ESTABLISH TECHNIQUES FOR SMALL SCALE INDIGENOUS MOLYBDENUM-99 PRODUCTION USING LEU FISSION OR NEUTRON ACTIVATION

Mushtaq Ahmad

Isotope Production Division, Pakistan Institute of Nuclear Science & Technology, P.O. Nilore, Islamabad, Pakistan e-mail: amushtaq1@hotmail.com

Abstract

Low enriched uranium foil (19.99% ²³⁵U) may be used as target material for the production of fission molybdenum-99 (⁹⁹Mo) in the Pakistan Research Reactor-1 (PARR-1). LEU foil annular targets or LEU foil plate targets can be irradiated in PARR-1 for the production of >100 Ci of ⁹⁹Mo at the end of irradiation. Neutronic and thermal hydraulic analysis for the fission ⁹⁹Mo production at PARR-1 has been performed. Power levels in the foil targets and their corresponding irradiation time durations were initially determined by neutronic analysis to have the required neutron fluence. Finally the thermal hydraulic analysis has been carried out for the proposed designs of the target holders using LEU foil for fission ⁹⁹Mo production at PARR-1. Data shows that LEU foil targets can be safely irradiated in PARR-1 for production of desired amount of fission ⁹⁹Mo. Although Pakistan is producing ⁹⁹Mo by the irradiation of HEU uranium alloy plates with aluminium clad targets in PARR-1 to meet the demands of ⁹⁹Mo/^{99m}Tc generators in the country, it intends to use LEU aluminide targets in coming years. In this perspective, R&D work on fabrication of LEU target for ⁹⁹Mo using natural uranium has been initiated at PINSTECH. Preliminary results of LEU target plate manufactured indigenously at PINSTECH show quite good separation of ⁹⁹Mo from target matrix activity.

1. INTRODUCTION

The prominent position of technetium-99m (99m Tc) in nuclear medicine practice has been due to its near ideal nuclear properties (140 keV gamma for imaging and no β^- particle emission), the ready availability in the form of convenient $^{99}Mo \rightarrow ^{99m}$ Tc generator systems and the rapid progress made in recent years in the development of 99m Tc radiopharmaceuticals for application in oncology, cardiology and other fields [1-7]. 99m Tc is the short-lived (T_{1/2} = 6h) daughter product of the parent molybdenum-99 (^{99}Mo , T_{1/2} = 66 h), which is mainly produced by the nuclear fission of uranium-235 (235 U).

Now-a-days Pakistan is consuming more than 35^{99m}Tc generators loaded with >16 Ci (reference date) of fission ⁹⁹Mo in nuclear medical centres. Under a technical cooperation program, the International Atomic Energy Agency (IAEA), Vienna has provided ⁹⁹Mo loading facility to PINSTECH, so that ^{99m}Tc generators conforming to the international standards can be produced locally. For the last eight years Pakgen ⁹⁹Mo/^{99m}Tc generators are prepared weekly and dispatched to 35 nuclear medical centres in the country. However, ⁹⁹Mo is still being imported from South Africa on a fortnightly basis. To overcome the problems associated with import of ⁹⁹Mo or ^{99m}Tc generators such as hard currency, increasing price of ⁹⁹Mo, import policies, delay and changes in supply schedules, etc., the indigenous production of fission ⁹⁹Mo in the country was planned. Currently fission ⁹⁹Mo is produced in ⁹⁹Mo

is used for manufacturing of ^{99m}Tc generators at PINSTECH. In January 2012, weekly production of ⁹⁹Mo will be started to fulfil all the demands of the country, whereas extra ⁹⁹Mo will be exported to neighbouring countries.

To reduce nuclear-proliferation concerns, the Reduced Enrichment for Research and Test Reactors (RERTR) program is working to limit the use of highly enriched uranium (HEU) by substituting low enriched uranium (LEU) fuel and targets. Low enriched uranium contains <20% ²³⁵U. A denser form of uranium is required in order to keep the target geometry the same when changing from HEU to LEU. Under a Coordinated Research Project sponsored by International Atomic Energy Agency (IAEA), Vienna, investigations were performed to adopt the technology developed by ANL (USA) for small scale production of ⁹⁹Mo using modified CINTICHEM process. For the preparation of safety documents, neutronic, thermal and hydraulic analysis for production of ⁹⁹Mo using LEU annular foil target and newly designed target holder in PARR-1 was performed. Neutronic and thermal hydraulic analysis for the fission ⁹⁹M production at PARR-1 using LEU foil plate target proposed by University of Missouri Research Reactor (MURR) was also performed. Power levels in foil target and their corresponding irradiation time durations were initially determined by neutronic analysis to have the required neutron fluence. Finally the thermal hydraulic analysis has been carried out for the proposed design of the target holder using LEU foil targets for fission ⁹⁹Mo production at PARR-1. Data shows that LEU foil targets can be safely irradiated in PARR-1 for production of desired amount of fission ⁹⁹Mo.

Argentina has successfully converted the HEU uranium alloy target into LEU uranium aluminide targets for the production of fission ⁹⁹Mo commercially. The same target technology is also being used by ANSTO Australia. Although Pakistan is producing ⁹⁹Mo by the irradiation of HEU uranium alloy plate with aluminium clad targets in PARR-1 to meet the demands of ⁹⁹Mo/^{99m}Tc generators in the country, it intends to use LEU aluminide targets in coming years. In this perspective, R&D work on fabrication of LEU targets for ⁹⁹Mo using natural uranium has been initiated at PINSTECH. Preliminary results of LEU target plates manufactured indigenously at PINSTECH show quite good separation of ⁹⁹Mo from target matrix activity.

2. NEUTRONIC AND THERMAL HYDRAULIC ANALYSIS

2.1 PAKISTAN RESEARCH REACTOR-1 (PARR-1)

Pakistan Institute of Nuclear Science and Technology (PINSTECH), Islamabad is operating PARR-1 to provide services to the users for the production of radioisotopes and for neutron irradiation. Since initial criticality, PARR-1 has rendered invaluable service in the training of manpower, production of radioisotopes and as a source of neutrons for basic and applied research. To reduce nuclear proliferation concerns, it became essential that its core be converted for operation with low enriched uranium (< 20% ²³⁵U) fuel. The PARR-1 is a swimming pool type research reactor originally designed for a thermal power of 5 MW. Its core has been redesigned to operate with LEU fuel at a power level of 9 MW in 1992 and 10 MW in 1998.

2.1.1 Reactor core assembly

The PARR-1 core consists of an assembly of standard and control fuel elements mounted on the grid plate. The fuel elements can be assembled in different core configurations. The core is immersed in demineralized water which acts as coolant, moderator and reflector. However, using specially designed reflector elements the light water can be replaced on one or more sides with other reflectors such as graphite, beryllium or heavy water.

2.1.2 Grid plate

The PARR-1 grid plate is made of 127 mm thick aluminium. It has 54 holes in 9 x 6 patterns with a lattice spacing of 81 x 77.11 mm. These holes accommodate the end fittings of the fuel elements. During operation the unused holes are closed using plugs so that maximum coolant passes through the fuel elements. In between the fuel element bearing holes there are 40 smaller holes (17.5 mm diameter) for coolant flow which passes through the side and the outer fuel plates. The PARR-1 core consists of LEU fuel having 19.99% ²³⁵U. The fuel material used is uranium silicide (U₃Si₂.Al).

2.1.3 Core configuration No. 98

Core configuration #98 is shown in Fig. 1. This core consists of 29 standard and 5 control fuel elements. There are two irradiation positions inside the core at the position C7 and C4. The core is reflected by graphite on two sides and water on other sides.



FIG. 1. Core configuration No. 98 (PARR-1).

3. LOW ENRICHED URANIUM (19.99% ²³⁵U) ANNULAR TARGET

The goal in the LEU target design was to produce 100 Ci of ⁹⁹Mo at the end of irradiation in PARR-1 to meet the demands of ⁹⁹Mo/^{99m}Tc generators in Pakistan. The LEU

foil target selected has the following characteristics. Uranium foil (19.99% 235 U) of 125 µm thickness is enveloped in 15 µm thick nickel foil and placed between two aluminium tubes that are welded from both ends. The dimensions of the annular target are given below.

- \blacktriangleright Outer Al tube external dia = 30.00 mm; Outer Al tube internal dia = 28.22 mm
- > Ni foil = 15 μ m; U foil = 125 μ m; Ni foil = 15 μ m
- > Inner Al tube external dia = 27.99 mm; Inner Al tube internal dia = 26.21 mm
- \blacktriangleright Al tube length = 162 mm
- U foil dimension = $76\pm 2 \times 88\pm 2 \text{ mm} = 16 \text{ g uranium}$, ²³⁵U contents = 3.19 g

3.1. ANNULAR TARGET HOLDER

The annular target holder is made of reactor grade aluminium metal. It has two tubes, one outer and the other inner which is larger. The extended part of the inner tube guides the insertion of annular target. The upper and lower plates have same dimension. Holes and groves have been provided in upper plate for fixing the stainless steel flexible wire. For steady flow of coolant (water) fins have been fixed on inner tube to separate the annular target from inner tube.

3.2. NEUTRONIC ANALYSIS

Neutronic analysis of the target holder for ⁹⁹Mo production was performed by computer codes WIMSD/4 along with BORGES and CITATION. In this study the target holder was placed at the central water box facility and was irradiated at five different axial positions. To generate the microscopic cross-sections for different regions of the core, the MTR-PC26 package was used. This package is a collection of standard computer codes and libraries for conducting reactor core static, depletion, transient, thermal hydraulic and shielding studies for MTR type reactors using personal computers. In current research, only the neutronic option for cross-section generation has been used from this package. For this purpose, computer codes WIMSD/4 along with BORGES were used from MTR-PC26 package [8-10]. Employing these codes, ten group microscopic cross-sections and number density calculations were performed. These calculations were performed for fuel, structure, control absorber, control follower, end plugs, water reflector, and graphite reflector regions. Also cross-sections were generated for the annular target holder. Representative unit cell models for the fuel meat of PARR-1 and for annular target holder are shown in Fig. 2 & Fig. 3. The incremental radii for each region are shown with their material specified in parenthesis. Burn up option of WIMSD/4 was used for fuel region to generate microscopic cross-sections for fission products. Computer code BORGES is used to read the output of WIMS/D4, as per instructions of the user, and then writes it in a form that is readily usable in the multidimensional, diffusion theory code CITATION. Hence these cross-sections and number densities at the output of BORGES were employed in CITATION. The equilibrium core No. 98 (Fig. 1) was modelled employing 3-D option of CITATION [11]. The target holder contains the annular target with 16 g of total uranium whereas contents of ²³⁵U are 3.19 g. Analysis of the target holder was performed by irradiating it in the central water box at C-7 location in core configuration #98 at the beginning of equilibrium cycle as shown in Fig. 1. The effect of irradiation was studied by placing target holder at five different axial positions. Excess reactivity in the core for these cases was calculated and is shown in Fig. 4. The target reactivities are summarized in Table 1. It is evident from the figure that maximum excess reactivity of 120 pcm is present in the core when the target holder is placed in the fourth plane from the top of the core. This is due to the neutron flux variation inside the core. It has been observed that location of maximum power density in the core remains unchanged by placing the target holder in any of five axial planes. However the value of maximum power density varies slightly by moving target holder in different axial planes. The variation in peaking factor calculated is from 2.928 to 2.961. This maximum peaking value is less than the safe value calculated for the PARR-1 core configuration [12]. Based on these calculations, the target holder can be irradiated in the central flux trap of the core. During reactor operation the target holder should not be moved because it can add reactivity in the system of the order of 120 pcm. No significant effect would be on peaking factor.



FIG. 2. Unit cell model employed in WIMSD/4 for annular target holder.



FIG. 3. Unit cell model employed in WIMSD/4 for fuel meat of PARR-1.



FIG. 4. Distance in cm of target holder from top of the core.

3.3. THERMAL HYDRAULIC ANALYSIS

Thermal hydraulic analysis has been carried out for the proposed design of the target holder for fission ⁹⁹Mo production at PARR-1. Parameter of interest is the maximum temperature gained when the reactor is in operation at full power (10 MW). It is intended to mount the target holder inside the water box in the core. Provision would be made that it could be inserted at any desired vertical position (with middle position for maximum neutron

targeting). Calculations were performed to assess its impact on the overall core coolant flow rate as well as to estimate its plate surface temperature rise.

Hydraulic calculations were performed to estimate the coolant flow rate through the target. Computer code DP was employed for this purpose. In the code, pressure drops, velocity distribution and flow rates through different channels of the core, effective and bypass flow are calculated. The code computes velocities through an iterative procedure after converging to the same pressure drop across the core for each channel.

The hole size at water-box exit was varied to check its impact on coolant velocity distribution in the core. Various sizes were considered in calculations. In order to minimize its effect on *standard fuel channel velocity*, hole size was kept the same as that of regular water-box (i.e. 1 cm diameter). Calculation of pressure drops along the holder in the water box revealed that a hole of such size would allow a coolant flow rate that would be less than 0.5% of total effective flow rate through the core [13]. Water velocity has been calculated along the active channel of holder to be 1.63 m/s.

Neutronic calculations predicted various power levels at different vertical positions as shown in Table 1. For the analysis, maximum power level (17.58 kW) has been considered. Axial distribution of power has been assumed to have cosine shape. To account for the uncertainties, an engineering hot channel factor (1.584) was incorporated using the conservative multiplicative method [14]. This factor is the product of three components: (i) a factor 1.2 for the coolant temperature rise due to manufacturing tolerances in the coolant channel spacing. (ii) a factor of 1.2 for the film temperature rise due to uncertainties in the heat transfer coefficient and inhomogeneities in 235 U distribution etc. and (iii) a factor of 1.1 for uncertainties in the calculated power distribution.

The target holder was modelled for one-dimensional hydrodynamics and onedimensional heat transfer. Local clad surface temperature was computed with the help of heat transfer coefficient (calculated by using Dittus-Boelter correlation) and the local coolant temperature variation as per axial power distribution. As the holder has good flow area (704 mm²) along active plate with appreciable coolant velocity, calculated maximum temperature rise is nominal ($T_{max, surface} = 44.1$ °C) with T_{inlet} as 38°C. Heat transfer coefficient along the plate was calculated to be 6910 W/m²-°K. Temperature distribution in axial direction is shown in Fig. 5.

At PARR-1, safety analysis performed earlier [14] considered various probable reactivity induced accidents such as a maximum start-up accident, drop of a fuel element on the core, flooding of beam-tube, removal of in-pile experiment, and movement of core towards thermal column. Amongst these accidents (with overpower trip setting at 115% of steady-state power level), a maximum of power level (\cong 40 MW) could be attained in case of accident due to drop of fuel element on the core. Calculations show that during such worst possible reactivity-induced accident at PARR-1 (resulting in heat flux increase by four times), plate temperature would only increase to a maximum of 62.5°C, which is far less than the saturation temperature (\cong 113°C) at the core pressure level. It is concluded that the proposed annular designs could be safely adopted for ⁹⁹Mo production project at PARR-1, without compromising reactor safety.

Annular T	arget Holder			
Plane	²³⁵ U(g)	Power (kW)	Net reactivity (pcm)	Irradiation Time (hrs)
1	3.19	5.42	7	37.77
2	3.19	10.05	34	20.36
3	3.19	14.37	73	14.25
4	3.19	17.41	120	11.76
5	3.19	14.62	81	14.00



FIG. 5. Temperature distribution along the plate surface.

4. LOW ENRICHED URANIUM FOIL PLATE TARGET

The goal in the LEU target design was to produce 100 Ci of ⁹⁹Mo at the end of irradiation in PARR-1 to meet the demands of ⁹⁹Mo/^{99m}Tc generators in Pakistan. The LEU foil plate target selected has following characteristics.

Uranium foil (19.99% ²³⁵U) of 125µm thickness is enveloped in 15µm thick nickel foil and placed between two aluminium plates that are welded from all sides. The geometry of foil plate target is shown in Fig. 6, while dimensions are given below.

- Aluminium plate (upper) = L 160 mm x W 60 mm x T 1 mm
- Aluminium plate (lower) = L 160 mm x W 60 mm x T 1 mm (groove to accommodate LEU foil enveloped in Ni foil; L 126 mm x W 41 mm x D 0.155 mm)
- ▶ LEU uranium foil = L125mm x W 40mm x T 0.125 mm
- ▶ Ni foil = $15\mu m$
- > Total weight of aluminium in LEU foil plate target = 51.84 g
- Weight of uranium in LEU foil plate target = $11.785 \text{ g} (2.357 \text{ g}^{235}\text{U})$
- Weight of nickel in LEU foil plate target = 1.336 g



FIG. 6. Geometry of foil plate target.

4.1. LEU FOIL PLATE TARGET HOLDER (IRRADIATION RIG)

The foil plate target holder is made of reactor grade aluminium metal. It is a rectangular assembly, capable of holding three foil plates vertically parallel to each other. Three aluminium plates along with two side plates provide the water channels for cooling of the target plates. Target foil plates will be loaded in the slots of the target holder with the help of tongs while placing the target holder under water in a tray in open pool. At the top of the holder two collars have been provided to attach it with a stainless steel (SS) string of about 10 m which will be used for locking and hanging the target holder for loading in the water box. After loading the required foil plate target, the holder will be loaded in the water box for irradiation purposes. Details and dimensions of foil plate target holder are depicted in Fig. 7 and Fig. 8.

Power levels in target fuel plates and their corresponding irradiation time durations were initially determined by neutronic analysis (Table 2) to have the required neutron fluence. It is intended to mount the target holder inside the water box in the core at grid position C-7. The inlet temperature of the coolant has been taken as 38°C. Hydraulic calculations were performed to estimate the coolant flow rate through the target fuel channels by code DP, which determines pressure drops, velocity distribution and flow rates through different channels of the core, effective and bypass flows. The code computes velocities through an iterative procedure after converging to the same pressure drop across the core for each channel. The coolant velocity in the water box with 5 cm diameter bottom hole would be 4.06 m/s.



FIG. 7. LEU foil plate target holder.



FIG. 8. Top view of LEU foil plate target holder.

TABLE 2. POWER IN TARGET FUEL PLATE (AT 10 MW STEADY-STATE REACTOR
POWER) OBTAINED FROM NEUTRONIC RESULTS

No. of	²³⁵ U	Power	Net Reactivity	Irradiation time
Foils	(g)	(kW)	(pcm)	(hr)
1	2.357	4.598	40.23	54.48
2	4.714	8.855	70.28	24.36
3	7.070	13.194	100.11	15.67

The target foils are just 125 mm in axial direction (much smaller than standard fuel plates) and will be used in a target holder that will be placed at top plane position in the water box. The axial power peaking in the plates has been computed to be 1.22. To account for the uncertainties, engineering factors were introduced: a factor 20% for the coolant temperature rise due to manufacturing tolerances in the coolant channel spacing; (ii) a factor of 20% for the film temperature rise due to uncertainties in the heat transfer coefficient and inhomogeneities in 235 U distribution etc.

Thermal hydraulics for the three cases (1, 2 or 3 target plates) was carried out using conservative correlations. Critical heat flux for the target fuel channel has been calculated to be 295 W/cm² and results show that use of one, two or three foils will have good safety margins (6.3) against Departure from Nucleate Boiling, DNB. For critical heat flux evaluation a conservative correlation by Mirshak et al. [15], has been employed. Table 3 shows the maximum clad surface temperature achieved during irradiation. The axial temperature profile in the target foil is shown in Fig. 9.

Before irradiating the LEU foil plate target, neutronic and thermal hydraulic calculations were performed for the production of 100 Ci (3700 GBq) ⁹⁹Mo in PARR-1. The calculated reactor irradiation time is 15.67 hours with three LEU foil plate targets and the maximum excess reactivity of 100.11 pcm may be generated with these target plates. The reactivity induced by these foil target is quite insignificant compared to excess reactivity of PARR-1 core which is about 5000 pcm. Critical heat flux for the target fuel channel has been calculated to be 295 W/cm² and results show that use of one, two or three foils will have good safety margin (6.3) against Departure from Nucleate Boiling, DNB. It is therefore concluded that the proposed plate target and its holder design could be safely adopted for a ⁹⁹Mo production project at PARR-1, without compromising reactor safety.

No. of	Maximum clad
foils	surface
	Temperature
	(°C)
1	80.6
2	79.0
3	78.8

TABLE 3. MAXIMUM CLAD SURFACE TEMPERATURE OF FOIL PLATE TARGET DURING IRRADIATION



FIG. 9. Distribution of clad surface temperature along the fuel foils.

5. PRODUCTION OF FISSION ⁹⁹Mo USING HEU TARGET

In all probability, the same criteria applied to evaluate the safety of the reactor fuel shall be used to evaluate the safety of the targets used for the production of fission ⁹⁹Mo. Highly enriched uranium (> 90% U-235) fuel was used from 1965 to 1989 for operation of PARR-1 and operators were confident in handling of HEU fuel. Hence characteristics of mini plate were selected as target material for generation of fission ⁹⁹Mo in PARR-1. The main features of the HEU target plate are given in Table 4.

Qualification criteria of the HEU target plate was set by fuel experts of the Pakistan Atomic Energy Commission Pakistan. Neutronic and thermal hydraulics calculations were performed to determine the maximum power of the targets, their uranium content, and the uniformity requirements for their loading [16-18]. Various techniques were employed for the testing of HEU target plates. Radiography technique is capable of characterizing both meat location and density. Specifically, target plates that meet fuel density specifications can be irradiated with little risk of power peaking and hot spots. An adequate characterization and qualification of target plate cladding is also critical, because cladding breaches will contaminate the reactor coolant. Bend testing is a dependable way of testing bond strength while Ultrasonic Testing (UT) examinations qualify both bonding homogeneity and minimum thickness of that cladding. The bonding quality is inspected by means of a blister test. Lastly, Optical microscopy is applied for clad thickness, which further supports the veracity of the UT characterization method.

Acidic and basic dissolution methods of uranium targets are described in the literature. In acid dissolution all the fission gases are released, while in basic dissolution valuable radioiodine remains in solution. Hence isolation of radioiodine from noble gases are easier and precipitation of uranium along with actinides with some fission fragments makes recovery of unburnt target material in the first step of ⁹⁹Mo separation. The disposal of

uranium is simple in basic dissolution technique. A method for extraction of iodine-131 (¹³¹I) from fission products has also been developed.

The irradiated HEU target plates are dissolved in a mixture of NaOH and NaNO₃. During dissolution uranium and other actinides are precipitated out and xenon is liberated which is trapped by freezing (liquid nitrogen). The solution contain radio-iodine, ⁹⁹Mo and some fission products like Ru and Rh. Ammonia gas produced during dissolution is distilled and trapped in H₂SO₄ solution whereas hydrogen is converted to water by action on CuO. Silver coated alumina is used for the removal of radio-iodine from the solution. Nitrates interfere with the adsorption of ⁹⁹Mo on alumina are changed to nitrogen by action of urea. Column chromatography technique using alumina and anion exchange resins are employed for the purification of ⁹⁹Mo from remaining contaminants. Final purification is performed by sublimation of ⁹⁹Mo, which is dissolved in NaOH.

TABLE 4. SPECIFICATIONS OF HEU PLATE TARGET

1	Target Plate Dimensions	Length = $160 \pm 2 \text{ mm}$
		Width = $60 \pm 1 \text{ mm}$
		Thickness = 1.3 ± 0.2 mm
2	Fuel core Location	Length = 125 ± 5 mm
		Width = $40 \pm 2 \text{ mm}$
		Thickness = 0.5 ± 0.05 mm
3	Surface Finish	Free of scratches, gouges and pits
		Max depth of defect at Meat Area = 0.0762 mm
		Max depth of defect at the rest of Area = 0.127 mm
4	Surface contamination	No loose contamination of U
4	Surface contamination	No loose contamination of U
4 5	Surface contamination % of U Contents	No loose contamination of U 20 ± 1
4	Surface contamination % of U Contents	No loose contamination of U 20 ± 1
4 5 6	Surface contamination % of U Contents % of ²³⁵ U content	No loose contamination of U 20 ± 1 93 ± 1
4 5 6 7	Surface contamination % of U Contents % of ²³⁵ U content	No loose contamination of U 20 ± 1 93 ± 1
4 5 6 7	Surface contamination % of U Contents % of ²³⁵ U content Blister	No loose contamination of U 20 ± 1 93 ±1 No blister is acceptable on core area Man size of blister on the rest of area - 2mm
4 5 6 7	Surface contamination % of U Contents % of ²³⁵ U content Blister	No loose contamination of U 20 ± 1 93 ±1 No blister is acceptable on core area Max size of blister on the rest of area = 3mm
4 5 6 7	Surface contamination % of U Contents % of ²³⁵ U content Blister Metallurgical bond	No loose contamination of U 20 ± 1 93 ±1 No blister is acceptable on core area Max size of blister on the rest of area = 3mm Interatomic diffusion between meat and cladding

6. SIDE PRODUCTS RECOVERY

Besides extraction of ⁹⁹Mo from large number of fission products and actinides, separation of some useful radioisotopes, like ¹³¹I, ¹³³Xe, and ¹³⁷Cs etc., will also be carried out for their application in medical field.

7. DEVELOPMENT OF LEU (UAl₂-Al) TARGET PLATES

R&D work on fabrication of LEU target plates is being carried out at PINSTECH. Uranium aluminide, predominated with UAl₂ phase, was prepared by arc-melting procedures and comminuted to required particle size. UAl₂ and Al powders were blended and compacted to required density. The picture-frame technique was used to clad the dispersions (UAl₂-Al) with aluminium. Table 5 presents the characteristics of target plates, while Fig. 10 depicts the geometry of target plate. A few target plates were fabricated by thermo-mechanical processing (hot rolling & annealing) of UAl₂-Al matrix contained in roll billet of Al. The fabricated plates were characterized by destructive and non-destructive testing techniques and then annealed to achieve required phase of uranium aluminide for proper dissolution in basic media. Preliminary experiments for dissolution of plates in basic media were successfully carried out. Small strips of target plates were sealed in aluminium capsule for irradiation in PARR-1 for subsequent separation of ⁹⁹Mo. The irradiated targets were dissolved in a mixture of NaOH and NaNO₃. More than 97% ⁹⁹Mo was recovered in filtrate.

TABLE.5. GENERAL SPECIFICATIONS OF LEU TARGET PLATE

Uranium Enrichment Wt. of ²³⁵ U in the Plate Wt. of LEU Metal in the Plate Surface Density of ²³⁵ U Homogeneity of ²³⁵ U in uniform Zone	$\begin{array}{c} 19.75 \pm 0.2 \ \% \\ 1.50 \pm 0.03 \ g \\ 7.595 \ g \\ 30 \ \mathrm{mg/\ cm^2} \\ \pm \ 20 \ \% \end{array}$
Homogeneity of 235 U in Dog bone	- 20% to + 30%
Zone Homogeneity of ²³⁵ U in Third	- 100% to + 30%
Materials of Meat	UAl ₂ Dispersed in
Material of Cladding Max. Weight of Aluminium in the Plate	the Matrix of Aluminium Aluminium 33.00 g



FIG. 10. Geometry of LEU target plate (All dimensions are in mm)

8. CONCLUSION

A facility for the separation of ⁹⁹Mo from fission products and actinides is operational. Due to non-availability of LEU foil, Pakistan could not adopt the modified Cintichem process; however Pakistan is willing to establish a ⁹⁹Mo processing facility using LEU targets. R&D work on fabrication of LEU target plates is being carried out at PINSTECH. Uranium aluminide, target plates are being developed. Preliminary work on dissolution of irradiated targets show good separation yield of ⁹⁹Mo from target matrix. Construction of hot cells and ancillary facilities are under planning stage.

ACKNOWLEDGEMENTS

Author is thankful to all scientists, engineers and supporting staff of Nuclear Engineering Division, Material Division, Health Physics Division, General Services Division, Isotope Production Division of PINSTECH and others involved in fission ⁹⁹Mo production program in Pakistan.

REFERENCES

- [1] MUSHTAQ, A., MANSUR, M.S., KARIM, H.M.A., KHAN. M.A., Hydrated titanium dioxide as an adsorbent for ⁹⁹Mo-^{99m}Tc generator. J. Radioanal. Nucl. Chem. Articles.147 (1991) 257-261.
- [2] MUSHTAQ, A., Underused Technetium-99m generators. Turk. J. Nucl. Sci 27, 2 (2001) 59-64.
- [3] MUSHTAQ, A., Quantification of pertechnetate in ^{99m}Tc-MIBI. Nucl. Med, Commun, 29, 9 (2008) 838.
- [4] MUSHTAQ, A., PERVEZ, S., HAIDER, I., MANSUR, M.S., JEHANGIR, M., A freeze dried kit for ^{99m}Tc(V) dimercaptosuccinic acid. J. Radioanal. Nucl. Chem. 243(3) (2000) 827-829.
- [5] PERVEZ, S., MUSHTAQ, A., ARIF, M., Technetium-99m direct radiolabeling of Lanreotide: A somatostatin analog. Appl. Radiat. Isotop. 55 (2001) 647-651.
- [6] ROOHI, S., MUSHTAQ, A., MALIK, S.A., Synthesis and biodistribution of [^{99m}Tc] Vancomycin in a model of bacterial infection. Radiochim Acta. 93 (2005) 415-418.
- [7] MANSUR, M.S., MUSHTAQ, A., JEHANGIR, M., Fractionation of lyophilized MIBI kit for ^{99m}Tc labeling. J. Radioanal. Nucl. Chem. 268, 1 (2006) 141-143.
- [8] DEEN, J. R., WOODRUFF, W. L., COSTESCU, C. I., "WIMS/D4 User Manual REV.0" ANL/RERTR/TM-23. 1995.
- [9] HALSALL, M. J., 1980. "A Summary of WIMSD4 Input Options", AEEW-M 1327.
- [10] RUBIO, R.O., 1993. INVAP SE "BORGRS v3.0".
- [11] FOWLER, T. B., VONDY, D. R., CUNNINGHAM, G. W., 1971. "Nuclear Reactor Core Analyses Code CITATION", ORNL TM 2496 Rev. 2.
- [12] Pakistan Research Reactor–1, "Final Safety Analysis Report (FSAR) for Conversion to LEU Fuel and Power Upgradation" Revision 1. Sep. 1992.
- [13] BOKHARI, I. H., ISRAR, M., PERVEZ, S., 'Thermal Hydraulic and Safety Analyses for Pakistan Research Reactor-1', Proceedings Of 22nd International Meeting on Reduced Enrichment for Research and Test Reactors (RERTR), 3-8 October, 1999, Budapest, Hungary.
- [14] BOKHARI, I.H., ISRAR, M., PERVEZ, S., 'Analysis of Reactivity Induced Accidents at Pakistan Research Reactor-1', Annals of Nuclear Energy. 29 (2002) 2225-2234.
- [15] MIRSHAK, S., DURANT, W.D., TOWELL, R.H., 1959, 'Heat Flux at Burnout', DuPont, DP-355.
- [16] MUSHTAQ, A., IQBAL, M., BOKHARI, I.H., TARIQ, M., TAYYB, M., ZAHOOR, A., QAMAR, Z., Neutronic and thermal hydraulic analysis for production of fission molybdenum-99 at Pakistan Research Reactor-1.
- [17] MUSHTAQ, A., IQBAL, M., BOKHARI, I.H., TAYYB, M., "Low enriched uranium foil plate target for the production of fission molybdenum-99 in Pakistan Research Reactor-1. Nuclear Instruments and Methods in Physics Research B 267 (2009) 1109– 1114.
- [18] MUSHTAQ, A., Specifications and qualification of uranium/aluminum alloy plate target for the production of fission molybdenum-99. Nucl. Eng. Des. 241 (2011) 163–167.