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Production of Tc-99m from Naturally Occurring Molybdenum Absent Uranium

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Abstract*—Technetium-99m (Tc-99m) is the world's most widely used medical isotope. Current production methods involve the irradiation of highly enriched uranium (HEU) and low enriched uranium (LEU) targets in nuclear reactors. Molybdenum-99 (Mo-99) is then extracted from these targets, which decays to Tc-99m. Currently, this process is centralized, as there are very few companies that manufacture Mo-99. In an effort to eradicate the need for uranium to produce this medical isotope, naturally occurring molybdenum was studied to produce Mo-99. Preliminary testing at Princeton Plasma Physics Laboratory included irradiating naturally occurring Mo coupons for varying amounts of time using a D-T neutron generator producing $1.5x10^8$ n/sec to produce Mo-99. Exploiting this technique, Tc-99m was successfully produced. Proof of principle testing is also underway to confirm the ability to produce Mo-99 from Mo-100 using high-energy gamma rays. Future work consists of creating a mobile device that is able to produce Tc-99m on demand, allowing for a distributed system of the medical isotope in hospitals and radio pharmacies worldwide.

Keywords: neutron activation, high energy gamma, medical isotope

I. INTRODUCTION

Technetium-99m (Tc-99m) is a radioactive tracer isotope, frequently used in the nuclear medical field for diagnostic imaging. In the United States alone, it is used in about 2/3 of all diagnostic medical isotope procedures[1]. Worldwide, Tc-99m is used in about 30 million procedures annually, making it a >billion dollar industry. The use of Tc-99m in particular is beneficial for various reasons. Most importantly, it has a relatively short half-life of 6.03 hours. This is especially ideal in diagnostic tests, for the patient only retains a minimal amount of radiation from the examination while the radiologist is still able to attain thorough results quickly and efficiently. Tc-99m's metastable state also ensures that the element will not transform into any other substance due to its own decay. For the diagnostic process, Tc-99m kits are created for producing radiopharmaceuticals. These kits are composed of small molecule ligands and proteins chemically bound to the pertechnate (TcO_4^-) form of Tc-99m, thus allowing for the compound to specifically bind with particular organs and tissues of the body. When Tc-99m decays, it emits 140.5 keV of gamma ray energy. Since the wavelength of these gamma rays is nearly identical to that of x-rays, scintillation equipment, such as gamma cameras, can be used to detect the photon energy emissions. The single gamma ray without the presence of beta emission also advantageously allows for more precise imaging.

Tc-99m is produced solely from the gamma decay of Molybdenum-99 (Mo-99), which is created by either of two methods. The first and most common method is by the fission of Uranium-235 (U-235). The U-235 within highly enriched uranium (HEU) targets is irradiated with neutrons in a nuclear reactor. The fission reaction results in Mo-99 as well as other medical isotopes Iodine-131 and Xenon-133 and extraneous fission fragments. The fission cross section is important to note, as it describes the cross section of an atom that is open to neutron scattering. For the thermal fission of U-235, the fission cross section is about 600 barns, where 1 barn = 1×10^{-24} cm^2. Only 6.1% of this results in Mo-99, which results in about 37 barns.

Because Mo-99 has a relatively short half life, it cannot be stored for an extended period of time. It must be continuously made, especially because it is in such high demand. Therefore, due to the limited number of facilities that are currently able to create the Mo-99, any unplanned interruptions of these facilities' functionality could have severe effects on patient care. In addition, the nuclear waste that is created as a byproduct of this fission reaction is clearly undesirable when considering clean environmental concerns. For these reasons, alternative sources of Mo-99 are being sought. Thus, a second, uncommon method is being further investigated which makes use of Molybdenum-98 (Mo-98) and Molybdenum-100 (Mo-100).

II. METHODOLOGY

Mo-98 is the most common molybdenum isotope and comprises 24.14% of all naturally occurring molybdenum. Its very long half life makes it ideal as target material. The Mo-98 is bombarded with neutrons and thus irradiated into Mo-99, which can then follow the same decay process into Tc-99m.

It is important to note that the Mo-99 created in this process is slightly different than that created in the fission process, as

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it has a lower specific activity. This is because due to the stable nature of Mo-98, most of the molybdenum in the Mo-99 is still comprised of Mo-98. Therefore, a larger technetium generator column and more liquid are needed to elute the Tc-99m. In addition, the production cross-section for this reaction is about 0.13 barns, compared to the 37 barn cross section from fission-produced Mo-99. This would require a high flux reactor that could compensate for this lower cross section in order to produce the same amount of Mo-99.

Despite these differences, this process requires no uranium, and thus, the amount of radioactive waste that is produced is greatly reduced, compared to the clean and natural method of the neutron capture process. In addition, the only four facilities, which supply Mo-99 for the rest of the world, use the fission process to create the Mo-99. Unfortunately, their age frequently causes sporadic breakdowns, resulting in an erratic supply for such a crucial element. Some countries, such as Japan, have begun using the neutron-capture process to provide a stable domestic supply, should unexpected halts in Mo-99 flow occur.

Mo-100 accounts for about 9.63% of naturally occurring molybdenum, making it another viable isotope to aid in the production of Tc-99m. When introduced to high-energy gamma rays, Mo-100 is irradiated to Mo-99, which will follow the normal decay of Mo-99 to the desirable Tc-99m. While about 1% of the Mo-98 reaction with neutrons will result in Mo-99, about 4% of this Mo-100 reaction with high energy gamma rays will result in Mo-99 (compared to the 6.1% of the fission reaction). This higher yield makes this reaction very desirable and worth investigation.

Though this process does produce a smaller amount of Mo-99 than the fission method, it has distinct advantages that would ultimately allow for a greater percentage of the world population to gain access to this crucial medical technology. This process may certainly have some disadvantages in terms of production, but the necessity of clean and dependable alternative methods is pressing.

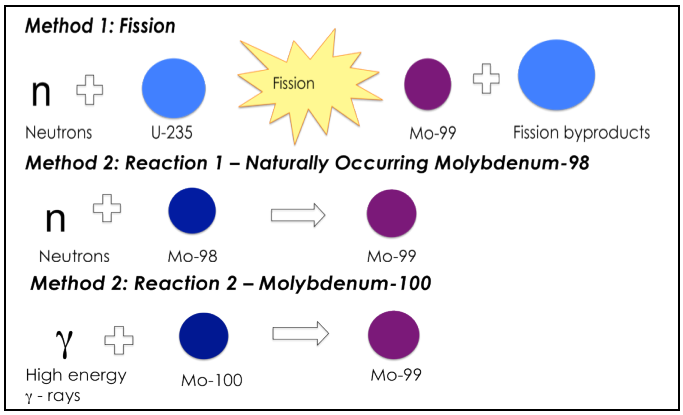

Figure 1: Graphical representation of reactions used to produce Mo-99.

III. Experimental Setup

Due to the use of two reactions for the production of Mo-99 from other molybdenum isotopes, two separate experimental setups were created and tested.

A. *Molybdenum-98 Experimental Setup*

A pulsed Deuterium-Tritium (D-T) Neutron Generator was employed for this reaction as a neutron source. The device emits 1.5 x 10^8 neutrons/sec. at 14 MeV, making it a viable source for neutrons for irradiation. Coupons of Mo-98 were placed 25 cm away from the head of the D-T generator, then the structure was surrounded by lead bricks to maximize the neutron exposure to the coupons. A neutron detector was placed adjacent to the structure to ensure proper neutron production. The setup was then encased in polyethylene to shield the outside from neutron exposure. This setup can be seen in Figure 2 below.

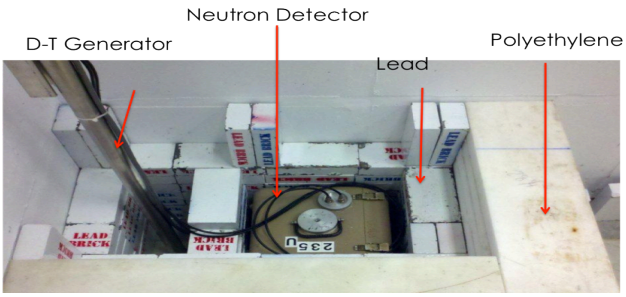

Figure 2: Experimental setup of neutron irradiation via the D-T Neutron Generator.

B. *Molybdenum-100 Experimental Setup*

As seen in Figure 1, in order to create the second method reaction that utilizes Mo-100, high-energy gamma rays are required. These high-energy gamma rays were produced using the D-T Neutron Generator by irradiating water. The O^{16} in water can be irradiated to N^{16} when exposed to neutrons, which emit 6.2 MeV gamma rays. The exploitation of this reaction is favorable for multiple reasons. In using the D-T Neutron Generator to produce high-energy gamma rays, it is possible to use the same device to create Mo-99 from Mo-98 and Mo-100. Also, O^{16} is obviously abundant since it is found in water, making it possible to create these gamma rays at low cost. In order to create high-energy gamma rays using water, tygon tubes filled with distilled water were wrapped around the head of the D-T Neutron Generator. The configuration of the devices can be seen in Figure 2. As used in the Mo-98 irradiation, the D-T Neutron Generator, neutron detector, lead bricks, and polyethylene were utilized.

IV. Monte Carlo N-Particle Modeling

Monte Carlo N-Particle Transport Code (MCNP) is used in this study to simulate neutron transport from the D-T generator to make molybdenum-99.

The input consists of a cell card, surface card, and data card. The cell card describes the problem space within respective boundaries. The surface card describes the problem geometry using Cartesian coordinates. The data card defines problem specifications, including information about the neutron source and energy distribution, as well as tally counts. Tallies are a crucial element of MCNP, for it provides the user with information about current across a certain point, fluxes, etc. The MCNP code that was used for these purposes focuses on the neutron tally over each surface, the average neutron flux over each surface, the average neutron flux within the molybdenum cell itself, and the energy deposition in the molybdenum cell. The energy distribution of the neutron source is normal Gaussian.

Previous experiments using this D-T generator have shown that the distribution of neutrons consists of a pluming effect. From this, the input code reflects these assumptions.

- Due to this pluming effect, only a proportion of neutrons will physically reach the molybdenum coupon. Thus, an estimate of 100,000 neutrons is predicted.
- The source of the neutrons is therefore placed directly in front of the molybdenum coupon, even though the experimental design has the neutron source placed 25 centimeters away. If the source were placed at this distance in the code, MCNP would show that too many neutrons were being lost due to the neutron distribution and isotropic scattering of the neutrons. Thus, these parameters were created to rectify this situation.

The tallies provide information regarding the percentage of neutrons hitting the target and creating a reaction. This gives an idea of the neutron distribution amongst the molybdenum target. Parameters in the code can easily be changed to provide updated information about the experiment. Current theoretical results from the MCNP coding have shown the ability to produce around 50 Milli Curies/day using both molybdenum reactions. This would allow for between 5 – 25 procedures/day, since a single procedures uses between 2 – 10 Milli Curies.

V. Results

A. *Molybdenum-98 Experimental Results*

The Mo-98 coupons were exposed to the neutron source for varying amounts of time. The data collected can be seen in Figure 3. As can be seen, detectable amounts of Tc-99m were found in the sample. Detectable amounts of Mo-99 were found in the sample as well. The majority of the remaining parts in the sample were Mo-98. This could potentially be extracted and reused for future reactions.

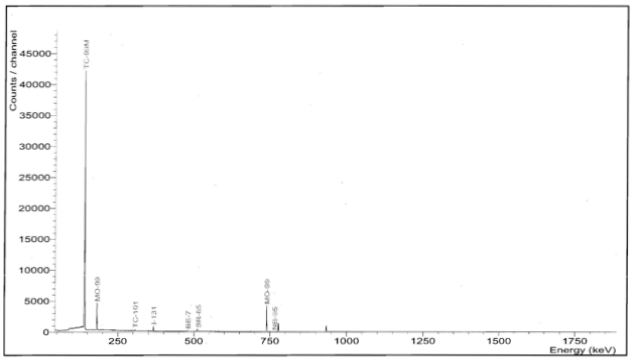

Figure 3: Results from Mo-98 exposure to neutrons. Tc-99m was detected, as well as Mo-99 and Mo-99.

B. *Molybdenum-100 Experimental Results*

As stated, high-energy gamma rays are required to irradiate the Mo-100 into Mo-99. These gamma rays came from a water activation study, whose results can be seen in Figure 4. This data shows that the gamma rays from N^{16} were detected at 6.2 MeV. These gamma rays can then be exposed to Mo-100, irradiating that material into Mo-99.

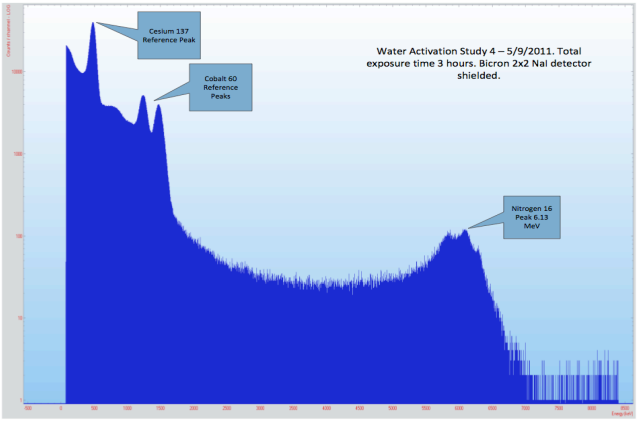

Figure 4: Results from the water activation study of exposing neutrons to O^{16}. Cesium and cobalt are shown as calibration curves.

VI. Future Work

Future work consists of continuing the development of MCNP modeling for the production modes. Through these models, it is possible to find the best combination of molybdenum reactions to maximize the Tc-99m yield. For the production of the high-energy gamma rays, other reactions are currently under investigation to produce prompt high-energy gamma rays in the 9-10 MeV range, which will increase the yield of Tc-99m. Once this maximum combination has been developed, the final configuration can be determined. After the final configuration has been developed and tested for safety and high production yield, it is possible to create a mobile device that allows for production in hospitals, radio pharmacies, and places where this type of imaging is not currently available. This device could be placed in hospitals and/or radio pharmacies to produce Tc-99m on demand. A technician will be able to turn on the machine and be able to extract the Tc-99m within a matter of days. This device would

also be able to travel to third world countries and places where these imaging capabilities are not currently possible. Since Tc-99m is used to image the heart, blood, and brain, the availability of this medical isotope could likely be used to diagnose many fatal diseases in these countries that may otherwise go untreated. This would allow for a very stable system, since the hospitals are responsible for their own yield, as well as a very distributed system in comparison to the extremely closed system that is currently in production.

REFERENCES

[1] Nuclear and Radiation Studies Board, *Medical Isotope Production without Highly Enriched Uranium.* Washington, DC: National Academies, 2009. Web. 28 June 2011.

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