Low Enrichment Mo-99 Target Development Program at ANSTO

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Abstract

The Australian Nuclear Science and Technology Organisation (ANSTO, formerly AAEC) has been producing fission product Mo-99 in HIFAR, from the irradiation of Low Enrichment Uranium (LEU) UO_2 targets, for nearly thirty years. Over this period, the U-235 enrichment has been increased in stages, from natural to 1.8% to 2.2%. The decision to provide Australia with a replacement research reactor (RRR) for HIFAR has created an ideal opportunity to review and improve the current Mo-99 production process from target design through to chemical processing and waste management options. ANSTO has entered into a collaboration with Argonne National Laboratory (RERTR) to develop a target using uranium metal foil with U-235 enrichment of less than 20%

The initial focus has been to demonstrate use of LEU foil targets in HIFAR, using existing irradiation methodology. The current effort focussed on designing a target assembly with optimised thermohydraulic characteristics to accommodate larger LEU foils to meet Mo-99 production needs. The ultimate goal is to produce an LEU target suitable for use in the Replacement Research Reactor when it is commissioned in 2005. This paper reports our activities on:

- The regulatory approval processes required in order to undertake irradiation of this new target
- Supporting calculations (neutronics, computational fluid dynamics) for safety submission.
- Design challenges and changes to prototype irradiation can
- Trial irradiation of LEU foil target in HIFAR
- Future target and rig development program at ANSTO

Target Development - LEUFR prototype

Australia is a signatory to the nuclear non-proliferation treaty and has a national commitment to the use of LEU targets. In collaboration with ANL, ANSTO has been investigating LEU (~19.8% U-235) metal foils as potential targets for future Mo-99 production at ANSTO.

Our program was structured to undertake initial feasibility studies before committing to larger scale trials. The initial feasibility studies were designed for the

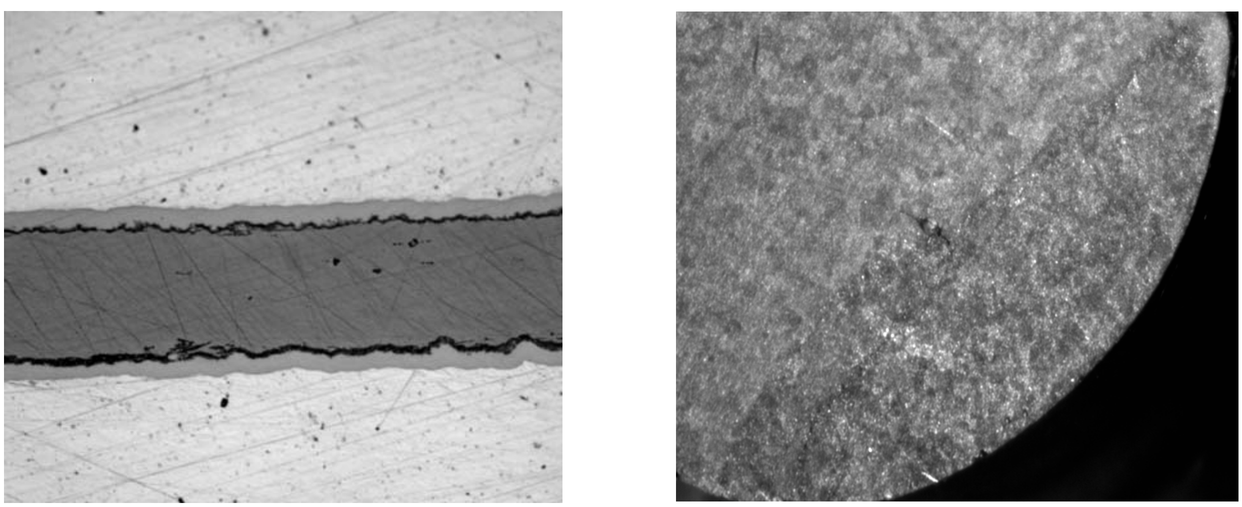
- HIFAR irradiation of a small piece of LEU foil under the existing HIFAR Operating Limits and Conditions (OLCs) for heat flux and power
- processing of the foil according to ANSTO's proven procedure
- preparation and testing of Tc-99m generators manufactured with Mo-99 produced from LEU foil.

The prototype target was designed to fit in the existing HIFAR irradiation rigs, thereby determining the external target dimensions and anchoring mechanism of the target within the rig. An annular target design was developed, in which the uranium metal foil was sandwiched between the outer and inner aluminium sleeves of the target can (Figure I (a) and (b)).

(a) (b)

Figure I (a) Annular target can components (b) Assembled annular target

Studies were also done on the number and dimensions of cap holes and the number and placement of base fins in order to optimise flow through and around the target. The manufacturing development addressed the issue of reproducible foil contact and weld quality for a given set of assembly conditions. Examples are below



(a) (b)

Figure II (a) Sectıon ot nickel plated DU foil (125 micron thick); note good contact of aluminium walls with DU foil (b) section of weld exhibiting low porosity and microcracks

Safety submission

The feasibility irradiations were subject to an internal safety assessment on irradiation can design, assessment of relevant safety issues and potential hazards, compliance with safety analyses considered in the HIFAR Safety Document (the reactor SAR) and HIFAR Operational Limits and Conditions.

Potential hazards identified and addressed in the safety analysis were:

1. heat flux and total heat limits being exceeded during irradiation or unload of the irradiated can
2. failure of the can welds and ingress of water into target region
3. over pressurisation of the can
4. Target coolant flow blockage
5. impact to the target and can resulting from a limiting reactor transient
6. impact on the reactor from inadvertent withdrawal or failure of the rig and target assembly
7. damage to the foil during loading to or removal from can and release of fission products
8. flotation of the can during irradiation and subsequent dropping off rig during transfer

Analyses, including neutronics calculations, thermal-hydraulic calculations, fluid dynamics experiments and reactor transient calculations, were completed. In the case of the thermal-hydraulic and fluid dynamics studies, information was incorporated into final target can design.

Neutronics calculations were undertaken to estimate the reactivity worth, energy deposition, the molybdenum production and heating rates in the target. The calculations were undertaken using the AUS neutronics code system[1] for irradiation of the target in the central position of the reactor at a thermal neutron flux of 1.3 x 10^{14} $n.cm^{-2}$. In order to undertake analysis of the trial irradiation, it was necessary to develop a model that accurately represented the annular can design. The acceptability of the model was verified by comparing results for a case where no rig or targets were loaded, with results from previous AUS models known to give good comparisons for irradiations with "rocket cans" which use 2.2% enriched uranium.

The model developed for this calculation assumes that the fuel element in the C3 position is highly irradiated (to 80 MWd, 73 g ^{235}U remaining from an initial mass of 170 g), providing a high thermal neutron flux, and high fission rate in the uranium target. The trial irradiation however, took place in an outer core position in a fuel element of low burn-up and thermal neutron flux of approximately 0.7 x 10^{14} $n.cm^{-2}.s^{-1}$. Under these conditions, the reactivity, production and heating rates calculated, over-estimated those in the LEUFR target, thereby yielding conservative results. The calculations used a ^{235}U target mass of 0.4 g, slightly greater than the mass of the actual target chosen for irradiation of 0.371 g, also providing another measure of conservatism.

Calculated results of powers generated in different targets (the standard "rocket" cans with 2.2% enriched uranium, and the 20% enriched uranium metal target) are shown below. For the trial irradiation powers were estimated by scaling the results by the ratio of measured thermal neutron fluxes in the respective positions.

Table 1 Heat Generated In Irradiation Cans and Targets (kW)

Irradiation Target Configuration	1 Rocket Can in C3	4 Rocket Cans in C3	LEUFR Can in C3	LEUFR Can in A1
Target only	3.27	13.08	1.75	0.94
Can & Target	3.66	14.64	2.17	1.17
Can, target, rig, coolant & liner	8.89	18.07	7.40	3.98

A series of measurements were made using ANSTO's Water Tunnel Facility. These measurements included, pressure loss characteristics, flow velocity measurement; and flow visualisation.

The measurements were used to validate the thermal-hydraulics calculations and provided input data for the optimisation of the annular can design. A number of design features were assessed during the measurements to optimise the thermal-hydraulics performance of the annular can. These included:

- the number and orientation of coolant exit holes in the can lid (4 or 8);
- the diameter of coolant exit holes (2,3 or 4 mm); and
- the number and configuration of fins on the base of the can. (6 or 8)

A full-scale liner, rig and can assembly were used under low flow conditions (flow rates between 0.13 and 0.23 $l.s^{-1}$ were studied; nominal flow through the rig and liner is 0.16 $l.s^{-1}$; maximum flow 0.17 $l.s^{-1}$) to determine the relationship between flow rate and pressure loss. Measurements were made with, the liner alone; the liner and an unloaded rig;and the liner, rig and various can designs. These measurements showed that approximately 90% of the pressure drop occurs at the liner entry, and that the can design and configuration of holes does not influence the pressure drop significantly beyond that produced by the liner and rig.

Point velocity measurements of flow were made using 1.9:1 scale models of the liner, rig and cans using the Laser Doppler Velocimetry (LDV) method,. Flow measurements with the 4-hole can showed that there was much greater flow through the central can region than between the can and rig (where the risk of burnout is greater), than was the case with the 8-hole model. This, together with a more demanding fabrication process associated with 4-hole design, resulted in a decision to pursue the 8-hole configuration, which provided a better distribution of flow between the central and outer channels than that of the 4-hole design.

Several alternatives of the can base-fins were considered, including 6 and 8 fin configurations with a variety of fin designs. The 6-fin designs were assessed to have an inherent limitation of the possibility that two fins could cover the coolant-flow slots in the rig platforms on which the cans sat, thereby reducing flow to the compartment containing the can. It was concluded that a can with 8 fins machined with a 10 mm end-mill producing slots tangential to the outside diameter of the can was preferred.

In order to identify the range of possible can temperatures, a number of cases were considered with different degrees of contact between the foil and inner can surfaces, including:

- continuous contact between the foil and can, and
- a continuous air-gap of 0.01, 0.05 and 0.1 mm between the foil and can (no contact)

Using the CFX4 computational fluid dynamics (CFD) computer code a complex mesh model of the detailed liner, rig and annular can design, were undertaken to simulate the thermal-hydraulics performance of the target in the HIFAR geometry. Models with coolant exit holes of 2 mm and 3 mm diameter were assessed using CFX4.

The model predictions of coolant flow made using the code were compared with the experimental data from the LDV flow measurements made using the water tunnel facility. Good agreement was obtained between the measured and calculated values, thereby providing validation of the CFX4 calculations for the model. Based on the CFX4 calculations, it was concluded that the optimum diameter for coolant exit holes for the annular can was 3 mm, as this provided the best balance of central coolant flow and flow through the can-rig channel, within the can rig compartment.

The thermal hydraulics calculations using CFX4 provided the following results for the trial irradiation in the proposed position using a target mass of 0.4 g (compared to an actual mass of 0.371 g), and a thermal neutron flux of 0.91 x 10^{14} $n.cm^{-2}.s^{-1}$ (compared to an expected flux of 0.7 x 10^{14} $n.cm^{-2}.s^{-1}$):

- a maximum temperature in the uranium foil of 138ºC;
- a maximum can wall temperature of 92ºC;
- a maximum surface heat flux at the can wall of 48.5 $W.cm^{-2}$; and
- coolant temperature at the can exit holes of 49ºC.

Two hypothetical accident scenarios were analysed to establish that there was no potential for damage to the LEUFR can and target or the reactor. The first was a loss of control arm accident -- a bounding –case, design basis accident. The second was the inadvertent withdrawal of the rig, can and target assembly and was assessed at high and low power, with and without Protection Signal System response. The analyses demonstrated that:

1. in the loss-of-control arm accident, the estimated energy released in the target and can would be ~2 kJ, resulting in a maximum temperature in the LEUFR can of approximately 250ºC; and some transitional boiling, and
2. the scenarios analysed relating to rig withdrawal were either within the capability of the Protection Signal System or resulted in acceptable consequences where the Protection Signal System failed to respond.

The second accident scenario is bounded by analysis in the HIFAR Safety Document, which demonstrates that withdrawal of the irradiation rig of maximum allowable reactivity worth, t the fastest achievable rate, is within the capacity of the Protection Signal System to provide adequate protection of the reactor. The reactivity worth of the trial irradiation rig, can and target assembly is much less than that of the limiting reactivity worth used in this assessment, and is on the lower range of reactivity worths of rigs routinely irradiated in HIFAR. It therefore follows that removal of this rig at the fastest achievable rate, is also within the capability of the Protection Signal System.

Trial Irradiation – LEUFR Prototype

The first irradiation of an LEU foil in the annular target can was conducted in September 2001. A internal safety submission was required for the first irradiation, with subsequent irradiations subject to its successful performance. The rig was positioned in an outer core position of low thermal neutron flux (~ 0.7 x 10^{14} $n.cm^{-2}.s^{-1}$). The first target was equipped with thermocouples for monitoring of can temperatures during reactor start-up, irradiation and unload. Eight thermocouples were placed on the target, four in each of the inner and outer walls. The positions were chosen to provide an indication of axial temperature variation. Can wall temperatures were measured during reactor start-up, throughout the eight-day irradiation and are shown in Figures III (a) and (b). Thermocouples T1 and T5 were positioned above the foil, and the lower temperature is as predicated by the CFX4 calculations; T4 and T7 were positioned midline to the foil at the rear of the irradiation rig, an area where constricted flow is expected – the higher temperature recorded in this position supports that prediction.; the remainder of the thermocouples were distributed midline to the foil in unrestricted flow positions.

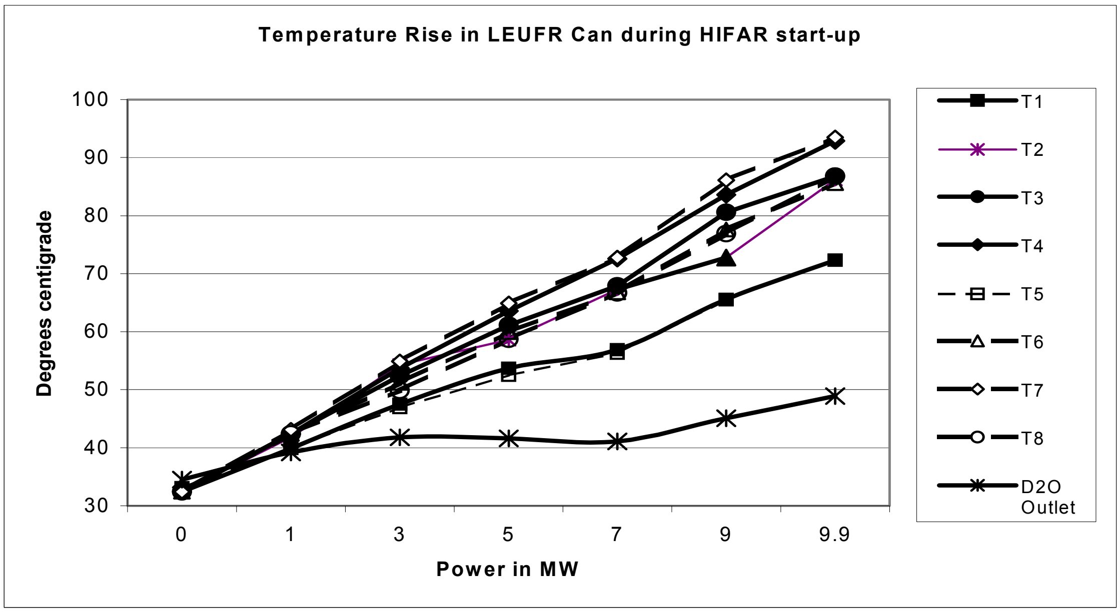


Figure III (a) Wall temperatures in LEUFR target during HIFAR start-up

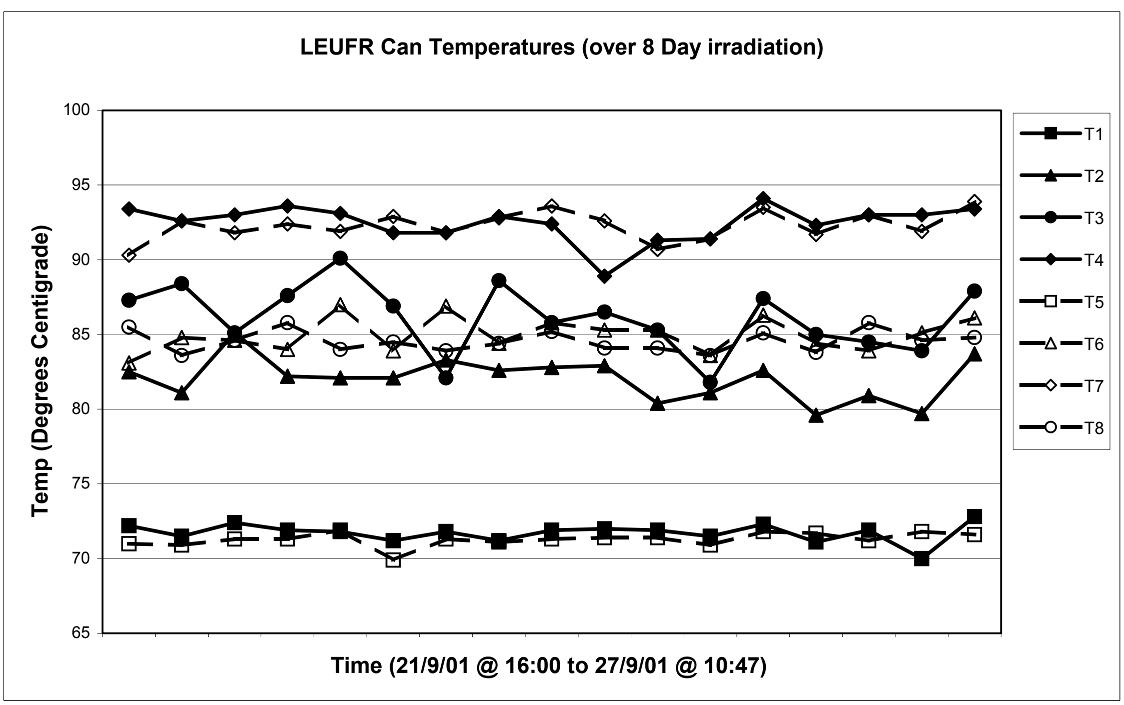


Figure III (b) Wall temperatures in LEUFR target during 8 day irradiation cycle

The HIFAR was shutdown in order to remove the experimental rig. The rig was transported to a hotcell and thermocouples cut and removed in order to obtain the LEUFR can. Subsequently, the can was transported to the Mo-99 production facility.

There was some difficulty during can opening, mainly due to the 3mm wall thickness of the LEUFR design. The foil was removed in predominantly one piece – breakage attributed to physical stress during can cutting (during subsequent irradiations, the irradiated foil was removed in one piece). The Mo-99 production facility has on-line monitoring of fission gases and no additional releases were detected during the can opening procedure. Fission gas release from the irradiated foil was not expected[1], however it must be noted that the relatively high background level of radiation within the cells could have masked any small releases.

The LEU foil was dissolved in 8M nitric acid, using a purpose built dissolver. The solution was then diluted with uranyl nitrate (depleted uranium) and loaded on an alumina column. The separation and purification of Mo-99 was done in the usual way. Three of the four LEU irradiations were processed in the same manner, and resultant Mo-99 purity given in Table X. Tc99m generators were prepared from all of these batches.

Table 2 Mo-99 purity obtained from irradiation of nickel coated LEU (19.81%) foils

	Specification		UEO3	UEO5	UEO6
Radionuclidic Purity (%)	^{99}Mo	>98	99.98	100	100
	^{131}I	<0.0002	ND	ND	ND
	^{132}I/^{132}Te	<0.002	ND	ND	ND
	^{112}Ag/^{112}Pd	<0.01	ND	ND	ND
	^{239}Np	<1.0	ND	ND	ND
	^{103}Ru	<0.05	ND	ND	ND
	^{127}Sb	<0.5	0.0143	ND	ND
	^{95}Nb/^{95}Zr	<0.01	ND	ND	ND
	^{140}La/^{140}Ba	<0.01	ND	ND	ND
	Others	<0.01	<0.01	<0.01	<0.01
Separated Iodines	^{131}I	<0.0002	ND	ND	6.78×10^{-5}
	^{132}I	<0.002	4.0×10^{-7}	1.8×10^{-6}	1.48×10^{-5}
Specific Activity	>180 TBq/g Mo		yes	yes	Yes
Appearance	A clear liquid		Yes	Yes	Yes
Pass/Fail			Pass	Pass	Pass

ND – not detected

Tc-99m Generator Analysis

ANSTO Radiopharmaceuticals and Industrials (ARI) Operations is a Therapeutic Goods Administration (TGA) licensed facility, accredited to ISO 9001/2000. Five generators were made from the Mo-99 obtained from irradiation of nickel coated LEU foils ranging. Generator size was typically 20 GBq for quality control testing purposes. All generators were manufactured and tested according to procedures evaluated and approved by the Drug Safety and Evaluation Branch of the TGA. A typical testing profile of a Tc-99m generator prepared from LEU origin Mo-99 is given below in Table 3

Table 3 Testing Results of ^{99m}Tc generator* from ^{99}Mo obtained from nickel coated LEU foil

Date	Time	^{99m}Tc Act (GBq)	^{99}Mo Act (GBq)	^{99}Mo (%)	^{131}I (%)	^{132}I (%)	^{103}Ru (%)	^{112}Ag (%)	^{239}Np (%)	Pass (P) Or Fail (F)
28/5	0635	14.00	17.00	1.7e-2	ND	3.0e-5	ND	6.0e-5	ND	P
29/5	0633	11.28	13.22	1.1e02	ND	ND	ND	1.0e-5	ND	P
30/5	0650	8.67	10.24	9.1e-3	ND	ND	ND	ND	ND	P
31/5	0638	6.68	7.98	9.6e-4	ND	ND	ND	ND	ND	P
2/6	0630	4.41	4.83	9.4e-3	ND	ND	ND	ND	ND	P
3/6	0645	3.17	3.74	3.7e-3	ND	ND	ND	ND	ND	P
4/6	0627	2.46	2.92	4.8e-3	ND	2.0e-5	ND	ND	ND	P
5/6	0630	1.89	2.27	6.0e-3	ND	ND	ND	ND	ND	P
6/6	0630	1.47	1.76	6.5e-3	ND	ND	ND	ND	ND	P
7/6	0645	1.14	1.37	6.2e-3	ND	ND	ND	ND	ND	P
9/6	0715	0.74	0.82	5.2e-3	ND	ND	ND	ND	ND	P
10/6	0654	0.52	0.64	5.3e-3	ND	ND	ND	ND	ND	P
11/6	0630	0.41	0.50	5.0e-3	ND	ND	3.0e-5	ND	ND	P
12/6	0630	0.31	0.39	4.0e-5	ND	ND	3.0e-5	ND	ND	P

* Radionuclidic purity – ^{99}Mo<0.1%; ^{131}I<0.0005%, ^{103}Ru<0.005%, Others <0.01%

As an additional test, gamma camera images of a rabbit were obtained from a standard bone imaging agent (MDP – mercapto diphosphonate). One batch was produced from LEU foil origin Tc-99m and the other from our standard Tc-99m product. The images were taken two hours after injection, and the same rabbit was used for both images (three day period between injections). There was no significant difference in purity of the prepared agents nor was there significant difference in the function of the agent, as evidenced by gamma camera images.

In conclusion, nickel coated LEU (19.81%) uranium metal foils produce Mo-99 equivalent in purity and function to that obtained from 2.2% uranium dioxide pellets. The use of LEU foils for production of Mo-99 using existing separation methodology is feasible at ANSTO.

Future Work

Our current program of work is focussed on demonstrating Mo-99 production from LEU foils on a routine basis leading to greater production capacity. This has required review of the current production methodology, as outlined in Figure IV, and then subsequent assessment of what changes will be needed as a consequence of changing targets from 2.2% uranium dioxide pellets to ~ 19.81% uranium metal foils.

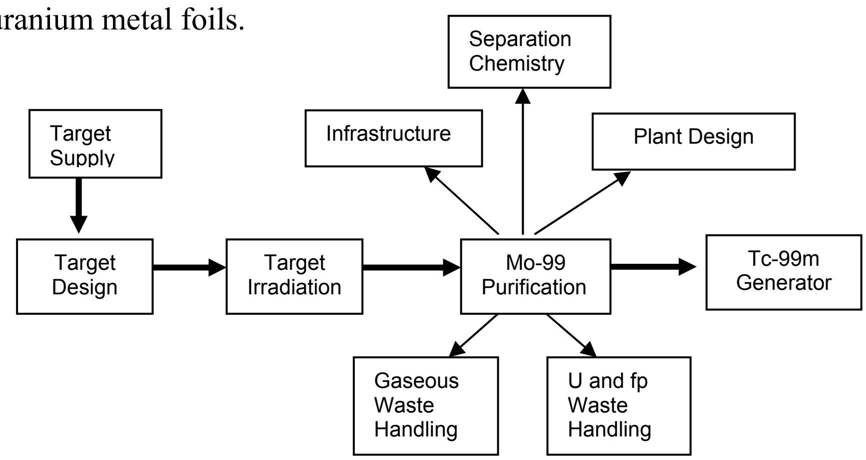


Figure IV Schematic of Mo-99 Production Methodology

Most work to date has been focussed on target and rig design. The prototype rig under evaluation has improved thermal hydraulic characteristics compared with ANSTO's existing irradiation rig. Experimental thermal hydraulic studies of the new rig, liner and target are in progress.

The other components of the production methodology such as liquid waste handling, fission gas trapping, target dissolution and plant design vary in development from conceptual design to prototype testing. The testing of these components for processing of irradiated LEU foils will constitute a change to our existing facility licenses, and as such will be subject to regulatory review by ARPANSA (Australain Radiation Protection and Nuclear Safety Agency). Consequently, large scale processing of irradiation LEU foils is scheduled for 2004, prior to RRR start-up.

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[1] F.J. Stubbs, G.N. Walton "*Emission of Active Rare Gases from Fissile Material During Irradiation with Slow Neutrons*". Proceedings of the International Conference on the Peaceful Uses of Atomic Energy Vol 7, p 163-168, 1995