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STUDY OF TARGETS TO PRODUCE MOLYBDENUM-99 USING 30 MeV ELECTRON LINEAR ACCELERATOR*

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Abstract

In this paper, two approaches to produce ^{99}Mo are studied using GEANT4 Monte-Carlo particle simulation software. In the first approach, converter target, bremsstrahlung photons are generated in a high Z target. The emitted photons hit ^{100}Mo, secondary target, producing ^{99}Mo through (γ, n) reaction. In the second approach, direct target, high energy electron beam hits ^{100}Mo target, where both (e, γ) and (γ, n) reactions take place simultaneously. A 30 MeV, 5-10 kW beam power electron linac is under development at Society for Applied Microwave Electronics Engineering and Research (SAMEER). The acceleration gradient required to achieve 30 MeV energy will be provided by two electron linear accelerators operated in series configuration and the high average beam power will be achieved by running the system at high duty operation. Main aim of this study is to optimize experimental parameters in order to maximize specific activity of ^{99}Mo. Since, ^{100}Mo is very expensive material therefore judicious use of the material is very important. Hence, optimization of electron beam energy and target dimensions are studied in detail in both the approaches. It is found that direct approach gives higher specific activity compared to the converter approach.

INTRODUCTION

^{99m}Tc is a widely used radioactive tracer isotope in nuclear medicine due to its short half-life. The parent radio nuclei of ^{99m}Tc, ^{99}Mo, has a half-life of 65.976 hours long enough to facilitate transportation to faraway locations. As of now, the production of ^{99}Mo depends mainly on reactor-based technology using Highly Enriched Uranium (HEU) target. But many of the big reactor facilities are either closed down or near their end of operation in coming few years [1].

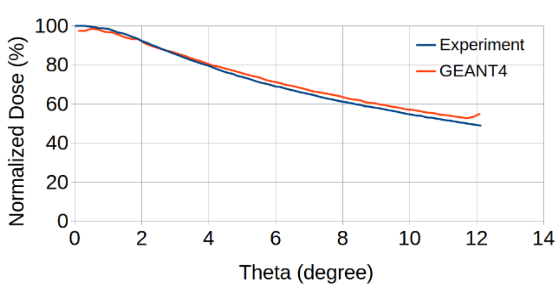

Figure 1: Comparison of 6 MeV linac experimental data with GEANT4 output.

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Therefore, a lot of efforts are being made worldwide to produce ^{99}Mo using alternate technologies [2-4]. Use of accelerator is proposed as one of the most effective future ideas for producing ^{99}Mo by means of photo-neutron conversion of ^{100}Mo. In this paper, we will be optimizing electron beam energy and target design to produce ^{99}Mo through ^{100}Mo (γ, n) ^{99}Mo reaction. Before we start with the optimization, we shall validate GEANT4 code which is used to obtain the final results.

GEANT4 OUTPUT VALIDATION

Validation of GEANT4 photon output is done by comparing it with the experimental data from SAMEER's 6 MeV linac.

In the experimental set up, photons coming out of the linac are collimated in a half cone angle of 12° in the forward direction and the dose measurements are done on Radiation Field Analyzer (RFA) placed at 1 m from target location. Target assembly consists of a 6 mm diameter circular pellet of tungsten with 1.5 mm thickness. It is backed by copper of 2.5 mm thickness. The beam diameter measured experimentally using indirect approach is ~3 mm. In GEANT4 simulations, a circular electron beam of 1.5 mm radius and 0.1 mm standard deviation with gaussian profile of 6 MeV energy and 0.5 MeV sigma is made to strike the tungsten target with copper backing, dimensions same as the experimental set up. Normalized dose output from GEANT4 is compared with the RFA data in Fig. 1. The plot shows maximum of 3% difference between experiment and GEANT4 output.

The validation of neutron output from GEANT4 is done by comparing standard neutron number data [5]. When an electron beam of 34.3 MeV hits Molybdenum target of 1 radiation length (r.l.), 12×10^{-4} neutrons per electron (n/e) are generated and neutron number simulated from GEANT4 gives a value of 12.28×10^{-4} n/e, which is comparable with the published results.

BEAM ENERGY OPTIMIZATION

When a high energy electron beam hits a high Z material it produces bremsstrahlung photons that can be used to trigger ^{100}Mo (γ, n) ^{99}Mo reaction. An optimum electron beam energy that gives maximum photons under the Giant Dipole Resonance (GDR) curve for ^{100}Mo (γ, n) ^{99}Mo reaction is determined using flux weighted average cross section (σ_a) [6]. Using Eq. (1), σ_a is calculated for various electron beam energies and plotted in Fig. 2.

$$\sigma_a = \frac{\sum[\Phi(E)\sigma_{GDR}(E)]}{\sum\Phi(E_{E_{th}>8.3MeV})} \quad (1)$$

It is observed that when electron beam energy increases from 15 to 70 MeV, total number of photons and the end

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point energy of bremsstrahlung increases. But the increase in the photons under the peak of GDR curve (12-17 MeV) is not appreciable hence σ_a decreases after 20-30 MeV.

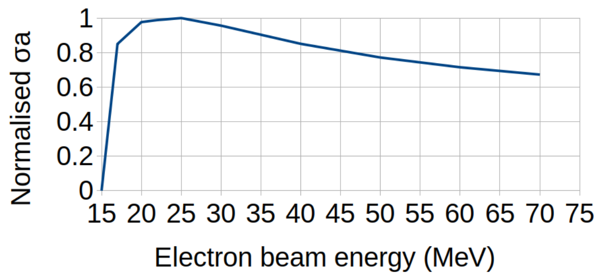

Figure 2: Flux weighted average reaction cross-section (σ_a) as a function of electron beam energy.

Threshold photon energy (E_{th}) required for ^{100}Mo (γ, 1n) ^{99}Mo reaction is 8.3 MeV and the peak of reaction cross section occur between 12-17 MeV. Whereas, the E_{th} for (γ, 2n) reaction is between 14-15 MeV. Therefore, the bremsstrahlung photons beyond 15 MeV of energy have some probability of producing ^{98}Mo by (γ, 2n) reaction. After comparing these cross sections, it is found that for 30 MeV electron beam, about 55% of the total number of neutrons are generated from (γ, 1n) whereas the rest of the neutrons are generated from (γ, 2n) reaction, hence not contributing to ^{99}Mo production [7]. Therefore, Activity of ^{99}Mo using neutron data is calculated as Eq. (2).

$$A = 0.55 \times N_{Mo} \tag{2}$$

Where, N_{Mo} is the number neutrons coming out of ^{100}Mo.

PARAMETERS OF LINAC SYSTEM

Electron linac with 15 MeV energy and 80 µA average beam current is successfully tested as a prototype development at SAMEER [8]. The acceleration gradient required to achieve 30 MeV energy can be provided by two linacs operated in series configuration. The main parameters of the system are given in Table 1.

Table 1: Linac Parameters

Parameters, Unit	Value
Energy, MeV	30
Average current, µA	350
Duty cycle, %	0.514
Peak current, mA	68
Beam power, kW	10.5
Repetition rate, HZ	400

IRRADIATION APPROACHES AND TARGET OPTIMIZATION

Two types of irradiation approaches, converter and direct irradiation, are studied in this paper.

Converter Target Irradiation Approach

In this approach, a tungsten target is placed before ^{100}Mo target to convert electron beam into bremsstrahlung photons to carry out ^{100}Mo (γ, n)^{99}Mo reaction.

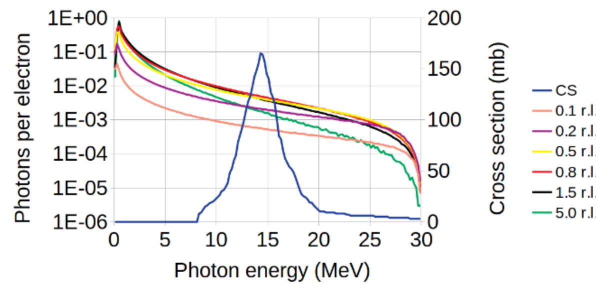

Figure 3: Bremsstrahlung photons for various tungsten thicknesses. The GDR curve (in blue) is associated with the Y-axis on right hand side showing reaction cross-section for ^{100}Mo (γ, n) ^{99}Mo reaction.

Thickness Optimization of Tungsten Figure 3 explains the behaviour of bremsstrahlung photons depending upon the thickness of tungsten target. For very thin targets many electrons come out of the tungsten without interacting completely thus producing less photons. As target thickness increases, number of photons also increases but it saturates after a few r.l. of thickness. This happens due to the self-absorption of photons inside the target. An optimized thickness of converter target is the one where there is maximum electron interaction and minimum self-absorption which is 0.8 r.l. in our case.

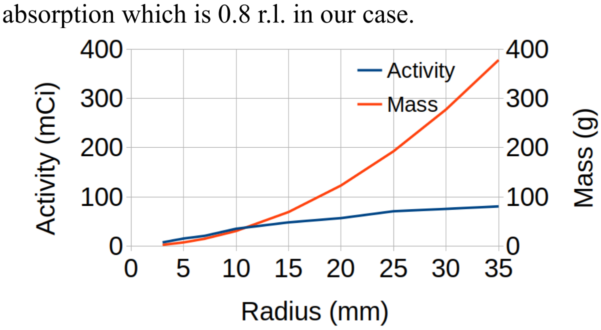

Figure 4: Comparison of increase in Mass and Activity as a function of radius of ^{100}Mo.

Radius Optimization of ^{100}Mo In converter target irradiation approach, (e, γ) reaction takes places in tungsten which is followed by a secondary ^{100}Mo target. The secondary target utilizes the photons coming from tungsten to produce ^{99}Mo through (γ, n) reaction. Simulations are done to observe the change in activity of ^{99}Mo with increasing radius of the secondary, ^{100}Mo, target. From Fig. 4 it can be seen that after a radius of 10 mm, increase in the activity saturates whereas the mass of ^{100}Mo keeps

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on increasing. Therefore, keeping the radius of ^{100}Mo as 10 mm will lead to its efficient use.

Direct Target Irradiation Approach

In this approach, an electron beam strikes ^{100}Mo target directly. Both (e, γ) and (γ, n) reactions take place in the same ^{100}Mo target.

Radius Optimization of ^{100}Mo In direct target irradiation approach, radius of cylindrical ^{100}Mo is changed from 1 mm up to 10 mm and it is found that beyond 4 mm of radius, increase in mass is 125%, 77% and 56% per 2 mm increase in radius, whereas the increase in activity is just 8%, 7% and 2% respectively. Angular distribution of photons suggests that most of the photons from 30 MeV electron beam are generated in forward direction. Therefore, the radius of cylindrical ^{100}Mo can be reduced to 4 mm in order to reduce overall mass without drastically affecting the activity.

Geometry Optimization The forward peaking bremsstrahlung pattern also suggests that an ellipsoidal or conical/frustum of a cone geometry will be better for the complete utilization of ^{100}Mo target. To compare these target geometries, viz. cylindrical, ellipsoidal and conical/ frustum of a cone, the specific activity of ^{99}Mo obtained from these targets is studied. Fig. 5 shows the change in specific activity for various shapes when 30 MeV electron beam hits their flat surface. The overall comparison shows that 1-1.2 r.l. thick conical target with front radius 4 mm gives the highest specific activity and hence utilizes ^{100}Mo with the best efficiency.

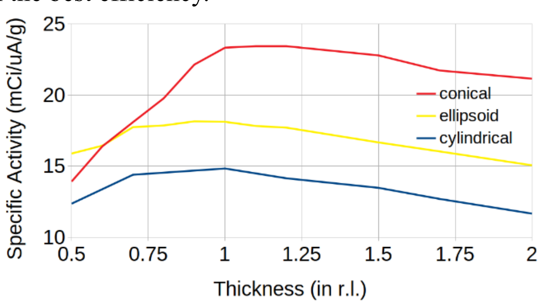

Figure 5: Comparison of specific activity for cylindrical, ellipsoidal and conical/ frustum of cone target geometry.

COMPARISON OF IRRADIATION APPROACHES

Activity obtained through direct target and converter target approach is compared in Fig. 6 to efficient ^{99}Mo production. The comparison shows that the activity from direct target is consistently more than the converter target irradiation approach.

RESULT

Activity calculation for converter and direct target approaches with various geometries is done using the simulation code GEANT4. For our system namely 30 MeV electron linac with average beam power of 5-10 kW and ^{100}Mo target, maximum activity estimated by converter target approach is 0.161-0.322 Ci/g. Whereas, in the case of direct target approach with conical target geometry maximum estimated activity is 3.904-7.808 Ci/g.

DISCUSSION

Geometry of direct target approach facilitates production and complete utilization of photons in all 4 π direction, which increases the overall activity, but cooling the molybdenum target becomes very crucial in this approach. Since the same irradiated target is used for radiochemistry later on therefore, designing the mounting of ^{100}Mo target for irradiation is very challenging. In converter target approach the entire beam energy is lost due to absorption in tungsten target therefore a cooling mechanism is provided only to tungsten. The variation in the activity values is large in most of the references either in converter target approach or direct target approach. There is no experimental data available to verify the final activity obtained after irradiation. Therefore, a good linac experimental setup to quantify the activity with any beam parameters is important.

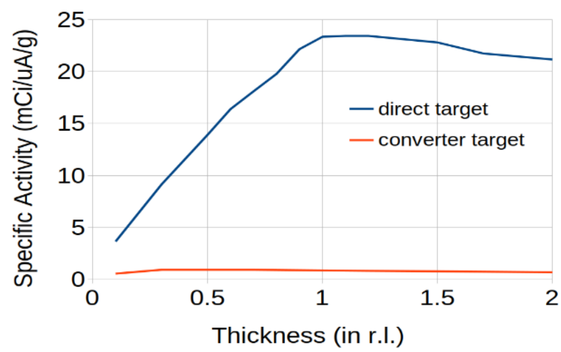

Figure 6: Comparison of ^{99}Mo activity obtained from direct and converter target irradiation approaches.

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