Production of Mo-99 Using 30 MeV Electrons and a Mo-100 Target

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INTRODUCTION

The sources of Mo-99 in the United States are primarily from the Canadian National Research Universal (NRU) reactor. Recent interruption in the reactor operation caused a shortage in the Mo-99 isotope which led to a shortage in Tc-99m, the most commonly used medical radioisotope [1]. The shortage in the Mo-99 supply prompted a search for alternative, domestic production methods [2]. One of the methods considered here is the use of the (gamma,n) reaction on a Mo-100 target. The advantages of this reaction are the relatively high reaction cross section and the elimination of the expensive fission products chemistry stream associated with irradiation of uranium. The (gamma,n) method for production of Mo-99 is not new and was described before in reference [3] and patented [4]. The purpose of this work was to compare experiments and calculations to quantify the accuracy of the Monte Carlo transport code MCNP 5.14 [5] and the current photoneutron cross section libraries. The results of this work provide an estimate of the expected Mo-99 yield and a guideline to selection of electron beam current and energy.

CALCULATIONS

Preliminary calculations of the yield of Mo-99 were done in order to find the best geometry for the experiment. Because the production is due to (gamma,n) reactions with Mo-100, two configurations were considered; electrons hitting a Mo target directly and electrons hitting a W converter placed in front of a Mo target. The converter has a higher Z and density and is thus expected to produce more Bremsstrahlung and increase the production of Mo-99. The target was a 0.4 cm radius by 3.49 cm long cylinder of Mo-100 made out of 22 disks each with a height of 0.159 cm. The calculations were done using the MCNP5 code starting with electrons at 30 MeV hitting the first Mo plate or a W converter placed in front of the first Mo plate. Calculations used the photoneutron production code library that comes with the MCNP 5.14 [5] code. The photoneutron cross sections in MCNP are based on a KAERI evaluation from 2000 [6]. The calculated quantity was the photon flux convoluted with the (gamma,n) cross section averaged in each disk. This data was then converted to activity A_t (in Ci) in disk *i* after an irradiation time *t* by:

$$A_i = \frac{n_e}{3.7\times 10^{10}} N \left\langle \int \phi(E)\sigma(E)dE \right\rangle_i \left(1-\exp(-\lambda\cdot t)\right) \quad (1)$$

Where n_e is the intensity of the incident electron beam [e/s], *N* is the number of atoms in each Mo-100 disk, λ is the decay constant of Mo-99 ($t_{1/2}$=2.75 d) and the integral quantity is the tally calculated by MCNP. The calculations indicate that for this geometry the addition of a 0.1 cm thick W converter increased the yield by about 3% which was near optimum.

Calculations with different electron energies indicate that the production yield of Mo-99 is proportional to the electron beam power (the product of the energy and beam current).

EXPERIMENTS

Experiments were done in order to verify the production yield for electron energy of 30 MeV, because this energy represented a break point in the cost of electron linear accelerators (LINACs). The experiment was not done in an optimum compact geometry suitable for chemical processing but rather was done to enable a simple comparison of experiments and calculations. The two configurations mentioned before were used in the experiment. The Mo sample was assembled from 16 plates of 2.54 cm x 2.54 cm x 0.159 cm of elemental Mo. For the indirect configuration a 0.159 cm thick W plate was added to the stack of Mo plates. For the two experiments the Rensselaer Polytechnic Institute LINAC was operated with electron beam energy of 27 MeV and electron beam current of 5 µA. The irradiation time was 15 minutes. Following the irradiations the plates were left to decay for 3.5 hours. Each Mo plate was placed at a distance of 100 cm from a calibrated HPGe gamma detector and was counted for 300 seconds. The spectrum was analyzed for the 181.4 keV and 740 keV Mo-99 gamma lines and the data was corrected for decay, detection efficiency and self absorption in the Mo plates such the activity at the end of irradiation was obtained. Calculations of the same geometry were done with MCNP using a 27 MeV electron energy and were scaled to the experimental electron beam current; the results are plotted in Fig. 1. The error in the detector calibration was about

5%; adding errors due to measurements of the electron beam current and energy resulted in an overall estimated error of 25%. There was a disagreement of up to 30% in the experimental activity calculated from the two different gamma lines as shown in Fig 1. In both cases the calculations agree better with the activity obtained from the 740 keV gamma line. It is possible that this disagreement is due to background under the peak which was as high as 25%-42% of the peak area for the 181.4 keV line vs. 3-4% for the 740 keV line.

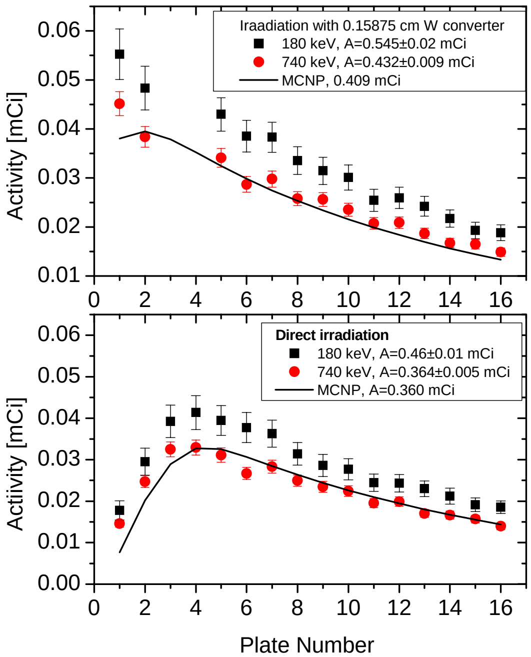

Fig. 1. Measured and calculate Mo-99 activity at the end of irradiation. The top plot is for the configuration with a W converter and the bottom for electrons incident directly on the first elemental Mo plate. The error bars represent counting statistics error and a 5% error due to the detector efficiency calibration.

CONCLUSIONS

Measurements and MCNP calculations of Mo-99 production using an elemental Mo target are in agreement within the experimental error. The data for the 740 keV Mo-99 gamma line is in good agreement with the calculations for both direct irradiation and the use of a W converter plate. For the experimental geometry the calculation indicates a 14% increase in the yield when a W converter was used compared to the experimental value of 19%. One of the reasons for this discrepancy could be the variation in the LINAC beam power between the two different experiments; better normalization of the beam power using an activation foil could solve this problem in future experiments.

Additional calculations for the distribution of the specific activity using a 30 MeV pencil beam of electrons incident on a 0.1 cm W converter indicate that 90% of the specific activity is produced in the first 2.2 cm of the target at a radius of 0.2 cm around the beam axis. For 24 hours irradiation of a Mo-100 target a maximum activity of 1.8 Ci/kW was calculated. Thus a high power accelerator capable of delivering 30 kW can produce 54 Ci (~4.8 Ci/g of Mo-99) every 24 hours. Assuming a target with a radius of 0.5 cm and length of 2.5 cm, a beam energy of 30 kW results in a high average power density of 15 kW/cm^3, much higher than any nuclear power reactor [7]. The calculated specific activity (~4.8 Ci/g) is in the middle of the range (1-10 Ci/g) given in reference [4]. The total activity (54 Ci) is in agreement with the value of 24.6 Ci calculated in reference [3] using 0.46 of the power used in calculation above (scaling our result to this power gives 24.8 Ci).

If such facility will irradiate a pure Mo-100 target for 3.5 days, followed by 12 hours chemical processing, then during one week two targets can be processed resulting in 27 six day Ci. To produce 3000 six day Ci as suggested by a recent DOE solicitation [8] about 55 LINAC beams will be needed. With current high power LINACs one can consider a raster beam and ~120 kW beam power which results in a requirement for 14 LINACs.

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