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DEVELOPMENT, QUALIFICATION, AND MANUFACTURING OF LEU-FOIL TARGETRY FOR THE PRODUCTION OF MO-99

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The Y-12 National Security Complex (Y-12), Argonne National Laboratory (ANL), and the University of Missouri (MU), are collaborating to develop and qualify a high density low enriched uranium (LEU) target suitable for large-scale production of 99Mo. This prototype annular target, which utilizes an LEU-foil, was designed by ANL as part of an IAEA Coordinated Research Project. An experiment test plan has been developed to irradiate LEU-foil annular targets and perform a post-irradiation examination of the targets at the Pitesti reactor facility, Romania. Other potential designs, such as plate geometry may also be explored in the future. The end goal of the project is to provide a design qualification package which can be utilized by current and prospective 99Mo producers for regulatory licensing and eventual adoption into their own site specific production processes. The high density LEU target promises yields comparable to current highly enriched uranium (HEU) and LEU dispersion type targets currently in use.

The objectives of the project are to:

1. Establish a target qualification methodology that is bounding for all ^{99}Mo target irradiators.

2. Develop the target qualification methodology by building upon the annular LEU-foil target design work and testing previously performed by ANL and ANSTO/CERCA.

3. Develop a final product in the form of a "generic" LEU-foil target qualification document that can be used by any Mo-99 target irradiator to support their facility specific "safety case."

4. Evaluate the technical feasibility of developing, qualifying, and manufacturing LEU-foil targetry.

5. Develop a set of "universal" target material specifications and target manufacturing quality control (QC) test criteria that are acceptable to all current target irradiators and potential future ^{99}Mo producers.

6. Optimize the target design, considering both reactor and processing facility safety, and the economics of manufacturing a cost-effective target to offset the inherent economic disadvantage of using LEU in place of HEU.

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1. Introduction

Both HEU and LEU dispersion type targets have a long standing pedigree as they are manufactured to industry accepted materials test reactor (MTR) fuel specifications. Furthermore, HEU dispersion type targets have a long history of successful irradiation without any significant cladding failures. Historically, thousands of dispersion targets have been safely irradiated to produce fission product ^{99}Mo.

In comparison, LEU-foil targets have a very limited irradiation history. This target type has been successfully irradiated in Argentina, Indonesia, Australia, and the United States (University of Missouri Research Reactor). It is estimated that less than fifty (50) LEU-foil targets have been irradiated to date. Furthermore, foil type targets are not currently manufactured to an industry accepted standard or specification.

As a consequence, a manufacturing standard or specification must be developed for this target type and a corresponding target qualification program must be successfully implemented. A manufacturing specification must be established and a qualification program must be completed before this target type will be considered as an HEU to LEU target conversion option by any large-scale ^{99}Mo producer. Sections 3.0 and 4.0 summarize the activities currently in progress to support manufacturing specification and qualification program development.

2. LEU-Foil Target Advantages

An LEU-foil target has several distinct advantages in comparison to an LEU dispersion type target. These advantages are:

1. On a per target basis having the same uranium mass, the time to chemically dissolve a foil target is significantly less than that of a dispersion target. As the foil is removed from its aluminum cladding, only the foil component of the target is dissolved in the first stage of the ^{99}Mo production process.
2. On a per target basis having the same uranium mass, the volume of liquid radioactive waste generated during the target dissolution phase is significantly less because only the foil component of the target is dissolved. The LEU-foil target's aluminum cladding is removed during target disassembly, allowed to decay, and disposed of as low-activity solid radioactive waste.
3. The uranium loading of a typical LEU dispersion target is in the range of 2.5 to 3.0 g U/cm^3. The density of the LEU metal foil is approximately 19 g/cm^3. As a consequence, a foil target of the same geometry can contain much higher amounts of uranium than the HEU or LEU dispersion type target. The uranium content will be limited by the ability of

the target's cooling system to remove heat (target power) during irradiation.

3. Analyses to Support Specification Development

Preliminary evaluations have been performed by the University of Missouri's College of Engineering to determine the thermally induced stress on the target's cladding which is caused by non-uniform heating. A series of analytic, numeric, and experimental tools have been developed in order to evaluate the annular target geometry and to establish a proof-of-concept for the plate geometry. More advanced 2-D analytic models have been developed to establish the property and geometry groups that will influence the target behavior during irradiation. A representation of the stresses developed in an annular target is shown in Figure 1 The colors represent the Von Mises stress that results from interfacial heating. The stresses generated as a result of macroscopic U-growth and fission gas pressure will also be analyzed. A preliminary analysis applicable to both the annular [1] and plate [2] target geometry has been performed.

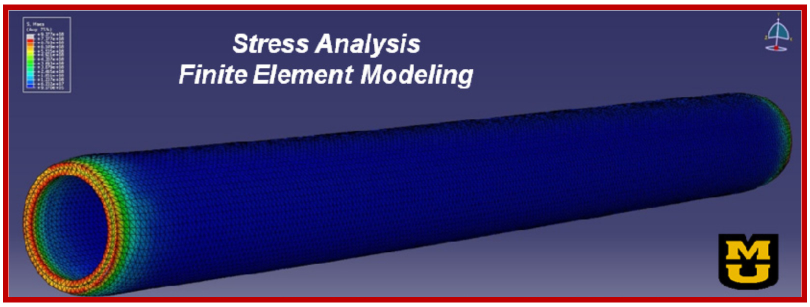

Fig. 1 ANL's LEU-foil annular target, thermal stress field

The results of the analyses and numeric simulations are being validated by experiment. A mock target, containing a heater element to provide an internal heat source, is placed in a flow loop test section as shown in Figure 2. The deformation of the target's cladding is measured by laser displacement.

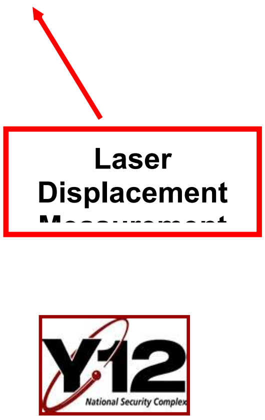

Fig. 2 Flow loop with test section

The temperature of the LEU-foil will depend upon the thermal contact resistance between the foil and the aluminum tube cladding. Contact resistance, in general, arises due to imperfect mating of components due to micro-scale surface roughness and macro-scale deformity. Any gaps that form between the target's components (aluminum cladding, LEU-foil, and nickel fission recoil barrier) contain relatively low thermal conductivity gases. The thermal conductance between the heat source (LEU-foil) and coolant (water) is thus reduced relative to perfect material contact between the components. A higher contact resistance implies a lower thermal conductance and a correspondingly higher LEU-foil temperature.

Experiments are currently being performed to evaluate the surface state (i.e., roughness) of the LEU-foil so that the thermal contact resistance for the annular target geometry can be characterized. Following this characterization, an envelope of thermal contact resistance for the target geometry can be established.

The surface texture of the foil is measured prior to target assembly and compared to the surface texture measurements following target disassembly to assess the magnitude of thermal contact resistance. The measurements are obtained using the instrument shown in Figure 3. A representative sample of uranium foil manufactured by KAERI using their cooling-roll casting method is shown in Figure 4. The surface texture of the uranium foil is visible.

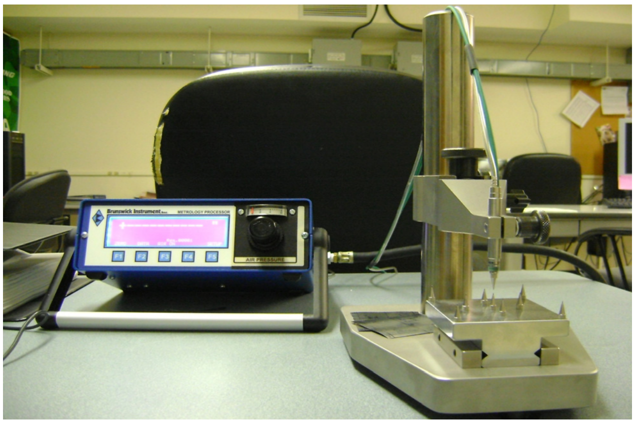

Fig. 3 Surface contact profilometer

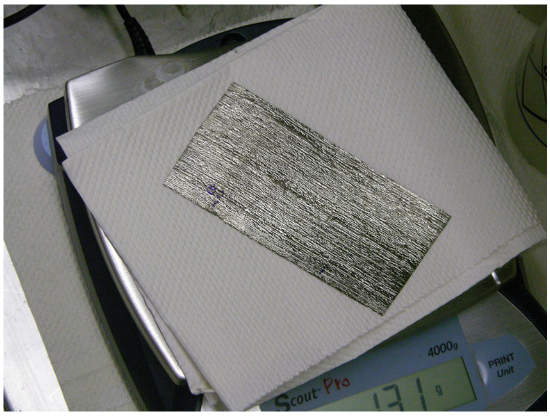

Fig. 4 LEU-foil supplied by KAERI

Strides were made in developing the baseline manufacturing process for LEU fission target foils during 2011. Most notably was the inclusion of an anneal step to bring the foils to necessary final dimension of 5 to 6 mils. The improved process map is depicted below.

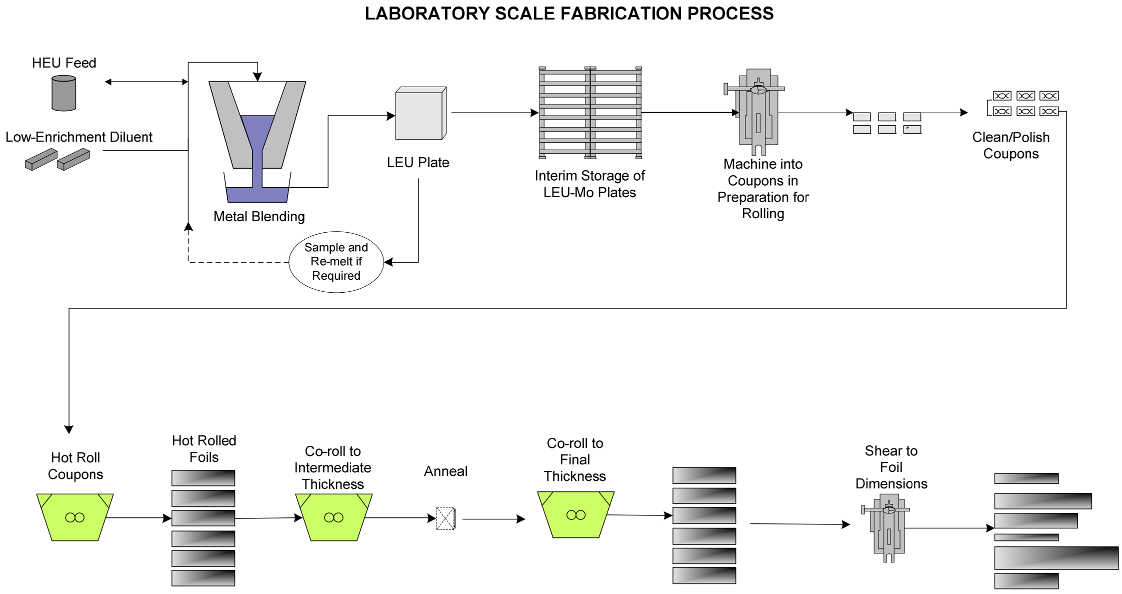

Figure 5: Baseline Process Map

The foil is cold rolled to near the final thickness and then annealed before cold rolling to the final dimensions. Extremely thin foils are possible but routinely limited to a minimum of 4 mils (100 μm). The intermediate anneal furnace is depicted below.

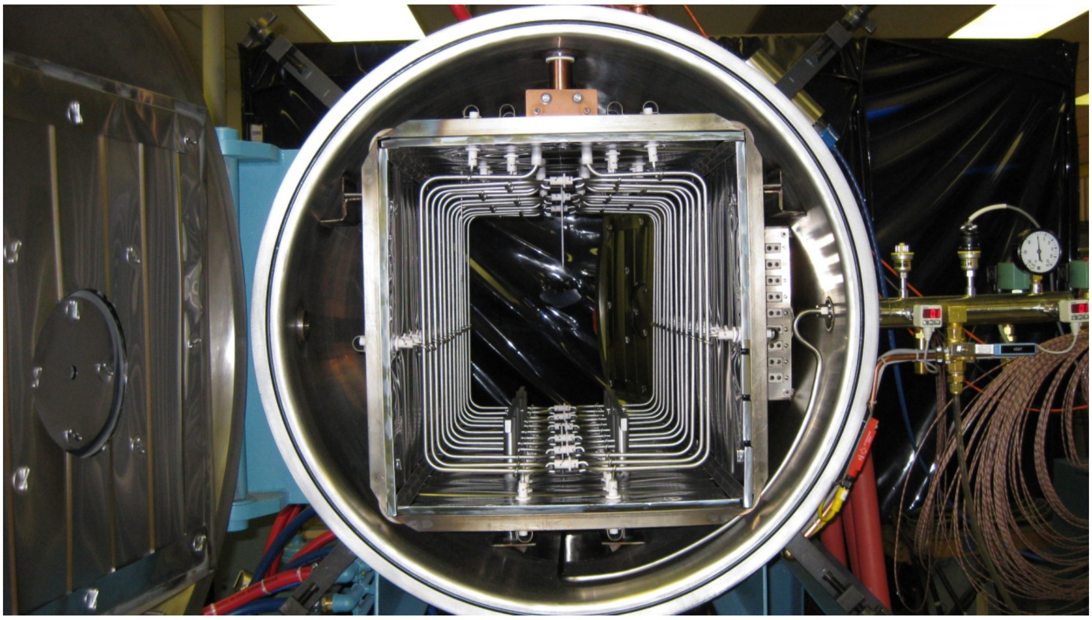

Figure 6: Developmental Anneal Furnace

The final result of the modified process is depicted below. The pictures illustrate the accuracy and consistency of the foil possible with this process.

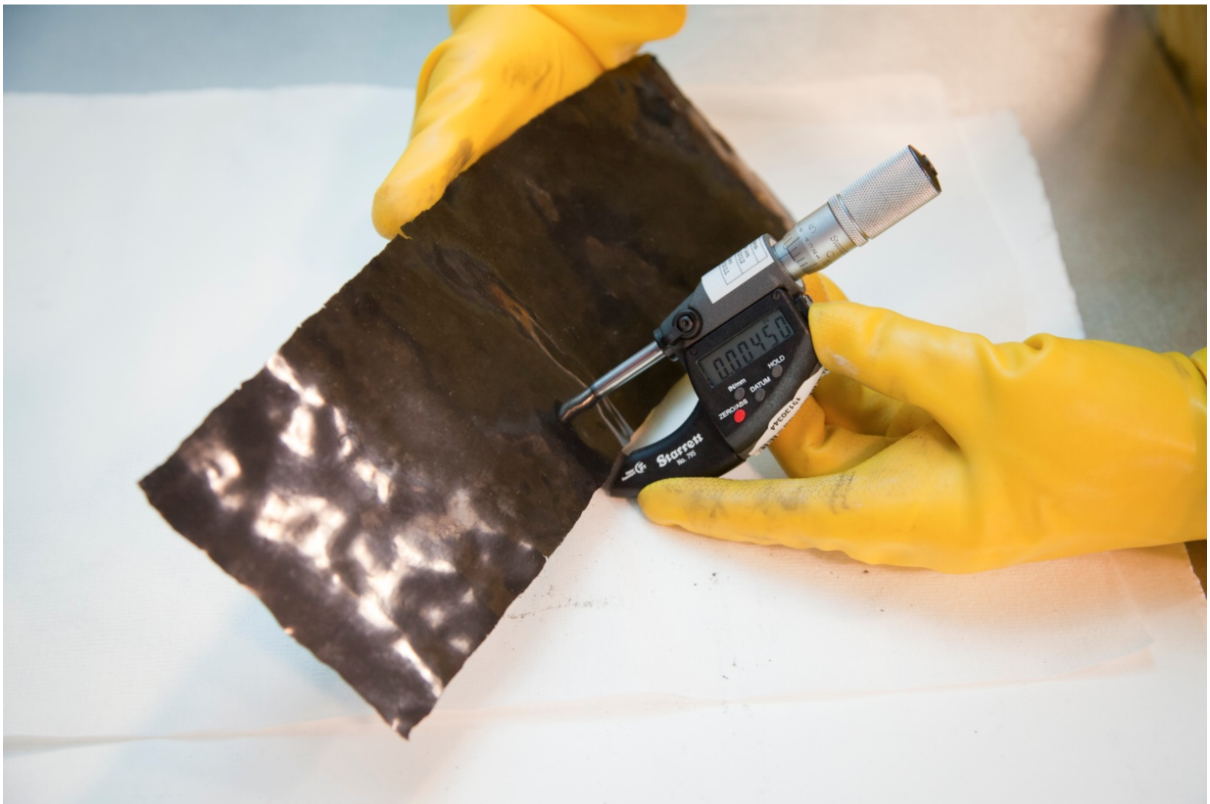

Figure 7: Finished foil at thickness of 4.5 mil (114 μm)

Currently LEU foil targets are manufactured with a wrapped nickel foil barrier. This barrier consists of a thin foil which is literally wrapped around the U foil to serve as a fission barrier so the foil can be removed from the clad if so desired. Experiments at Y-12 have shown that a nickel clad barrier can be applied to the foil through the electroplating process. Depleted uranium surrogates have been electroplated and shown strong promise of advancing manufacturing readiness levels (MRLs). Figure 8 below depicts the electroplating bath optimized for coating uranium.

Fig. 8: Nickel electroplating bath for plating uranium metal foils

In addition to the standard aqueous electroplating process, development work began on a nonaqueous process in FY11. A basic corrosion study of uranium metal immersed in an ionic liquid yielded encouraging results which suggest that the ionic liquid could be used for nonaqueous electropolishing of uranium to prepare it for nickel plating.

4. Testing to Support Qualification Strategy

There are limited experimental data published that characterizes the physical behavior of thin uranium foils during irradiation. Macroscopic U-swelling and fission gas generation data is needed to construct models capable of predicting the behavior of LEU-foil Mo-99 targets during irradiation. The models will be used to demonstrate the structural integrity of LEU-foil targets during irradiation.

A test plan has been developed that outlines a strategy for acquiring LEU-foil irradiation behavior data using the Pitesti reactor's post irradiation examination (PIE) facility shown in Figure 9. The PIE data will be used to support the development of a universally qualified LEU-foil based target that can be used by any current or future Mo-99 producer who desires to evaluate the LEU-foil targetry option. A series of tests will be performed under irradiation conditions that represent the maximum irradiation parameters (i.e., thermal neutron flux and irradiation time) of dispersion type targets that are now being irradiated to produce Mo-99. Parameters such as macroscopic U-swelling and fission gas generation will be quantified.

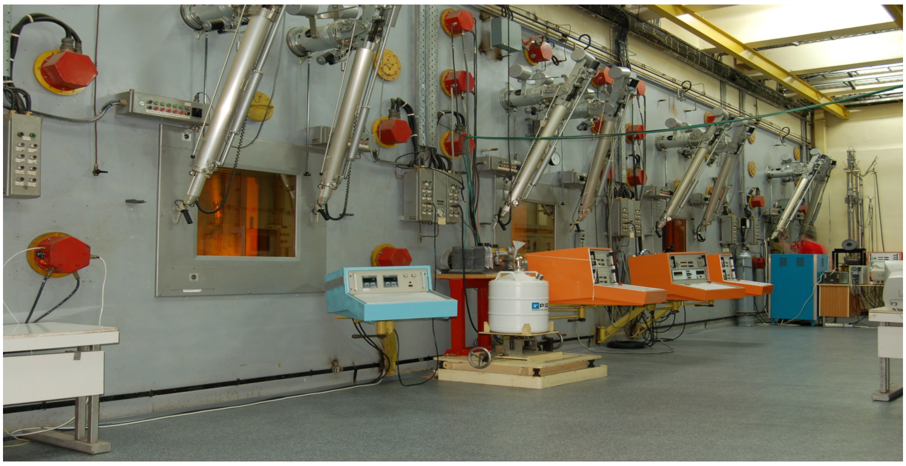

Fig. 9 Pitesti Reactor PIE Facility

5. Acknowledgements

We would like to thank Dr. George Vandegrift of Argonne National Laboratory, Dr. Jonathan Morrell and Mr. Justin Holland of Y-12 National Security Complex.

6. Technology Readiness Assessment

A Technology Readiness Assessment (TRA) is an industry recognized and accepted method used to assess the technological maturity of a product (in this case, an LEU-foil target) under development. This assessment is formal, systematic, and metric-based. The metrics are defined by the Technology Readiness Levels. A TRA: 1) provides a common language to communicate the maturity of a technology, 2) enables a disciplined approach to evaluate technology readiness, and 3) provides an effective tool and metrics to assess technology risk. It is simply an analysis for determining the technology maturity with respect to meeting product realization goals. Using this assessment methodology [3], the LEU-foil target is assigned a maturity of TRL 5 for the high-volume production (meaning mass production in sizable lots) [4].

7. References

[1] IMECE2009-13238, "Thermal-Mechanical Analysis of Annular Target Design for High Volume Production of Molybdenum-99 Using Low-Enriched Uranium", K. Turner, G. Solbrekken, C. Allen; Proceedings of the ASME 2009 International Mechanical Engineering Congress & Exposition; November 13-19, Lake Buena Vista, Florida, USA

[2] IMECE2010-39393, "Thermal-Mechanical Analysis of Varying Boundary Conditions on a LEU Foil Based Molybdenum-99 Plate Processing Target", K. Turner, G. Solbrekken, C. Allen; Proceedings of the ASME 2010 International Mechanical Engineering Congress and Exposition; November 12-18, 2010, Vancouver, British Columbia, Canada

[3] DOE G 413.3-4, U.S. Department of Energy, Technology Readiness Assessment Guide, October 12, 2009

[4] "Engineering of LEU-Foil Based Mo-99 Target for High Volume Production", G. Solbrekken, K. Turner, C. Allen; Proceedings from the 2009 RERTR, 31st International Meeting on Reduced Enrichment for Research and Test Reactors; November 1-5, 2009, Beijing, China

[5] “Electroplating of Uranium Foils FY11 Report for PD100274”, J. M. Holland, September 2011.