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Electron accelerator-based production of molybdenum-99: Bremsstrahlung and photoneutron generation from molybdenum *vs.* tungsten

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ABSTRACT

A new "one-stage" approach for production of ^{99}Mo and other radioisotopes by means of an electron linear accelerator is described. It is based on using a molybdenum target both as a bremsstrahlung converter and as a radioisotope producing target for the production of ^{99}Mo via the photoneutron reaction ^{100}Mo(γ, n)^{99}Mo. Bremsstrahlung characteristics, such as bremsstrahlung efficiency, angular distribution, and energy deposition for molybdenum targets were obtained by means of the EGSnrc Monte Carlo simulation code system. As a result of our simulations, it is concluded that a 60 MeV electron beam incident on a thick Mo target will have greater bremsstrahlung efficiency than the same thickness (in units of r_0) W target, for target thickness $z > 1.84r_0$, where r_0 is the electron range. A 50 MeV electron beam incident on a Mo target will result in greater bremsstrahlung efficiency than the same thickness W target (in units of r_0) for target thickness case: $z \geqslant 2.0r_0$. It is shown for the one-stage approach with thicknesses of (1.84–2.0)r_0, that the ^{99}Mo-production bremsstrahlung efficiency of a molybdenum target is greater by ~100% at 30 MeV and by ~70% at 60 MeV compared to the values for tungsten of the same thickness (in units of the appropriate r_0) in the traditional two-stage approach (W converter and separate ^{99}Mo producing target). This advantage of the one-stage approach arises from the fact that the bremsstrahlung produced is attenuated only once from attenuation in the molybdenum converter/target. In the traditional, two-stage approach, the bremsstrahlung generated in the W-converter/target is attenuated both in the converter in the ^{99}Mo-producing molybdenum target.

The photoneutron production yield of molybdenum and tantalum (as a substitute for tungsten) target was calculated by means of the MCNP5 transport code. On the basis of these data, the specific activity for the one-stage approach of three enriched ^{100}Mo-targets of a 2 cm diameter and thicknesses of 1, 2, and 3 radiation lengths (RL) were calculated to be: 19.54 Ci/g, 23.05 Ci/g, and 21.23 Ci/g, respectively. These results were compared with the evaluation presented in by Diamond et al. [19] for the same diameter and thickness ^{100}Mo-targets in a two-stage approach. The comparison demonstrates that for all thicknesses under consideration, the specific activities at equilibrium in the one-stage approach are substantially greater than those evaluated in Diamond et al. [19] for the two-stage approach. More specifically, the specific activities at equilibrium in the Mo converter/target approach are greater than those in the standard W converter/Mo target approach at ^{100}Mo target thicknesses of 1RL, 2RL, and 3RL by 28.9%, 82.5%, and 80.1%, respectively.

1. Introduction

The metastable nuclear isomer of ^{99}Tc, ^{99m}Tc, is the most extensively used radioisotope in nuclear medicine. It is employed in approximately 85% of nuclear medicine procedures numbering more than 40 million procedures annually worldwide [1]. ^{99m}Tc emits low-energy (140.5 keV) easily-detectable γ photons with a half-life of 6 h. This short physical half-life and its short biological half-life (1 day) make it possible to employ diagnostic procedures that minimize the radiation exposure to the patient. It is equally important that the precursor of ^{99m}Tc, ^{99}Mo, has a half-life of 66 h, facilitating its transport and distribution over very long distances.

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Presently, the state-of-the-art technology for producing the radioisotope ^{99}Mo is based on the irradiation of highly enriched, weapons grade (≈93% ^{235}U) uranium targets in dedicated nuclear reactors. This implies that weapons grade uranium from national defense stockpiles (up to tens of kg per year) must be transported within and between countries, endangering non-proliferation nuclear safety in general.

Currently, more than 95% of the world's production of ^{99}Mo employs the thermal neutron fission (n,f) process on ^{235}U. It uses an intense thermal neutron flux of up to 10^{15} n/cm^2/s in a nuclear reactor irradiating highly enriched uranium (HEU) targets ($\sigma_{(n,f)}$ = 586 b) producing ^{99}Mo in 6.161% of all fission events (fission yield). The combination of high thermal neutron flux, high enrichment of uranium targets, high thermal neutron fission cross section and high fission yield of ^{99}Mo results in a ^{99}Mo specific activity of $\geqslant 1.85 \times 10^5$ GBq/g ($\geqslant 5.00 \times 10^3$ Ci/g) [2]. However, this method of production requires very elaborate and very expensive processing facilities. Extreme precautions must be taken (hot cell processing) to avoid contamination of the ^{99}Mo with highly toxic products and transuranics. This results in production costs that are more than four times higher than other methods.

This approach has two more challenges. The first is that all five major producers of ^{99}Mo in the world (NRU reactor in Canada, HFR reactor in Netherlands, BR2 reactor in Belgium, Osiris reactor in France, and SAFARI reactor in South Africa), that combine to produce roughly 90% of the global ^{99}Mo supply, have an average age of more than 50 years. Consequently, these reactors are shut down frequently for unscheduled and time-consuming repairs as well as routine maintenance. All of them are under threat by decommissioning. The second problem is that the US administration now opposes the use of HEU, especially weapons grade uranium, for production of radioisotopes because of the proliferation concerns.

There is no generally accepted scientific and technological strategy to avert this crisis, despite a number of alternative methods having been proposed [3,4]. The most developed of these is based on the neutron capture process, (n,γ), whereby an intense thermal and/or epithermal neutron flux bombards a ^{98}Mo (isotopic abundance 24.13%) target producing the ^{99}Mo radioisotope. The main disadvantages of this approach are that a nuclear reactor would still be required, and a major change in the ^{99}Mo/^{99m}Tc generator would be needed because of the much lower specific activity product and the need to separate ^{98}Mo from ^{99}Mo [3].

Another method is based on the photofission process on ^{238}U, *i.e.* the ^{238}U (γ,f) reaction. This photofission process is based on a high-powered electron linear accelerator (linac) employed to generate high-energy bremsstrahlung photons produced as the electron beam interacts with a high-Z converter target, usually water-cooled tungsten (W). The bremsstrahlung photons are then used to irradiate a natural uranium target placed outside the linac just behind the high-Z converter to produce photofission. In this case, 6.161% of the fission yield will be ^{99}Mo. The main advantages of this approach [3] include using low-cost natural uranium targets, no criticality issues, low-cost waste storage, and use of the existing ^{99}Mo/^{99m}Tc generator technologies. The main disadvantages of this approach include higher waste volume than the existing reactor-based HEU-fission technology, and high operating and capital costs for the linac [3].

Of the non-reactor and non-fission methods for production of ^{99}Mo, the most promising one is the method based on the use the photoneutron process, *i.e.* the (γ,n) reaction, for ^{99}Mo production. [3,5–9]. In this process, the heaviest stable isotope of molybdenum (Mo), ^{100}Mo (isotopic abundance of 9.63%), is irradiated by bremsstrahlung photons from a linac. The reaction occurring is ^{100}Mo(γ,n)^{99}Mo, an endothermic reaction with threshold, E_{th} = 9.1 MeV.

Just as in the previous case of the (γ,f) process, the source of the γ-radiation in the (γ,n) reaction is a linac with an electron energy up to 50 MeV and an electron beam power up to 500 kW irradiating a metal target. The target, that converts the kinetic energy of an accelerated electron beam into bremsstrahlung should be chosen from the high atomic number metals, such as $_{74}$W, $_{73}$Ta, natural, or depleted uranium, in order to maximize bremsstrahlung yield. In this case, the radioisotope ^{99}Mo producing target (^{100}Mo) has to be located in close proximity, downstream of the bremsstrahlung converter. Owing to the considerable self-absorption of the bremsstrahlung in the high-Z body of the bremsstrahlung target, the target must be cooled by distilled water under pressure, increasing the distance between the bremsstrahlung source and the sample to be irradiated (^{100}Mo), thereby decreasing the efficiency of ^{99}Mo production.

There were a number of attempts to calculate, by means of different Monte Carlo codes, the main design parameters of thick bremsstrahlung targets. The most comprehensive work on the subject is that of Berger and Seltzer [10]. That work presented the results of Monte Carlo calculations of electron–photon cascades in thick W and tantalum (Ta) targets bombarded by monoenergetic electrons with kinetic energies between 2 and 60 MeV. As a result of those calculations, the following important information pertaining to the design of the thick W targets have been obtained: (1) the spectra of the bremsstrahlung emitted in various directions, (2) the angular distributions of the emitted bremsstrahlung intensity, (3) the bremsstrahlung efficiency as a function of target thickness and of the electron energy, taking into account self-absorption of the bremsstrahlung in the target, (4) the transmission of primary and secondary electrons through the target, (5) the energy deposition as function of the depth in the target, and (6) the yield of photoneutrons as a function of the incident electron energy and of the target thickness.

Recently, the World Intellectual Property Organization (WIPO) and the United States Patent and Trade Office (USPTO) published a patent application of Tsechanski [11]. This patent proposes to utilize a bremsstrahlung producing Mo target/converter in a linac. The converter and target are combined, thus maximizing production of ^{99}Mo via the ^{100}Mo(γ,n)^{99}Mo. In addition, the use of Mo directly as a bremsstrahlung target (converter) makes it possible to utilize the neutrons produced by the reactions like (γ,n), (γ,$2n$), (γ,pn), and so on for complementary production of ^{99}Mo via the (n,γ) reaction on the isotope ^{98}Mo (isotopic abundance of 24.13%). Therefore, ^{99}Mo will be produced simultaneously from two (of seven) stable isotopes of Mo: both from ^{100}Mo via the (γ, n) reaction and from ^{98}Mo via the (n,γ) reaction. To maximize the second channel for the ^{99}Mo production via the (n,γ) reaction, the neutrons from the first (neutron producing) channel should be slowed down to the epithermal/thermal energy interval. For this purpose, distilled water, intended primarily for cooling down of the target assembly of the linac, can be used for thermalizing neutrons as well.

According to this proposal, production and accumulation of the radioisotope ^{99}Mo has been carried out in the Mo-target/converter itself located in the target assembly inside the linac. Therefore, intense fluxes of the high energy bremsstrahlung photons and neutrons (many MeV's energy range) will be found around the target assembly outside the linac. These high energy bremsstrahlung photons can be used to produce some other very important radioisotopes via the (γ,n) reaction on the appropriate target materials placed outside the linac target assembly and adjacent to it. For example, an external target of the lightest stable isotope of xenon, ^{124}Xe, used in this way would make it possible to produce additionally two other very important radioisotopes: ^{123}I via the (γ,n)