High Power Targets for Cyclotron Production of $^{99m}$Tc†


aTRIUMF, Vancouver BC, Canada; bLawson Health Research Institute (LHRI), London ON, Canada; cCentre for Probe Development and Commercialization (CPDC), McMaster University, Hamilton ON, Canada; dBritish Columbia Cancer Agency (BCCA), Vancouver, BC, Canada

Introduction

Technetium-99m, supplied in the form of $^{99m}$Mo/$^{99m}$Tc generators, is the most widely used radioisotope for nuclear medical imaging. The parent isotope $^{99m}$Mo is currently produced in nuclear reactors. The NRU reactor in Chalk River Canada currently produces 35-40% of the world supply and the HFR, the Petten reactor in the Netherlands produces another 30%, in total covering about 70% of global requirements. However, both reactors are aging and have experienced extended shut downs, particularly in 2009-2010, which caused disruptions in the $^{99m}$Mo supply chain [1]. In 2010 the Canadian government made the announcement that after 2016 the production of Mo-99 at the NRU reactor would no longer be supported. This has prompted the development of non-reactor based technologies for producing Tc99m. A number of technologies have been investigated for the production of Mo-99 or via the direct production of $^{99m}$Tc.

Our approach involves the $^{100}$Mo(p,2n)$^{99m}$Tc reaction on isotopically enriched molybdenum using small medical cyclotrons ($E_p \leq 20$ MeV), which is a viable method for the production of clinically useful quantities of $^{99m}$Tc [2]. Multi-Curie production of $^{99m}$Tc requires a $^{100}$Mo target capable of dissipating high beam intensities [3]. We have reported the fabrication of $^{100}$Mo targets of both small and large area targets by electrophoretic deposition and subsequent sintering [4]. However, we initially encountered thermal transfer issues with the EPD fabricated GE-Pettrace targets, and ultimately a redesign of this target was put into place. As part of our efforts to further enhance the performance of these high power targets, we have developed a novel system based on a pressed and sintered $^{100}$Mo plate brazed onto a dispersion-strengthened copper backing.

Materials and Methods

In the first step, a molybdenum plate is produced similarly to the method described in [5] by compacting approximately 1.6 g of commercially available $^{100}$Mo powder (Isoflex; 99.815%) using a cylindrical tool of 20 mm diameter. A pressure between 25 kN/cm$^2$ and 250 kN/cm$^2$ is applied by means of a desktop hydraulic press.

The pressed molybdenum plate is then sintered in a reducing atmosphere (Ar/2% H$_2$) at 1,700 $^\circ$C for five hours in a Carbolite tube furnace using the same temperature profile as previously reported [4]. The resulting $^{100}$Mo plates have about 90-95% of the bulk density of molybdenum with an average thickness of 0.63 mm.

The $^{100}$Mo plate is brazed in a GCF1100 inert gas furnace at ~750 $^\circ$C onto a backing manufactured from a dispersion strengthened copper composite (e.g. Glidcop AL-15) using a high temperature silver-copper-phosphorus brazing filler.

This process yields a unique, mechanically and thermally robust target system for high beam power irradiation [Fig. 1].

Materials and Methods

In the first step, a molybdenum plate is produced similarly to the method described in [5] by compacting approximately 1.6 g of commercially available $^{100}$Mo powder (Isoflex; 99.815%) using a cylindrical tool of 20 mm diameter. A pressure between 25 kN/cm$^2$ and 250 kN/cm$^2$ is applied by means of a desktop hydraulic press.

The pressed molybdenum plate is then sintered in a reducing atmosphere (Ar/2% H$_2$) at 1,700 $^\circ$C for five hours in a Carbolite tube furnace using the same temperature profile as previously reported [4]. The resulting $^{100}$Mo plates have about 90-95% of the bulk density of molybdenum with an average thickness of 0.63 mm.

The $^{100}$Mo plate is brazed in a GCF1100 inert gas furnace at ~750 $^\circ$C onto a backing manufactured from a dispersion strengthened copper composite (e.g. Glidcop AL-15) using a high temperature silver-copper-phosphorus brazing filler.

This process yields a unique, mechanically and thermally robust target system for high beam power irradiation [Fig. 1].

Figure 1. Brazed Mo-Cu Target Disc for GE PETtrace Cyclotron

Irradiations were performed on the GE PETtrace cyclotrons at LHRI and CPDC with 16.5 MeV protons and beam currents ≥100 µA [Fig. 2]. Targets were visually inspected after a 6 hour, 130 µA bombardment (2.73 kW/cm$^2$, average) and were found fully intact. Up to 4.7 Ci of $^{99m}$Tc have

†Patent pending.  ‡Corresponding author; e-mail: zeisler@triumf.ca
been produced to date. The saturated production yield remained constant between 2 hour and 6 hour irradiations.

Figure 2. Enriched molybdenum target in capsule after irradiation on the GE PETtrace cyclotron.

Results and Conclusion
It was noted during the Mo-plate fabrication that particle size does not have a large effect on the final molybdenum pellet. The grain size certainly plays a role during sintering and densification; however, the molybdenum plates do not require a near theoretical density, and therefore the particle size variation was not considered to be an issue. It was also discovered that some porosity is desirable as it aids in the dissolution of the target in preparation for the separation of the technetium from the molybdenum.

The finer grain sizes, particularly in the low micron range, were found to affect the flatness of the Mo-plate. The densification forces are strong enough to pull up the edges of the pellet, thus creating a bowl shaped piece [Figure 3]. This problem was remedied by placing a weight on top that does not bond to the molybdenum plate, such as a flat piece of alumina [5]. The flatness of the Mo-plate is crucial to ensure a good mechanical bond is made to the target backing and for good thermal conductivity during irradiation, particularly at high power densities.

We chose to utilize a thick molybdenum plate for this particular target design to allow for the entire beam energy to be deposited into it, due to the fact that copper is highly activated and produces long lived $^{65}$Zn. Initially we had designed the EPD style target [4] utilizing a tantalum backing, which allows for the deposition of 10 MeV of protons into it, as tantalum is not significantly activated. Due to thermal transfer issues with this particular design, we chose a copper composite as the target backing for its good thermal conductivity and its relative lower cost compared to tantalum.

Figure 3. On the left a flat molybdenum plate versus the right, which contains a curved, sintered molybdenum plate.

The size of this makes it suitable for high throughput manufacturing. Many pellets can be placed in an alumina boat for sintering and for brazing [Fig. 4]. The process was found to be very robust and highly reproducible.

Figure 4. Mo-targets in alumina boats

The results demonstrate that our brazed target assembly can withstand high beam intensities for long irradiations without deterioration. The molybdenum target appears to have no defects after irradiation with visual inspection [Fig. 3]. The silver-copper braze is a high temperature material with a good thermal conductivity, which creates a firm bond between the molybdenum plate and the backing. This assembly allows for high beam power on small orthogonal targets to be efficiently dissipated. Efforts are currently underway to determine maximum performance parameters.

References


**Acknowledgements**
Funding for this work was provided by Natural Resources Canada under the NRCAN-ITAP program.