# Uranium-Molybdenum Fuel Foil Fabrication Development Activities at the Y-12 National Security Complex


## ABSTRACT

The Reduced Enrichment for Research and Test Reactors (RERTR) Program was initiated by the United States Department of Energy (DOE) with the goal of developing the technologies necessary for research reactor core conversions from highly enriched uranium (HEU) to low enriched uranium (LEU). Many of the remaining HEU research reactors have fissile atom density requirements too high to be met by existing LEU fuel types, thus forcing new fuel developments. The most promising of which has been the production of high-density, LEU, monolithic uranium-molybdenum (U-Mo) fuels, in which the fuel meat is comprised entirely of a single foil of the U-Mo alloy. In support of RERTR program goals, efforts are underway at Y-12 to develop and validate a production oriented, monolithic U-Mo foil fabrication process adaptable for potential implementation in a manufacturing environment. These efforts also include providing full-scale prototype enriched U-Mo foils to the RERTR program in support of qualification testing. The work will be approached in three phases. The initial phase of the work at Y-12 consists of the development and demonstration of foil fabrication process techniques utilizing DU-10Mo source material. The next phase of the work will be to transition the production techniques from depleted to enriched uranium and to provide enriched U-Mo test foils. The final phase of the work will evaluate full-scale implementation of the developed production techniques. Y-12 has completed the initial phase of the work via the development of production techniques and the successful fabrication of DU-10Mo foils. A description of Y-12's endeavors regarding the execution of these efforts is presented herein.



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## INTRODUCTION

The Reduced Enrichment for Research and Test Reactors (RERTR) Program was initiated in 1978 by the U.S. Department of Energy (DOE) to develop the technical means for the conversion of research reactors from HEU to LEU. The RERTR program cooperates with the research reactors operators to achieve this goal of HEU to LEU conversion while maintaining reactor performance. One hundred six (106) research reactors around the world have been identified for conversion. The goal of the RERTR program is to complete the conversion of all targeted research reactors by 2014. Many of these reactors cannot convert utilizing today's low enriched fuels, thus forcing new developments in LEU fuels.

The RERTR program has focused recent efforts in new LEU fuel development on the production of high density, low-enriched, monolithic U-Mo fuel for the remaining HEU to LEU research reactor core conversions. This monolithic fuel would be theoretically capable of converting every research reactor currently operating in the world today. In monolithic U-Mo fuel plates, the entire plate fuel matrix is comprised of a single U-Mo alloy foil clad in metal (e.g. aluminum). The existence of the single alloy fuel provides the maximum uranium loading per plate, limited only by the fuel alloy density as the fuel is absent of the dispersing material matrix found in dispersion fuel. This result in higher uranium loading densities than are available in the traditional LEU fuels and would potentially allow HEU research reactors to maintain their existing performance without major modifications. Table 1 illustrates the comparative densities of various alloys and typical research reactor fuel matrices.

**Table 1. Densities of Typical LEU Alloys and Fuel Matrices**

| Fuel Matrix | U Density ($g\text{-}U/cm^3$) |
|---|---|
| $U\text{-}Al_x$ | 2.3 |
| $U_3O_8$ | 3.2 |
| $U\text{-}ZrH_x$ | 3.7 |
| $U_3Si_2$ | 4.8 |
| U-10Mo[1, 2] (dispersion) | 6 |
| U-10Mo[1, 2] (monolithic) | 15.3 |
| Mo | 10.22 |
| U | 18.95 |

The development, testing, and qualification of new LEU fuel technologies is being pursued very aggressively as the current goal of RERTR is to achieve full fuel qualification by the end of 2010.

[1] 90 weight percent uranium - 10 weight percent molybdenum alloy.
[2] Various other uranium-molybdenum weight percentage ratios are also under consideration by RERTR programs.

## MONOLITHIC U-MO FUEL FOIL DEVELOPMENT AT Y-12

In support of RERTR program goals, efforts are underway at Y-12 to develop and validate a production oriented, monolithic U-Mo foil fabrication process adaptable for potential implementation in a manufacturing environment. These efforts also include providing full-scale[3] prototype enriched U-Mo foils to the RERTR program in support of qualification testing. The work will be approached in three phases. The initial phase of the work at Y-12 consists of the development and demonstration of foil fabrication process techniques utilizing DU-10Mo source material. The next phase of the work will be to transition the production techniques from depleted to enriched uranium and to provide enriched U-Mo test foils. The final phase of the work will evaluate full-scale implementation of the developed production techniques.

## DEPLETED U-MO FOIL PRODUCTION AT Y-12

Y-12 has completed the initial phase of the work via the development of production techniques and the successful fabrication of DU-10Mo foils. An update on Y-12's efforts regarding the execution of the initial phase of the work is presented herein.

### *SOURCE MATERIAL CHARACTERISTICS*

A previously cast U-10Mo casting was used as the source material. The original casting was made by co-melting molybdenum powder and uranium plate scrap in a vacuum induction furnace. Two samples were taken from the casting (at the top and bottom, 180$^{o}$ apart); the results are shown below.

**Table 2. Source Material C and Mo Content**

| Parameter | Top | Bottom |
|---|---|---|
| Ending C Content (ppm) | 57 | 56 |
| Ending Mo Content (wt %) | 10 | 8.27 |

A portion of this casting was re-melted into a 4" x 4" x 3/8" rolling billet in a vacuum induction furnace. Chemistry samples of the final rolling billet were not taken, however, it is anticipated that the C content did not increase significantly and, at a minimum, remained between 80 to 100 ppm.

### *FOIL PRODUCTION PROCESS*

The rolling billet was cut into multiple coupons for subsequent rolling. The coupons were approximately 1" x 4" x 3/8".

After some trial and error resulting in various foil failures, thin DU-10Mo foils were successfully rolled. Figures 1, 2, and 3 present some examples of foils which failed during fabrication or annealing.

[3] The full-scale dimensions of the monolithic foil, length and width, will be analogous to existing, corresponding research reactor core dimensions. The required thickness of the monolithic fuel foil will be calculated volumetrically based on density of the U-Mo alloy used in fabrication and the equivalent $^{235}U$ loading required in the fuel.

**Figure 1. This specimen was hot rolled to 0.040" on a standard gross rolling mill, intermittently heated, and then warm-rolled on a finishing mill to 0.030". The specimen appeared to be brittle. It was then cold rolled and developed significant tears. Local pressure points were created during rolling from salt bath deposits on the surface of the specimen.**

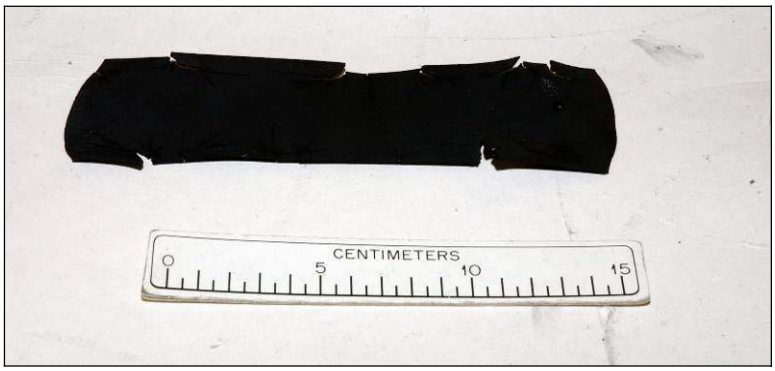


**Figure 2. This specimen had been vacuum heat treated and was cold rolled to ~0.025". Large edge cracks formed during rolling, then propagated along the length of the strip within the day. By the second day, fine cracks had formed radiating in toward the center of the foil. No further crack propagation has been observed.**

**Figure 3. This strip was hot-rolled, intermittently heated, cleaned, and cold-rolled to 0.01". The specimen then underwent an extended final anneal. Upon removal from the salt bath the specimen showed significant distortion from stress relief.**

Successful rolling of the DU-Mo foils from the cast coupons was eventually accomplished via a two stage rolling process: 1) hot-rolling, with intermediate heat treatment, to ban intermediate thickness and 2) cold-roll finishing to the final thickness.

Individual coupons were repeatedly heated in a salt bath and hot-rolled at temperature to achieve a desired intermediate thickness. The desired intermediate thickness for the hot-rolling process was determined based on rolling mill parameters/limitations as well as limitations on the viable percent reduction in thickness for subsequent cold-rolling steps.

Hardness testing was performed on as-cast and hot-rolled material to examine the brittleness of the material. Hardness values for both the as-cast and rolled material were in agreement, indicating that the worked specimen has not undergone excessive brittleization as a result of hot-working.

The hot-rolled specimens were allowed to cool and then cleaned with a grit blaster to remove the deposited layer of salt and provide a clean surface for cold-rolling. Prior to cold-rolling any small edge cracks were removed (by shearing) in order to eliminate crack initiators. Multiple passes were used to cold-roll to the desired final thickness[4] of approximately 0.010”. Any edge cracks that initiated during final rolling were also removed (by shearing).

The process steps were successfully repeated for numerous coupons to ensure reproducibility. All processing was performed on the same parent batch of cast material. Some of the foils were annealed after final rolling, while others were not given a final anneal. These finished foils have not shown any indications of residual stress cracking after final rolling (this applies for both the annealed foils and those that were not annealed).

All of the foils produced are still pliable. All have been stored in open (humid) atmosphere (approximate 4 month storage time). They have a shiny brown oxide finish, which has not changed since they were rolled. Two scraps that had been stored coiled were uncoiled, then recoiled in the opposite direction 4 months later and have shown no indication of brittleization.

**Figure 4. Finished DU-10Mo Foil, (1.5” width, 20” length, 0.01” thick)**

[4] The minimum finished foil thickness achieved was 0.007”.

## *METALLOGRAPHY*

Several metallographic specimens were collected from the U-10Mo material. For comparison, a metallographic sample was taken from the "hot top" of the as cast material and was polished by standard metallographic techniques. The sample was then electroetched with 50% Phosphoric acid with 6 volts for about 30 seconds and observed under a microscope. Figure 5 is a 200x image of the etched, as-cast U-10Mo material. The sample was slightly overetched which brings out a darker "starred" type pattern in the material which is typical of dendritic segregation, i.e. a higher Mo concentration in those areas.

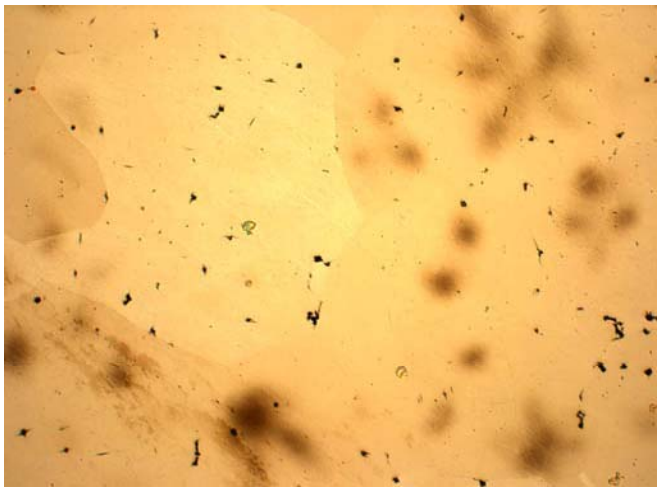

**Figure 5. As-Cast U-10Mo Material (200x)**

Micrographs were also taken of some initial rolling trials with this material. The grain structure can easily be resolved in the as-rolled and annealed micrograph. A slight overetch was given to this sample shown in Figures 6 (100x) and 7 (200x). These micrographs are taken so that the viewing angle is perpendicular to both the rolling direction and to the thickness of the foil. The rolling direction or z-direction is denoted by the blue arrow in the micrographs from hereafter.

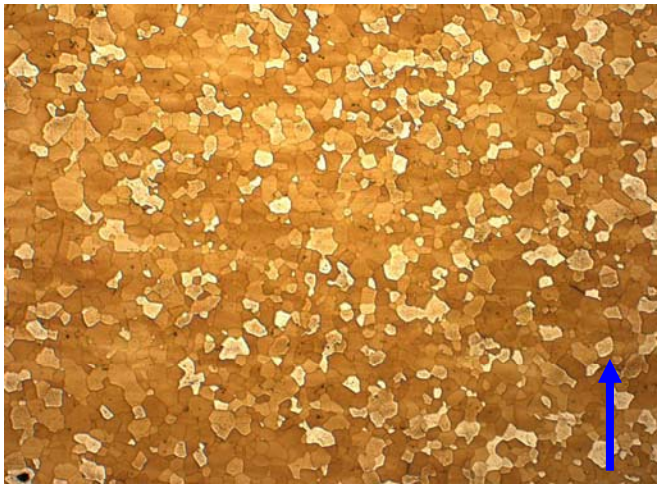

**Figure 6. U-Mo Material Initial Rolling (100x)**

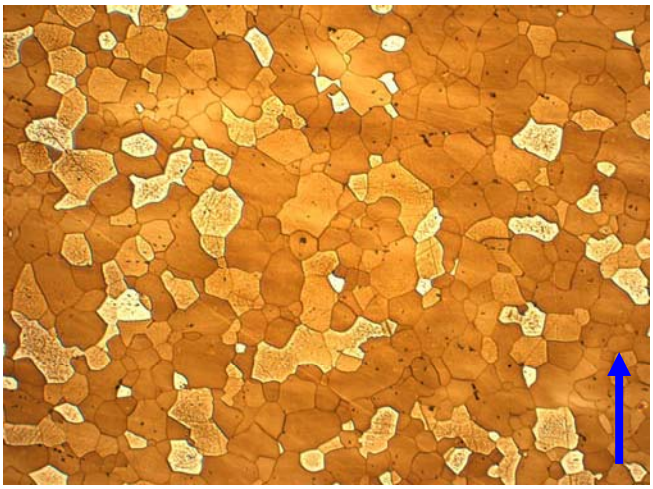

**Figure 7. U-Mo Material Initial Rolling (200x)**

Viewing a metallographic sample in the as-polished condition at 100x we observe the severe stress cracking of an initial rolling attempt on the U-10Mo foil. These types of intergranular cracks are seemingly brought on by crack initiator sites along the edges of the foil. It has been found that this problem can be avoided or at least mitigated by simply shearing off any crack initiator sites along the edges of the foil to keep a nice smooth edge on the foil as it is being rolled.

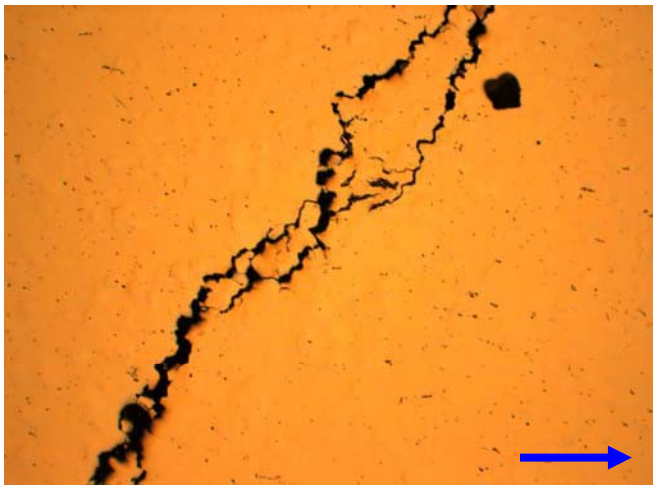

**Figure 8. U-10Mo Material Stress Crack, Initial Rolling Attempt (100x)**

Viewing this sample in the etched condition, we observe a layered type of microstructure with interchanging dark and light areas. This suggests that certain areas are preferentially etched which would indicate a difference in Mo content. However, an odd observation is that this segregation does not seem to go along with the grains or grain boundaries as typical solute segregation exhibits.

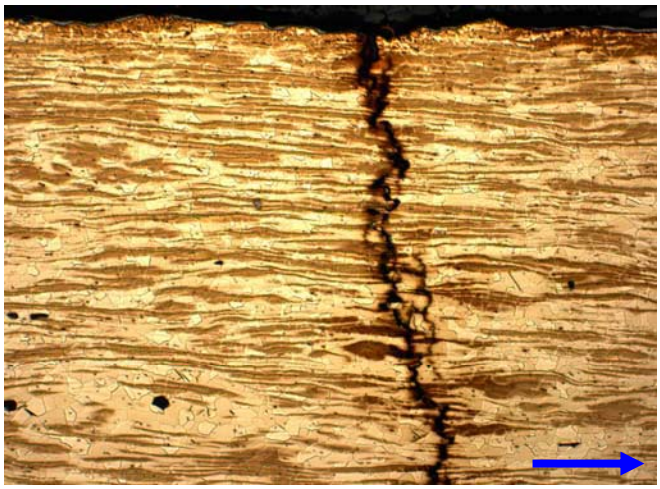

**Figure 9. U-10Mo Material Stress Crack, Initial Rolling Attempt (etched, 100x)**

Other subsequent attempts were performed at rolling the U-10Mo into foils that meet specifications. Some attempts were shown to have some cracking problems but most attempts were completely successful. Figure 10 shows an as polished specimen at a 100x view perpendicular to the rolling direction which cracked during initial rolling. Notice the clear definition of the grains even without etching, which is believed to be from grain stress relief. Also note that there are not many inclusions in the material due to the cleanliness of the starting feedstock material.

**Figure 10. U-10Mo Material, Initial Rolling Attempt (100x)**

A 500x as polished view in Figure 11 shows how the cracks follow along the grain boundaries and tend to "unzip" the grains. This is termed "intergranular cracking".

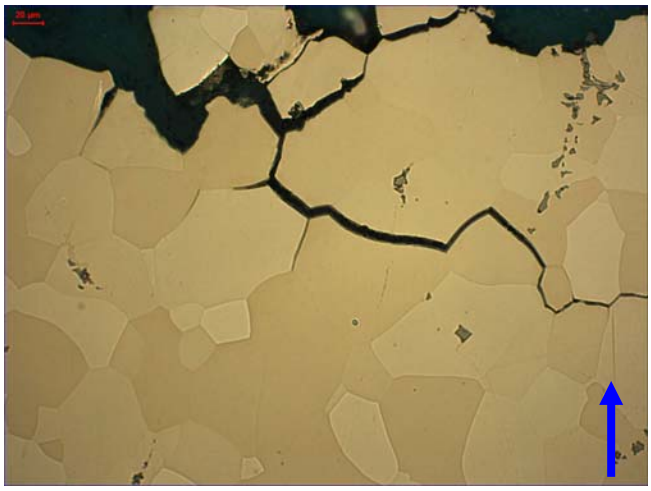

**Figure 11. U-10Mo Material, Initial Rolling Attempt (500x)**

The information gained on these samples aided Y-12's efforts in producing foils that are void of cracks as shown in Figure 12. Figure 12 is an as-polished 100x view of a foil rolled down to specified thickness (0.01"). Notice the textured microstructure along the rolling direction as well as inclusions that have been strung out in the rolled direction which are called "stringers". Also notice the nice sharp edge on the foil which has been discovered to be paramount in crack prevention of the U-10Mo foils.

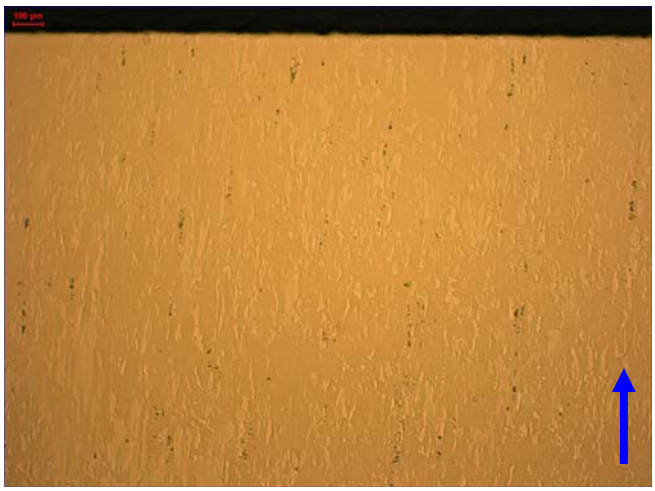

**Figure 12. U-10Mo Material, Rolled to Specified Thickness (100x)**

A 200x as polished view of the foil rolled to final thickness shows the textured grain pattern through the thickness of the foil as it has been cold rolled down to this specified thickness. Also, again it can be seen that this material is fairly clean i.e. very few inclusions.

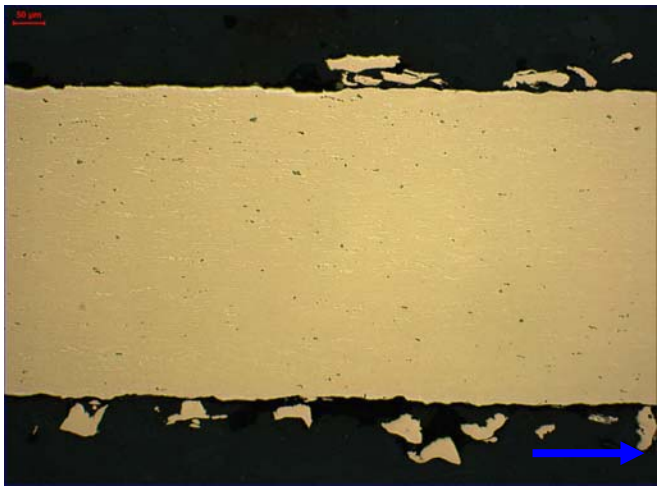

**Figure 13. U-10Mo Material, Rolled to Specified Thickness (200x)**

## FUTURE U-MO DEVELOPMENT WORK AT Y-12

Future work at Y-12 consists of the transition from the DU-10Mo work to the fabrication of enriched U-Mo fuel foils. Along with the development of test foils, the casting process, cast homogeneity, examination of process parameters, and detailed material characterization will be performed to assess the effects on the material response of LEU versus DU during forming. This work will also provide parameters for the development of an initial product specification for enriched U-Mo foils.

A detailed evaluation is also required, regarding the requirements for U-10Mo fuel foil fabrication scale-up to production level at Y-12. This evaluation will examines all aspects of implementation including production requirements, process implementation, and transportation options.

## SUMMARY

Y-12 is an operating production facility with the resources for enriched uranium material handling, processing, and transportation. Y-12 intends to support to the goals of the RERTR programs through leveraging the site capabilities and expertise.

DU-10Mo foils have been successfully roll-formed at Y-12 utilizing essentially production scale equipment which indicates that a transition to production level capacity of U-Mo foil fabrication is feasible. However, the U-10Mo fabrication process requires further development, validation, and optimization utilizing fuel material analogous to typical research reactor requirements. Work with enriched U-Mo is planned and will serve to further validate the developed fabrication techniques, as well as to provide prototype monolithic U-Mo foils for testing. Y-12 is evaluating the logistics and technical aspects involved in implementing a production line for monolithic U-Mo fuels on-site. Significant planning and evaluations need to be carried out to support the implementation, however no new or different, general process requirements, other than those currently utilized at Y-12, are anticipated.