STATUS OF ANSTO MO-99 PRODUCTION USING LEU TARGETS

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ABSTRACT

The Australian Nuclear Science and Technology Organization (ANSTO) has produced Mo-99 using Low Enriched Uranium (LEU) UO_2 targets for nearly thirty years. The Replacement Research Reactor (RRR) provides ANSTO with a good opportunity to review and improve the current Mo-99 production process. Uranium target design improvements were performed through a collaborative effort with Argonne National Laboratory under the auspices of the RERTR program. The ANSTO program was also focused on identifying a manufacturer for LEU foil targets. To this end, ANSTO contracted CERCA to develop the methods for LEU foil target manufacture This paper, presents the latest results and conclusions of this program.

Introduction

ANSTO currently produces Mo-99 using uranium dioxide pellets enriched to 2.2% uranium-235. This method consists of irradiating the pellets in the HIFAR reactor at an average flux of 8×10^{13} $n/cm^2/sec^2$. This process has been in operation for over 20 years but is limited by the amount of Mo-99 which can be produced with the 2.2% enrichment level of the target. With a new research reactor being built it was decided to review this process and develop an alternative target with a higher enrichment level. It was important that any new target had an enrichment level of less than 20% uranium-235 to comply with Australia's non-proliferation commitments.

ANSTO and Argonne National Laboratory (ANL) entered into collaboration in 1999. The collaboration focused on the development of ANL's uranium metal foil target concept for application in the HIFAR reactor. ANL had undertaken some early irradiations of this target and were keen demonstrate the target technology for Mo-99 production. The target consisted of a uranium metal foil sandwiched between concentric cylinders of aluminium.

The uranium metal foil is removed from the target body for processing. This removes a bulk of aluminium which then does not need to be dissolved, thus reducing the volume of liquid waste from any processing technique. This is an important consideration for any manufacturer but in particular for those converting HEU targets to LEU. In order to prevent spot welding of the foil to the aluminium cylinders by fission fragments the uranium target foil was wrapped in another foil of appropriate thickness. For ANSTO's process using nitric acid dissolution of the target the uranium foil was wrapped in nickel foil.

The Mo-99 target development program was conducted in two phases. The purpose of the concept phase was to evaluate the use of LEU foils for Mo-99 production. A prototype target (LEUFR) was irradiated, processed and Tc-99m generators prepared and evaluated according to ANSTO protocols. This phase was regarded as successful. The second phase of the program was directed at HIFAR validation of an annular LEU target that would be similar to that used in the Replacement Research Reactor (RRR) and establishing the long-term manufacturability of the LEU foil targets.

Concept Testing

In order to test the target concept ANSTO developed four targets which were given the acronym LEUFR. The nickel electroplated uranium metal foils were provided by ANL with U-235 enrichment of 19.81%. The LEUFR prototype target was designed to fit existing HIFAR irradiation rigs. The initial irradiation was thermocoupled to validate the thermalhydraulic codes used to predict behaviour of the target during irradiation at a flux of 0.7×10^{14} $n.cm^{-2}s^{-1}$ for 6 days. This and subsequent LEU foil irradiations were processed in the usual manner, and Tc-99m generators produced and evaluated. Whilst not used for medical applications, the Tc-99m produced met all specifications. The results of this investigative program were reported at the RERTR 2002 meeting in Bariloche, Argentina [[i][1]].

One problem experienced during this concept testing was the post-irradiation removal of the uranium foil from the target. Testing prior irradiation indicated target disassembly was easily achieved in three cuts - two for the can ends and one longitudinal cut. The longitudinal cut through the outer aluminium sleeve allowed the two aluminium cylinders to slide apart permitting easy foil removal. However, after irradiation, this proved not to be the case. The aluminium cylinders did not come apart and considerable effort was required to separate the uranium foil from the target. It was determined that any future development of the target should be accompanied by the development of an effective can opening device. It was also established that thinner walls than those used in the LEUFR concept aid disassembly.

Scale-up Activities

Initially, the program of work was to test "full-size" uranium metal foil targets in HIFAR and then convert production over to the new target, contingent on successful irradiation trials, regulatory approvals and necessary infrastructure being in place. As the program of work proceeded this aim was found to be impossible since (a) power loads and unloads in HIFAR were not possible with new targets and rigs (b) no post-irradiation target cooling facilities existed in HIFAR i.e. post-irradiation target cooling required HIFAR shutdown for in-core cooling. Such an interruption to normal operations could not be sustained while maintaining other reactor services for radioisotope production and neutron beam use. Therefore the scope of the investigation in HIFAR was modified in order to demonstrate the irradiation of a small number of "full-size" LEU targets in HIFAR followed by processing.

Approval from the Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) would be required prior to undertaking irradiation and processing experiments. Thus, the development program at ANSTO focused on studies required to
(a) address all safety aspects associated with irradiation of 19.75% enriched metal foils in HIFAR (target and rig design, operation, excursion conditions, etc)
(b) demonstrate compliance with Operating Limits and Conditions for heat flux and power for HIFAR, and additional HIFAR license conditions
(c) address all safety aspects and infrastructure modifications required for the processing of 19.75% enriched metal foils for Mo-99 production.

Target Design Studies

In order to derive a meaningful specification for target manufacture, ANSTO conducted a series of investigations to ensure optimum target design characteristics.

The water tunnel facility at ANSTO was used to conduct flow visualisations and measurements at discrete locations within the narrow spaces in a transparent mock-up fuel element irradiation rig model. The use of Laser Doppler Velocimetry (LDV) enabled collection of non-intrusive point velocity measurements. Additionally, a three-dimensional computational fluid dynamics (CFD) model was used to investigate the hydraulics behaviour within a HIFAR fuel element model of the liner and irradiation rig, located in the central portion of the HIFAR fuel element**.** The flow visualisations and measurements substantially assisted in checking the CFD model predictions of the fluid flow. The predicted flow behaviour was comparable to the experimental observations. Predicted velocities were also found to be in good agreement with LDV measurements.

CFD simulation of the fluid flow through the various components of the mock-up fuel element model that included the irradiation rig and annular target cans was performed. Based on the experimental flow rate of 1.6965 kgs^{-1} and a base diameter of 0.3 m of the mock-up fuel element model, a very low flow velocity outside of the liner nose cone was predicted as portrayed in Figure 1(a). The fluid directed through the small bottom and side holes of the liner nose cone caused merging flows to interact, yielding a highly complicated flow structure. This flow structure consisted of multiple vortices of recirculating flows (see Figure 1(b)).

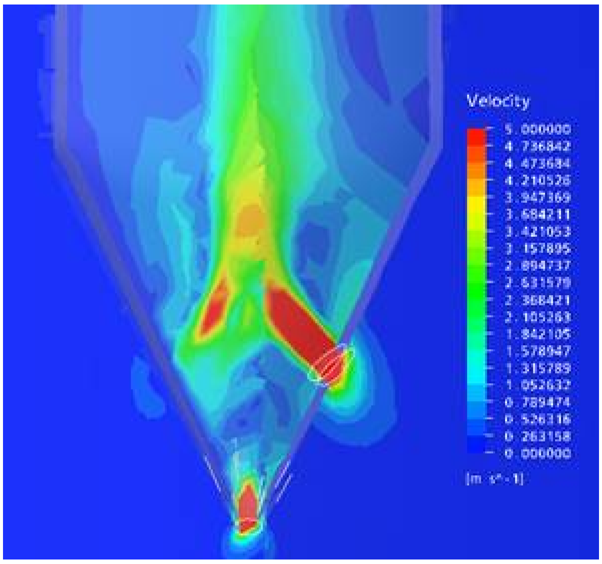

Figure 1 (a): Flow distribution velocity contours predicted inside and outside of the liner nose cone.

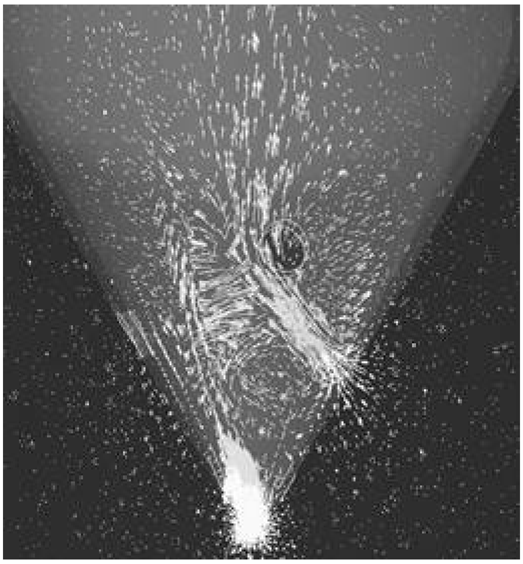

Figure 1 (b): Flow distribution velocity vectors predicted inside and outside of the liner nose cone.

Near the bottom hole of the liner nose cone, the CFD model predicted a normal velocity of approximately of 3 ms^{-1}. This predicted value was found to be in good agreement with the experimental LDV measurement of 2.6 ms^{-1}, thus providing confidence in the reliability of the models employed in the CFD computer program.

The rig and annular target prototypes pictured below are the result of a design approach integrating experimental and theoretical thermalhydraulic data with manufacturability and safety features for the HIFAR situation. Nominal uranium foil dimensions were provided to CERCA at this stage. In parallel, further CFD calculations were undertaken in order to ensure that an LEU foil of nominal thickness 120 micron would comply with HIFAR operating limits and conditions during irradiation and excursion scenarios.

Figure 2: ANSTO prototype rig and annular targets for HIFAR

It was established through neutronics calculation that a total power of 32.7 kW (15.3 kW for the upper target and 17.4 kW for the lower target) would be produced during irradiation. The heat generation of 15.3 kW and 17.4 kW was provided as input of volumetric heat sources to the heat transfer calculations in CFX5.6 for the upper and lower foils respectively. Constant thermal conductivities for the aluminium and uranium were employed for the heat transfer calculations. The inlet temperature of the heavy water in all the calculations was taken to be 45°C. The mass flow rate entering the fuel element was 16 kgs^{-1} while the amount of bypass coolant flow in the liner was taken to be 5% obtained through the water tunnel experiment[ii[2]] .

The temperature distributions of the upper and lower uranium foils, which are located within the aluminium targets are pictured in Figure 3. Since the lower foil was irradiated at a higher power than the upper foil, the temperature in the lower foil achieved a maximum temperature of approximately 367°C. The maximum temperature of the upper foil was significantly lower reaching a temperature of 322°C. Figure 3 also illustrates the varying temperature distribution along the two targets. Owing to the high conductivity of aluminium, there exist only very small temperature differences within the finite thickness of the two targets.

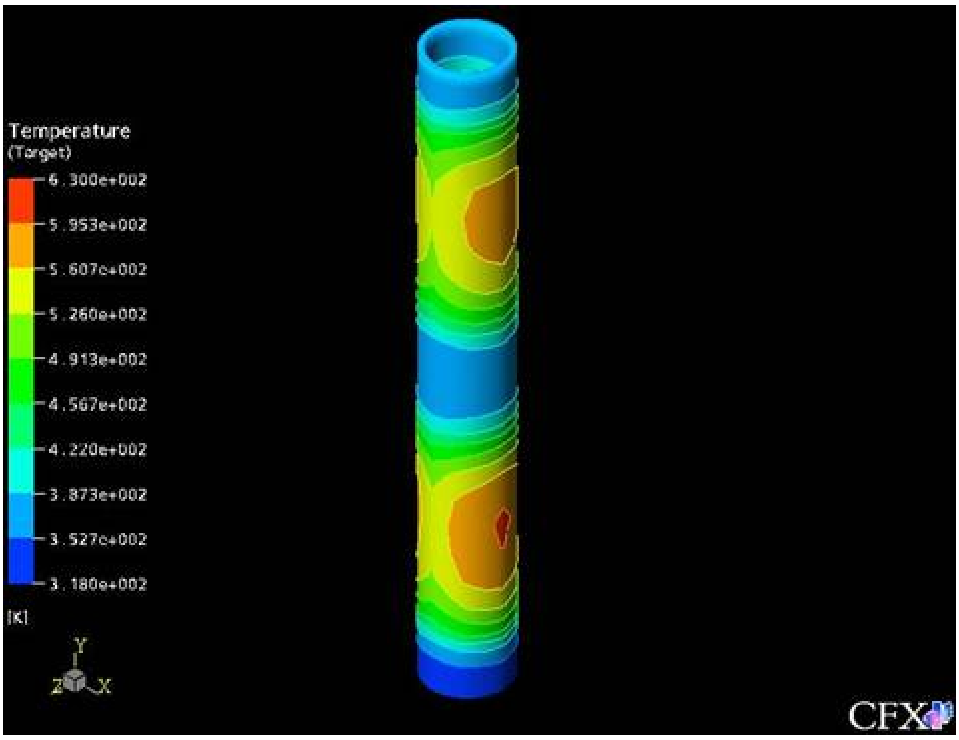

Figure 3: Uranium metal foil temperature (K) distribution calculated by CFX5.6

The temperature profiles of aluminium walls adjacent to uranium metal foils are illustrated in Figure 4. The maximum temperature, predicted through computations, at the upper target is 92°C. The bypass flow is seen to be rather effective in cooling the lower target. As more heat is deposited into the coolant flow, the temperature increases as it progressed downstream as depicted by the higher temperature profiles for the upper target.

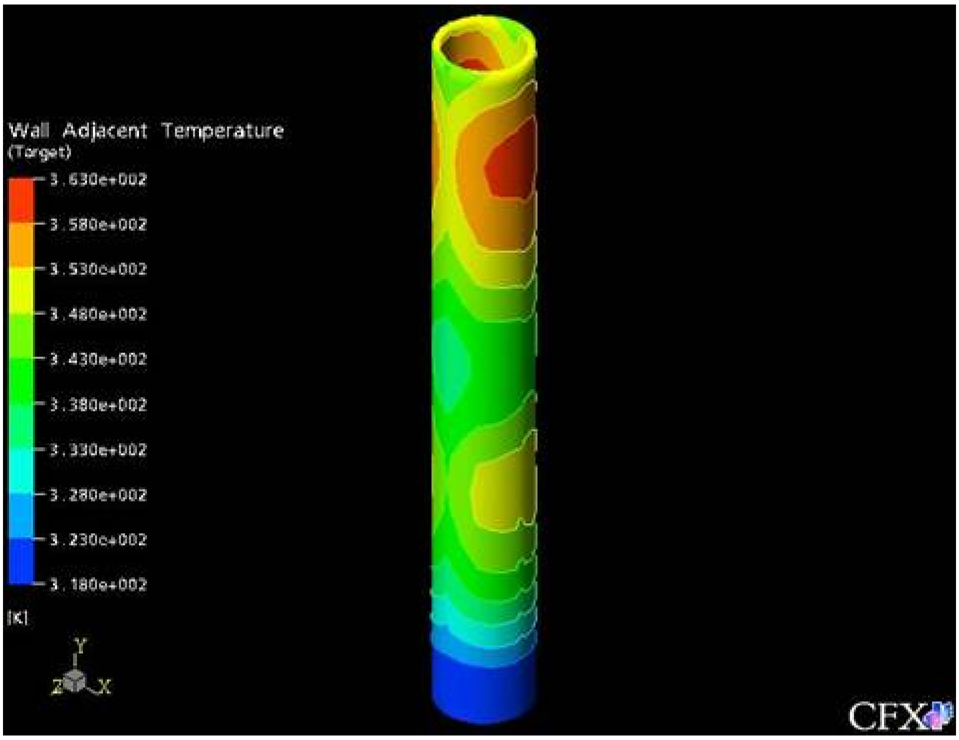

Figure 4: Aluminium wall temperatures (K) distribution calculated by CFX5.6

The wall heat flux distribution for the two targets is represented in Figure 5. Here, the model predicts a local maximum heat flux of 92.5 W/cm^2. In summary, it was calculated for the base case (10 MW reactor power) with 5% bypass flow that the maximum wall temperature predicted was 92 ^{o}C with a local maximum heat flux of 92.5 W/cm^2. The downstream exit temperature near the fins of the X216 rig is determined to be 60^{o}C. The maximum temperature in the foil is 367^{o}C. These results complied with the HIFAR operating limits and conditions for heat flux and power.

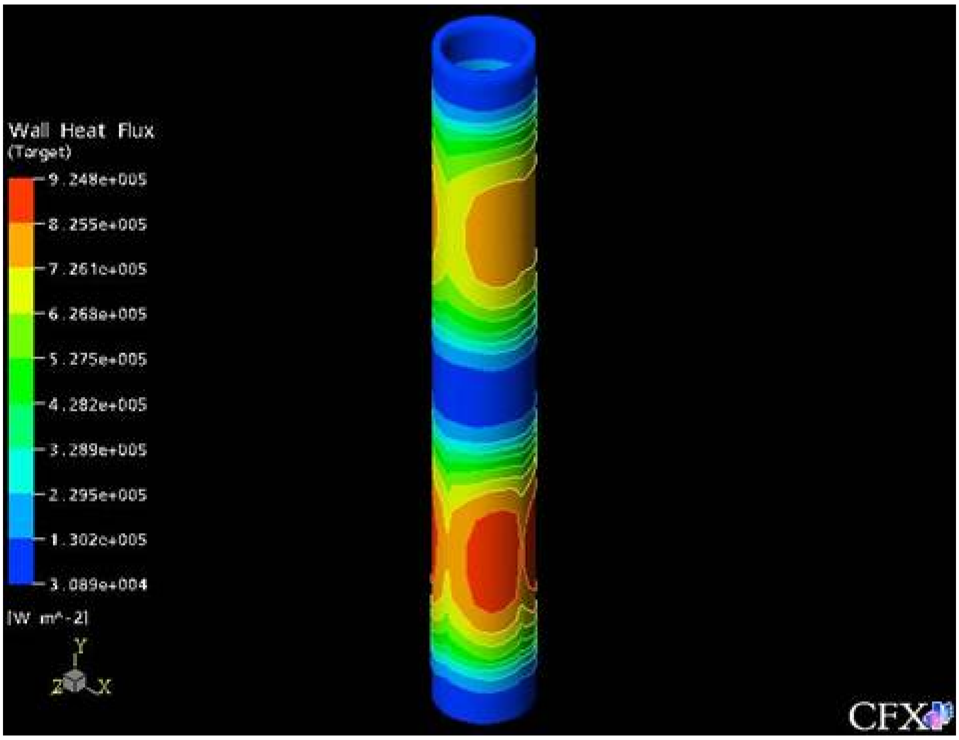

Figure 5: Heat flux distribution for LEU target walls as calculated by CFX5.6

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3. CERCA Manufacturing Development

Under the auspices of the U.S/DOE and through an agreement with ANSTO, CERCA has developed an industrial scale production process for manufacturing annular cans targets. The manufacturing process flow consists of four main stages and the scheme is outlined in Figure 6. Each stage of the process is explained here after.

Target design requires that the uranium foils should not have a preferential grain orientation to ensure that growth due to neutron irradiation does not occur in any specific direction and cause physical target failure. CERCA has access to technology for uranium foil production that grows isotropically under neutron irradiation. The foils are produced by cooling roll casting and have a thickness range between 100 and 120 µm. The foils quality which is not yet stabilized has to be optimized. The main issue is the high thickness variation which has to be reduced as an acceptable range. This quality parameter plays a key role for obtaining the right U_{235} mass per target - ^{99}Mo yield production as well - as achieving a perfect thermal contact of the components after assembling.

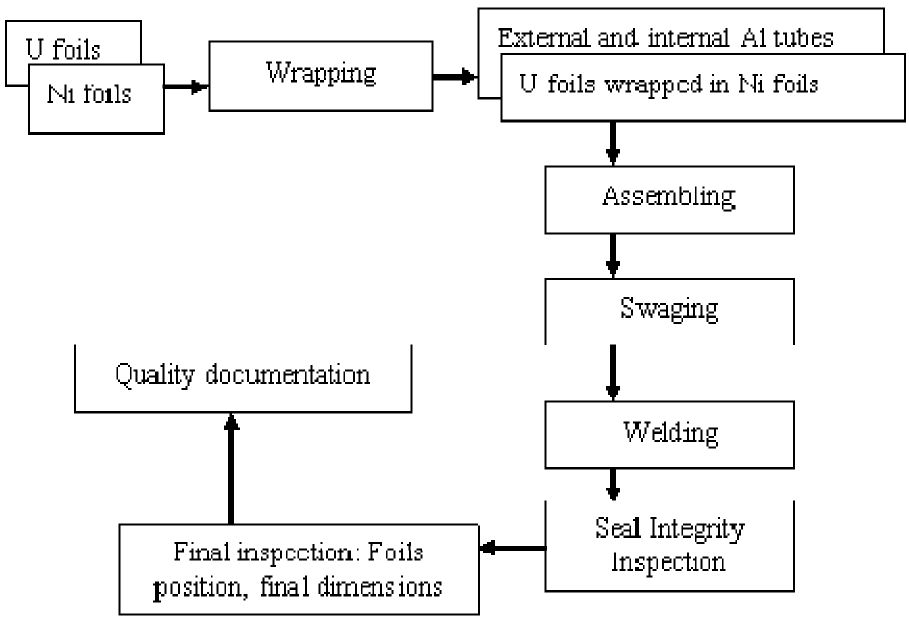

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Figure 6 : Main Stages of CERCA's Annular Target Manufacturing Process

The uranium foils were wrapped in nickel foil then placed in a longitudinal housing along the inner surface of the aluminium cylinder. The cylinders were swaged by drawing a plug along the cylindrical structure and then welded together along the circumference at both ends. Swaging causes plastic deformation of the internal aluminium cylinder and elastic deformation of the external cylinder which ensures a maximum contact between the aluminium surfaces and the nickel-wrapped uranium foils. This operation is now fully qualified and meets all the requirements.

The assembled targets must provide a hermetic seal to fission products under irradiation conditions. This is assured by using a TIG welding process. CERCA demonstrated and validated the preparation and welding sequence on prototype samples, depleted and low enriched uranium targets. Target sealing integrity inspection is assured by a CERCA well known technology based on Helium leak test. This method was also qualified through sampling a significant number of annular targets. Moreover, a new industrial method for controlling the intimate contact of aluminium and uranium compounds after assembly was developed and validated by CERCA. All the manufactured uranium targets were found in compliance with the thermal transfer criteria requested for performing the irradiation planned by ANSTO.

Mastering thermocouples implantation for in core instrumentation, specific instrumented annular cans targets were also manufactured. These instrumented targets were planned to be used by ANSTO for thermal code validation and safety demonstration during preliminary irradiations.

From manufacturing point of view, the industrial annular can target production process was developed and important know-how was acquired. Uranium foil quality remains to be improved by reducing the thickness variation along the foil.

Summary

The targets CERCA has manufactured meet ANSTO specifications. The theoretical calculations indicate that these LEU uranium foil targets could be safely irradiated in HIFAR. Preliminary irradiation and small-scale processing of these LEU uranium foil targets indicates that the resulting Mo-99 and Tc-99m generators meet specification.

In parallel with the target development effort, the current Mo-99 production facilities and process were evaluated to identify what changes were required as a consequence of using the low enriched targets. This evaluation also had to take into account anticipated scale of production in the hot-cell facilities. Key issues to address were: control and minimisation of radioactive gaseous emissions, criticality safe design of process equipment, criticality safe design of uranium containing liquid waste streams, compliance with ALARA for all equipment and facilities to be used at increased production levels, long-term management of uranium and fission product containing wastes

The detailed design of required infrastructure was significantly more complex than had been anticipated from the preliminary design estimates. Many of the design complexities were linked with the chemistry of ANSTO's production process – an acidic process. Thus, due to budgetary reasons, it has been decided not to progress implementation of LEU foil targets as this time.

References

[i[1]] Donlevy, T.M., Anderson, P.J., Beattie D., *et al* "Low Enrichment Mo-99 Target Development Program at ANSTO" RERTR 2002, Bariloche, Argentina

[ii[2]] Yeoh, G.H., 2003 "Comparison of Computational "Comparison of Computational Fluid Dynamics Model Predictions for the X216 Rig and Target Design with Water Tunnel Observations and Measurements," NTD/TN 281