Results of the Six-Day Electron-Accelerator Irradiation of Enriched Mo-100 Targets for the Production of Mo-99

Experimental Operations and Facilities Division

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by

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ABSTRACT

For six days, a 12-mm-diameter metal molybdenum target containing 15 enriched Mo-100 disks was irradiated at Argonne's electron linac. This report describes the irradiation conditions and the means used to process the targets for shipment to NorthStar Medical Isotopes, LLC, for feed to their RadioGenix™ technetium generator.

1 INTRODUCTION

NorthStar Medical Radioisotopes, LLC (NorthStar), plans to produce an important medical isotope, Mo-99, by using a photonuclear path. In this new method of producing Mo-99, a target consisting of enriched Mo-100 press-and-sintered metallic disks is irradiated with a high-current electron beam. Argonne National Laboratory (Argonne), in collaboration with Los Alamos National Laboratory (LANL) and NorthStar, and with support from National Nuclear Security Administration's Office of Material Management and Minimization, is developing technology for the production of Mo-99 via electron accelerator irradiation (Chemerisov et al. 2014; ROTSCH-2014; Gromov et al. 2015a). Argonne has developed technology to dissolve and process the irradiated Mo targets to produce a feed that can be used by NorthStar's RadioGenix™ Tc-99m generator (Messina et al. 2014).

In support of these activities, we conducted five irradiations of enriched Mo-100 targets at the Argonne low-energy accelerator facility (LEAF) and produced up to 15 Ci of Mo-99. We reported the results of those irradiations in (Chemerisov et al. 2016a,b). To test the performance of RadioGenix with an amount of radioactivity similar to what will be used in radio-pharmacies, we designed an experiment to provide an appropriate amount of Mo-99. The aim of the experiment was to produce ~60 Ci of Mo-99, and to ship three source vessels containing several Ci of Mo-99 to NorthStar for testing on the RadioGenix™ Tc-99m generator.

2 DESCRIPTION OF IRRADIATION EXPERIMENTS

The target assembly and He-cooling loop for the target were designed by LANL, and are discussed elsewhere (Dale 2014; Dale et al. 2013). Figure 1 shows the main components of the irradiation setup and Figure 2 is a photograph of the irradiation setup with beamline and shielding. Argonne's low-energy and high-power electron linac, was used as the source of the electron beam for the experiments. The parameters of the accelerator appear in (Gromov et al. 2015a,b).

FIGURE 1 Placement of Mo disk target and He cooling system in the irradiation room of the Argonne linac.

FIGURE 2 Beamline with installed Mo disk target and He cooling system in the irradiation room of Argonne linac.

The molybdenum target consists of 25 metallic disks (1 mm thick, 12 mm diameter) that are separated by 1-mm gaps for gaseous He cooling (Figure 3). In order to produce ~60 Ci of Mo-99 by the end of 6 days and to place the resulting solution into three source vessels, we used 15 enriched Mo-100 disks placed in positions 3–17. This placement provided the highest production of Mo-99.

FIGURE 3 Close-up of open half-shell Mo disk target and drawings of the target inside lead shielding. Arrows in the left panel point to Mo-100 disks (position 3–17).

The He gas flow was supplied by a closed-loop cooling system. The cooling system consists of a roots blower, a pair of heat exchangers, a mass-flow meter, an oil filter, and piping. The cooling system is pressurized to 280–290 psig, and the roots blower can provide a flow of ~100 g/s of He gas through the target.

Placement of the beam and the beam shape on the target window was monitored using optical transition radiation (OTR). OTR is a visible light generated when relativistic electrons pass through a boundary between two materials with different characteristics. The OTR image, therefore, provides the location and distribution of the electron beam at the entrance window to the target. The image is captured by a charge-coupled device (CCD) camera Basler™ acA640-120gm equipped with a 180-mm focal length lens. A significant amount of shielding is required to protect the camera from the background radiation. The camera is placed at a significant distance from the target (approximately 2 m) and by shielded with ~15 cm of lead and 10–15 cm of borated polyethylene. Lead provides shielding from x-rays, while borated polyethylene provides shielding from neutrons. The camera is located near the floor, and a pair of mirrors is used to provide an optical line of sight from the camera to the target window, while avoiding direct line of sight to the target (source of x-rays and neutrons). The camera is synchronized to the electron beam and communication is provided via an Ethernet connection to the control system. Figure 4 shows an example of OTR images of an electron beam on the target.

FIGURE 4 OTR beam image detected by CCD camera. Red circle: 12-mm-diameter target window area. Green and blue rectangles: integration regions providing horizontal and vertical beam profile. Beam is shifted off the target window to flat plate for profile tuning.

The shielded cart described in [CHEMERISOV-2016] provides a way to insert, remove, and transport the target assembly from the target housing attached to the beamline to a shielded cell located in an adjacent room. All operations are performed with adequate shielding around the target, and all cart operations are performed manually.

The 99.815% enriched Mo-100 material used for 15 enriched Mo disks was obtained from Isoflex, in the United States. Table 1 lists the composition of the enriched Mo-100 material. Impurities present in the enriched Mo-100 material are listed in Table 2.

TABLE 1 Isotopic composition of enriched Mo-100 material

Isotope	Content (%)
Mo-92	0.003
Mo-94	0.003
Mo-95	0.003
Mo-96	0.003
Mo-97	0.003
Mo-98	0.17
Mo-100	99.815

**TABLE 2 Chemical impurities
present in enriched Mo-100 material**

Element	Content (ppm)
Mn	<0.1
Cr	1.16
Cu	5
Fe	16
Sn	1.80
Ni	0.50
Si	15
Na	6.0
Mg	1.00
Ti	0.26
Al	2.16
Co	<0.1
Zn	<1.0
W	14

3 BEAM PARAMETERS FOR 6-DAY IRRADIATION RUN

We performed the 6-day irradiation run with an electron beam energy of 40 MeV. Linac's spectrometer has an upper limit for beam energy measurements of 40 MeV; therefore, the beam energy profile tuning was performed at an energy of 36 MeV (Figure 5). After that, we decreased the injector current to reduce beam load for the accelerating structure and to increase the energy gain of the electron beam up to 40 MeV according to the linac's load-line data presented in [CHEMERISOV-2016]. The transverse beam size was tuned to 5.9×5.8 mm at the target window.

We focused and aligned the beam using a low beam power (about 0.2 kW). The beam was placed on the flat part of the target housing. After appropriate beam shape had been achieved, the beam was moved to the center of the window. Because the window is concave, the beam size measured from the OTR signal from the window is much smaller. After a tune-up, the repetition rate was slowly increased to 180 Hz, which corresponds to an average beam current of about 380 μ A and an average beam power of about 15 kW (Figure 6). This power was maintained most of the time, except during a few interruptions due to arcing in the accelerator structures.

We monitored the average beam power during the experiment with fast current transformers. The cause of some variation of the power is due to the injector's thermal cathode instability. Keeping the power in the desired range required periodic correction by the linac operator. At the beginning of the irradiation, the plan was to set the beam power at 18.5 kW, but during the first hour one of the modulator high-voltage power supplies failed. This issue limited the maximal repetition rate of the linac up to 180 Hz, so the beam power was limited to 15 kW. The total power delivered on the target was 2,200 kW·h.

FIGURE 5 Beam energy spectrum at about 36.5 MeV. The full-width at half-maximum (FWHM) of the beam energy spectrum is ~1.2 MeV.

FIGURE 6 Beam power history

4 GAMMA COUNTING

Gamma counting of irradiated and dissolved enriched Mo-100 targets was performed by taking a small aliquot from final potassium molybdate solution in ~5M KOH. The aliquot was counted using an high purity Ge detector (HPGe) that was calibrated using multi-nuclide gamma standard from Eckert & Ziegler. Mo-99 was detected using a 739-keV line. The presence of side reaction products such as Nb-95, Nb-96, Nb-97, Nb-98m, and Zr-95, and impurity activation products such as Cr-51 ($^{52}\text{Cr}(\gamma, n)^{51}\text{Cr}$), Mn-54 ($^{55}\text{Mn}(\gamma, n)^{54}\text{Mn}$), and Co-57 ($^{58}\text{Ni}(\gamma, p)^{57}\text{Co}$) was also monitored.

5 CHEMICAL PROCESSING

5.1 COLD Mo DISSOLUTION TESTING

In preparation for the 60-Ci Mo-99 production run, a small dissolution setup was designed for use in the Argonne hot cell. A 500-mL Erlenmeyer flask was used for dissolution, and water vapor was condensed by a series of two condensers. To minimize the release of Mo-99 into a hot cell atmosphere, oxygen that formed during the dissolution was released from the dissolution setup after running it through glass beads mixed with Drierite to capture any droplets of moisture that may have carried Mo particles.

Nine dissolutions with 15 g of sintered Mo disks (using broken 26-mm $\times$ 1-mm disks) and one dissolution using 12-mm $\times$ 1-mm disks were performed with 50% hydrogen peroxide (Fisher Chemical) to test the apparatus (Figure 7). An average consumption of peroxide for complete dissolution of ~15 g of Mo was 172 ± 8 mL, with an average dissolution time of 21 ± 3 min, giving a dissolution rate of 0.74 ± 0.14 g/min. The goal of the cold dissolution runs was to determine whether a 500-mL flask is sufficient for dissolution of 15-g batches. We determined that if H_2O_2 is added in 10-mL aliquots, a 500-mL flask provides more than sufficient head space to contain the vigorous reaction.

Another goal of these runs was to determine whether any Mo aerosols could escape the dissolution apparatus, and potentially contaminate the hot cell or the ventilation system. We successfully demonstrated that a combination of two condensers, with Drierite in the last condensation flask, prevented Mo from escaping the dissolution apparatus. As an extra step, we installed a HEPA filter (high-efficiency particulate air filter). We tested it during the final cold dissolution, and then implemented it while processing the irradiated Mo targets. The total processing time, including dissolution, evaporation, and filtration, was about 90 min.

FIGURE 7 Dissolution setup developed for dissolution of ~15 g of enriched Mo-100 disks in hot cell

5.2 DISSOLUTION OF IRRADIATED Mo-100 TARGETS

Fifteen 99.815% enriched Mo-100 12-mm × 1-mm disks were irradiated (positions 4–18). Their characteristics are summarized in Table 3. In addition, nine sintered natural Mo disks were irradiated, but were not dissolved. After transferring the irradiated target into the hot cell, we observed some corrosion on the target housing. The first three natural Mo disks were black, with some rusty coloration similar to the corrosion observed on the target housing (Figure 8). Enriched Mo-100 disks seemed to be less affected by corrosion. Similar corrosion was observed in previous irradiations, and we believe that it is a product of corrosion of the carbon-steel shielding enclosure or the He cooling loop that supported the target assembly.

Fifteen Mo-100 enriched disks (15.17 g) were separated from natural Mo disks, placed into a 500-mL Erlenmeyer flask, and dissolved in ~170mL of 50% hydrogen peroxide (Figure 9). The water vapor coming from the dissolution reaction was condensed into a collection flask. After complete dissolution, we evaporated the solution using a hot plate to concentrate Mo and destroy excess peroxide. After the solution turned orange, a saturated solution of KOH was added to convert Mo into potassium molybdate. The solution then turned colorless, and evaporation continued.

TABLE 3 Characteristics of 15 Mo-100 enriched disks used in irradiation

Disk	Mass (g)	Theoretical Density	Diameter (mm)	Thickness (mm)
190124-1	1.03	86%	11.99	0.998
190124-2	1.01	85%	11.99	0.986
190124-3	1.01	85%	11.99	0.985
190124-4	1.01	85%	11.99	0.987
190124-5	1.00	84%	11.99	0.989
190124-6	1.01	86%	11.99	0.982
190124-7	1.01	85%	11.99	0.985
190124-8	1.01	85%	11.99	0.984
190124-9	1.01	88%	11.99	0.956
190124-10	1.01	85%	11.99	0.991
190124-11	1.01	85%	11.99	0.986
190124-12	1.01	86%	11.99	0.980
190124-13	1.01	85%	11.99	0.988
190124-14	1.01	85%	11.99	0.985
190124-15	1.02	86%	11.99	0.992

FIGURE 8 Corrosion observed on opened target housing after irradiation containing 25 disks

FIGURE 9 Photographs of dissolution setup in hot cell (left), Mo solution after complete dissolution (middle), and clear solution of K_2MoO_4 in ~ 5 M KOH obtained after filtration and sent to NorthStar (right)

During the evaporation step, Mo solidified as potassium molybdate salt, and was extremely difficult to convert back to solution. Due to time limitations, we decided to add more water to help re-dissolve the Mo solid. However, we expect that ~5% of Mo did not re-dissolve. Then, KNO_3 was added, followed by $\text{Fe}(\text{NO}_3)_3$ to co-precipitate Zr and Nb isotopes, and the solution was filtered. After filtration, a final addition of KOH was made. An additional volume of water led to the dilution of the final Mo concentration. The final solution had a density of ~1.34 g/mL instead of the desired 1.55g/mL. The estimated total volume of the Mo solution in KOH was ~95 mL, while the desired volume was ~76 mL.

A small aliquot of Mo product solution (30 uL) was transferred for gamma counting. Because dose rates of the aliquot were very high (~22 mR/hr at 30 cm), a second dilution had to be performed. We counted 2.07 uL aliquot at 181cm from the HPGe detector, giving ~3.4% dead time. Gamma counting results are shown in Table 4.

TABLE 4 Gamma counting results of Mo-99 product

Radionuclide	Total Activity at EOB (mCi)	Total with 5% Undissolved (mCi)	X/Mo-99	1 Sigma
Mo-99	5.15E+04	5.40E+04	100%	3.0%
Zr-95	7.39E-01	1.44E-05	0.0014%	3.2%
Cr-51	2.93E+00	5.68E-05	0.0057%	13.8%
Nb-95	4.41E-01	8.57E-06	0.0009%	4.0%

Based on gamma counting of the aliquot, we determined that at EOB (end of bombardment) we had produced ~54 Ci of Mo-99 in 15 enriched Mo-100 disks. This is in very good agreement with the predicted activity of ~57 Ci.

Three shipping vessels were loaded with about 10.4 Ci of Mo-99, and a total of ~31.1 Ci at time of shipment on Tuesday, October 15, 2019, at 10:00 A.M. Two vessels were delivered to NorthStar's Madison facility, and one vessel was delivered to NorthStar's Beloit facility. After about 6 weeks of decay, the Mo-99 product aliquot was counted again to determine whether any long-lived side-reaction or activation products were present.

A very small presence of Zr-95 and Nb-95 was observed, with a total fraction of less than 0.002% of Mo-99 activity at EOB. This indicates that co-precipitation with Fe effectively removes Zr and Nb isotopes from the Mo-99 product. The absence of activation products Mn-54 and Co-57 correlates well with their very low content in the starting enriched Mo-100 material (Table 2). Data in Table 3 could indicate some presence of Cr-51, but due to very low activity and bad counting statistics (counting time of ~15.5 h), its presence should not be of significant concern.

6 CONCLUSION

For six days, 12-mm $\times$ 1-mm sintered Mo metal disks containing 15 enriched Mo-100 disks, and 10 natural Mo disks were irradiated at Argonne's electron linac, LEAF. Due to the failure of one of the high-voltage power supplies at the beginning of irradiation, the beam power was limited to 15 kW, and the total power delivered on the target was 2,200 kW·h. After transferring the irradiated target into a hot cell for processing, corrosion on the target housing was observed. Similar corrosion was observed in previous irradiations, and we believe that this is a product of corrosion of the carbon-steel shielding enclosure or the He cooling loop.

During the chemical processing of irradiated Mo-100 disks, the Mo solution solidified as potassium molybdate salt, and was extremely difficult to convert back to solution. Because of that, more water was added to re-dissolve the Mo salt. However, we expect that ~5% of Mo did not re-dissolve. Based on gamma counting results, we determined that ~54 Ci of Mo-99 was produced in ~15 g of Mo-100 disks. Three vessels containing about 10.4 Ci each were shipped to NorthStar for testing on the RadioGenix™ Mo-99/Tc-99m generator. A very small presence of Zr-95 and Nb-95 was observed, with a total fraction of less than 0.002% of Mo-99 activity at EOB.

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