# Production of Molybdenum-99 by Heterogeneous and Homogeneous Uranium Fueled Reactors

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## Abstract

The use of radioisotopes for various procedures in the health care industry has become one of the most important practices in medicine. At the forefront of the medical isotope list is molybdenum-99 and its daughter isotope technetium-99m, which encompass over 80% of radiopharmaceutical procedures [1]. Fission of uranium-235 to produce molybdenum-99 is the most widely used method for producing this radioisotope. The heterogeneous reactor and the aqueous homogeneous reactor are looked at here with emphasis on the use of low enriched uranium as the fuel source. Methods of technetium-99m generation and its medical use are also reviewed.

## 1. Background

Radioisotopes can be found both in nature and in nuclear reactions performed synthetically. They are the product of the over enrichment or shortage of nucleons (protons or neutrons) within an atom. Over the course of the last century, methods to produce an assortment of radioisotopes have been discovered and their decay mechanisms studied. The uses for radioisotopes range from medical care to industrial testing.

The most popular method for producing radioisotopes is through the irradiation of atoms with neutrons. By subjecting an element, X, to a flux of neutrons, the element can absorb a neutron increasing the neutron-to-proton ratio in the element, shown in Equation 1 below. This increase creates an imbalance in the neutron-to-proton ratio for element ${}^{A+1}_{Z}X$ that could be important enough to cause the element to either alpha or beta decay, or in some cases, undergo fission in an attempt to correct the non-preferred state. Equation 2 below depicts a negative beta decay characterized by an increase in the number of protons and decrease in the number of neutrons in the element, moving it closer to its original neutron-to-proton ratio.

$$ {}^{A}_{Z}X + {}^{1}_{0}n \rightarrow {}^{A+1}_{Z}X \quad (1)$$

$$ {}^{A+1}_{Z}X \xrightarrow{t_{1/2}} {}^{A+1}_{Z+1}X + {}^{0}_{-1}\beta^{-} + \bar{\nu} \quad (2)$$

where Z is the atomic number, A is the atomic mass number, n is a neutron, $\beta^{-}$ is a negative beta particle, and $\bar{\nu}$ is an anti-neutrino.

As mentioned above, in some cases the irradiation of an element can cause the fission of the nucleus. This is mostly restricted to a small list of heavy elements, mainly: uranium-233, uranium-235, and plutonium-239. When fission occurs in these elements, 2 fission products are produced as well as 2 or 3 neutrons. Equation 3 depicts the fission of uranium-235.

$$^{235}_{92}U + \ ^{1}_{0}n \xrightarrow{fission} X + Y + \sim 2 \cdot \ ^{1}_{0}n + \ \gamma \tag{3}$$

where X and Y are fission products and γ is a gamma ray. The two fission products are usually radioisotopes and have a ratio of neutrons to protons similar to that of the fissioning nucleus, meaning a high number of neutrons to protons, which is not preferred. To reduce this ratio, a radioactive decay occurs such as beta decay depicted in Equation 2 above.

Radioisotopes have found very useful application in the medical industry for diagnosis and therapy practices such as imaging and cancerous tumor destruction. The most commonly used radioisotope in the medical industry is the radioisotope molybdenum-99, accounting for about 80% of all radiopharmaceutical procedures [1]. This radioisotope is mainly prepared via the fission of $^{235}$U in fission reactors and is the parent nuclide to the radioisotope used in the imaging process, technetium-99m. $^{99}$Mo has a 66 hour half-life [2], which allows enough time for transportation from its production site to the consumer without losing too much of the isotope's radioactive value (activity).

The production of molybdenum-99 is performed in select reactors around North America and the world. The most popular method for $^{99}$Mo production is through the use of **highly enriched uranium** (HEU) targets, however, more research is beginning to be put into the use of **low enriched uranium** (LEU) as the target fuel, as well as the use of LEU in homogeneous reactor based production.

## 2. Production of Molybdenum-99 and Technetium-99m

At this time, the production of $^{99}$Mo has mainly been delegated to fission reactors utilizing weapons grade $^{235}$U (~93% $^{235}$U) in HEU targets as fission reactors provide an efficient source of thermal neutrons. The use of HEU has been preferred due to more extractable $^{99}$Mo produced from less material in a shorter time. $^{99}$Mo is produced from fission 6.1% of the time, so the more fissions that occur, the more $^{99}$Mo can be extracted and sold [2]. The reaction mechanism for fission is displayed below.

$$^{235}_{92}U + \ ^{1}_{0}n \xrightarrow{fission} \ ^{99}_{42}Mo + \ Other\ Fission\ Product(s) + \sim 2 \cdot \ ^{1}_{0}n + \ \gamma \tag{4}$$

When producing $^{99}$Mo, to reach the greatest concentration in the uranium target, it is necessary to irradiate the sample until saturation activity has been reached between production of $^{99}$Mo from fission and the radioactive decay of $^{99}$Mo. This saturation activity is essentially reached within 5 to 7 days with any irradiation time beyond that not producing a significantly higher concentration of molybdenum in the target [3]. When $^{99}$Mo is priced, it is based on units of radiation typically referred to as "activity". Activity is a measure of the decay rate of a nuclide species and in this case the unit is the curie (Ci) which is equal to 37 billion decays per second. As of 2009, the global demand for $^{99}$Mo is approximately 12,000 6-day curie per week [3]. Since this "6-day curie" unit is a measurement 6 days after production of $^{99}$Mo, reactors producing this radioisotope must produce approximately 71,000 Ci per week to meet this demand. To produce this 6-day Ci value, it would require approximately 0.11 grams of $^{99}$Mo after processing of spent fuel [3].

Due to the important proliferation and safety issues created by the use of weapons grade uranium, a strong push is being made to convert isotope production reactors to using LEU or uranium containing 20% or less uranium-235. The use of LEU to produce molybdenum and other radioisotopes could be of much benefit to developing countries that do not have access to HEU but want to supply themselves with these useful isotopes. Initial research was performed on LEU isotope production starting in 1986 within the U.S.'s Reduced Enrichment for Research and Test Reactors program aimed at developing fuel designed for minimum modifications to the most widely used processes [4].

The use of aqueous homogeneous fission reactors using liquid fuel with LEU has also been proposed to allow for small research reactors to produce radioisotopes in multiple locations all over the world. This could provide production pathways with less capital cost investment and a potentially lower operating cost [5].

### 2.1 Heterogeneous Reactor

Heterogeneous fueled reactors, the prevalent reactor type for molybdenum production, rely on the fission of solid uranium oxide fuel to produce the necessary flux of neutrons. The uranium targets that are used to produce molybdenum are specially created in varying geometric shapes ranging from thin sheets to pins or cylinders that are highly enriched with fissile $^{235}$U (~93%). The National Research Universal (NRU) reactor in Chalk River, Ontario utilizes targets of HEU-aluminum alloy pins within aluminum cladding allowing for irradiation of up to 20 targets at a time [6]. Once the target is removed from the reactor, it is cooled and moved to a "hot cell" where the molybdenum and other useful radioisotopes are separated. Due to a short irradiation time (~7 days), only 3% of the $^{235}$U is fissioned in the target leaving 97% to go to waste or partial recycling.

Utilizing low enriched uranium (LEU) or uranium containing 20% or less uranium-235 by weight greatly reduces proliferation issues arising from the use of weapons grade uranium. Since most current reactors will not support a change in the geometry of the targets they can accept, it is necessary to manufacture LEU targets with the same physical dimensions as the HEU targets currently being used. To allow for a LEU target to yield equivalent amounts of $^{99}$Mo, approximately five times as much target uranium is necessary and therefore requires an increase in the density of the uranium target. Chemical processing methods should also be kept the same as for HEU for cost saving reasons.

### 2.2 Aqueous Homogeneous Reactor

The development of the homogeneous reactor which combines moderator/coolant and uranium fuel in liquid solution was among the first nuclear systems to be developed after the discovery of fission. Renewed interest in the technology has been spurred in an effort to produce radioisotopes cost effectively and without the use of HEU. An aqueous homogeneous reactor is very different in design from a traditional heterogeneous reactor with solid uranium oxide fuel. The aqueous reactor relies on the use of a liquid fuel consisting of a solution of enriched uranium salt, traditionally either uranyl nitrate $(UO_2NO_3)_2$ or uranyl sulfate $(UO_2SO_4)$ acid to repress hydrolysis in the solution, and water (moderator/coolant). Listed below are some of the benefits to the use of a liquid fuel over a solid fuel for producing radioisotopes:

1. There is no need for fabrication and transportation of fuel elements and target;
2. Reactor design becomes more flexible in terms of parameter and geometry variation;
3. Molybdenum is produced directly from the reactor fuel, eliminating the $^{99}$Mo waste that heterogeneous reactors create in their fuel elements;
4. Fission product waste is 1/100$^{th}$ of that produced using a uranium target [7];
5. Uranium consumption and heat production are 1/100$^{th}$ that of using a uranium target [7];
6. Extraction of molybdenum is simplified by eliminating the need for uranium dissolution required when using solid target material;
7. Possibility for removal of neutron poisoning fission gasses allowing for higher burn-up of the fuel;
8. Inherent safety as these reactors often display strong negative temperature coefficients;

The flexibility of reactor design is a very important attribute that aqueous homogeneous reactors (AHR) possess. This flexibility makes it possible for a heterogeneous core to be replaced with a homogeneous core with minimal modifications to original core dimensions. Good examples for this conversion are the SLOWPOKE-2 research reactors at the Royal Military College of Canada and École Polytechnique de Montréal - These reactors could continue their research duties while producing medical isotopes.

The reduction in the amount of uranium needed on site and the increase in molybdenum extraction efficiency has also contributed largely to the attention that is being given to AHR's. An AHR uses its fission fuel to produce radioisotopes rather than a fabricated target. This provides a huge savings in uranium requirements and target fabrication costs as well as reduces the amount of uranium required per curie of $^{99}$Mo.

## 3. Medical Application of Molybdenum-99

Technetium-99m's (molybdenum-99's daughter) main use comes from medical diagnostic imaging of organ structure and detection of disease using the 140 keV decay gamma from $^{99m}$Tc. This decay gamma is ideally matched to the Anger camera which has worldwide use in nuclear medicine [8]. Images that medical physicians utilize can reveal the blood flow in complex organs such as the brain and also the presence of tumors such as head and neck carcinoma[9]. Technetium-99m's appeal comes not just from its ideal decay gamma energy but also from its parent nuclide's 66 hour half-life which allows for transfer of the radioisotope from production facilities to medical facilities with minimal loss in decay activity.

## 4. Future Isotope Production Pathway

The design of a small inherently safe aqueous homogeneous reactor could prove crucial to continuing the supply of medical radioisotopes. An aqueous homogeneous design of the

Canadian built research reactor SLOWPOKE could help in the development of a small-scale dispersed radioisotope supply. Installation of this reactor in various universities could allow for the distribution of isotope sources across the country while providing a source of income for the school's reactor program.

Neutronic calculations have already begun on a homogeneous SLOWPOKE [10] [11]. Thermal hydraulic calculations are in progress to ensure a pool type aqueous homogeneous reactor will retain the proven safety characteristics of the SLOWPOKE-2. Initial estimates of potential Mo-99 activity that a 20 kWt aqueous SLOWPOKE could generate are around 202 $_{6day}$Ci week$^{-1}$ resulting in an annual revenue from sales of 4.9 M$ year$^{-1}$ [12] (at $470 $_{6day}$Ci$^{-1}$).

## 5. Conclusion

The use of radioisotopes in medical procedures has become a very important part of the medical industry. The upcoming paramount shut down of the largest provider of these isotopes, the NRU, has escalated the importance of the development of alternative sources; evident by the recent supply crises during prolonged outages of the NRU reactor.

Most popularly, the production of isotopes to fuel the radiopharmaceutical industry has been from the fissioning of highly enriched uranium. In an effort to reduce proliferation concerns and to allow for developing countries to produce their own source of radioisotopes, research into the use of low enriched uranium targets is ensuing with promise.

The use of aqueous homogeneous fission reactors could offer further benefits over solid fueled reactors. Studies have shown that less uranium would be required for every curie of molybdenum-99 produced, and that the separation of molybdenum-99 from the uranium fuel could be less complicated [7]. The flexibility in core geometry when designing a homogeneous core allows for more institutes to be able to adopt this design to replace the solid core reactor they may be currently using. AHR's also offers the ability for easy radioisotope production for institutes or companies not already doing so.

The continued study of AHR's to accurately detail the inherent safety characteristics of a liquid system are crucial to establishing this technology in the medical isotope industry. The development of the Canadian SLOWPOKE reactor into a homogeneous system could help augment the supply deficit the shutdown of the NRU will cause when it ends operation in the next decade.

## 6. References

[1] I. A. E. A. IAEA, "Canada: Research Reactor Details," 2011. [Online]. Available: http://www-naweb.iaea.org/napc/physics/research_reactors/database/rr%20data%20base/datasets/report/Canada%20%20Research%20Reactor%20Details%20-%20SLOWPOKE-2,%20RMC.htm. [Accessed 18 11 2011].

[2] KAERI, "42-molybdenum-99," 2000. [Online]. Available: http://atom.kaeri.re.kr/ton/nuc8.html. [Accessed 24 11 2011].

[3] National Research Council of the National Academies, "Medical isotope production without highly enriched uranium," *National Academies Press*, Washington, D.C., 2009.

[4] J. L. Snelgrove, G. L. Hofman, T. C. Wiencek and C. T. V. G. F. Wu, "Development and processing of LEU targets for Mo-99 production -- overview of the ANL program," Argonne National Laboratory, University of Illinois at Urbana-Champaign, Indonesian National Atomic Energy Agency, 1995.

[5] IAEA, "Homogeneous aqueous solution nuclear reactors for the production of Mo-99 and other short lived radioisotopes," IAEA, 2008.

[6] C. Conner, E. F. Lewandowski, J. L. Snelgrove, M. W. Liberatore and D. Walker, "Development of annular targets for 99Mo production," IAEA, Argonne, IL, 1999.

[7] R. Ball, V. Y. Khvostionov and V. A. Pavshook, "Present status of the use of LEU in aqueous reactors to produce Mo-99," in *International Meeting on Reduced Enrichment for Research and Test Reactors,* Sao Paulo, Brazil, 1998.

[8] National Research Council and Institute of Medicine of the National Academies, "Advancing nuclear medicine through innovation," *National Academies Press*, Washington, D.C., 2007.

[9] C. Van de Wiele, C. Lahorte, H. Vermeersch, D. Loose, K. Mervillie, N. D. Steinmetz, J. Vanderheyden, C. A. Cuvelier, G. Slegers and R. Dierck, "Quantitative tumor apoptosis imaging using technetium-99m-HYNIC annexin V single photon emission computed tomography," *American Society of Clinical Oncology*, pp. 3483-3487, 2003.

[10] R. Gagnon, "Safety analysis of a homogeneous SLOWPOKE reactor," Royal Military College of Canada, Kingston, Ontario, Canada, 2009.

[11] P. J. F. Busatta, "Homogeneous SLOWPOKE reactor: feasability study of transforming the SLOWPOKE-2 reactor for homogeneous fuel for the production of commercial radio-isotopes," Royal Military College of Canada, Kingston, Ontario, 2005.

[12] H. Bonin, P. Busatta, R. Gagnon and J. Hilborn, "The homogeneous SLOWPOKE nuclear reactor: a small and convenient nuclear reactor for the production of medical radioisotopes," *Submitted to Nuclear Technology,* 2011.