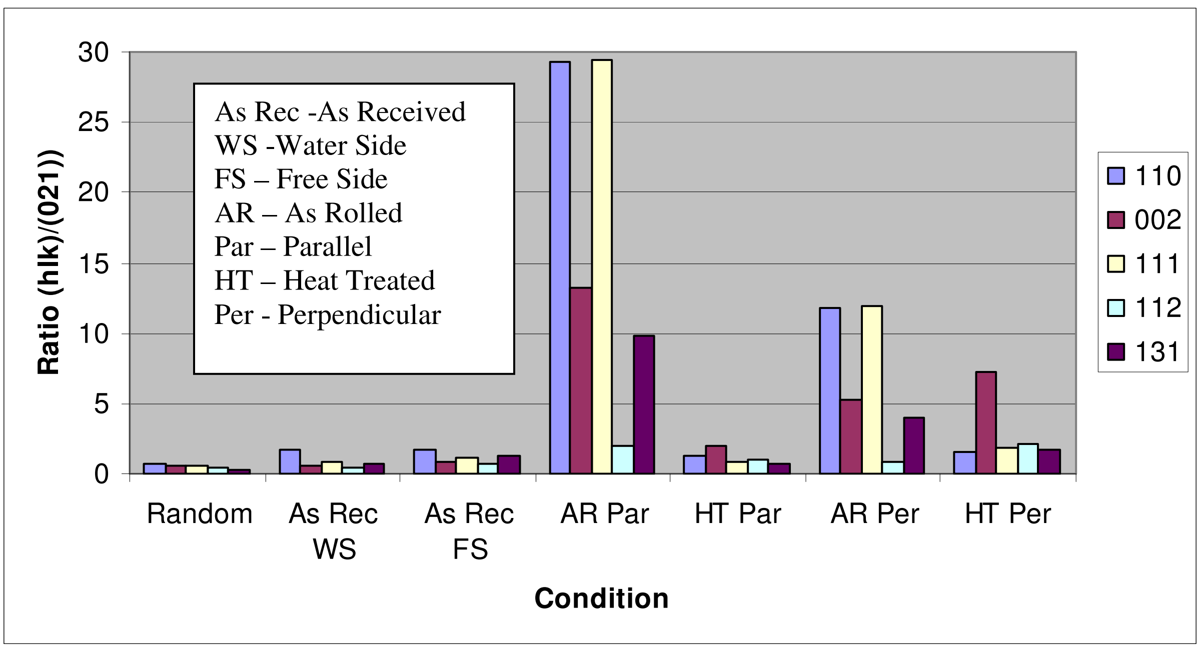

Figure 10. Relative ratios of (hlk) peak to (021) peak for various LEU foil conditions

Target Fabrication with Direct-Cast KAERI Foil

Targets were produced using the standard Argonne production tooling. As can be seen from Figure 11, where the foil (dark area) is uniform in thickness, the contact is quite good between the foil, the nickel recoil barrier (light grey) and the 3003 aluminum cladding. However, gaps between the free side of the direct-cast foil and the recoil barrier were also observed in different areas of the same sample (Figure 12). Calculations done by ANSTO[12] (Australian Nuclear Science and Technology Organization) and MURR[13] show that small gaps within the annular targets will add little or no risk to the irradiation step of the ^{99}Mo production process. However, no irradiation data of direct-cast foils is currently available.

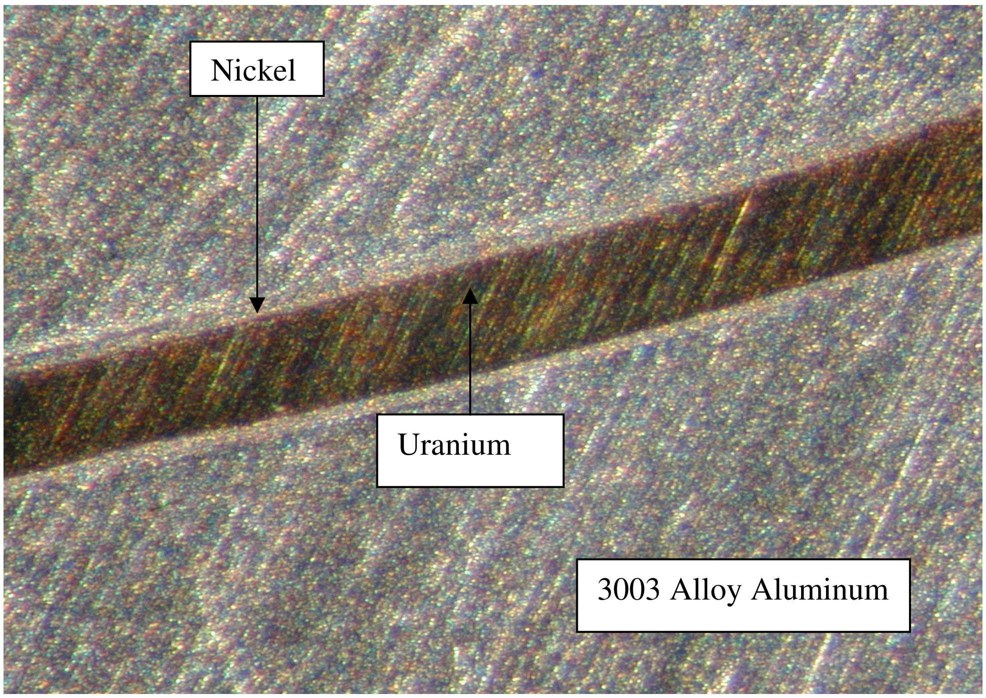

Figure 11. Optical micrograph of a cross section of an annular target using direct-cast LEU foil. Note the excellent contact along each interface.

Other Activities

Production of LEU Foil for MURR ^{99}Mo Target

As discussed in other papers at this meeting, we have been cooperating with MURR to test an LEU target for the production of ^{99}Mo.[14,15] Because of the concern of possible gaps in a target using direct-cast LEU foil, Argonne was asked to fabricate LEU foil for MURR by the standard hot and cold rolling method. A 75-g ingot was rolled and a 5-g section was inserted into a target, which was shipped to MURR. The nominally 5 gram foil had a range in thickness of 38 µm (0.0015 in.) and is shown in Figure 13.

Fabrication of LEU Ingots for BATAN, Indonesia

As part of our cooperative research with BATAN, Indonesia, Argonne has agreed to ship 1 kg of pure LEU to BATAN. The LEU was sized into 75 g ingots to minimize shipping costs and to match their production plans. Figure 14 shows six completed ingots before shipping. A total of thirteen ingots were produced and it is expected that the final shipment of six ingots will be made soon.

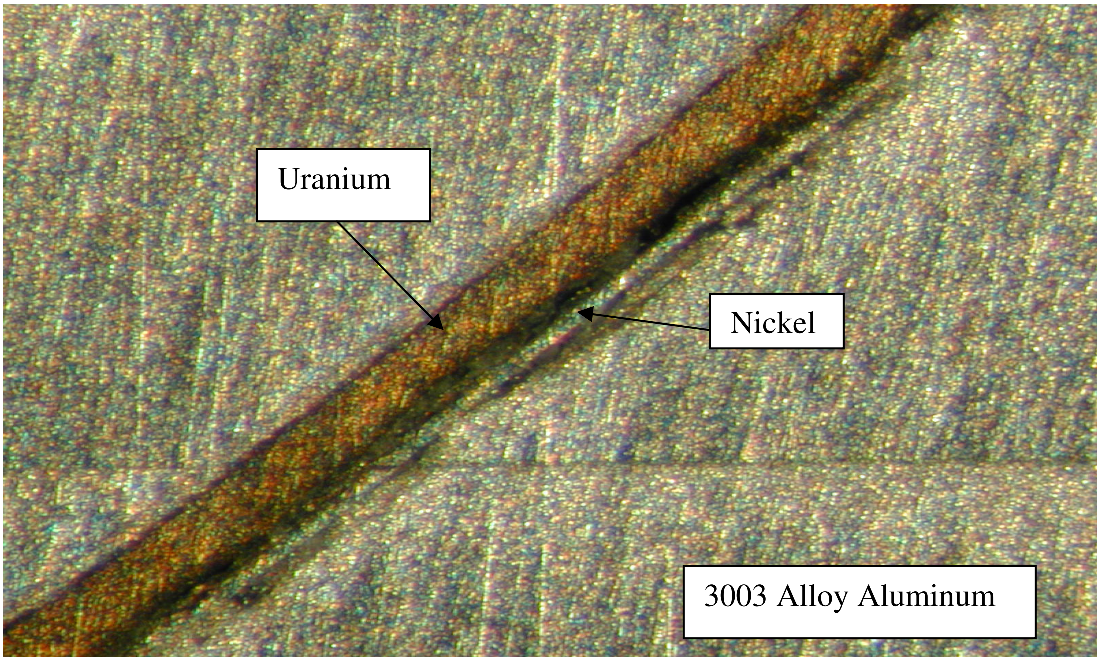

Figure 12. Optical micrograph of a cross section of an annular target using direct-cast LEU foil. Note the poor contact along the lower interface between the uranium and the nickel interface.

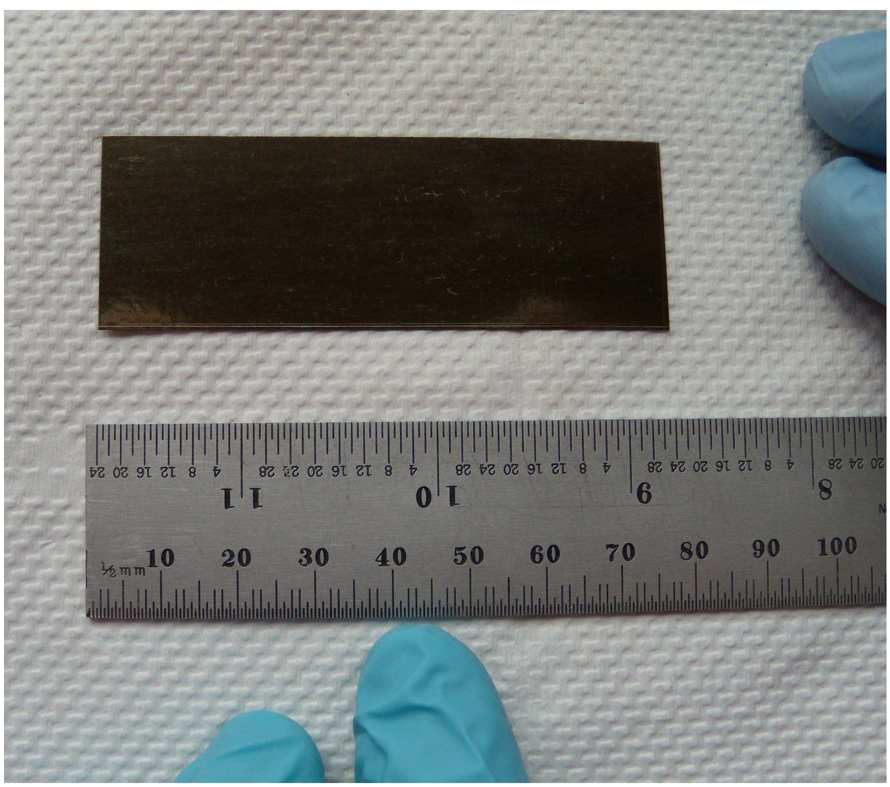

Figure 13. Hot and cold rolled LEU foil produced by ANL for MURR

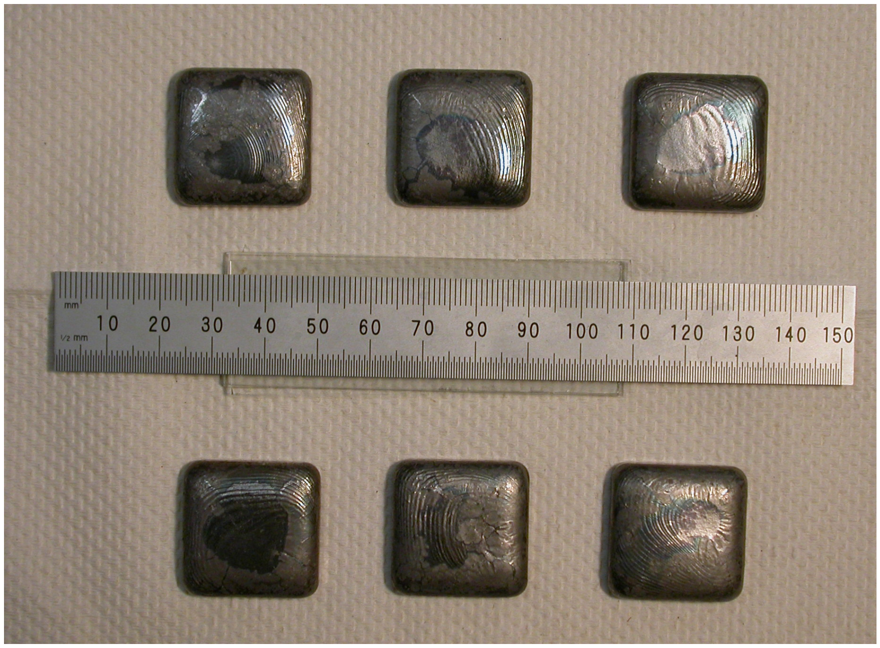

Figure 14. Arc-cast LEU ingots produced by ANL for BATAN

Pre-Conceptual Design of an LEU-Foil and Target Production Capability

Argonne is preparing a high-level preliminary study of the requirements (equipment and footprint,) related to producing rolled foil and targets for industrial-scale production of ^{99}Mo using the Argonne annular LEU-foil target. To meet 100% of the current US demand, approximately 6,000 20-g. foils are required. The design will be based on a production capability of 13,000 20-g. foils to allow for growth in the US demand.

There are multiple processes that can be used to produce foils. A few assumptions are made regarding the process. First, the incoming material will be supplied as LEU, the fabrication method must be a proven technology, 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, and melted. A defect-free ingot is then heated (protected from oxidation) 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 waiting further processing, they are stored under a protective atmosphere. A flow chart of this general foil making process is given in Figure 15.

The cladding of the LEU foil begins with the addition of the nickel recoil barrier. 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 tube is inspected for leaks and dimensions. The tubes that pass inspection are then stored under vacuum until they are shipped. Figure 16 shows a flow chart of the target making process.

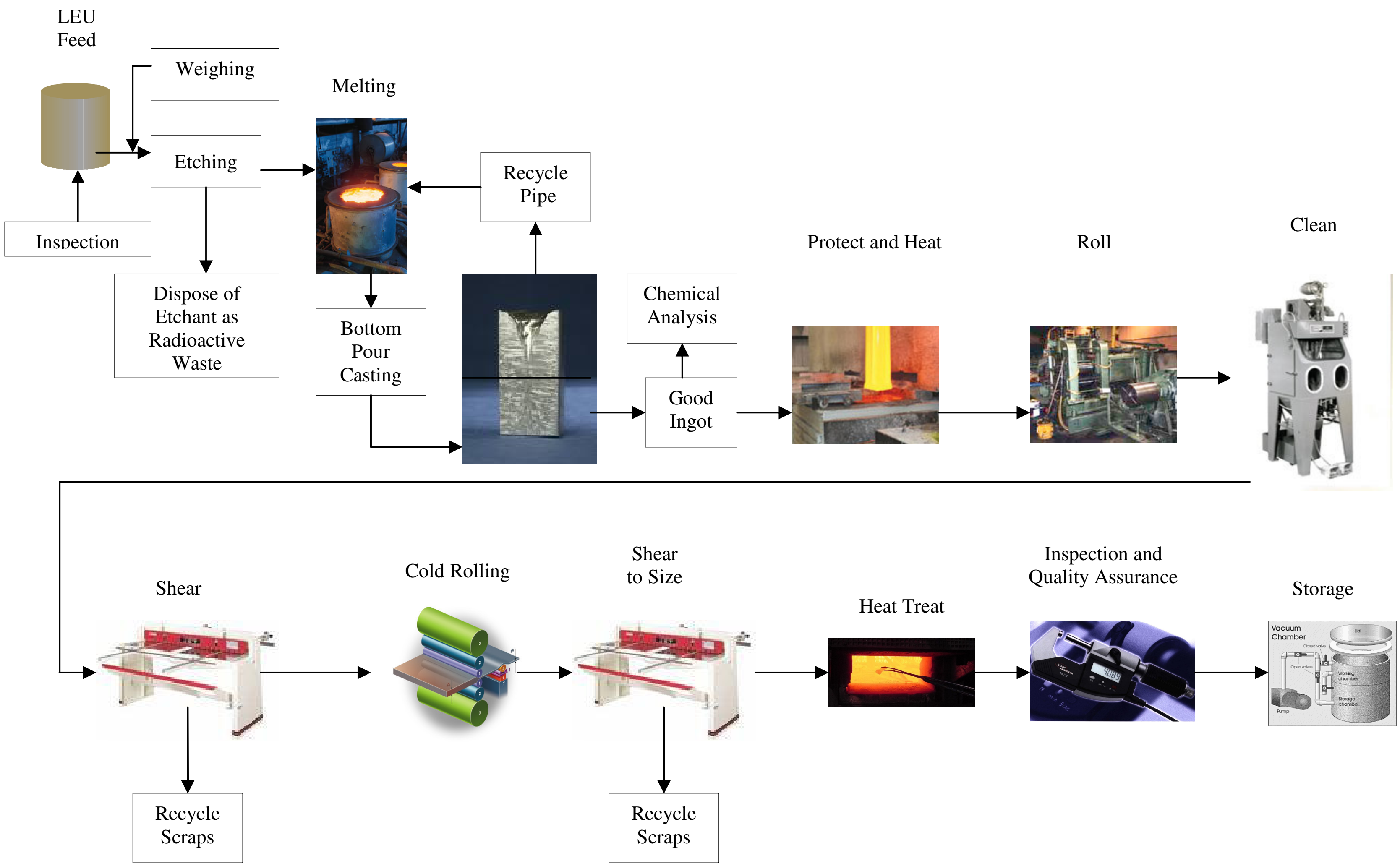

Figure 15. Flow chart of foil making process

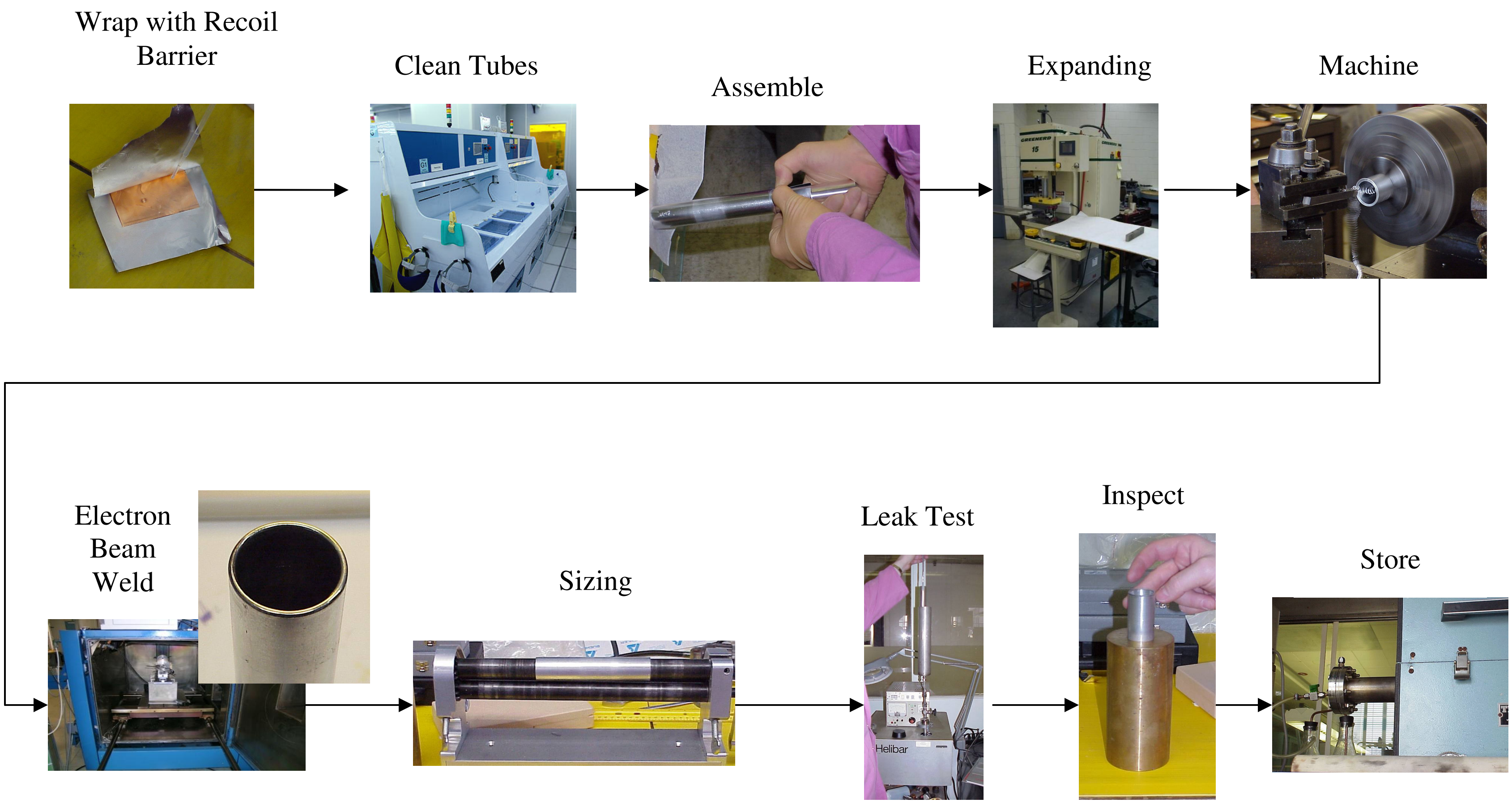

Figure 16. Flow chart of target assembly

3. Conclusions

KAERI's direct-cast LEU foils intended for use in annular targets exhibit more surface roughness and variations in thickness than previous foils that were hot and cold rolled at Argonne. Preliminary analysis of the direct-cast foils showed little preferential grain orientation, which eliminates the need for any post-casting heat treatment. Cold rolling the direct-cast foils resulted in highly oriented grains and necessitated a heat treatment. Heat treating the cold rolled foils randomized the grain orientation. The addition of these two steps would significantly increase the cost of producing a foil.

Targets were fabricated using the direct-cast foils contained some gaps between the uranium and nickel foils. Calculations from ANSTO and MURR suggest that the gaps observed might not affect the irradiation behavior of the targets. However, no irradiation data of direct-cast foils is currently available.

These preliminary findings confirm the feasibility of using these direct-cast foils for demonstration purposes. If irradiation shows that the surface roughness does affect behavior of the KAERI foils, further treatment may be necessary or an advanced method of direct roll-casting may need to be developed.

4. Acknowledgments

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