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ENGINEERING OF LEU-FOIL BASED MO-99 TARGET FOR HIGH VOLUME PRODUCTION

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The University of Missouri Research Reactor's (MURR) business development objective is to supply 50% of the US domestic Mo-99 demand. To establish a competitive Mo-99 market price, a cost-effective target must be developed. Existing dispersion targets utilize relatively expensive 'meat' containing uranium-aluminum alloys in an aluminum matrix. The dissolution of dispersion type Mo-99 targets generate a relatively large volume of high-activity liquid waste. The proposed strategy is to use a low cost LEU-foil wrapped in a nickel fission recoil barrier to reduce the amount of liquid waste. The primary challenge with this design strategy is ensuring the target cladding sufficiently conforms to the LEU-foil during irradiation, allowing proper cooling. This paper provides a review of target-based analytic, numeric and experimental activities that have been completed on annular and plate geometries to date. The results do not appear to suggest technical barriers in pursuing a foil-based target for high-volume production of Mo-99.

1.0 INTRODUCTION

The University of Missouri continues to move forward with its plans to design, license, and build a facility to process low-enriched uranium (LEU) targets to produce molybdenum-99 (Mo-99). The design of the facility will be based on processing irradiated LEU-foil targets to produce fission product Mo-99.

An LEU-foil target has several distinct advantages in comparison to the commonly used uranium-aluminide 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 type 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 Mo-99 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 (by a factor of at least 15) because only the foil component of the target is dissolved. The LEU-foil target's aluminum cladding is disposed of as solid radioactive waste.
3) The cost per gram of LEU-foil target material is less than the cost per gram of LEU dispersion target material.
4) On a per target basis, the cost to fabricate an LEU-foil target is projected to be cost-competitive to fabrication costs for dispersion type targets containing the same uranium mass.
5) The total uranium loading of a typical dispersion target is in the range of 2.5 to 3.0 g U/cm3. The density of an LEU metal foil is approximately 19 g/cm3. As a consequence, a foil target can be made more compact in size. This economizes limited available reactor irradiation space.

To support a large-scale weekly Mo-99 production goal, a facility must be placed in operation to produce the LEU-foil and to manufacture the targets. There are multiple processes that can be used to produce foils. Five assumptions are made regarding the process for manufacturing LEU-foil targets:

- The incoming material will be supplied as LEU metal
- The fabrication method must be a proven technology
- Recycling of scrap U-metal will be an important part of the manufacturing process
- The plant will produce a final product of LEU targets ready to be irradiated
- The targets must be manufactured under an established quality assurance program

The University of Missouri will collaborate with B&W Y-12, Oak Ridge, Tennessee, and AREVA-CERCA, Romans, France, to design and manufacture LEU-foil Mo-99 targets. Both of these companies have the manufacturing experience and capability to manufacture the number of targets required to meet the facility's weekly Mo-99 production goal.

An LEU-foil target has been designed by Argonne National Laboratory (ANL) to be used as the target for the LEU-Modified Cintichem process. ANL has designed and fabricated LEU-foil targets for irradiation in Indonesia and Argentina. Target dimensions were set by their individual irradiation positions and Mo-99 yield requirements. These targets were successfully irradiated and processed to produce Mo-99 on a demonstration trial basis.

The geometry of the LEU-foil target developed by ANL is annular. AREVA-CERCA fabricated an LEU-foil target based on ANL's annular target design for ANSTO (Australia). This target was also successfully irradiated and processed to produce Mo-99 on a demonstration trial basis. The target contained 20.5 grams of LEU-foil. MURR has also successfully irradiated two LEU-foil targets of the ANL annular design. Each of these two targets, irradiated and processed on a demonstration trial basis, contained approximately 5 grams of LEU-foil.

The University of Missouri will work with B&W Y-12 and AREVA-CERCA to develop LEU-foil targets in the geometry of a plate. The objective of this target development work is to transition ANL's proven annular geometry to a plate geometry.

The University of Missouri has filed provisional U.S. Patent No. 61/185,077, entitled, "Apparatus and Methods for Producing Radioisotopes." This provisional patent is applicable to the development of a U-foil target in the geometry of a plate. Using the DOE's established Technology Readiness Level (TRL) assessment methodology, the development of the plate LEU-foil target is TRL 3.

The University of Missouri has performed analyses to validate the design of ANL's LEU-foil annular target. The results of these analyses will be presented later this year at the Proceedings of the ASME 2009 International Mechanical Engineering Congress & Exposition. A paper entitled, "Thermal-Mechanical Analysis of Annular Target Design for High Volume Production of Molybdenum-99 Using Low-Enriched Uranium," will be presented at the proceedings. The development of the ANL's LEU-foil annular target is between TRL 5 and 6.

It is planned that MURR will commence Mo-99 production using the annular LEU-foil target developed by ANL. A Manufacturing Level Readiness (MRL) assessment of both the LEU-foil annular and plate target geometries must be performed. It is anticipated that AREVA-CERCA's collaborative participation will be in the form of a "Cooperation Agreement." Efforts are currently underway by AREVA-CERCA and the University of Missouri to establish this agreement.

The purpose of this paper is to review the current state of the mechanical design of an LEU foil-based target that is appropriate for a high-volume manufacturing environment. The scope of the analysis is confined to the steps identified in the process flow shown in Figure 1. The process effectively starts with the definition of the target component specifications and ends in the hot cell where the target is disassembled. The TRL methodology is used to evaluate the design maturity of engineered component concepts. The review is conducted for both the annular and plate target geometries.

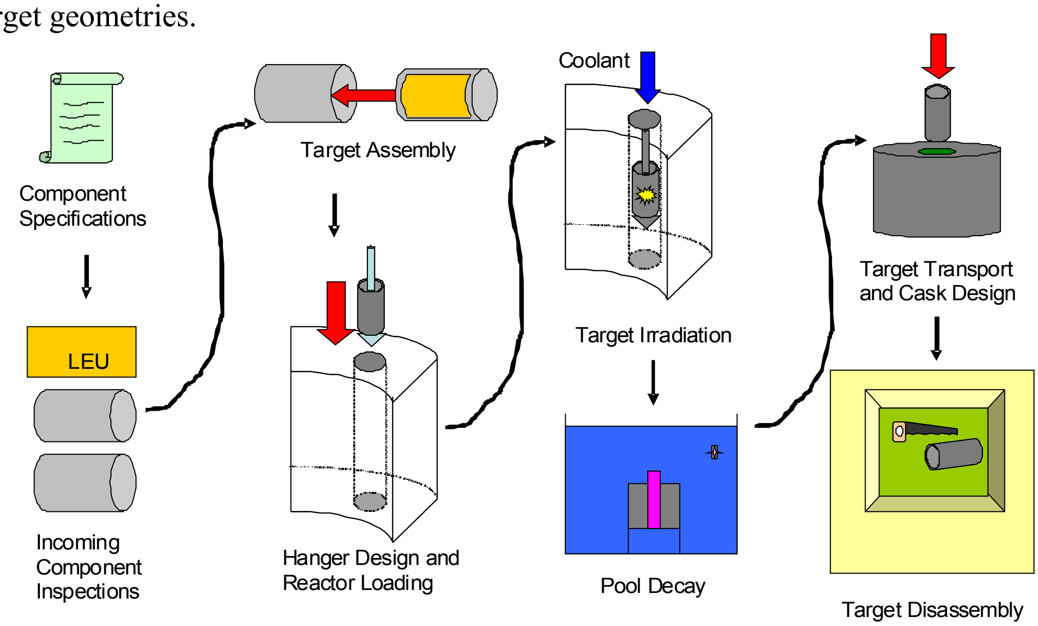


Figure 1. Process Flow for LEU Foil-Based Target

3.0 TECHNOLGY READINESS LEVEL (TRL) OVERVIEW

Product design methodologies are used to some extent in all industries to ensure that the technologies required to put a product into market are in place in a timely manner. The utility of these methodologies is that they provide a common language to describe the extent to which the product design is complete. They also provide a guide for designers to follow, allowing them to focus only on the tasks defined for a given development level. This prevents superfluous studies that do not contribute to the advancement of the product.

The Managing & Operating Contractor Agreement that defines the TRL process is used as the methodology for LEU foil-based target development. The TRL assessment consists, in sequence, of deriving product specific TRL requirement, defining TRL milestones, assigning TRL's, comparing the TRL assignment with the target, and then documenting the results.

The TRL's are shown in Figure 2 where each box contains the title for each TRL. It can be seen that the general progression of the TRL's trends from research and development activities to design demonstration in a production environment. It should be noted that there is a partner concept that describes Manufacturing Readiness Levels (MRL's) which is to be used in concert with the TRL's. The MRL's synchronize with the TRL's in a manner illustrated in Figure 3. It can be seen that during the first four technology development levels there is limited manufacturing development. After that point significant manufacturing development needs to take place to help propel the technology development. For purposes of this paper, MRL's will not be addressed specifically.

As will be explored more thoroughly in the next sections, two specific target designs for foil-based LEU production of Mo-99 are under development. The annular geometry target developed primarily by Argonne National Lab is estimated to be at TRL 6, although the corresponding MRL's have not necessarily been satisfied. A plate geometry is currently being explored as a potential cost-reducing alternative. At this point in time it is estimated that the plate geometry is at TRL3. These TRL estimates are shown in Figure 3.

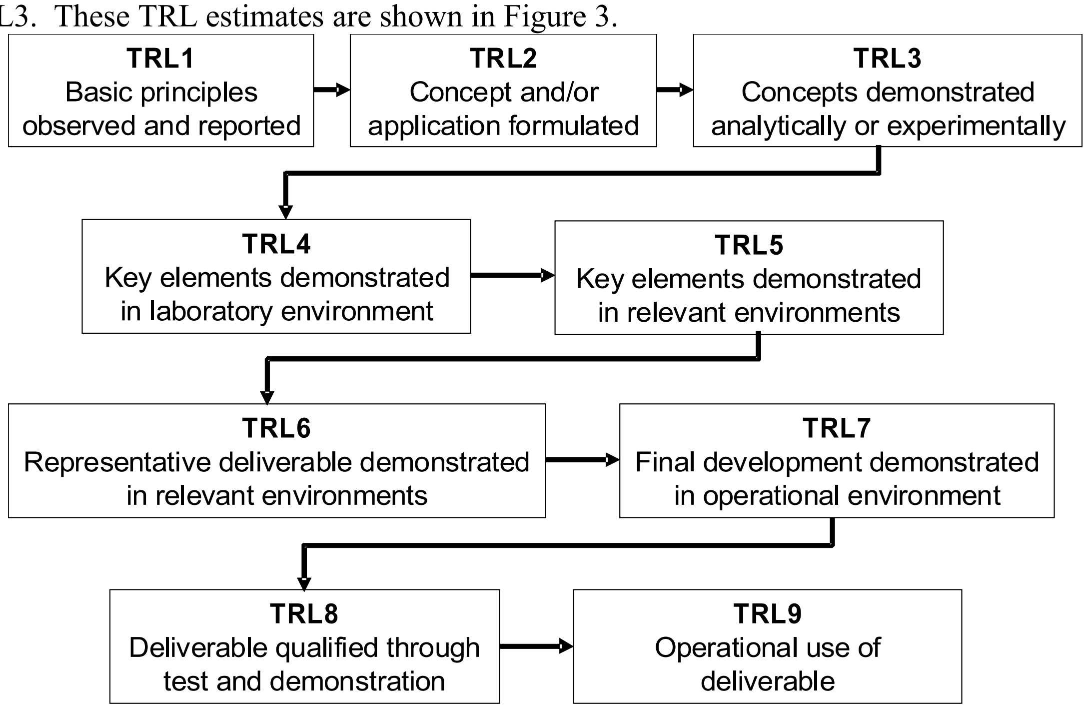


Figure 2. Description of Technology Readiness Levels

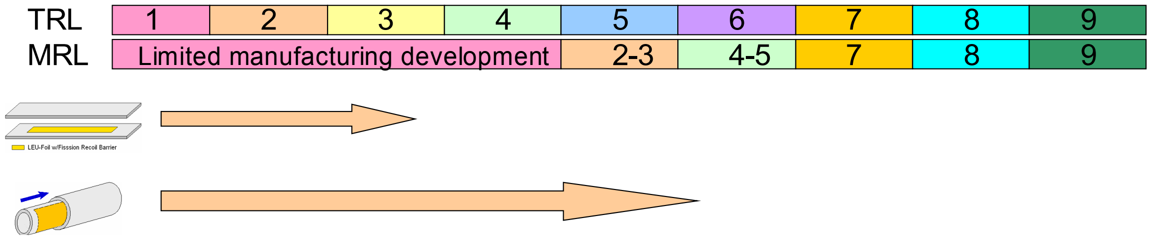


Figure 3. Synchronization of TRL's and MRL's with Level Estimates for Plate and Annular Geometries

4.0 TRL ASSESSMENT

The TRL assessment presented here will be limited up to TRL 5 as significant MRL development needs to take place in order to move from that level. A short description of each level will be given along with the exit criteria. Some technical discussion will be provided when available. The first two TRL's will be treated at the same time as they are very fundamental in nature, as will TRL 4 and 5 as the annular target has already achieved those level reported elsewhere.

4.1 TRL 1 and TRL 2

TRL 1: This is the first level of technology readiness and includes fundamental scientific research. At this level, basic scientific principles are being studied analytically and/or experimentally. Examples might include paper studies of a technology's basic properties.

Exit criteria: A fundamental concept, innovation or scientific principle has been identified

TRL 2: Practical applications are beginning to be invented or identified. Applications are still speculative and there is no proof or detailed analysis to support assumptions. Examples might include applied research in a field of potential interest.

Exit criteria: Potential practical applications for this research and/or innovation are identified

The fundamental technology that is being evaluated is a foil based target, where the target is the cladding and fission recoil barrier which encase the LEU foil. In other words, the LEU foil development is outside the scope of this work. The practical application is the support of high-volume production of fission product Mo-99.

4.2 TRL 3

Active research and development is initiated. This includes analytical and laboratory-based studies to physically validate analytical predictions of key elements of the technology. These studies and experiments should constitute "proof-of-concept" validation of the applications/concepts formulated at TRL 2. Examples include the study of separate elements of the technology that are not yet integrated or representative.

Exit criteria: 1) Analytical models and/or laboratory prototypes demonstrate 'proof-of-concept' for key elements of the intended or potential applications. These elements are not necessarily integrated 2) Key elements identified 3) Intended or potential applications identified 4) Potential customers identified

This is the estimated TRL for the plate target geometry. The third and fourth exit criteria have been satisfied, where the intended application is high volume fission product Mo-99 production and MURR is a potential customer.

The technology key elements have been identified by establishing the fundamental performance requirements in a reactor and interpreting those requirements applied to the target. The fundamental requirements for MURR are that 1) the target must contain all fission products, 2) the maximum temperature of any component in the target may not exceed ½ the melting temperature of the lowest melting point material used in the target, and 3) the temperature of the reactor pool coolant may not exceed its saturation temperature.

The first requirement implies that the mechanical integrity of the cladding and the target bonding must be maintained throughout the irradiation process. The technical metric that will be used to evaluate the cladding mechanical integrity is the yield stress for the cladding material. The bonding integrity will be similarly defined although the maximum allowable stress will need to be defined by the sealing manufacturing process. The fission recoil barrier should not play a role in satisfying this requirement.

The second technical requirement implies that the LEU temperature must be kept below ½ the melting temperature of the cladding material, as the likely cladding candidate is aluminum and aluminum has a lower melting temperature than uranium and the fission recoil barrier material nickel. The temperature of the LEU material will be dependent on the thermal contact resistance between the LEU and cladding material. A first order approximation of the LEU temperature can be estimated by considering a 1-D thermal analysis where the LEU foil is completely surrounded by a uniform air gap. The plot in Figure 4 clearly shows that the temperature of the LEU will depend on the heat flux and the air gap thickness.

The in-service heat flux will depend on the details of the LEU foil and the target location in the reactor. Figure 4 is constructed in a way that defines the maximum allowable heat flux for a given air gap when those details become available. The gap thickness requires more involved analysis. When one completes that analysis they find that the key parameters that govern the amount of plate deflection are the plate geometry, the plate thickness, and the heat flux.

A series of analytic, numeric, and experimental tools have been developed in order to establish a proof-of-concept for the plate geometry. More advanced 2-D analytic models are currently being developed to establish the property and geometry groups that will influence the target performance. Those will not be discussed at this point in time. Numeric simulations have been conducted to understand the influence of boundary conditions and geometry on performance. An example of simulation that explores the impact of edge mechanical constraint on the thermally induced stress can be seen in Figures 5a and 5b. For the example it is assumed that the LEU creates a uniform heat flux on the cladding surfaces. The three different edge constraints are shown in Figure 5a. The 'fully constrained' condition can be envisioned to be where the entire target is rigidly held in place while the 'only z constrained' condition would be where the edges of the target are welded together to prevent their separation, but the edges may expand in the plane of the plate.

The generated Von Mises stress fields are shown in Figure 5b. The locations of red coloration indicate areas of high stress while the blue areas are relatively low stress areas. From the color plots it can be seen that for the fully constrained case the central area of the plate experiences high stress while the bonded edges have low stress. This is in contrast with the only z constrained case where the bonded edges indicate high stress and the central areas have low stress. The partially constrained case shows high stress in the corners where the free edges meet the constrained edges.

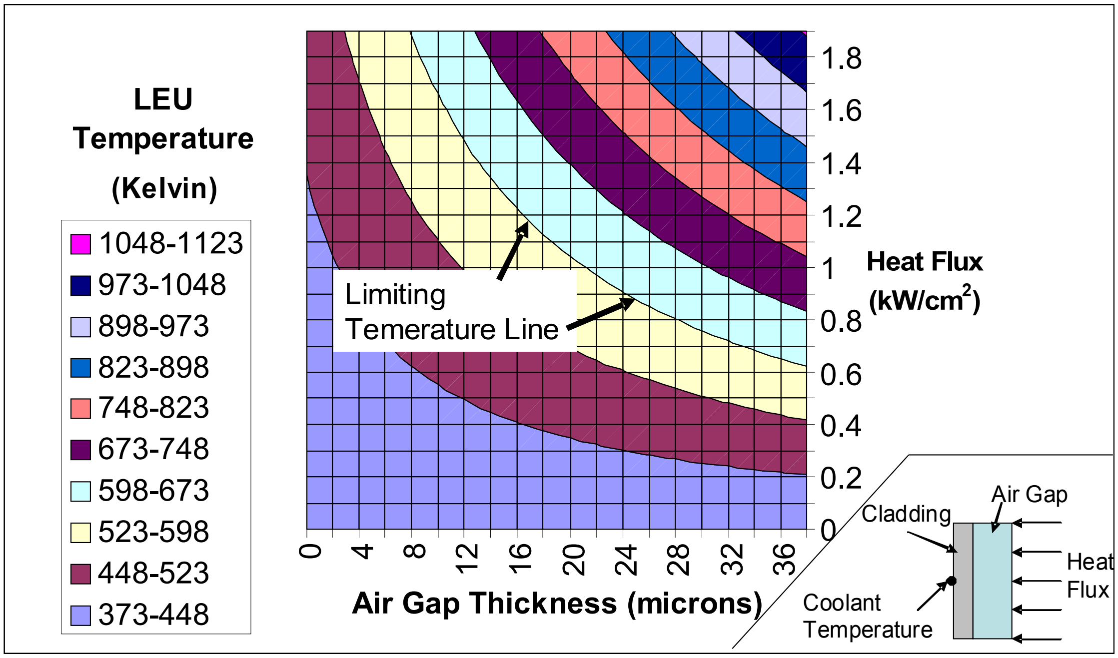


Figure 4. LEU Temperature Assuming Uniform Air Gap between Cladding and LEU
Note: The cladding is assumed to be 1mm thick aluminum and the coolant temperature is 373K.

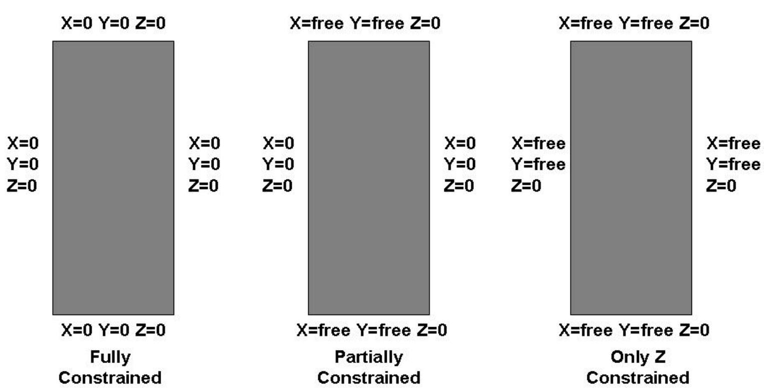


Figure 5a. Mechanical Constraint Boundary Conditions for Plate Geometry

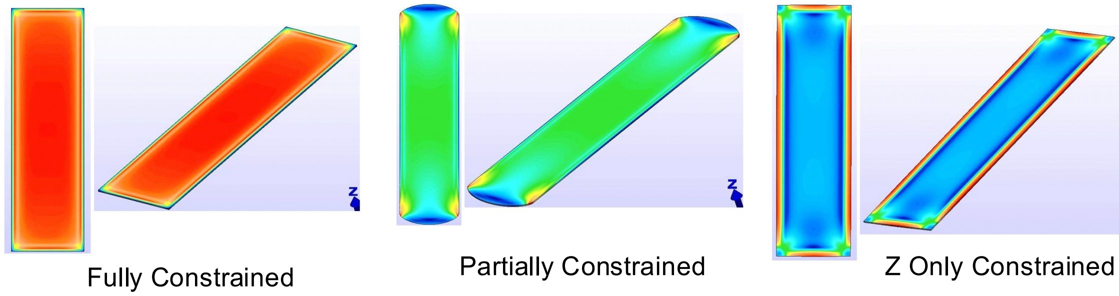


Figure 5b. Von Mises Stress Field for the Mechanical Constraint Boundary Conditions

While the numeric modeling has provided a means to understand qualitatively the impact of different mechanical constraints, the quantitative stress values can only be interpreted as indicative. Experimental calibration is necessary to establish that all necessary features are accounted for in the numeric model. An experimental test facility was built at the University of Missouri to allow such calibration measurements to take place. A photograph of the flow loop is shown in Figure 6.

The flow loop has a test section where sample plates, such as that shown in Figure 7, can be placed and tested. For one of the experiments to determine if a gap does indeed open up during heating, a heater was fabricated by placing Ni-Cr wire between two pieces of Kapton tape. Five thermocouples were positioned as near to the heater wire as possible in each of the four corners and in the center of the heater. The heater assembly was placed between two aluminum plates that were 1mm thick. The plates were welded fully on three sides and partially on the fourth side, leaving an open gap for the wires to pass through. Temperature measurements were taken while power to the heater was slowly increased until the heater burned out due to localized heating of the wire where it exited the plate.

A plot of the thermal resistance from the heating experiment can be seen in Figure 8. The plot shows that the thermal resistance, defined in the figure, actually decreases as the heater input increases. The explanation for the improvement is that the Kapton tape tends to soften as it gets hotter so better contact is made with the heater wire. More importantly, however, there is no indication that a gap increases as the heater power increases before the heater burned up. Using the measured thermal resistance of about 0.012 K/W, the estimated heat input where the heater temperature reaches ½ the melting temperature of aluminum, 330 ^{o}C, is 13.5 kW.

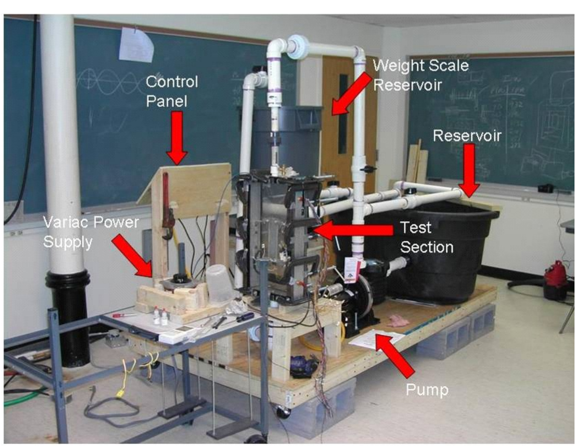


Figure 6. Photograph of Flow Loop

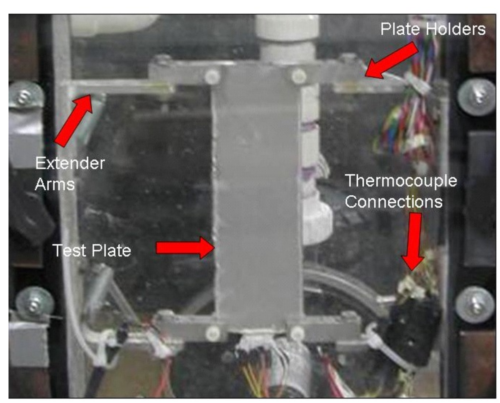
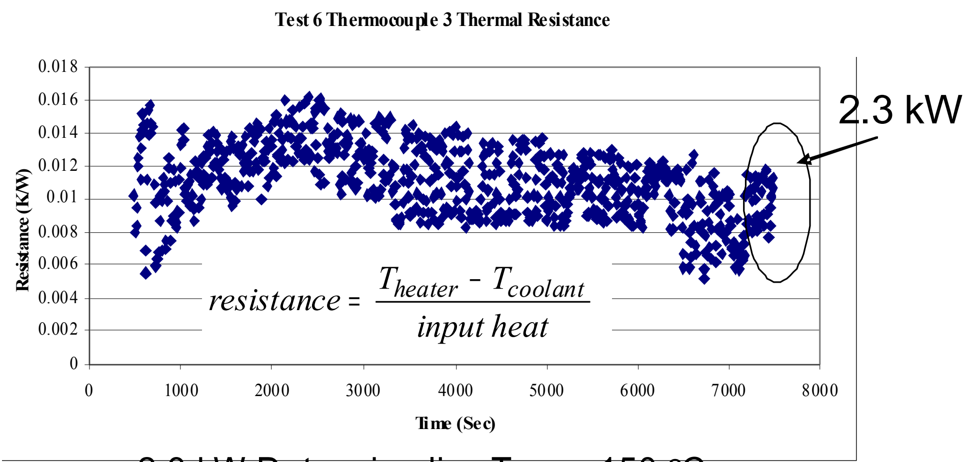


Figure 7. Photograph of Test Target



2.3 kW Data – implies $T_{LEU} \cong$ 150 °C
Linear scaling implies 13.5 kW possible

Figure 8. Experimental Data from Initial Thermal Experiment in Flow Loop

Calibration quality experiments are expected to take place on another flow facility built at Y-12. That facility will allow direction measurement of the plate deflection due to heating. Obtaining that data will be considered the milestone that transitions the plate geometry from TRL 3 to TRL 4.

4.3 TRL 4 AND TRL 5

TRL 4: The key elements must be integrated to establish that the pieces will work together. The validation should be consistent with the requirements of potential applications, but it is relatively low-fidelity when compared to a final product. Examples include integration of ad-hoc hardware or software or with mock material in the laboratory such as breadboards, low-fidelity development components, and rapid prototypes.

Exit criteria: 1) A laboratory prototype has been created which integrates all key elements necessary to address a particular problem or application 2) Demonstrated in lab environment 3) Key elements integrated with mock material or in a mock assembly 4) Metrics for successful performance have been defined and met 5) Potential customer agrees that this provides a solution

TRL 5: Fidelity of the key elements increases significantly. Key elements are integrated with realistic supporting elements so that the technology can be tested and demonstrated in simulated or actual environments.

Exit criteria: 1) A prototype has been built and used to successfully demonstrate required functionality and performance before, during, and after exposure to the customer's environments. Prototype may be built with mock materials and/or in a mock assembly. 2) The demonstration includes all functionality and performance metrics that the customer expects 3) Key and support elements integrated in a realistic manner. 4) Metrics for successful performance in relevant environments have been defined, met and documented. 5) End user customer known, and customer agrees that testing environment is relevant 6) Customer and technology supplier are working together to formally define/document requirements.

As noted above the annular target has achieved TRL level 5 as reported in [1] where an LEU sample has been irradiated at various sites. In order for the annular target to progress further, high volume manufacturing integration needs to be taken into consideration. We are currently completing a study on the impact of manufacturing variation on the gap between the LEU foil and the target cladding. The study consists of measuring the foil thickness variation before assembly and then taking apart the target and measuring the foil thickness after assembly. The goal is to understand the degree of plastic deformation. The goal will then be to modify the assembly process to see if looser manufacturing tolerances can be used to ease the assembly.

Test irradiations and post-irradiation examination is necessary in order for the plate target to proceed through TRL levels 4 and 5. A test coupon would be sufficient to establish the proof-of-concept.

5.0 CONCLUSIONS

The current status of LEU foil-based target design for high-volume production of fission product Mo-99 has been discussed. TRL's have been used to assess the maturity of the technology development. The analysis has shown that the plate geometry is at TRL 3 while the annular target is at least at TRL 5. Further model calibration and test irradiations are necessary for the plate design to proceed further while integration with manufacturing practices needs to take place before the annular target can proceed to higher TRL's.

6.0 ACKNOWLEDGEMENTS

We would like to thank Dr. George Vandegrift of ANL and Lloyd Jollay of Y-12 for their support.

7.0 REFERENCES

[1] C. Conner, E. F. Lewandowski, J. L. Snelgrove, M. W. Liberatore, D. E. Walker, T. C. Wiencek, D. J. McGann, G. L. Hofman, and G. F. Vandegrift, "Development Of Annular Targets For 99 Mo Production," 1999 International Meeting on Reduced Enrichment for Research and Test Reactors, Budapest, Hungary, October 3-8, 1999.