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ORIGINAL PAPER

Development of the technique embedded into a Monte Carlo transport system for calculation of photonuclear isotope yield

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Abstract. The paper describes two methods that use Monte Carlo transport code for computing the photonuclear isotope yield for arbitrary target activation conditions. One of them is based on a direct simulation of new nuclei generation events (DSE method). The other one involves a step-by-step calculation of isotope microyield along the photon trajectories (SBSM method). The techniques have been realized in the computer codes using the PENELOPE package of 2001, 2006 and 2008 versions. The program benchmarking was performed using experimental data on the activity distributions of ^{67}Cu isotope produced in the ^{68}Zn(γ,p)^{67}Cu reaction in thick zinc targets. Both approaches have shown to give similar results at an appreciably greater speed of the SBSM method. The results of simulation based on the PENELOPE 2006 and 2008 codes are in excellent agreement with all experimental data. At the same time, the PENELOPE 2001 computations give good agreement with the experimental results for target activation by the electron beam, but in the case of target exposed to bremsstrahlung systematic underestimation of about 15% has been observed.

Key words: electron accelerator • bremsstrahlung • photonuclear reaction • Monte Carlo (MC) simulation • isotope yield

Introduction

Practically all radionuclides employed in medicine, science and engineering are produced in reactors and heavy particle accelerators [1]. Meanwhile, recent investigations have demonstrated that it may appear promising to produce some isotopes at moderately-priced electron accelerators via photonuclear reactions $A(\gamma,x)B$, where A is an isotope-target, B – an isotope-product and x may represent n, p, np, 2n, 2p, α etc. (see, e.g. Refs. [2–4, 6, 7]). The method is based on pre-conversion of the accelerated electron beam into bremsstrahlung that activates the isotopic target. Generally, for bremsstrahlung generation a special intermediate target-converter (or radiator) is needed. However, this process can also be realized immediately in the isotopic target directly exposed to the accelerated electron beam [3, 6].

The calculation of the photonuclear isotope yield from a thick production target exposed to a substantially non-uniform mixed flow of electrons and bremsstrahlung photons is a rather complicated task. The problem is best solved by the computer simulation based on the Monte Carlo transport systems MCNP [5], PENELOPE [12], etc. It should be noted that the PENELOPE simulates only electromagnetic processes, while the MCNP also enables computation of neutron transport.

The validity of simulation results is determined by the accuracy of particle transport calculation as well as the reaction cross section description. However, even application of validated codes for the solution of the problem sometimes gives a disagreement (up to 50%

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and more) with the experimental data (e.g., see Refs. [6, 10]). In the case of a great amount of computations (in particular, in the optimization problems) apart from the accuracy of the method, its speed of operation also plays an important role.

The PENELOPE code in its basic package provides simulation of electron, positron and photon cascades. To determine the isotope yield, we have modified the package by adding the algorithms for two essentially different computing methods and also a number of procedures which provide:

- Input of data on the photonuclear cross section, the isotope-target concentration and half-life of the isotope-product.
- Tracing each step of a photon in the target as well as computation of the reaction cross section for the current photon energy and summation of isotope microyields over all photon trajectories.
- Final calculation of the isotope-product distribution in the target volume with determination of the total target activity, etc.

The paper contains the description as well as comparative analysis of the accuracy and operating speed for both methods of the photonuclear isotope yield simulation based on the PENELOPE 2001, 2006 and 2008 versions.

Calculation technique

Energies k of bremsstrahlung photons are distributed continuously over the range

$$0 < k \le k_{\max} = E_0 \quad (1)$$

where E_0 is the initial electron energy (see, e.g. Ref. [8]). Each photonuclear reaction has its energy threshold $E_{\text{threshold}}$ (in most cases, $E_{\text{threshold}} \ge 10$ MeV, Ref. [9]). So, for realization of the reaction it is necessary to have $k > E_{\text{threshold}}$. Therefore, in the algorithms for both methods of the isotope yield determination particle transport is calculated up to the moment when their energy decreases down to $E_{\text{threshold}}$ for electrons and photons or to $(E_{\text{threshold}} - 2m_ec^2)$ for positrons. Final result of the calculation represents the yield of new nuclei summed over all particle trajectories in the target volume and normalized to one electron of the primary beam. To determine the target activity, this quantity is multiplied by the total amount of accelerated electrons with allowance for the decay of nuclei-products during exposure.

DSE method

The simulated trajectory of the photon of initial energy k in the target presents a sequence of linear segments (steps) between the points of its interaction with atoms. The length l_s of the s-th step of photon trajectory in the target randomly varies in the neighborhood of the free path value $r(k)$ or

$$l_s \sim r(k) = \mu^{-1}(k) \quad (2)$$

where $\mu(k)$ is the photon attenuation coefficient [8]. To determine the isotope yield at the end of each step, the probability of a single reaction (i.e., $A(\gamma,x)B$ event) is calculated through the comparison of the reaction cross section with the cross sections for all other electromagnetic interactions of the photon – the direct simulation of events (DSE) method. It should be noted that the contribution of photonuclear processes to the total cross section is generally not more than several percent [8]. Consequently, at the given statistical uncertainty the calculating speed of the isotope yield by the DSE method depends on the reaction cross section and such simulation takes much more time than the determination of space-energy distribution of the photon flow.

SBSM method

The photon energy k_s along each s-step of its trajectory may be assumed to be constant. Therefore, the total isotope yield Y can be represented as a sum of microyields from all the steps along the trajectories of all the photons that have crossed the target in any direction, wholly or in part, or have been produced in it. Then, Y may be written as

$$Y = \sum_s (1 - e^{-n_A \cdot \sigma(k_s) \cdot l_s}) \quad (3)$$

where n_A is the nuclear density of the isotope A in the target material; $\sigma(k_s)$ is the reaction cross section for the photon of energy k_s.

Benchmarking experiment

To verify developed codes we have calculated by the both methods the spatial distribution of ^{67}Cu produced in the ^{68}Zn(γ,p)^{67}Cu reaction under different target photoactivation regimes. The reaction cross section (Fig. 1) has been taken from the reference data [9].

The simulation conditions corresponded to the experiment described in Ref. [6]. Namely, two configurations of the output devices of accelerator were reproduced: i) target directly exposed to accelerated electrons, and ii) with an intermediate target-converter. The electron beam was assumed to be parallel and monoenergetic with uniform particle distribution within

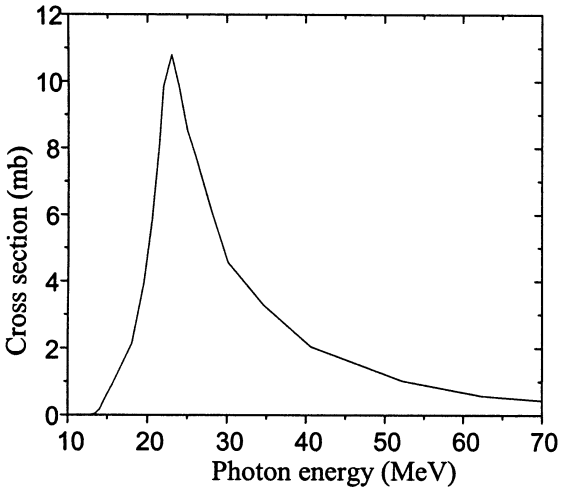

Fig. 1. ^{68}Zn(γ,p)^{67}Cu reaction cross section.

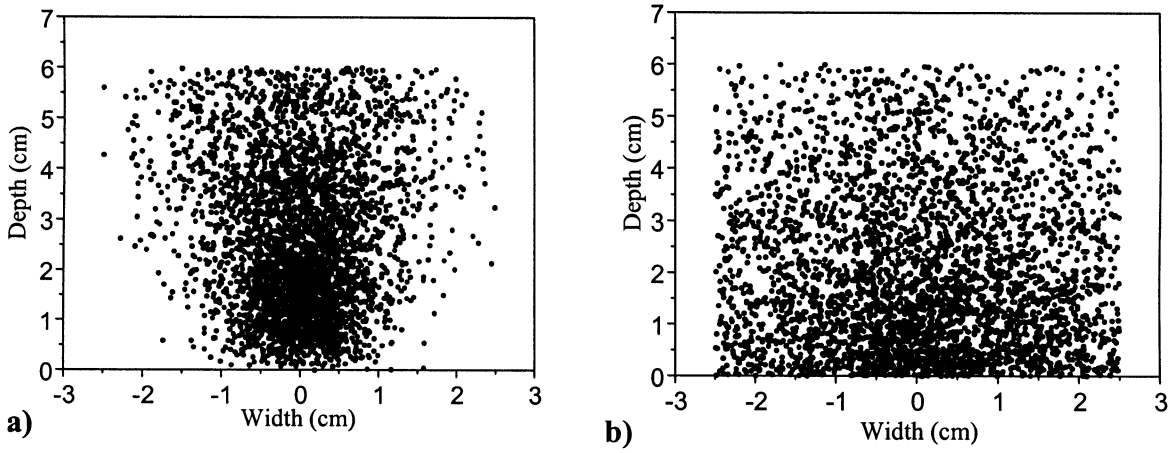

Fig. 2. ^{67}Cu nuclear distributions in the target under irradiation with: a – electron beam; b – bremsstrahlung.

the 10 mm circle. The Ta converter consisted of successive four plates (two 1.6 mm thick and other two 3.2 mm thick) separated by 1.6 mm gaps for cooling water. The plates were placed into a casing made from 1.6 mm thick aluminum. Each target had a form of parallelepiped consisting of closely stacked 36 square plates, 50.8 mm in width and 1.59 mm in thickness. Natural zinc (density 7.13 g/cm^3, ^{68}Zn isotope-target abundance 18.8%) was used as a target material.

Direct target activation was performed by the electron beam (55 MeV, 4 μA). In case of bremsstrahlung activation the converter was exposed to the (49 MeV, 100 μA) electron beam. The distance from the target to converter *d* was chosen to be 75 mm. The target exposure time was 40 min in both cases. The experimental data on the ^{67}Cu activity in separate Zn plates along the target axis under its activation by electrons and bremsstrahlung photons have been presented in Refs. [3, 6], respectively. The total uncertainty of activity measurements was estimated to be 10%.

Thus, the reasons for choosing given experiment to test developed codes were the following:

- Realization of various modes of target activation.
- Utilization of thick targets with substantially non-uniform distribution of particle fluxes and produced isotope.
- Detailed measurements of activity spatial distribution in the irradiated targets.

Comparison of simulation results with a great number of experimental data increases the reliability of code estimate.

Results and discussion

Comparison with experimental data

As an illustration of the DSE method operation, Figure 2 shows the ^{67}Cu nuclei density distribution in the targets obtained by the use of that method.

Figure 3 shows the experimental data on ^{67}Cu activity distribution along the target axis at EOB (end of bombardment) as well as the results of simulation based on the DSE method and the PENELOPE 2001 and 2006 versions. As in Ref. [6], the final value of produced activity has been normalized to 1 MJ of the electron beam energy.

The results of simulation with the use of the given algorithm and the PENELOPE 2008 fully coincided with the ones obtained on the basis of the PENELOPE 2006 version.

The results of the SBSM method based simulation using the PENELOPE 2008 are presented in Fig. 4. In particular, the computations show that the photons leaving the converter have an average exit angle of about 17 degrees. Therefore, the converter-target dis-

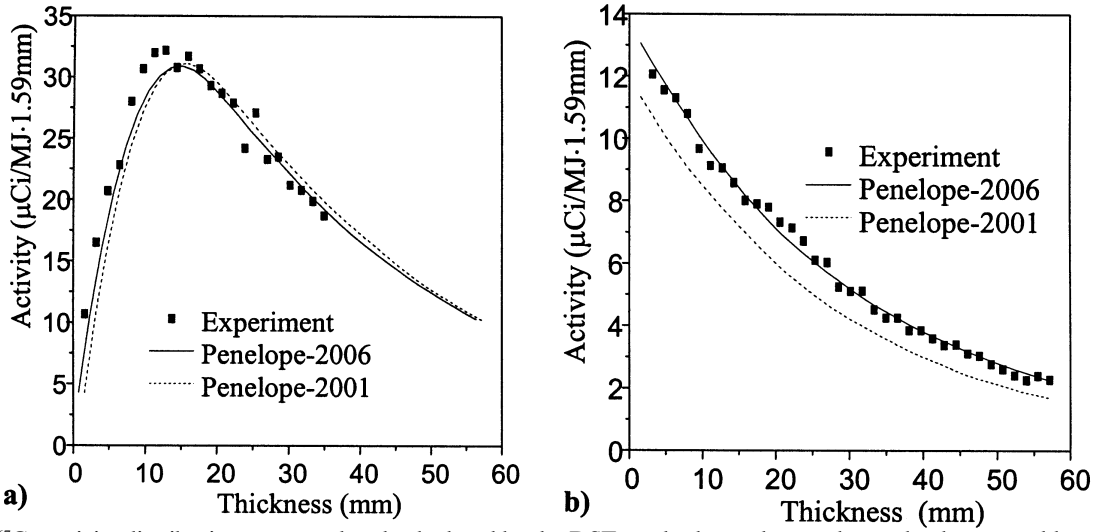

Fig. 3. ^{67}Cu activity distribution measured and calculated by the DSE method: a – electron beam; b – bremsstrahlung.

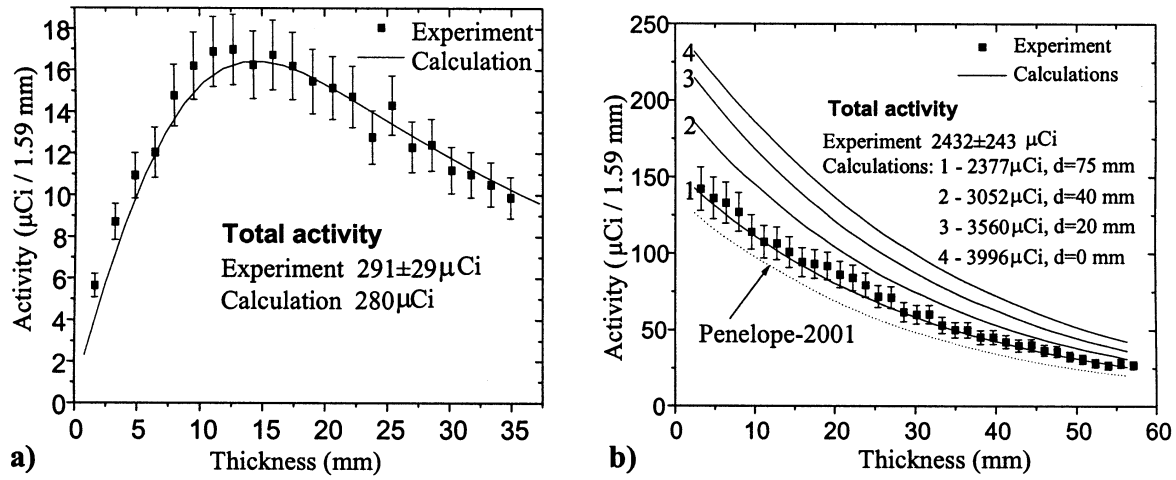

Fig. 4. ^{67}Cu activity distribution calculated by the SBSM technique: a – electron beam; b – bremsstrahlung.

tance d exerts an essential effect on the value of activity produced in the target. Thus curves 1 to 4 in Fig. 4b correspond to d = 75, 40, 20 and 0 mm, respectively. It can be seen that the maximum yield is attained at d = 0 mm. In this case the total target activity is 68% higher than the value obtained in the experiment.

We have also performed similar computations by the SBSM technique with the use of earlier PENELOPE 2001 and 2006 versions. Comparison of the obtained results (see Fig. 5) indicates that, as in the case of the DSE method, all the codes provide good description of activity distribution in the target exposed to a direct electron beam (Fig. 5a). As to the case of activation by bremsstrahlung (Fig. 5b), the data obtained with the PENELOPE 2006 and 2008 versions are practically coincident and show very close agreement with the experimental results. On the other hand, application of the PENELOPE 2001 for simulating bremsstrahlung activation gives the same underestimated (by ~ 15%) result for both methods (see Fig. 3).

The difference between two modes of target activation lies in the fact that under direct action of the electron beam the processes of bremsstrahlung generation and isotope production by photons take place only in the target, while in the second case the photon transport occurs successively in the thick converter, in the converter-target air gap, and in the target itself. It becomes obvious that the PENELOPE 2006 and 2008 versions solve the problem more correctly.

Comparison of the efficiency

It should be noted that the algorithms of the developed techniques provide computation of photonuclear processes simultaneously with the simulation of particle cascades by the basic package. Meanwhile, all additional arithmetical operations involved in the computation of isotope yield take negligible time in comparison to that of particle transport simulation. For comparative estimation of adequacy and efficiency of the DSE and SBSM methods we have performed on their basis a combined modeling of the both modes of target activation using the PENELOPE 2008. The computations were carried out by a PC (3.0 GHz Intel Core 2; 2 GB RAM). The computation time of target activation by the electron beam and by bremsstrahlung was 97 and 87 h, respectively.

The obtained data are presented in Table 1 and in Fig. 6. The table gives the number of events of ^{67}Cu nuclei generation in the target for a period of computation, the normalized nuclear yield per electron of the primary

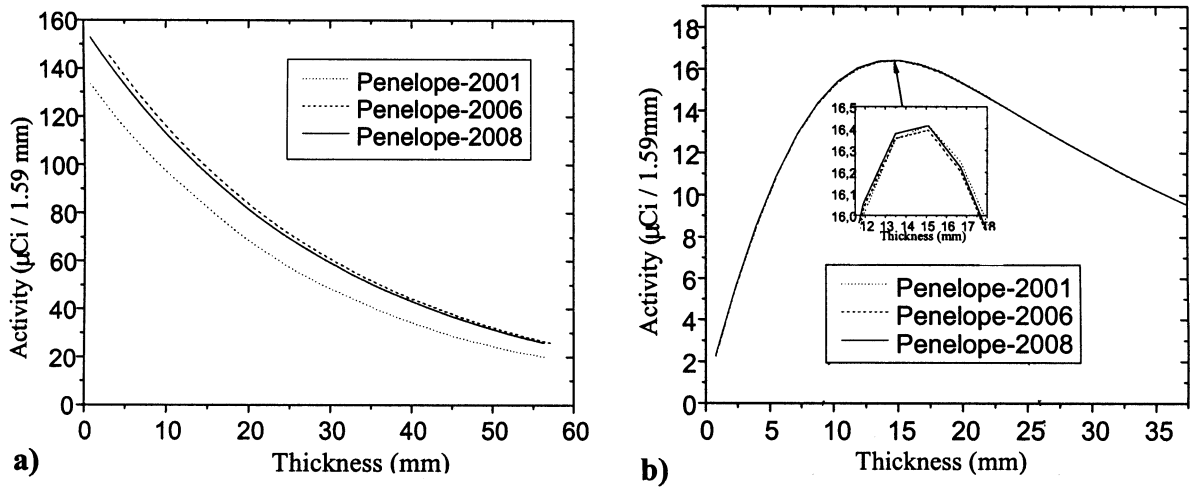

Fig. 5. Comparison of target plate activity distributions computed on the basis of different PENELOPE versions (SBSM method): a – electron beam; b – bremsstrahlung.

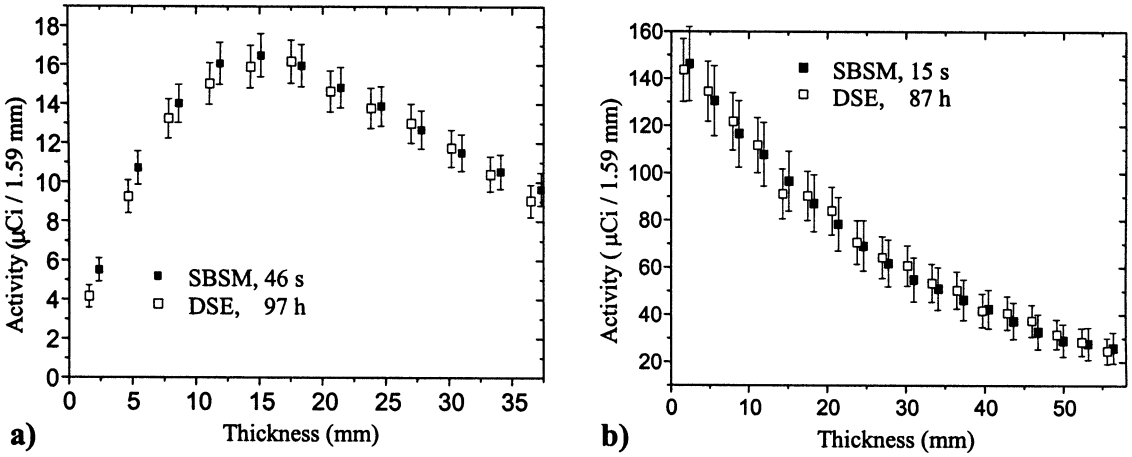

Fig. 6. Illustration of adequacy and efficiency of DSE and SBSM methods: a – electron beam; b – bremsstrahlung.

Table 1. Characteristics of isotope yield computation algorithms

Method	Number of events	Nuclear yield/e^- (10^{-5})
Electron beam activation		
DSE[a]	24 601	7.708 ± 0.034
SBSM[b]	24 545	7.690 ± 0.00039
Bremsstrahlung activation		
DSE	9 781	2.043 ± 0.062
SBSM	9 824	2.052 ± 0.00043

[a]DSE – direct simulation of events.
[b]SBSM – step-by-step method.

beam, and the statistical uncertainty. The data fit for the total yield of the ^{67}Cu (accurate to 0.2 and 0.4% at direct electron irradiation and under bremsstrahlung, respectively) as well as the agreement between the activity distribution data given in Figs. 6a and 6b indicate that the methods are mutually adequate.

The efficiency (operating speed) of the methods was estimated by comparing their computation time required to provide the same statistical uncertainty of results. As is seen from the data given in Table 1, in the case of target exposure to a direct electron beam the SBSM technique appears more efficient than the DSE by a factor of $(0.034/0.00039)^2 = 7569$. In other words, at the same statistical uncertainty the result of the DSE-based computations attained over a period of 97 h is provided by the SBSM technique within 46 s (Fig. 6a).

In simulation of target activation by the bremsstrahlung the SBSM calculation appears more efficient than the DSE by a factor of $(0.062/0.00043)^2 = 20\,736$. That is, the result obtained by the DSE for 87 h is provided by the SBSM technique in 15 s (Fig. 6b). For example, the time of SBSM-based computation of curves 1 to 4 in Fig. 4b ranged from 10 to 30 min at a statistical uncertainty between 0.4 and 0.2%.

Conclusion

The two methods developed for calculating the photonuclear yield of isotopes are embedded into the basic package of the MC transport system. Therefore, they allow to compute simultaneously not only the isotopic product yield, but also the absorbed radiation power in the exit device components of the accelerator, and hence, enable one to optimize the mode of target activation with due regard for thermal stability of the target. This possibility is of crucial importance in view of the prospects offered by the development of photonuclear technology at high-power electron accelerators (see, e.g. Ref. [11]).

With the use of PENELOPE 2006 and 2008 codes, the results of simulation based on the developed methods are in good agreement with the experimental data for both modes of target activation, namely, by the electron beam and the bremsstrahlung. At the same time the SBSM technique provides ~ 10^4 times higher computation speed, and in contrast to the DSE, this speed is independent of the reaction cross section.

It should be also noted that the SBSM approach is universal in character. This is specified by versatility of formula (3) forming the basis for the technique. Thus, the given method can be basically applied for calculating the isotope yield in any nuclear reaction of $A(a,x)B$ type. For this purpose the computation algorithm must be properly embedded into the package for modeling a-particle transport.

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