Comparison of calculated and measured yields of medical isotopes produced by electron bremsstrahlung

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Abstract

Certain medical isotopes can be produced with high radionuclide purity via (γ, n) and (γ, p) reactions using bremsstrahlung photons at electron accelerators such as microtron MT25, for example, or linac of the new IREN facility at the Joint Institute for Nuclear Research. We perform the MCNP modeling of the energy spectra, angular distribution and fluxes of bremsstrahlung photons from a W target and subsequent calculations of yields of isotopes of interest. We find an agreement between the calculated and published yields of ^{99}Mo, ^{236}Pu, ^{237}U within a factor of ~ 1.5. Specifics for medical isotopes production at the IREN electron linac is surveyed in conclusion.

Introduction

Since all major hospitals use radioisotopes for diagnosis, biochemical analysis and radiotherapy, the demand for them is rapidly increasing. Most of these isotopes are produced as activation products in nuclear reactors, some – in cyclotrons. Electron accelerators, however, provide an alternative possibility for producing radioisotopes, especially attractive in view of certain nuclear medicine regulations governing radiopharmaceutical quality. Electron accelerators produce high-energy bremsstahlung photons that can induce photoneutron reactions, and linacs like that of IREN, with electron energy variable up to about 50 MeV, promise a higher radioisotope purity which in turn limits human radiation exposure.

The yields of radioisotopes are usually obtained empirically. Even though comprehensive photonuclear data exists, yield calculations are rarely performed because they require a good knowledge of bremsstrahlung yields and spectra. The Bethe-Heitler theory and Schiff's analytical approximation of the photon spectrum were widely used in thin target calculations as reviewed by Koch and Motz [1]. A bremsstrahlung target is defined as thin if the multiple electron scattering and energy losses have negligible effects on the produced photon spectrum, less than 0.01mm. At the other extreme, highest bremsstrahlung production have been obtained for targets such as tungsten of several mm. The theoretical description of charged-particle processes in thick targets has a long, a half-century history with perhaps the most recent by Bielajew [2]. The developed theoretical approaches and models were implemented in several Monte Carlo codes for simulatting electron scattering and bremsstrahlung in thick targets. The codes EGS4, GEANT and PENELOPE have been used recently by Faddegon et al. [3] for calculating photon spectra and yields and comparing them with measurements on thick Be, Al and Pb targets in the electron energy range from 10 to 30 MeV. The calculations of the bremsstrahlung characteristics were found to be in a $\simeq 10\%$ agreement with measurements.

Our goal has been to model the bremsstrahlung energy spectra, angular distribution and fluxes from a tungsten target and to subsequently calculate the yields of several radioisotopes for which there are existing measured data in order to validate the MCNP (Monte Carlo N-Particle) transport code [5] for such application.

Calculating activity

We calculate the activity A (the number of decays per second) induced by a beam of photons of flux density $\Phi(E, E_0)$ after the irradiation time t_{irr} by using a standard expression from activation analysis:

$$A = \lambda N(t_{irr}) = N_T(1 - \exp - \lambda t_{irr}) \int_{E_{th}}^{E_0} \sigma(E)\Phi(E, E_0)dE. \tag{1}$$

Here N_T is the total number of nuclei of a chosen isotope in the target under irradiation, λ is the decay constant for the daughter nuclei $N(t_{irr})$, $\sigma(E)$ is the cross section for the particular photoreaction (with threshold E_{th}) at the photon energy E, while $\Phi(E, E_0)dE$ is the photon flux density in the energy interval between E and $E + dE$, produced by the electron beam with the end-point energy E_0. Assuming that the electron beam irradiates the whole area S of the target, introducing the electron intensity (the number of electrons per second) $I_e(E_0)$ and converting from electrons to photons we write

$$\Phi(E, E_0)dE = \frac{I_e(E_0)}{S} n_B(E, E_0)dE \tag{2}$$

and correspondly

$$A = \lambda N(t_{irr}) = N_T \frac{I_e(E_0)}{S}(1 - \exp - \lambda t_{irr}) \int_{E_{th}}^{E_0} \sigma(E) n_B(E, E_0)dE, \tag{3}$$

where $I_e(E_0)$ is the number of electrons crossing the area S per second and $n_B(E, E_0)$ is the number of photons per unit energy interval at the energy E produced by one electron with the energy E_0 in a particular bremsstrahlung target B.

The experimental cross sections $\sigma(E)$ can be found in major photonuclear data bases [4] while the differential photon spectrum $n_B(E, E_0)$ can be calculated directly by the MCNP code [5] for a desired target B thickness.

Results

We have performed the MCNP modeling of bremsstrahlung for a set of tungsten targets subject to a parallel beam of monochromatic electrons with energies E_0 ranging from 20

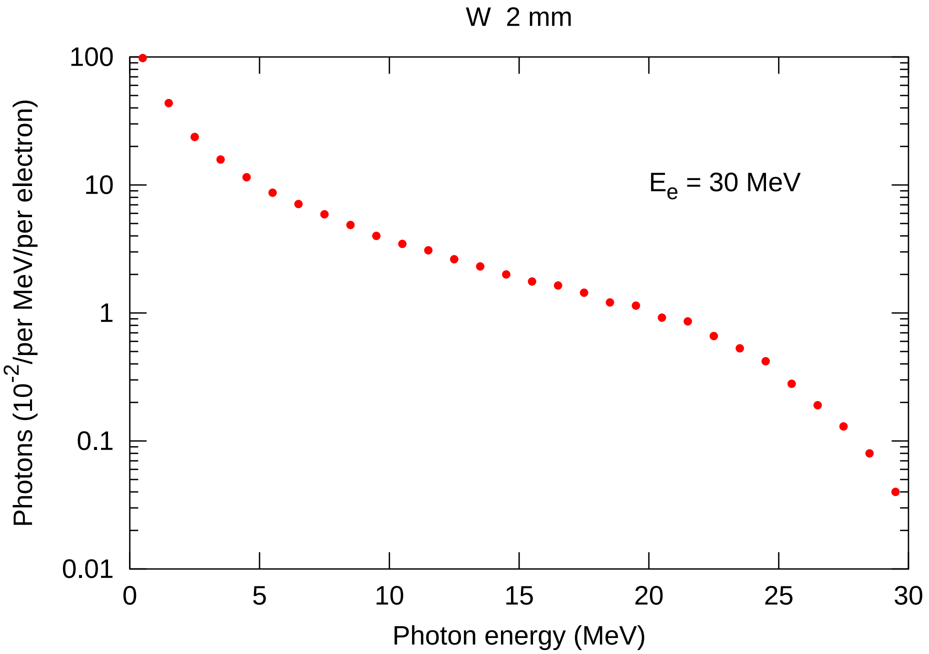

Figure 1: The bremsstrahlung photon energy spectrum. This is the quantity $n_B(E, E_0)$ calculated by the MCNP code for the 30-MeV electrons incident on the 2-mm tungsten target.

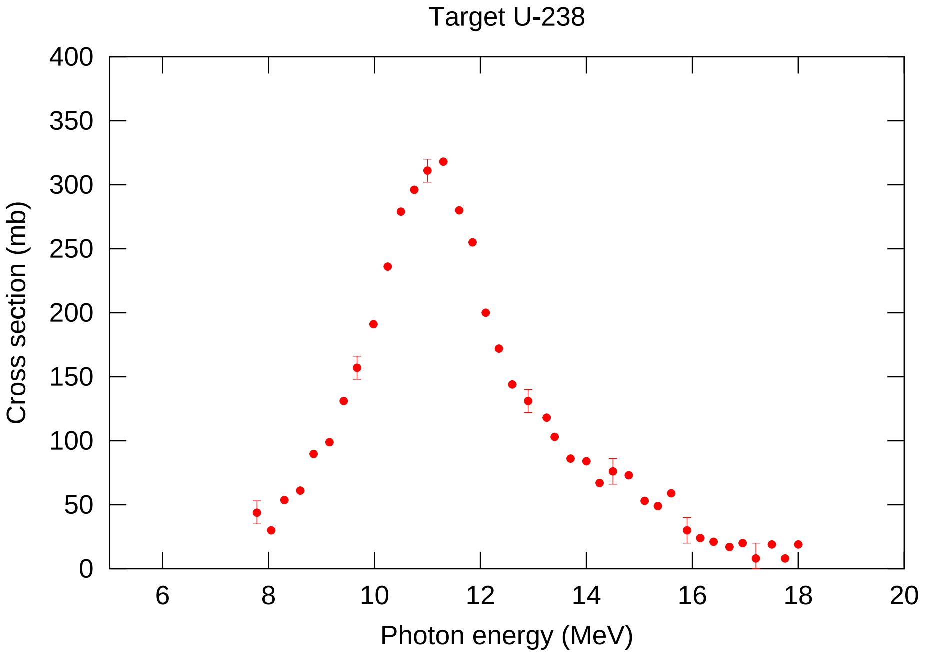

Figure 2: The cross section of the (γ, n) reaction for the ^{238}U isotope target.

to 80 MeV. Targets were of 2-cm diameter disks 0.2 to 4.0 mm thick. The number of photons per incident electron was sampled in 1-MeV width photon energy bins inside several $\cos\theta$ intervals corresponding to the photon exit angle θ between 0° to 90° with the respect to the incident electron direction. With a 2-mm target, the angular accumulated yield of photons with $E > 8$ MeV was found to be 85% up to 30° and 92% up to 45°. We therefore present the differential energy spectrum summed over angles from 0°to 45° as a MCNP-calculated quantity $n_B(E, E_0)$ shown for example in Fig. 1 for the 2-mm thick tungsten target with $E_0 = 30$ MeV. It is useful to compare photon yields from various targets with different electron energies E_0, in which case we should use the effective photon yield $I_B^{ef}(E_0)$ integrated over E from $E_{th} = 8$ Mev to $E_{max} = 20$ MeV:

$$I_B^{ef}(E_0) = \int_8^{20} n_B(E, E_0) dE. \tag{4}$$

The 8−20-MeV photon window is the energy range where the Giant Diplole Resonance (GDR) for photonuclear cross sections $\sigma(E)$ is concentrated for most of middle and heavy mass nuclei. We illustrate this by example in Fig. 2 for the ^{238}U isotope target with only the cross section for the (γ, n) reaction (the $(\gamma, 2n)$ and photofission channels are not included). Because this GDR strength concentration, the integral photon yield below ∼ 8 MeV, which is about one order of magnitude higher than the yield $I_B^{ef}(E_0)$, does not contribute to the isotope production.
The next two figures, Fig. 3 and Fig. 4, show a saturation of the effective yield $I_B^{ef}(E_0)$ within several millimeters of the tungsten target and how the saturated value of $I_B^{ef}(E_0)$ depends on the electron energy E_0 for a monochromatic electron beam.

Producing radioisotopes with lifetimes on the order of several days (using irradiation times of an order of hours) means that $\lambda t_{irr} << 1$ and allows us to perform a series expansion of the exponential function in Eq. (3). We can then express N_T through the mass m and the molar mass M of the isotope target and introduce the half-life T instead of the decay constant λ to obtain

$$A \simeq \frac{m}{M} N_{Av} \frac{I_e(E_0)}{S} \frac{t_{irr}}{T} \ln 2 \int_{E_{th}}^{E_0} \sigma(E) n_B(E, E_0) dE. \tag{5}$$

To get the *specific activity rate* $A[Bq]/(\text{mg}\cdot\mu\text{A}\cdot\text{h})$, which is used in practice, we express

Table 1: Comparison of calculated and experimental specific activity rates

Isotope	Reaction	$T_{1/2}$, h	$A/(\text{mg}\cdot\mu\text{A}\cdot\text{h})^{calc}$, kBq	$A/(\text{mg}\cdot\mu\text{A}\cdot\text{h})^{exp}$, kBq	Refs.
Mo-99m	$^{100}\text{Mo}(\gamma, n)$	66	2.2	3.2	[6]
U-237	$^{238}\text{U}(\gamma, n)$	162	0.97	1.0	[7]
Pu-236	$^{237}\text{Np}(\gamma, n)$	$2.45\cdot10^4$	$3.0\cdot10^{-3}$	$4.4\cdot10^{-3}$	[8]

the electron beam intensity in μA (6.2x10^{12} particles per second for $1\mu A$), the irradiation time in hours and the target mass m in mg:

$$A/(mg \cdot \mu A \cdot h) = \frac{2.9 \cdot 10^9 \text{ s}^{-1}}{S \cdot M_{[g]} \cdot T_{[h]}} \int_{E_{th}}^{E_0} \sigma(E) n_B(E, E_0) dE. \tag{6}$$

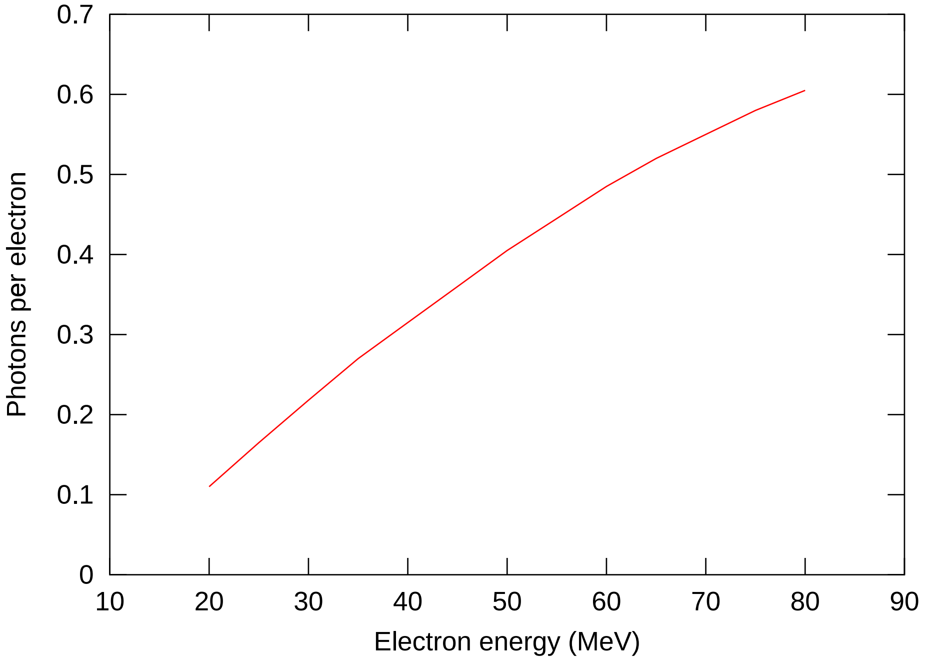

Figure 3: The effective yield $I_B^{ef}(E_0)$ of photons in dependense of the W-target thickness for electrons with the end-point energy of 30 MeV.

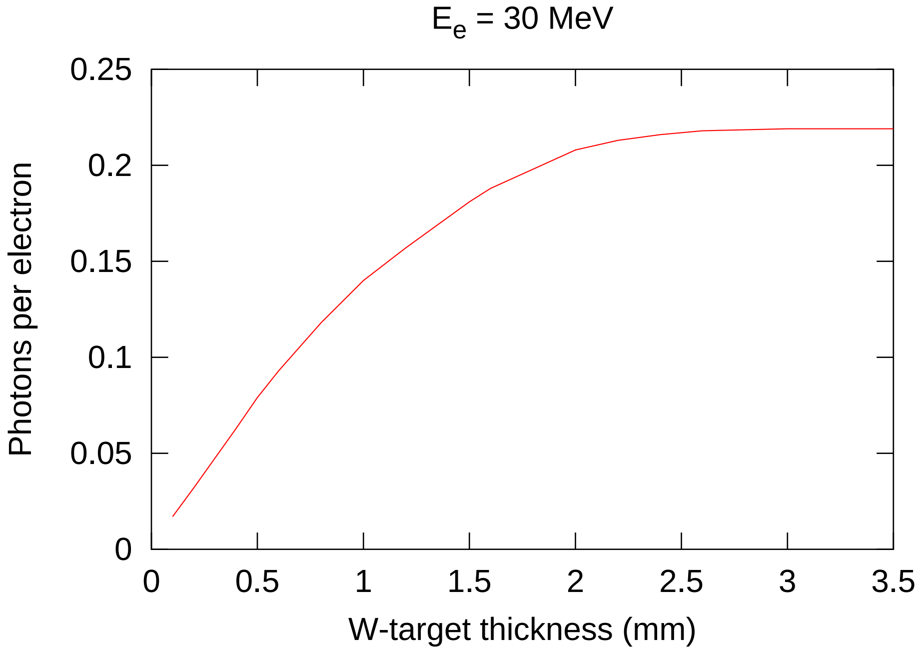

Figure 4: The effective yield $I_B^{ef}(E_0)$ of photons in dependense of the electron end-point energy E_9 for the W-target.

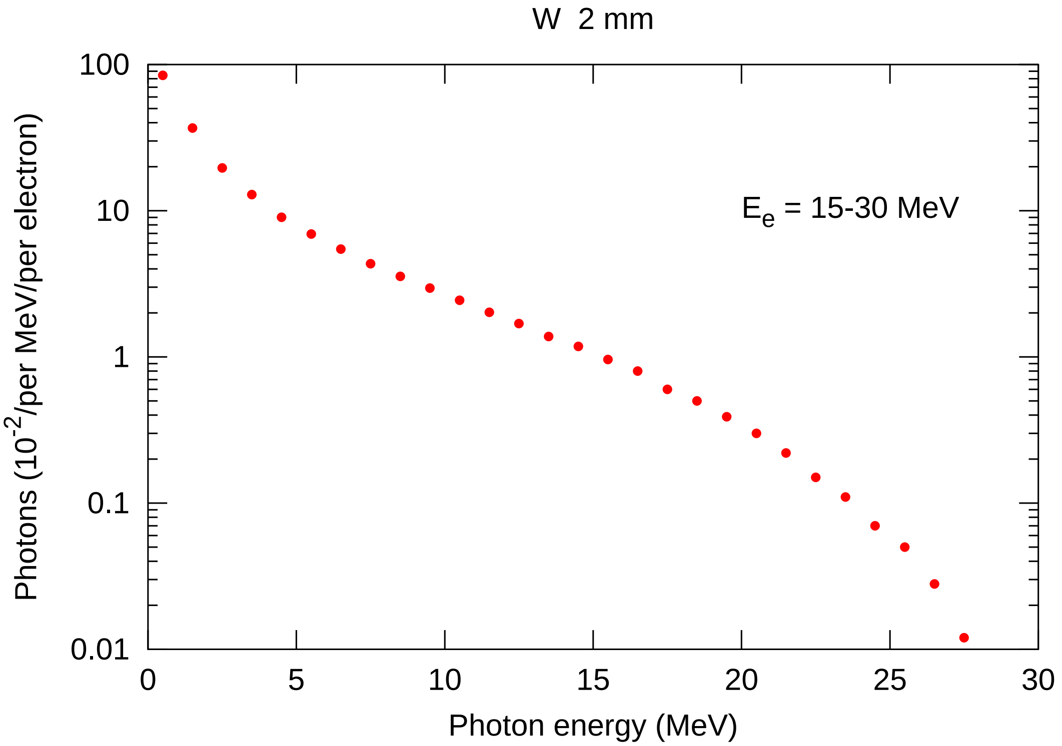

Figure 5: The bremsstrahlung photon energy spectrum $n_B(E, E_e)$ calculated by the MCNP code for electrons with the end-point energies E_e uniformly distributed from 15 Mev to 30 MeV.

The quantities $M_{[g]}$ and $T_{[h]}$ in this formula are dimensionless although expressed in grams and hours, as written explicitly. The dimensions of the cross section $\sigma(E)$ (here in barns) and the area S (cm^2) mutually cancel.

The results for calculated and experimental activities rates are shown in Table 1. The reported numbers are for the *mg* of the *target isotopes*. An agreement within a factor of 1.5 can be thought satisfactory in a view of several factors which may be present in experiments but not taken account of in calculations. In particular, the work [9] of the Kharkov Institute of Physics and Technology indicates a role of the reaction (ee', n) when the electrons are not removed from the beam and penetrate into the target under study. The Kharkov Institute had developed and put into operation the 20-kW beam power Linac for medical isotopes production. The electron energy can be varied in the interval of 10-25 MeV keeping the energy spread within 3% [10]. With this accelerator it is possible, for example, to produce the Mo-99m with activity 4 Curie per day using a natural Mo target.

Discussion and Conclusion

Physical startup of the first stage of the IREN (Intense REsonance Neutrons) source for neutron time-of-flight applications had took place in January 2009 [11]. Its present 50-MeV linac can also be used as a source of the bremsstrahlung γ-rays produced by a 1-kW electron beam. Specifics of this linac is a broad electron energy spread due to a high pulse current loading. Preliminary study [11] indicate several possible shapes of the electron spectra depending on the acceleration regime. In order to estimate how this feature will influence the isotope production we performed calculations of bremsstrahlung of electrons with the end-point energies uniformly distributed from 15 Mev to 30 MeV. The result is shown in Fig. 5. Comparing with Fig. 1 we conclude that the effective (in the range of 8-20 MeV) photon yield is reduced by a factor of about 2. A loss of about the same factor in the yield of radioisotopes can always be compensated by an increase of the target thickness and/or increase of the beam energy. We also conclude that our results validate the MCNP code for a "medical" application.

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