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UPDATE ON URANIUM-MOLYBDENUM FUEL FOIL FABRICATION ACTIVITIES AT THE Y-12 NATIONAL SECURITY COMPLEX

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ABSTRACT

The Y-12 National Security Complex (Y-12) is a key participant in the NNSA NA-21 Convert Program, also known as RERTR. In 2010, Y-12 continued to fabricate uranium-molybdenum material to support Fuel Development and Fuel Fabrication Capability activities. In 2010, Y-12 began implementing a production based zirconium clad foil fabrication process, using depleted uranium-molybdenum (DU-10Mo) as a low enriched uranium–molybdenum (LEU-10Mo) surrogate material. This work performed by the Y-12 Technology Development Group provides a direct transition to the Y-12 Uranium Production facilities and allows Y-12 to demonstrate process parameters in a pilot facility scale. In addition to production development activities, Y-12 examined and improved the chemical analysis techniques used in the process. The purpose of this report is to update the RERTR audience on Y-12 efforts to implement a production process to support the RERTR U-Mo program goals.

1. Introduction

The Reduced Enrichment for Research and Test Reactors (RERTR) Program was initiated by the U.S. Department of Energy (DOE) to develop the technical means for the conversion of high powered research reactors (HPRRs) from HEU to LEU. The RERTR program cooperates with the research reactor community to achieve this goal of HEU to LEU conversion while maintaining reactor reliability and performance. The goal of the RERTR program is to complete the conversion of all HPRRs.

In support of the RERTR Program, Y-12 supports both the Fuel Development and Fuel Fabrication Capability initiatives. Y-12 provides both source material and fabricated foils to support testing by the Fuel Development program. Y-12 also supports the Fuel Fabrication Capability in transitioning the program from a laboratory scale operation to a production scale operation.

2. Foil Fabrication Activities at Y-12

2.1 Baseline Foil Fabrication Process

This year Y-12 implemented the baseline foil fabrication process development by the Idaho National Laboratory (INL). A general schematic of the baseline fuel process is shown in **Figure 1**.

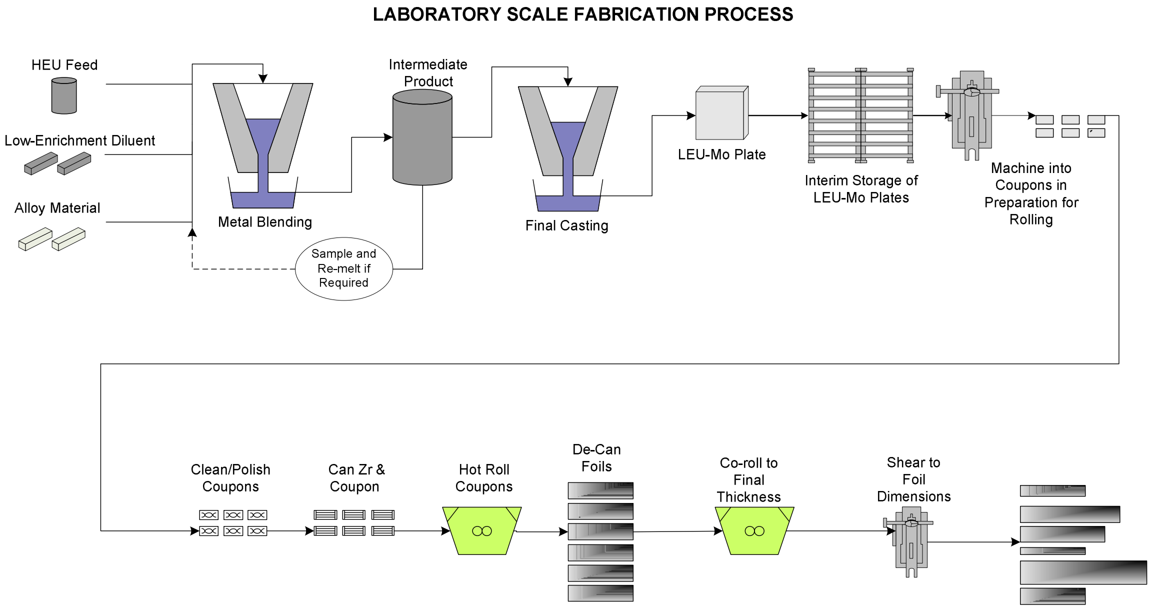

Figure 1: Laboratory Scale Fabrication Process

The feed material used in the fabrication of monolithic U-Mo foils begins as uranium metal and may be of DU, LEU or HEU enrichment. At this time, no significant quantities of EU-Mo alloys currently exist to use as feed material. The initial step in the fabrication process is the blending or alloying of feed material to obtain a homogenous U-10Mo alloy. This is completed in two separate casting steps. In the pre-melt casting, the uranium metal is combined with the molybdenum powder to form an intermediate product. At this point, the product is sampled in multiple locations to ensure the homogeneity of the alloy. The final casting provides a plate shaped casting, which is then cut into coupons. Typically, one coupon is used to fabricate one U-Mo foil.

Each coupon is machined/milled to provide a smooth surface for zirconium adhesion. Furthermore, the coupons are machined with a specified radius to remove any sharp edging. The radius prevents the coupon from puncturing the can assembly during the rolling process. Once the coupon is machined, it is cleaned and welded into a carbon steel can assembly.

The next step of the fabrication process is the initial sizing. The coupon assembly is then reduced to an intermediate thickness by hot-rolling (with intermediate heating in a salt-bath) and/or machining. At this point, the intermediate foil is de-canned and the can is discarded. The foil is then cold rolled to a final thickness. Once the desired thickness is obtained, the foil is sheared to the final dimensions.

The laboratory scale process was implemented in the Technology Development area of Y-12. This area allows for laboratory and pilot scale activities that will provide a direct transition to production area operations. By using the Technology Development area, Y-12 is able to estimate potential throughput and identify process constraints prior to production scale implementation.

2.2 Production Scale Foil Fabrication Process

In preparation for reactor conversions, Y-12 will need to completely transition from a pre-production (or interim production) mode to a full scale production mode.

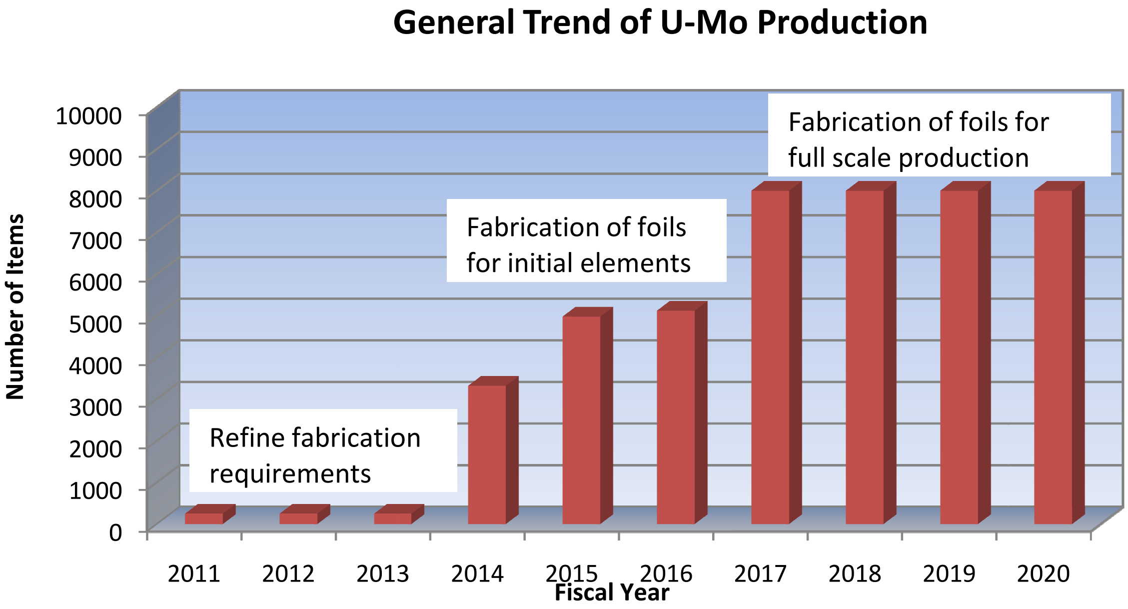

Figure 2: Trending for Production Planning

Figure 2 displays a general trend of U-Mo production. This type of trend allows Y-12 to forecast production levels in both equipment and resource loadings. In the early stages of readiness, the fabrication requirements and the production process are refined. The production equipment is ready for use; however, initial barriers in the transition (e.g. procedure refinement) to production are identified and resolved. The process then transitions to fabricate the foils for the initial elements. During this stage, production throughput is significantly increased and production strategies (i.e. campaigns, etc.) are determined. The final stage is full scale production.

As shown in Figure 2, full scale production is needed to support reactor conversion. To achieve full scale production, adjustments to the baseline process are needed to increase throughput.

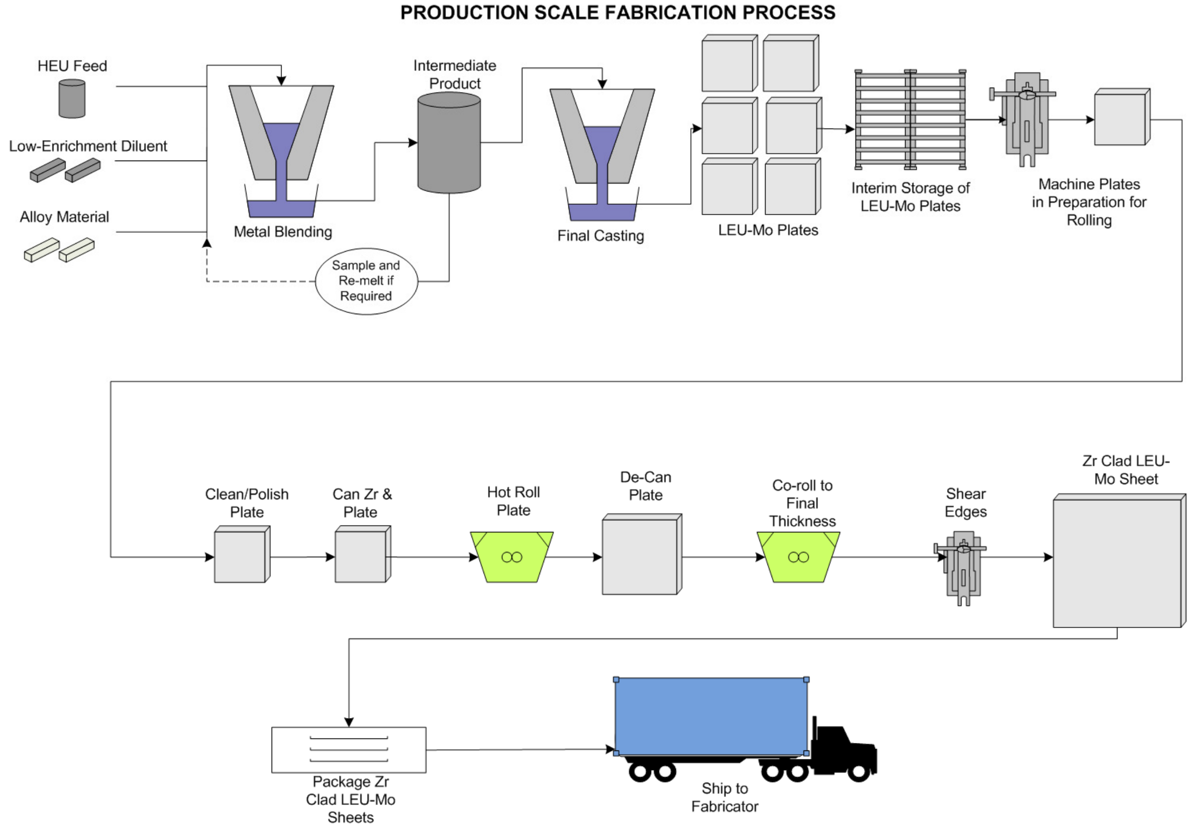

Figure 3: Production Scale Fabrication Process

Figure 3 displays a potential process for achieving the desired throughput. First, a multi-plate bookmold would be used during the final casting stage. Instead of cutting and machining individual coupons, the entire plate would be machined, cleaned and welded into the canned assembly. The plate would then be rolled into sheets and shipped to a fabricator.

2.3 Transitional Activities

2.3.1 Casting Activities

In 2010, Y-12 continued to fabricate DU-10Mo, LEU-10Mo and HEU-10Mo coupons. These castings supported both the Fuel Development research experiments and the Fuel Fabrication Capability objective of transition to production. An area of focus this year was the improvement of blend plans. Blend plans are used during the pre-melt stage to identify the amount of materials needed to achieve the desired product. As a result the improvement, Y-12 decreased the number of intermediate castings needed to achieve the specified Mo content.

Figure 4: Removal of Mold from Furnace

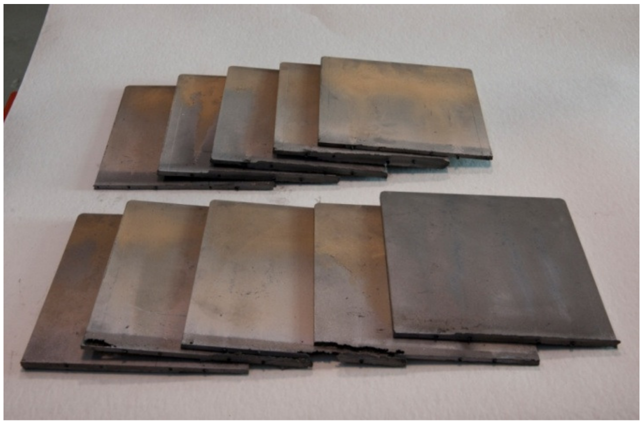

Figure 5: Plates Cast from Multi-plate Bookmold

Figure 4 displays the furnace and multi-plate bookmold used in the Technology Development area. Figure 5 display U-Mo plates cast using a multi-plate bookmold. The plates display the material directly after the cast, prior to machining activities. Y-12 performed multiple experiments using a twenty plate configuration and a ten plate configuration. The book mold can be adjusted to accommodate the quantity of plates desired. While the twenty plate bookmold allowed for larger quantities of material, the thinness of the plate did not allow for adequate machining of the material, which is required to provide a smooth surface for zirconium adhesion. Furthermore, the mold can be adjusted to accommodate downstream process constraints. For example, the dimensions of the mold can be adjusted to accommodate the throat size of the salt bath used downstream during the rolling process. Data collected during these experiments will be used to transition the mold for production level operations.

2.3.2 Machining/Milling Activities

Figure 6: Machined HEU-Mo Coupon

Figure 6 displays an HEU-Mo coupon machined for research experiment. The picture displays the multiple passes used to machine the coupon to the desired thickness. In 2010, Y-12 installed new tooling to reduce the number of passes required per coupon. This resulted in a significant reduction of cycle time. In the transition to production level operations, the tooling can be modified to machine an entire plate.

2.3.3 Sampling Techniques

In 2010, Y-12 examined how the sampling techniques affect the reliability of the analytical results. For example, larger samples are needed to conduct the carbon analysis. The preparation of the sample for the carbon analysis requires that the sample be rinsed in an acetone solution and then dried to clean the sample prior to the analysis. If the sample contains fines (or material with the consistency of "dust") the acetone may not be completely removed prior to the analysis. Thus, the analysis indicates a higher level of carbon due the carbon contribution during the rinse.

Y-12 also transitioned to using inductively-coupled plasma (ICP) methods for the elemental impurity analysis. While multiple analytical groups have developed methods for complete isotopic and chemical characterization, Y-12 focused on the determination of percent-molybdenum by ICP-optical emission spectroscopy (ICP-OES) and the determination of trace (metal) impurities in U-Mo by a combination of ICP-OES and ICP-mass spectrometry (ICP-MS).

For the analysis, U-Mo samples were dissolved in nitric/hydrochloric/ hydrofluoric acid solutions for analysis. The resulting solution was then split into three aliquots for: (i) Mo content, (ii) pre-chromatography analysis and (iii) post-chromatography analysis. Chromatography was used to remove the uranium from the third aliquot to aid in the determination of trace-level elemental impurities because the optical emission from uranium is complex and can interfere with optical emission lines from analytes of interest. The Mo content was determined by diluting the first aliquot and analyzing the solution directly by ICP-OES. The pre-chromatography analysis uses ICP-MS to measure a select group of elements that are not eluted quantitatively from the column (e.g. gold, erbium, thullium, ytterbium, thorium, etc.). Final analysis consists of the measurement of selected groups of elements by ICP-OES (primarily main group and first-row transition metal elements) and ICP-MS (second- and third-row transition metal and lanthanide elements). In total, sixty elemental impurities are currently being determined in the uranium-molybdenum alloy utilizing the two ICP techniques.

With the uranium removed and the Mo concentration determined, the Mo then presented a set of obstacles for the analysis of many other elements on both spectrometers. Some of those problems included:

- The isotopic composition of Mo presented a significant challenge to ICP-MS analysis for cadmium determination because the formation of Mo-oxide ions in the inductively-coupled plasma process interferes with the measurement of all but one, minor cadmium isotope.
- OES must be used for measuring titanium concentration because doubly-charged ions from Mo interfere with ICP-MS analysis.
- The spectral complexity of Mo required careful emission line selection, which was accomplished by studying the emission of various elements in the presence of Mo.
- Multiple inter-element correction (IEC's) factors were determined for elements that had no emission lines available without spectral interference from the Mo.

The use of high resolution inductively coupled plasma mass spectrometry (HR-ICPMS) is also being investigated as a possibility to determine impurities without the laborious sample preparation of the uranium removal.

2.3.4 Rolling Activities

In the past, Y-12 has provided coupons and bare foils for experimentation. In 2010, Y-12 implemented the INL baseline process to apply the zirconium barrier with the co-rolling process. The initial process was implemented using DU-10Mo as a surrogate material.

Figure 7: First Co-rolled Foil Fabricated at Y-12

Figure 7 displays the first foil fabricated by the co-rolling process. The process was slightly adjusted to accommodate differences in processing equipment between INL and Y-12. For example, Y-12 uses a salt bath as the heat source during the hot rolling stage. The close proximity of the equipment to the mill allows for multiple reduction passes prior to reheating.

To prepare for the transition from the Technology Development area to production level, Y-12 is experimenting with campaigning material deliverables. During a campaign, operations will focus on one project from beginning to completion. This activity can quickly identify process constraints. For example, campaigning can identify barriers in material movement. Barriers can range from the quantities of material physically located in a certain area, such as a glovebox, to resource loadings.

Figure 8: Co-rolling Activities

Figure 8 shows a snapshot of the campaigning activity. The picture shows that as one foil completes a reduction pass, a technician is waiting hand the foil another technician (not shown in picture) to perform measurements. While one is being measured, another foil will complete a reduction pass. The continuous relay and parallel operations reduce the cycle time needed to fabricate the foil. Furthermore, once fabrication is complete the foil is immediately packaged for shipment. This reduces the cycle time of pack/ship operations.

2.3.5 Equipment Installation

To support the transition to production level operations, Y-12 needed to procure and install a new rolling mill to support the cold rolling process in the production operation. In 2010, Y-12 procured and installed the new mill shown in Figure 9. The start-up activities for the new mill will occur in early 2011, with production availability expected in June.

Figure 9: Installation of New Rolling Mill

2.4 Continued Support of Fuel Development Initiatives

2.4.1 Source Material

Y-12 continues to support Fuel Development initiatives. In 2010, Y-12 provided source materials (DU-Mo, LEU-Mo and HEU-Mo) and foils for examination. This is a continuous effort to help define process parameters and material requirements needed for the production level operations.

2.4.2 Alternative Processing for Zirconium Application

Y-12 is examining physical vapor deposition (PVD) as an alternative process for zirconium application. PVD uses a sputtering process to clean the foil. This is less labor intensive than alternative methods and is performed directly before the zirconium application. Therefore, there is no downtime between cleaning and zirconium application. PVD provides an intermetallic bond, giving an enhanced adhesion. In 2010, Y-12 continued data collection on barrier thickness and deposition rates. In 2011, the collected data will be used to optimize the process and will be used on HEU-Mo foils that will be tested during a reactor experiment. This test will ultimately decide if PVD is a viable option for production level activities.

3. Summary

In 2010, Y-12 continued to support the Fuel Development initiatives, such as providing source material for reactor testing and experimenting with alternative processes for zirconium application. This will continue into the future as the production process evolves. In support of the Fuel Fabrication Capability, the initial campaign experiments performed in 2010 indicate the U-Mo foil fabrication process shown is fundamentally feasible and scalable to support production levels. For the transition to production levels, process parameters and material requirements are essential and they continue to be defined as the production process evolves. This transition will require some minor adjustments to the current baseline process. However, these are equipment related adjustments and are not expected to adversely affect material compositions or properties. The transition should occur in 2011 and 2012 to support the anticipated production levels.