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Target optimization for the photonuclear production of radioisotopes

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HIGHLIGHTS

- We have simulated distribution of specific activity (SA) of ^{67}Cu produced via ^{68}Zn(γ,p)^{67}Cu reaction.
- Among different target shapes we studied the semi-ellipse had the highest SA.
- Cylindrical geometry with radius to height ratio of 0.2–0.25 was found to be optimum.
- Power deposition into the target and target heating for different target shapes was addressed.

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ABSTRACT

In this paper we discuss the optimum shape of a target for photonuclear production of radioisotopes using an electron linear accelerator. Different target geometries such as right cylinder, conical frustum, Gaussian volume of revolution and semi-ellipsoid have been considered for the production of ^{67}Cu via ^{68}Zn(γ,p)^{67}Cu photonuclear reaction. The specific activity (SA) of ^{67}Cu was simulated for each target shape. Optimum ratio of radius to height for cylindrical targets was found to be between 0.2 and 0.25 for target masses ranging from 20 g to 100 g. It was shown that while some unconventional target shapes, such as semi-elliptical volume of revolution, result in slightly higher specific activities than cylindrical targets, the advantage is not significant and is outweighed by the complexity of the target production and handling. Power deposition into the target was modeled and the trade-off between the maximization of ^{67}Cu yield and the minimization of target heating has been discussed. The ^{67}Cu case can easily be extended for production of many other isotopes.

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1. Introduction

Novel methods to produce radioisotopes at lower costs than conventional reactor-based technology could be a great alternative to satisfy growing demand for medical and industrial isotopes. The idea of using electron linear accelerators (LINACs) for production of many high-demand isotopes such as ^{67}Cu, ^{99}Mo, and ^{225}Ac is not new and was described before (Ayzatskiy et al., 2006; Bennett et al., 1999; Maslov et al., 2006; Danon et al., 2008, 2010; Starovoitova et al., 2014) and patented (Lidsky and Lanza, 1998). Electron LINACs operate at much lower costs than nuclear reactors and produce far smaller waste streams; however, historically, LINACs have not been widely used for isotope production. Recently the interest in domestic, accelerator-based isotope production methods has heightened (NSAC, 2009; US-DOE, 2014). To increase the isotope production rate using electron LINACs both mass and geometry of the target need to be optimized. High surface area for efficient heat removal is also an important criterion which we will address later in the paper.

2. Physics of bremsstrahlung photons and photonuclear production

An essential component of photonuclear production of radioisotopes with electron LINACs is a bremsstrahlung converter, which transforms electron's energy into photons. Bremsstrahlung photons form a forward cone and are incident on the sample, typically placed after the converter. The bremsstrahlung interaction produces a continuous spectrum of photon energies, ranging from zero up to the energy of the incident electron beam. The photon flux decreases monotonically with increasing photon energy and drops down to zero at the maximum photon energy. Due to the strong dependence of the conversion efficiency on the atomic

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number of the material, converters are typically made of high Z metals, such as tantalum or tungsten, which also have excellent engineering properties.

The conversion efficiency between electrons and bremsstrahlung photons is usually defined as a fraction of the kinetic energy of the incident electrons converted into photons' energy and depends on the electron energy, the converter material and converter thickness. Because the bremsstrahlung both produced and attenuated by the converter, for any given converter material and electron beam energy, an optimum converter thickness exists, whereby the bremsstrahlung efficiency reaches a maximum value. This optimum converter thickness corresponds approximately to the 0.3–0.5 of the electron range in the material (Berger and Seltzer, 1970). For example, the optimum thickness for tungsten converter for 40 MeV electrons is about 2 mm and the mean electron range is about 7 mm.

During the first stage of photonuclear reaction a bremsstrahlung photon is absorbed, and a nucleus becomes excited. During the second stage, the excitation energy is released – usually in the form of a photon, or a neutron thus producing a daughter nucleus. The yield of the radioisotope $\boldsymbol{A}$ depends on several parameters. Such parameters include the number of target nuclides that are irradiated $\boldsymbol{N}$, the threshold energy of the nuclear reaction $\boldsymbol{E_{threshold}}$, the maximum energy of photons $\boldsymbol{E_{max}}$, the photon flux density $\boldsymbol{\varphi(E)}$, the cross-section of the photonuclear reaction $\boldsymbol{\sigma(E)}$, the decay constant of the daughter nuclide $\boldsymbol{\lambda}$ and irradiation and cooling time intervals $\boldsymbol{t_i}$ and $\boldsymbol{t_c}$ (Eq. 1):

$$A(t_i) = N\cdot \int_{E_{th}}^{E_{max}} \varphi(E)\cdot\sigma(E)\, dE\cdot(1 - e^{-\lambda\cdot t_i})\cdot e^{-\lambda\cdot t_c} \quad (1)$$

While yield dependence on the sample mass, time intervals of irradiation, and cooling is straightforward, the photon flux (weighted by the cross-section) is a much more interesting parameter, defined by the geometry and material of both the converter and the target.

3. ^{67}Cu distribution in the Zn target

Spacial distribution of the photon flux density is not trivial. Fig. 1 shows a photon flux distribution, created by a 40 MeV electron beam incident on a 2 mm thick tungsten converter. The photon flux intensity decreases rapidly as the distance from the converter increases. At the same time, the radial flux density profile changes. Right next to the converter the gradient of flux density is high as it drops off very fast, but as the distances from the converter increases the profile flattens out. Variations in photon flux density result in variations in ^{67}Cu activity density within the target. To investigate distribution of ^{67}Cu generated via ^{68}Zn(γ,p)^{67}Cu reaction in the Zn target, Monte-Carlo simulations were performed to model the photon flux weighted by the photonuclear cross-section.

A 40 MeV electron beam with Gaussian shape (FWHM = 1 mm) was assumed. Such electron beam parameters are commonly used at the 10 kW, 48 MeV electron LINAC located at the Idaho Accelerator Center. An “infinitely large” right cylindrical natural zinc target (ρ = 7.14 g/cm^3) with ^{68}Zn abundance being 18.8% was used for the simulations. The length of the target was assumed to be 10 cm and the radius to be 5 cm, which clearly exceeds the optimum size of the target. A tungsten converter optimized for 40 MeV electron beam (2 mm thick) was separated from the target by a 1 cm air gap to accommodate for converter and target holder (see Fig. 2). Note that for different target material different the optimum converter thickness might vary so it needs to be optimized for every photonuclear production route.

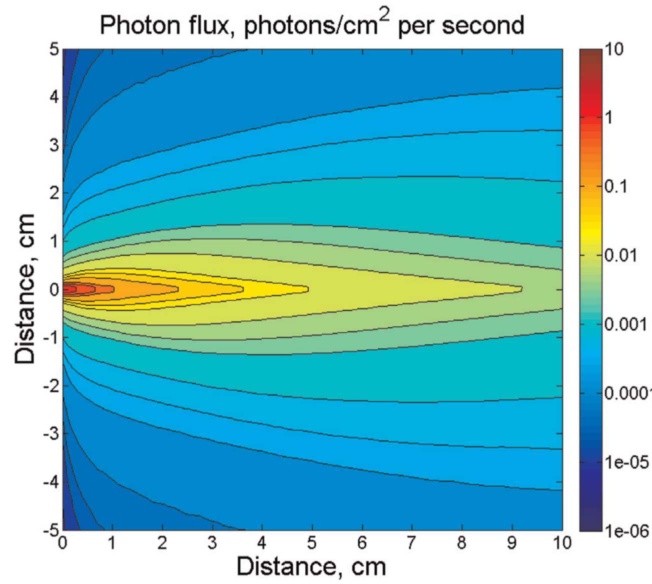

Fig. 1. Photon flux produced by a 40 MeV 25 μA electron beam incident on a 2 mm thick tungsten converter.

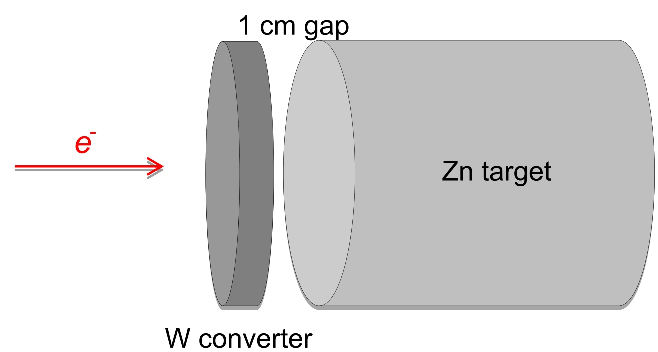

Fig. 2. Simulation setup.

The calculations were performed using MCNPX software (Pelowitz, 2008), which utilizes the Bethe–Heitler Born approximation to sample bremsstrahlung photons (Koch and Motz, 1959). Photon flux was calculated by the standard MCNPX volume-averaged flux estimate (f4 tally) for 40 MeV electron energy beam. MCNPX track-averaged cylindrical mesh tally (type 1) followed by keyword MFACT was utilized to visualize the conversion rate distribution. Cylindrical mesh voxels had different volumes as their inner and outer radii were ranging from 0 to 5 cm in 0.1 mm increments (keeping the arch thickness to be 0.1 mm). The length of each voxel was 0.1 mm and azimuthal angle was 2π. The reaction cross-section for the simulations was adopted from the IAEA Handbook on Photonuclear Data (IAEA, 2000). Note that this cross-section has not been measured experimentally and only modeled function exists, which poses high uncertainty in the ^{67}Cu specific activity calculations. Energy multiplier card was used to weight the photon flux by the ^{68}Zn(γ,p)^{67}Cu cross-section.

Fig. 3 shows the resulting distribution of ^{67}Cu specific activity (SA) in the target and a forward peak is clearly seen. The irradiation time for all the calculations was assumed to be 1 h. The highest SA (at the point where beam hits the target) reached 170 MBq/g; however it drops off very quickly to few MBq/g and even less. Integrating over different volumes, the average yield and specific activity of ^{67}Cu were found for different target shapes and sizes. In particular, to compare our results with similar simulations done for 40 MeV electron beam hitting 4 mm thick tungsten