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

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ABSTRACT

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 targets utilize relatively expensive 'meat' containing uranium-aluminum alloys in an aluminum matrix. Those designs necessarily result in a large amount of costly 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 an overview of the target assembly and disassembly processes using low cost foil with a large thickness variation. Preliminary models establish the allowable design envelope for thermal contact resistance and coolant requirements. Alternate target geometries are discussed.

1.0 INTRODUCTION

The US market for the radioisotope Mo-99 is estimated to be 6000, six-day Curies per week. In order to supply 50% of the US demand, the University of Missouri Research Reactor (MURR) must establish the means to produce about 27,000 Out-of-Reactor Curies of Mo-99 each week. This includes developing a target, an irradiation process, and chemical process that can be scaled to accommodate approximately 2300 20-g low-enriched uranium (LEU)-foil targets each year. This paper will focus primarily on the target design capable of economically producing Mo-99 and yet be capable of satisfying the safety requirements set forth by MURR and the Nuclear Regulatory Commission (NRC).

The strategy to develop a cost effective target is to minimize the volume of processing waste. The LEU-Modified Cintichem process developed by Argonne National Laboratory (ANL) [1] is the proposed technique to recover Mo-99 from the LEU fission products. The rationale for using the Cintichem process is that the liquid waste from processing the LEU foil is minimized as long as the LEU foil can be removed intact from the target (there will be no need to dissolve the cladding material). This implies that the LEU foil must not bond to the target during the irradiation process, unlike current dispersion target designs.

While reducing the amount of liquid waste, LEU foil that does not intimately bond with the target cladding has the potential for gaps to form. Gaps may also form during irradiation due to fission product gas generation, non-uniform thermal expansion of the cladding material, LEU growth, and LEU swelling [2]. If the gaps become sufficiently large the thermal contact resistance could cause the LEU temperature to increase beyond the value established by the MURR technical specifications.

An annular LEU-foil target has been designed by ANL [3] to minimize the thermal contact resistance while still being easy to disassemble. The design has been successfully irradiated on a demonstration trial basis at BATAN (Indonesia), CNEA (Argentina), and ANSTO (Australia). The target utilizes concentric aluminum cylinders as the cladding material. The LEU foil is fabricated using a series of cold-rolling and annealing steps until a very flat foil has been produced. The LEU foil, wrapped in a nickel foil recoil barrier, is placed between the two aluminum cylinders. The inner cylinder is plastically expanded into the LEU foil and outer cylinder by a drawing process. The ends of the target are sealed by welding.

In order for MURR to utilize the ANL target design concept in a high volume production environment, the cost-per-target needs to be reduced. Three specific cost reduction activities are being pursued. The first is to utilize LEU foil that has a large variation in thickness as compared to rolled foil, but significantly cheaper. The second is to explore different target geometries, including the size of the LEU foil, cladding shape, and simultaneously irradiating multiple targets in the graphite reflector region at MURR. The third is to define a simpler fabrication technique.

Irradiation of LEU targets will be the first time that MURR has irradiated fissile material in the pool area. As such, a safety case must be documented to obtain a reactor license amendment from the NRC. The safety case must establish the mechanical integrity of the target design, while still meeting the MURR Technical Specification requirements. An engineering design process is utilized to efficiently establish a technically and economically viable target.

2.0 TARGET DESIGN PROCESS

To satisfy the MURR safety case, the proposed target must contain all fission products. The target must also satisfy the MURR reactor technical specification requirements that state the temperature of all target materials must be kept below ½ their melting temperatures and the pool coolant must be kept below its saturation temperature. To establish that the chosen design satisfies these constraints, a strict engineering analysis and design process is being followed. The process followed will establish performance limiting design envelopes that are based on physical models and experiments. This is in contrast to analyzing only a specific target design. To facilitate identifying requisite analysis, a schematic identifying the process steps that embody the 'life' of a target is shown in Figure 1.

The first step of the process flow shown Figure 1 is the development of a list of component specifications. The list of component specifications is provided to material suppliers so that it is clear what acceptable incoming material tolerances are met. The quoted tolerances will be developed based on material supplier capabilities and an engineering sensitivity analysis that shows how performance is impacted by the variation. As an example, the LEU foil thickness variation will be specified based on what level of variation can be tolerated, and still maintain the LEU temperature below the MURR Technical Specification requirements.

An incoming material inspection plan, the second process step, needs to be developed to ensure supplier quality control. The rationale is to ensure that the targets will meet performance requirements after going through the developed assembly process, step three. The inspection plan will use statistics to establish the sampling rate and use a fast measurement technique that has been correlated to use conditions.

For the foil based target used in the LEU-Modified Cintichem process, minimizing the thermal contact resistance between the foil and the target cladding is of primary importance. The thermal contact resistance is driven by gaps created during the initial assembly of the target, as well as fission gas, uranium swelling, uranium growth, and non-uniform temperature thermal expansion. Therefore the assembly process will be a critical step to ensure proper thermal performance. Plastic deformation of the aluminum cladding to conform to the LEU foil will be the primary metric to evaluate different assembly processes.

The fourth and fifth steps in the overall target life cycle are loading the target into the graphite reflector region and providing sufficient cooling to the target during irradiation. In the Figure, only a single target in a single channel is shown. However, in production, multiple targets in multiple channels are likely to be utilized. The design challenge is to ensure that proper coolant is supplied in order to ensure that coolant boiling does not take place and that the thermal contact resistance between the LEU foil and target cladding is low enough to keep the LEU temperature below ½ of its melting temperature.

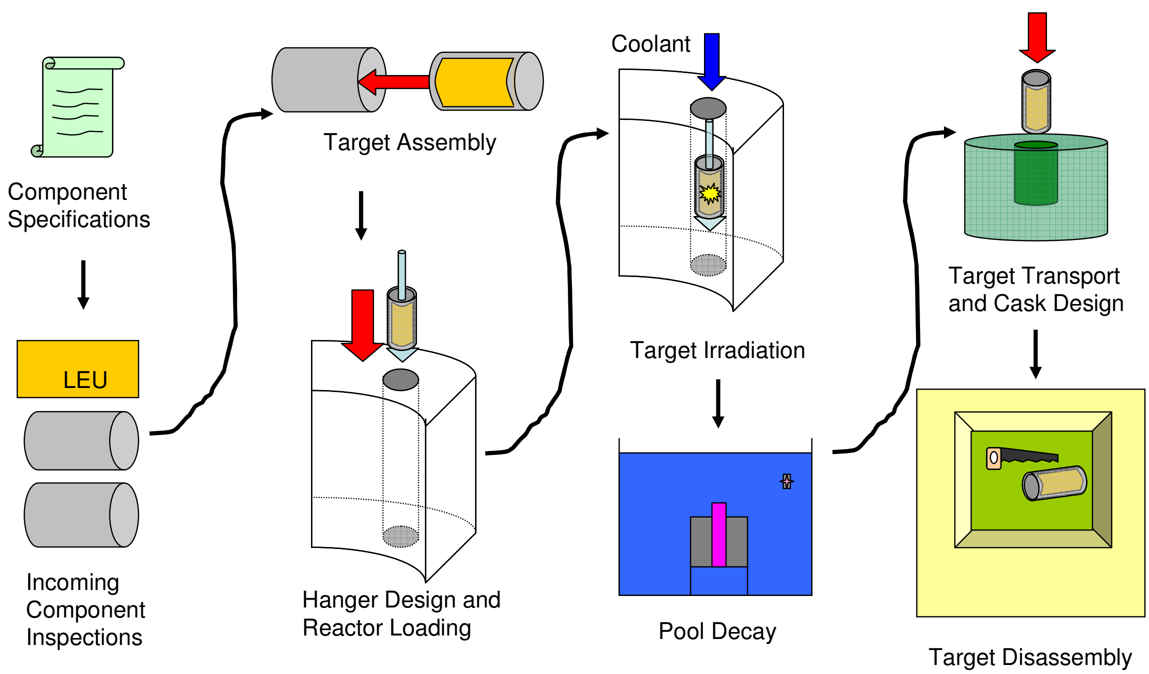

Figure 1. Process Steps for High Volume Irradiation of LEU Foil for Mo-99 Production

After the target has been irradiated, it is necessary to allow short half-life fission products to decay before removing from the pool. In these conditions, the coolant is the stagnant pool water that provides natural convection cooling. The target is then transported to a hot cell for LEU processing in a cask. Any residual heating during the cask transport process needs to be dissipated through the cask. Within the hot cell, the target needs to be cut open to remove the LEU material for chemical processing. The cutting process ideally will not cause chips or dust to be generated.

3.0 THERMAL TECHNICAL REQUIREMENTS

The MURR has two specific thermal technical requirements. The first is that no material may experience a temperature greater than ½ any target material melting temperature. The second is that the local temperature of the coolant may not exceed its saturation value. A top-view sketch of the proposed cylindrical target design is shown in Figure 2. The sketch illustrates that the two technical requirements effectively split the analysis problem into two pieces – one of conduction from the LEU foil to the outside of the aluminum cladding, and the second of convection from the outer surface of the target to the reactor coolant (water).

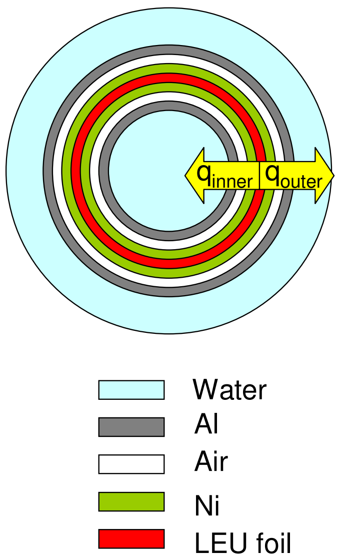

Figure 2. 1-D Model of Analyzed Geometry

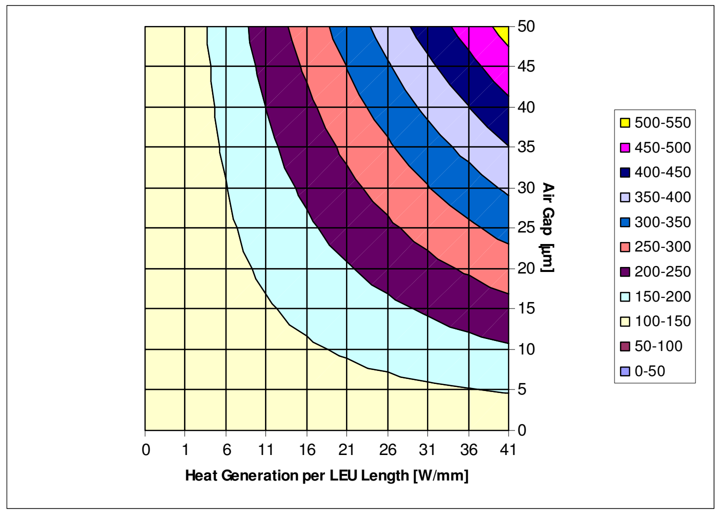

Figure 3. LEU Foil Temperature Map for Uniform Air Gap and Cladding Wall Temperature of 113 °C

3.1 Melting Temperature Requirement

Heat is generated in the LEU foil while it is irradiated, raising the temperature of the foil and the target. Since the LEU is the heat generating component of the target, the LEU will experience the hottest temperature in the target and be subject to the Melting Temperature Requirement. The lowest melting temperature for any of the proposed target materials is that of the aluminum cladding, whose melting temperature is 660 °C. Therefore 330 °C is the maximum allowable temperature for the LEU.

The sketch in Figure 2 suggests that the heat generated in the LEU can be dissipated to the coolant that is flowing through the inside or outside of the target. In either case, the heat must conduct through the nickel recoil barrier, the aluminum cladding, and the interface between the nickel and aluminum, identified as an air gap in the sketch. In reality, a uniform air gap on either side of the LEU foil will not exist in a well built target. The simplified model allows for an initial assessment of the impact of the interface between the nickel and aluminum materials.

The temperature of the LEU foil during irradiation is plotted in Figure 3 for a given air gap and heat load-per-unit length of LEU foil. In the figure, it is assumed that the aluminum cladding that is in contact with the water coolant is at 113 °C, the water saturation temperature. It is further assumed that the temperature drop across the nickel and aluminum is negligible. This last assumption is made based on the fact that the temperature drop across the nickel and aluminum is about two orders-of-magnitude smaller than the temperature drop across the air gap.

Finally, it is assumed that the target diameter will be at lease 20 mm. The figure shows that increasing the air gap or increasing the heat load increases the LEU temperature. If one considers the 330 ^{o}C temperature limit imposed by the MURR Technical Specification, it can be seen from the plot that there is a trade-off between the heat load and allowable air gap. For example, if the heat load that needs to be dissipated is 31 W/mm (total power is 3.1 kW for a 100 mm long LEU foil), the maximum air gap that can be tolerated is about 35 μm.

While the previous analysis assuming a uniform air gap is extreme, it does illustrate an upper limit as to the level of LEU foil flatness variation and maximum LEU foil size that can be clearly tolerated from a thermal performance standpoint. The thickness variation of a sample piece of LEU foil was measured to understand the thickness variation present in a low-cost LEU foil. A photograph of the foil measured is shown in Figure 4. The photo clearly shows that there are significant ridges caused by the manufacturing process. In addition to the ridges, there is a large variation in thickness as shown in Figure 5 where the thickness was measured in 18 different locations.

The measurements shown in Figure 5 indicate that there is a variation in the LEU foil thickness of about 50 μm. By comparing this variation with Figure 3, it is not immediately clear what level of heat dissipation can be effectively removed. Therefore it is necessary to complete a more thorough study of interfacial heat transfer to conclusively establish what the acceptable target design limits are for the NRC safety case. Specifically, it is not clear as to the level of deformation of the aluminum cladding and the LEU foil during assembly that could close air gaps. It is also not clear what the full impact of intermittent contact between the LEU foil and the aluminum cladding will have to the interfacial heat transfer.

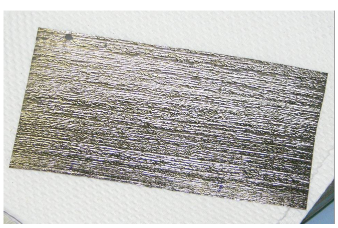

Figure 4. Photograph of Cost Effective LEU Foil

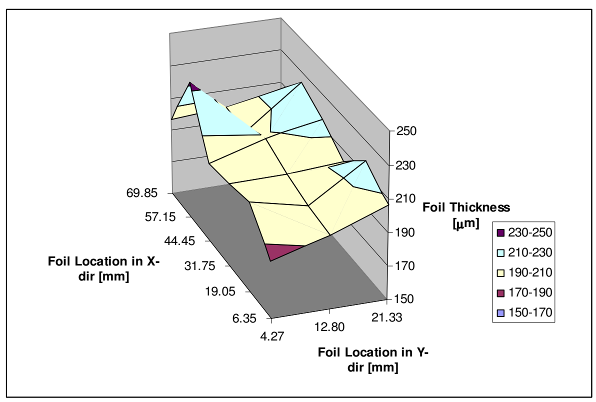

Figure 5. Topography of Cost Effective LEU Foil

3.2 Coolant Saturation Temperature Requirement

The saturation temperature of water at a depth of 6.1 m (20 feet), where the pressure is 158 kPa (23 psi), is about 113 ^{o}C. The coolant water temperature must be kept below this value in order to satisfy the second MURR technical requirement. This implies that the outer target surface needs to be kept below 113 ^{o}C.

It can be demonstrated from heat exchanger theory that the maximum heat flux which can be dissipated from the target surfaces depends on the water flow velocity, the target aspect ratio (LEU length/Cylinder diameter, or L/D) and the temperature difference between the target surface and the coolant inlet [4]. The maximum heat flux effectively defines the size of the LEU foil that can be irradiated. It should be noted that the total heat generated within the LEU foil can be dissipated from the interior and exterior target surfaces.

The maximum heat flux for a range of water flow velocities is shown in Figure 6. The calculations used for creating the plot assume that the heat transfer rate from the internal and external surfaces of the cylindrical target is the same. The analysis also assumes that the target surface temperature is 113 °C while the water inlet temperature is 50 °C. The plot shows that the heat flux increases with a power of about 0.8, based on the Nusselt number correlation used to determine the heat transfer coefficient.

A total heat dissipation map is created in Figure 7 assuming the target length to be 10 cm. The target length is based on the neutron flux distribution in MURR. For a 20 g target, roughly 20 kW will need to be dissipated. At a flow velocity of just over 3 m/s, this implies a cylinder with an L/D ratio of about 5 needs to be used.

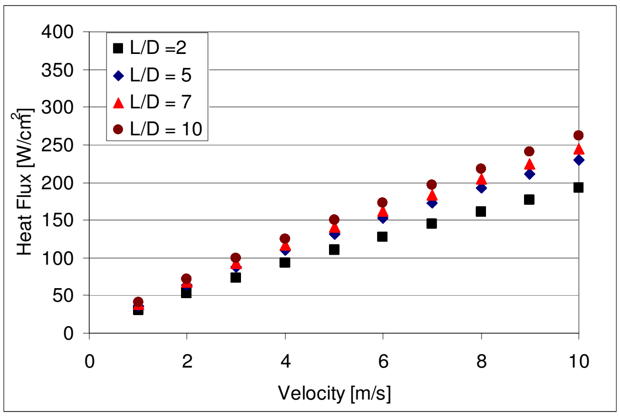

Figure 6. Maximum Target Heat Flux to Ensure Coolant Temperature Stays Below Saturation Temperature (113 °C)

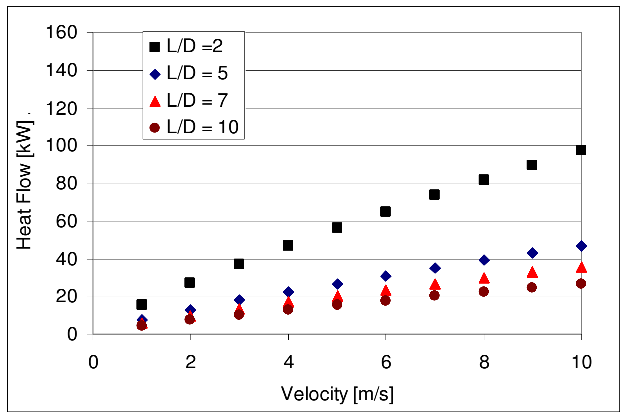

Figure 7. Total Heat Dissipation from Annular Target into Coolant Water at 50 °C.

It is clear that the amount of heat that can be dissipated from a single target, and hence the size of the LEU foil that can be irradiated, depends on the water flow velocity. Flow measurements were taken using a hot wire anemometer to obtain an estimate of what might be expected in MURR. The measurement process is explained in detail in Solbrekken, et al [4], however the flow velocity in the targeted irradiation channel was found to be 3.4 ± 0.8 m/s at a 95% confidence interval. The rather large uncertainty can be traced to the extreme sensitivity of the hot wire probe to temperature and turbulence fluctuations. Additional flow measurements are planned in order to establish a more accurate performance estimate. Multiple channel flow studies are currently under way.

4.0 TARGET MECHANICAL DESIGN

The primary mechanical requirements for the target are to contain all fission products and to facilitate the satisfaction of the thermal technical requirements. As such, one can envision multiple possible target geometries ranging from the Argonne annular design to a plate configuration. The final design configuration will provide the lowest overall production cost of Mo-99.

The Argonne target geometry is annular to provide superior mechanical strength. The LEU foil is wrapped in a nickel foil that acts as a fission recoil barrier. The nickel allows the LEU to be easily removed from the aluminum tube cladding. The assembly of the target starts with wrapping the LEU foil with nickel foil of 0.015 mm thick. The wrapped foil is then inserted between two concentric aluminum tubes (annular target) as shown in Figure 8. To prevent foil slip during insertion, a grove is made in the inner tube. A longitudinal line was scribed on the outer tube for the foil gap indication. In order to assure good contact between the foil in the concentric tubes, a draw press is used to draw the whole assembly of the annular target as shown in Figure 9.

The current target sealing process is by TIG welding. It is expected that the welds will hold under fission-gas pressure that develops during irradiation. However, this design envelope needs to be evaluated. Further, the probability of the target structure failure during irradiation due to U-growth and U-swelling needs to be established for the safety case.

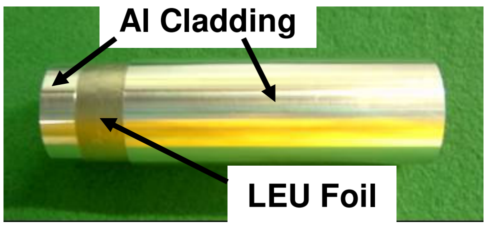

Figure 8. Annular Target Before Assembly

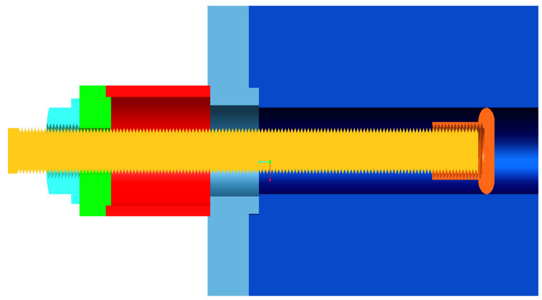

Figure 9. Sketch of Drawing Press Used for Target Assembly

El-Gizawy et al [5] have analyzed the mechanics of the ANL drawing process using finite element method. The objective of this analysis was to explore the safe operating conditions that would result on sound consolidated target without any defects. Their results show that using conical punches with certain geometry under specific deformation conditions would assure plastic deformation through the inner tube wall thickness but only elastic deformation in the foil and the outer tube. This would be sufficient to establish full contact at the interface between tube surfaces and the foil materials. For alternate target geometries, such as plate or curved plate, additional structural analysis needs to be completed.

4.1 Disassembly

The University of Missouri modular design process is used to develop disassembly device for removing the irradiated LEU foils from their annular targets remotely inside the hot cell [6]. All previously reported designs are evaluated in order to gain knowledge of the limitations existed with available devices. The new design called “UM Disassembly Device for Annular Target”, is based on slitting mechanism using commercially available cutters. The final configured design is displayed in Figure 10

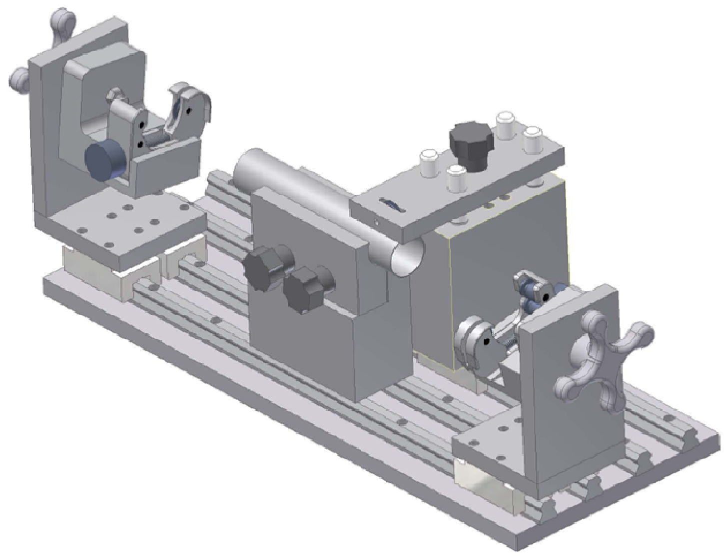

Figure 10. Configured Design of the UM Disassembly Device

UM Disassembly Device has the following unique features and capabilities: 1- it uses one holder and three cutters in succession, therefore eliminating time consuming and unsafe re-handling of the delicate assembly; 2- it is compact in order to fit available hot cells with allowances for tooling and manipulators; 3- the developed process does not generate radioactive chips, therefore eliminating expensive and time consuming disposal processes; 4- the design is modular and uses standard parts for simple construction and repair operations; 5- it is flexible and can be easily modified for disassembling annular targets with different dimensions. A fully automated version of the present design using stepper motors and programmable logic controller is recommended for future development for high production fission product Mo-99.

CONCLUSIONS

An outline of the design process for a high volume production target that utilizes LEU foil has been described. A design methodology based on performance maps has been presented with examples provided for the Thermal Technical Requirements. Further studies are identified that will need to be completed in order to fulfill the documentation requirements for an NRC license amendment to begin production of Mo-99.

ACKNOWLEDGEMENTS

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