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 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% then finally 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, right from target design through to chemical processing and waste management.

The major focus at this point in time is the development of a LEU target, initially suitable for irradiation in HIFAR and, with subsequent modification, suitable for irradiation in the RRR. We have entered into collaboration with Argonne National Laboratory (RERTR) to develop such a target using uranium metal foil with U-235 enrichment of less than 20%. The first trial irradiation of this target is planned in December 2000 and it will be processed using the current Mo-99 production process at ANSTO. The review of this trial will determine the program for further irradiations.

Mo-99 Production at HIFAR - current

The target containment for Mo-99 production at ANSTO is familiarly referred to as a "rocket" can (Figure 1). Each can contains thirteen UO_2 pellets (2.2% U-235) with MgO dispersed between pellets and the outer walls of the target to aid in heat transfer. Four of these targets are irradiated in a rig assembly that fits in the centre of HIFAR hollow fuel elements. Target cooling is achieved by diverting ~1% of the element cooling flow through the irradiation rig.

Figure 1 Irradiation (or "Rocket") Cans for UO_2 Pellets

Targets are irradiated an average of seven days and a maximum of six rocket cans can be processed in one batch. The chemical purification method for the Mo-99 is very simple, based on the original alumina column separation of fission Mo-99 from other by-products (Figure 2).

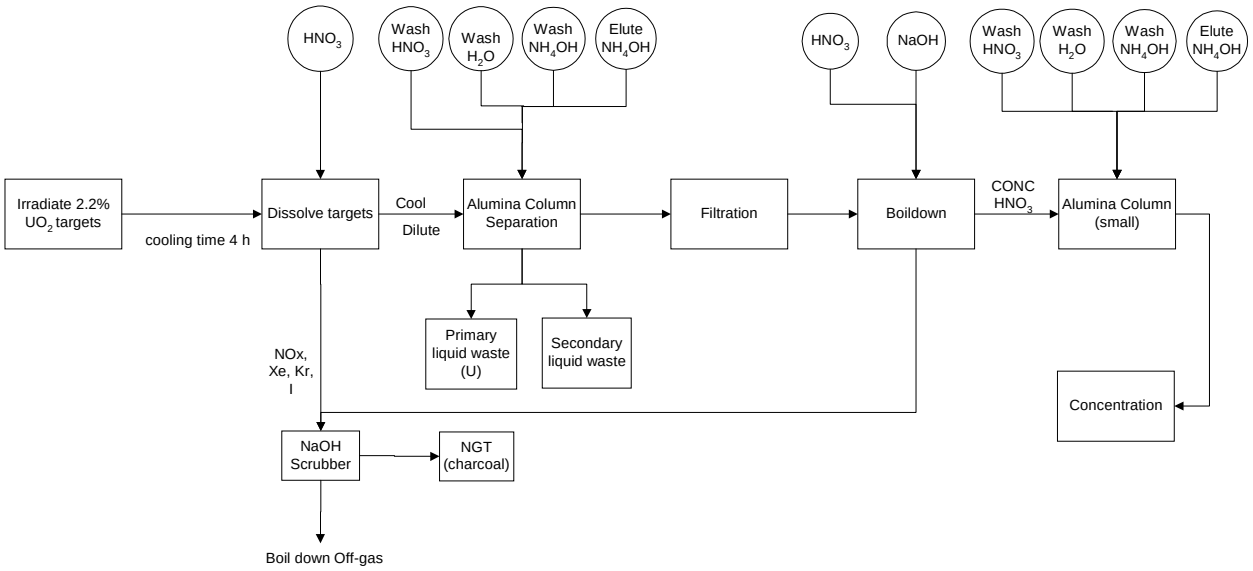


Figure 2 Simplified Flowchart of ANSTO Mo-99 Process

ANSTO would like to increase the efficiency of Mo-99 production in order to meet future demands for Tc-99m for the Asia-Pacific region. The approval for a replacement reactor for HIFAR has created an ideal opportunity to review the Mo-99 process.

Target Development and Trial Irradiation/s

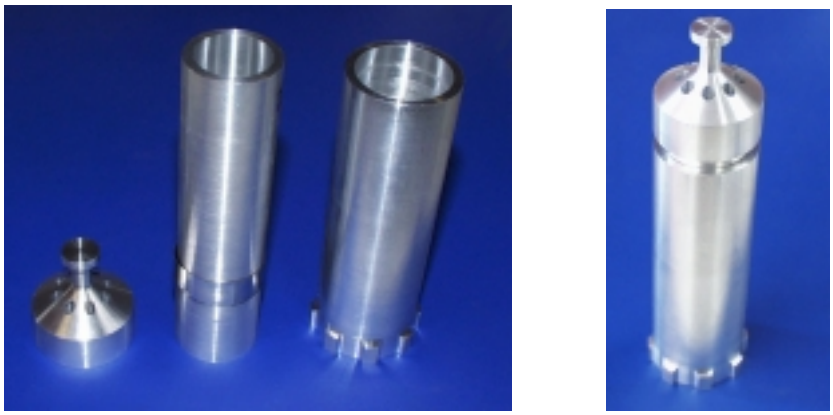
Australia is a signatory to the nuclear non-proliferation treaty and has a national commitment to the use of LEU targets.

When increased target enrichment was first discussed as an option for increasing Mo-99 production, the possibility of using higher enriched (greater than 2.2% U-235) UO_2 pellets was explored. It was found that the centre-line temperature of these pellets, under irradiation conditions would be extremely high, and increases beyond the existing 2.2% could not be approved under HIFAR's Operating Limits and Conditions. Thus, a target with higher U-235 enrichment would require a different chemical form.

Around this time, ANSTO became aware of the work being done on LEU uranium metal foils by Argonne National Laboratory. For ANSTO, increasing target enrichment from 2.2% up to ~19.8% U-235 is a very attractive option. Use of higher enriched targets offers the potential to reduce liquid waste volumes per curie of Mo-99.

In collaboration with ANL, ANSTO is investigating LEU (~19.8% U-235) metal foils as potential targets for future Mo-99 production at ANSTO. Initially, the target must be designed to

fit in the existing HIFAR irradiation rigs. Therefore, the external dimensions of the target and anchoring mechanism are predetermined. An annular target design has been developed, in which the uranium metal foil is sandwiched between the outer and inner aluminium sleeves of the target can (Figure 3 (a) and (b)). The optimum wall thickness for effective heat transfer was generated from heat transfer calculations.



(a) (b)

Figure 3 (a) Annular target can components (b) Assembled annular target

Two options for holes in the cap of the target are under consideration (Figure 4). Results of computational flow dynamic studies and water tunnel experiments will determine which design is best.

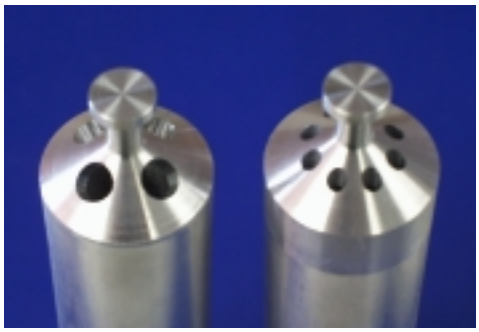


Figure 4 Cap variations for annular target

Thermocouples will be attached to inner and outer surfaces of the target can during irradiation. Temperature readings during irradiation will be recorded and compared with theoretical temperature calculations. A special rig has been built to accommodate this.

Provided safety approvals are in place, the first irradiation of LEU foils in the annular target can is planned for December 2000. A small foil with approximate U-235 content of 0.5 g will be irradiated. After irradiation the foil target will be processed using the method outlined in Figure 2. Real-time monitoring of fission gas emissions will be used to detect if fission gases are released during target opening. A Tc-99m generator will be prepared from the resultant Mo-99 and checked in the usual way by quality control. The results of quality control monitoring will be compared to generators arising from current Mo-99 production methods.

Future Work

Work is proceeding to determine the maximum U-235 content per target and per rig (or irradiation position) that can be irradiated under HIFAR Operating limits and conditions. In the short term, this limit will determine the potential throughput of Mo-99 in HIFAR. Prior to production transferring to the RRR, modified target designs will be developed to maximise production efficiency and to take full advantage of the improved target cooling capability of the RRR. Effort will also focus on optimisation of rig design in collaboration with INVAP for the RRR.

Optimisation in chemical processing of Mo-99 from LEU foils is under investigation. This includes investigation of more effective means for the capture/delay of fission gases and handling of fission wastes arising from LEU (19.8% U-235) targets.

Regulatory approval from the Australian Radiation Protection and Nuclear Safety Authority (ARPANSA) is required before any changes or modifications to Mo-99 production operations are made. ARPANSA has been informed of the proposed investigations so that regulatory concerns can be identified and addressed before final approval is sought. Approval from the Therapeutic Goods Administration must also be obtained before Mo-99 produced by a new method (target and/or process), can be used on Tc-99m generators. It is proposed that a period of simultaneous Mo-99 production in both HIFAR and the RRR will simplify the TGA approval process.