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Full Length Research Paper

Investigations of proton beam energy of the MC-50 cyclotron at KIRAMS

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The accuracy of the measured excitation functions of nuclear reactions largely depend on the precise measurements of the exposed beam energy in activation experiment. We investigated the proton beam energy of the MC-50 cyclotron at the Korea Institute of Radiological and Medical Sciences (KIRAMS) employing the method $^{nat}Cu(p,xn)^{62}Zn$ / $^{nat}Cu(p,xn)^{65}Zn$ together with a stacked-foil activation technique. The beam energy along with the stacked samples was also theoretically calculated using computer program SRIM-2003. The measured beam energy showed generally a good agreement with the calculated ones, and this fact demonstrated that the energy (<30 MeV) of the proton beam could be determined by irradiating thin metallic Cu foil target with natural isotopic compositions. Hence, this may be considered as a useful technique for beam monitoring purposes in activation experiment.

Key words: Proton beam energy, stacked foil technique, $^{nat}Cu(p,xn)^{62,65}Zn$ reactions, KIRAMS.

INTRODUCTION

The precise measurement of beam energy makes a great impact on the accuracy of excitation functions for the production of medically and technologically important residual radionuclides by activation experiment. Beam energy monitoring plays an important role not only in maximizing the production yields of interested radionuclide but also in minimizing unwanted radionuclide impurities. Nowadays, numerous research groups are involved in the production and applications of medical radionuclides using medium energy cyclotron facilities all over the world. To obtain the optimum production circumstances of medically and technologically important radionuclides, an accurate knowledge of production cross-sections are required. Since cross-sections of nuclear reactions have dependence on exposed beam energy, therefore determination of exact beam energy is crucial for radioisotope production. Owing to this fact, we investigated the proton energy of the MC-50 cyclotron at the KIRAMS using the cross sections ratio of the monitor reactions products [$^{nat}Cu(p,xn)^{62}Zn$/ $^{nat}Cu(p,xn)^{65}Zn$ method] in conjunction with a stacked-foil activation experiment, and found a consistency of the measured energy points to the calculated ones by a computer program SRIM-2003 (Ziegler et al., 2003). This work provides an additional technique about the determination of proton energy in a stacked foil activation experiment.

EXPERIMENTAL

The irradiation technique, the activity determination and the data analysis were similar to those described in detail elsewhere (Khandaker et al., 2006; Khandaker et al., 2007). The most salient features relevant to the present work have been presented in here. A high purity copper foil having natural isotopic composition [^{63}Cu(69.17%), ^{65}Cu(30.83%)] with 100 µm thickness was used as targets for determination of beam energy. Aluminum (Al) and Molybdenum (Mo) foils were also inserted in the stack for additional measurements. The stacked-target was formed by a total of 15 foils, following the order (Cu→Mo→Al), repeatedly. The schematic diagram of stacked foils is shown in Figure 1.

The thickness of each foil in the stack was same (0.1 mm). In order to irradiate the samples, the stacked-foil was placed in an aluminum holder, where the incident beam energy (35 MeV) was degraded initially to as 27.5 MeV by 1 mm thick of metal (aluminum) window. The stacked samples were then irradiated by an external beam line of the MC-50 cyclotron at the KIRAMS, with 27.5 MeV nominal proton beam energy. The irradiation of the stacked samples was done (with a 0.1 cm diameter beam and 45 to 50 *nA*

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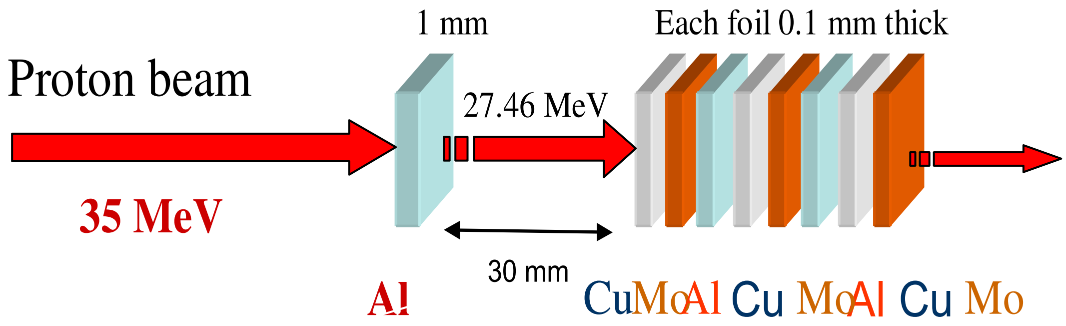

Figure 1. Schematic diagram of the stacked target used in the present experiment.

Table 1. Gamma-ray standards for detector calibration (IAEA (1991) TECDOC-619).

Nuclide	Decay mode	Half-life [d]	γ-ray energy [keV]	Emission probability (I_γ)
^{109}Cd	EC	462.6(7)	88.0341(11)	0.0363(2)
^{57}Co	EC	271.79(9)	122.0614(3)	0.8560(17)
			136.4743(5)	0.1068(8)
^{133}Ba	EC	3862(15)	80.998(5)	0.3411(28)
			276.398(1)	0.07147(30)
			302.853(1)	0.1830(6)
			356.017(2)	0.6194(14)
			383.851(3)	0.08905(29)
^{137}Cs	β^-	1.102(6) ×10^4	661.660(3)	0.851(2)
^{54}Mn	EC	312.3(4)	834.843(6)	0.999758(24)
^{60}Co	β^-	1925.5(5)	1173.238(4)	0.99857(22)
			1332.502(5)	0.99983(6)
^{22}Na	EC	950.8(9)	511	1.8101
			1274.542(7)	0.99935(15)

intense current) for 30 min. The beam intensity was kept constant during irradiation. It was necessary to ensure that equal number of incident particles interact with the monitor and target foils. The irradiation geometry was kept in a position so that the foils get the maximum beam line.

DATA PROCESSING

The activities of the produced radioisotopes in the Cu foils were measured nondestructively on the basis of their gamma radiation energy by using a high purity germanium (HPGe) γ-ray spectrometry. The γ-ray spectrometer was an n-type coaxial ORTEC (PopTop, Gmx20) high-purity germanium (HPGe) detector with a diameter of 55.1 mm and a thickness of 52.2 mm. The HPGe -detector was coupled to a 4096 multi-channel analyzer (MCA) with the associated electronics to determine the photo peak-area of gamma-ray spectra by using gamma vision (*EG&G ORTEC*) computer program. The energy resolution of the detector was 1.90 keV at full width half maximum (FWHM) for the 1332.5-keV peak of ^{60}Co. The photo-peak efficiency curve of the γ-ray spectrometer was calibrated with a set of standard point sources (Table 1). The detection efficiencies as a function of the photon energy were measured at 5 to 20 cm distances from the end-cap of the detector to avoid coincidence losses, to assure a low dead time (<10%) and a point like geometry (Wyttenbach, 1971).

Figure 2 represents the counting efficiencies of the used HPGe detector for the entire source to detector distances (5 to 20 cm). In this work, we were interested only with the Full energy peak (FEP) efficiency which is independent of the detector geometry, and was calculated by the formula given in Equation 1:

$$\text{Efficiency (\%)},\quad \varepsilon = \frac{CPS}{A_0 \times I_\gamma \exp(-\lambda t_d)} \times 100\% \qquad (1)$$

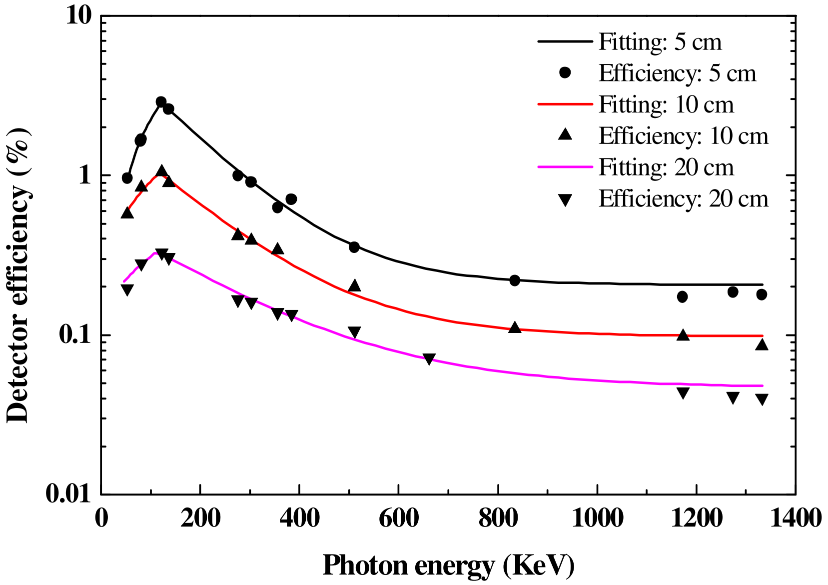

Figure 2. Measured efficiencies at different source to detector distances.

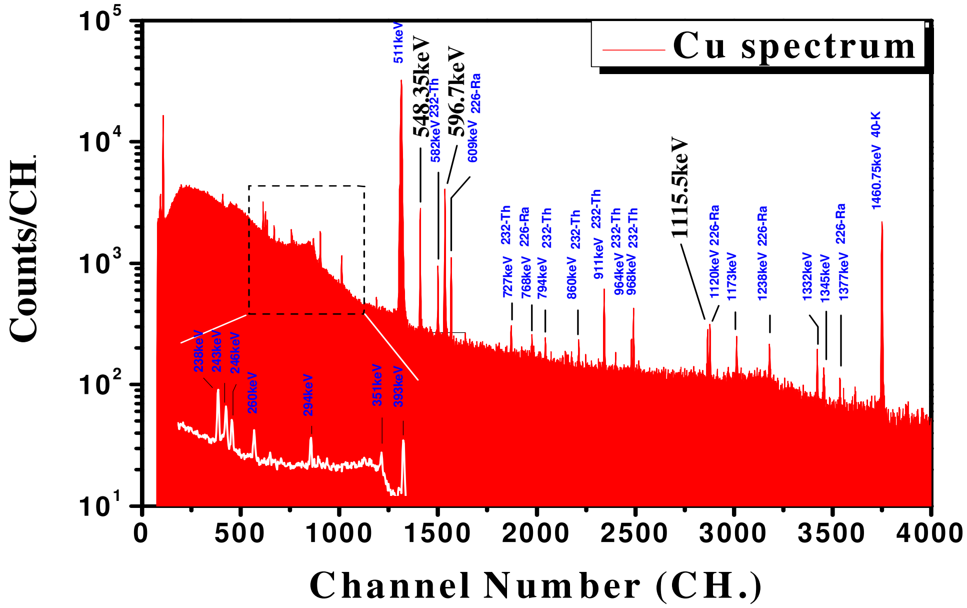

Figure 3. Typical Cu spectrum obtained by using ORTEC- Gamma vision software after a cooling time of 12 h.

where, *CPS* is the counts per second under a photo peak, I_γ is the gamma-ray intensity per decay, A_0 is the initial activity of the standard calibrated sources, λ is the disintegration constant of the sources, and t_d is the decay time. The uncertainty of the full-energy-peak efficiency was obtained by using the uncertainty propagation formula and assuming that all the measured quantities are independent. Efficiency uncertainties range from 0.3% up to 3%.

Measurement of cross sections ratio OF ^{62}Zn to ^{65}Zn radioisotopes

Figure 3 represents a typical Cu spectrum obtained by using the gamma vision 5.0 (EG&G Ortec) program. The production cross-

Table 2. Decay data of the investigated reaction products.

Nuclide	Half-life	Decay mode (%)	Gamma energy (keV)	Intensity (%)	Reaction	Q-value (MeV)	Threshold energy (MeV)
^{62}Zn	9.186 h	EC+β^+ (100)	548.35	15.2	^{63}Cu(p, 2n)	-13.2626	13.4750
			596.7	26.0	^{65}Cu(p, 4n)	-31.0889	31.5715
^{65}Zn	244.26 d	EC+β^+ (100)	1115.5	50.6	^{65}Cu(p, n)	-2.13431	2.16744

Table 3. Principal sources of uncertainties used in this measurement.

Principal sources of errors	Uncertainties (%)
Statistical error	1-5
Error due to detector efficiency curve	-4
Error due to γ-ray intensity	1-2
Overall uncertainty	4.2-6.7

sections ratios of ^{62}Zn and ^{65}Zn radioisotopes were measured using their characteristics gamma lines of 548.35 keV and 1115.5 keV with the formula given in Equation 2:

$$\frac{\sigma_i}{\sigma_j} = \frac{C_i}{C_j} \times \frac{\lambda_i}{\lambda_j} \times \frac{\Lambda_j(t_i, t_c, t_m)}{\Lambda_i(t_i, t_c, t_m)} \times \frac{\varepsilon_j}{\varepsilon_i} \times \frac{I_j}{I_i} \tag{2}$$

where

$\Lambda_k = \Lambda_j(t_i, t_c, t_m) = (1 - e^{-\lambda_k t_i}) e^{-\lambda_k t_c} (1 - e^{-\lambda_k t_m})$, σ_k is the formation cross section of k-isotope of zinc, C_k is the gamma-ray peak area assigning the k-isotope of zinc, λ_k is the decay constant of the k-isotope of zinc, ε_k is the detector efficiency of the k-isotope of zinc corresponding to the characteristics gamma line, I_k is the gamma-ray abundance of the k-isotope of zinc, and t_i, t_c, t_m are the irradiation, cooling, measurement time, respectively.

The contributing reactions and relevant information for this measurement is given in Table 2. The decay data used in the calculation were taken from (Brown and Firestone, 1986). The threshold energies in Table 2 were calculated by using the Los Alamos National Laboratory, T-2 Nuclear Information Service on the internet (NuDat data base, 1994-99).

In the present experiment, all the errors were considered as independent. Consequently, they were quadratically added according to the laws of error propagation to obtain the total errors. However, some of the sources of errors are common to all data, while others individually affect each reaction. The estimated major sources of errors considered in deduction of cross-sections ratio are summarized in Table 3. The total uncertainties of the measured cross-sections were calculated by combining the statistical uncertainties (δ_{sts}) and other uncertainties (δ_{oth}).

Determination of proton energy using the measured cross sections ratio

The standard cross-sections (IAEA-TECDOC-1211, 2001; Kopecky, 1985) of the ^{nat}Cu(p,xn)^{62}Zn and ^{nat}Cu(p,xn)^{65}Zn nuclear reactions and their ratios [^{nat}Cu(p,xn)^{62}Zn/ ^{nat}Cu(p,xn)^{65}Zn] have been reproduced in the Figures 4(a) and 4(b), respectively. Since at least two of the monitor radionuclides are produced simultaneously in the energy region of interest (15 to 26 MeV), the cross-sections ratio of the two radionuclides ^{62}Zn and ^{65}Zn can be used to determine the beam energy for the interested region (Kim et al., 2006). In the Figure 4(b), it is clearly shown that the peak ratio correlates linearly with the proton energy up to our region of interest (15 to 26 MeV). Since, the ^{62}Zn and ^{65}Zn nuclide cross-sections ratio formed a linear correlation with the proton energy, therefore the proton energy of each copper foil could be estimated by using the measured cross-sections ratio of the same radionuclide. Based on this fact, and using the measured cross sections ratio, the proton energy of each Cu foil in the irradiated stack was determined.

Theoretical calculations of proton beam energy by using SRIM-2003 computer program

Stopping and Range of Ions in Matter (SRIM) is a simulation programs which can calculate the stopping and range of ions (up to 2 GeV/amu) in matter using a quantum mechanical treatment of ion-atom collisions (assuming a moving atom as an "ion", and all target atoms as "atoms"). It is, of course, impossible to predict how a given charged-particle will interact with any given atom of the absorber medium. Also, when we consider that the coulombic forces of charged particles will interact simultaneously with many atoms as it travels through the absorbed medium, we can only predict an average effect of energy loss per particle distance of travel. Taking into account the charge, mass and speed (energy) of the particle, density and atomic number of the absorbing medium, Tsoulfanidis (1995) modified the Bethe and Ashkin (1953) formula for calculating the stopping power resulting from the coulombic interactions of heavy charged particles traveling through absorber media as in Equation 3:

$$\frac{dE}{dX} = 4\pi r_0^2 z^2 \frac{mc^2}{\beta^2} NZ[\ln(\frac{2mc^2}{I}\beta^2\gamma^2) - \beta^2] \tag{3}$$

where $\frac{dE}{dX}$ is the particle stopping power in units of MeV/m, r_0 is

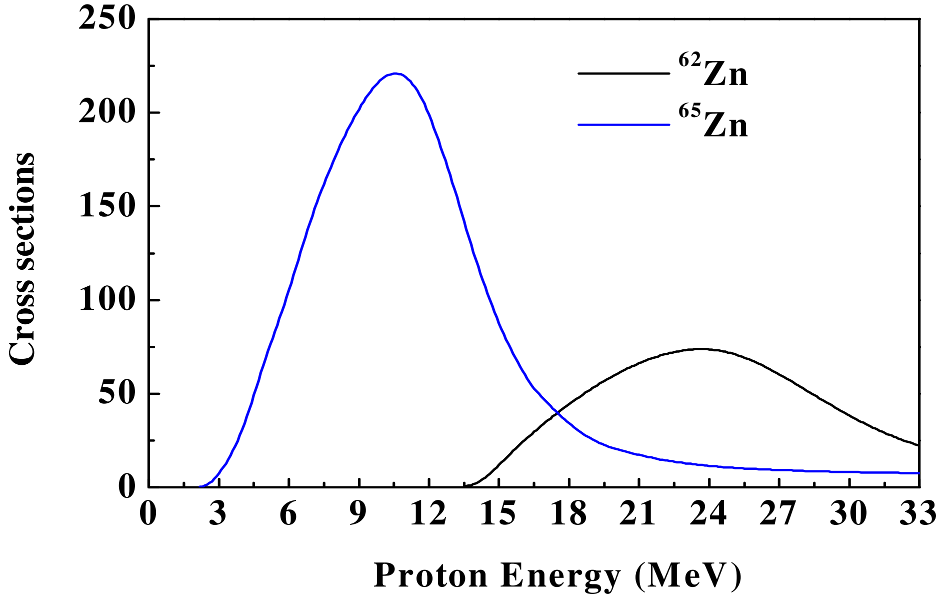

Figure 4(a). Standard cross-sections of ^{nat}Cu(p,xn)^{62}Zn and ^{nat}Cu(p,xn)^{65}Zn reactions.

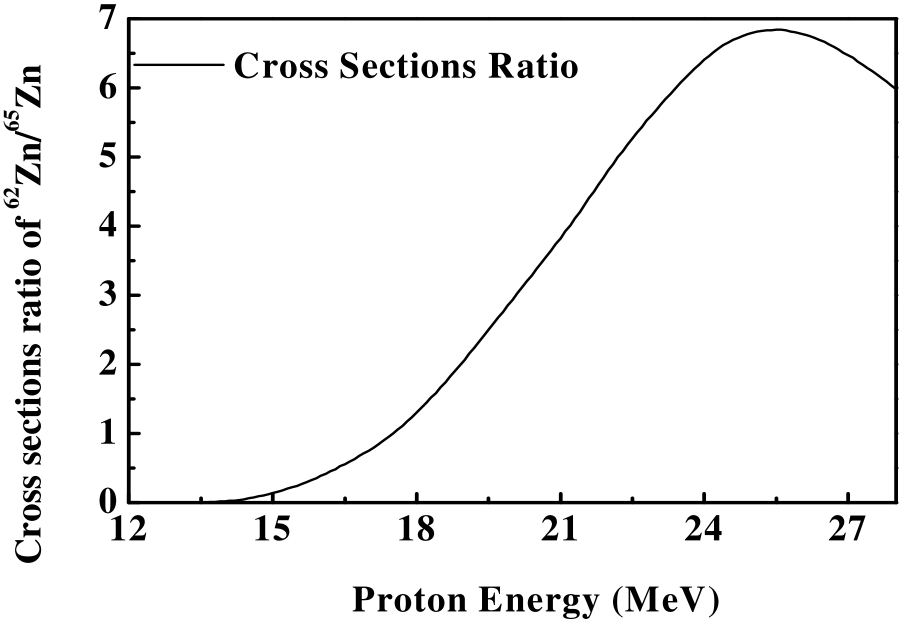

Figure 4(b). Standard cross-sections ratio of ^{62}Zn to ^{65}Zn radionuclides.

the classical electron radius = 2.818×10^{-15} m, z is the charge of the incident particle ($z=1$ for p, d, β$^-$, β$^+$, and $z=2$ for α, mc^2 is the rest energy of the electron = 0.511 MeV, N is the number of atoms per m^3 in the absorber material through which the charged particle travels [$N=\frac{\rho\times N_A}{A}$, ρ is the absorber density, N_A is the Avogadro's number], $A \text{ and } Z$ are the atomic weight and atomic number, respectively, of the absorber, $\gamma = \frac{T+Mc^2}{Mc^2} = \frac{1}{\sqrt{1-\beta^2}}$ where T is the particle kinetic energy in MeV and M is the particle rest mass (for example, proton = 931.5 MeV/c^2), and $\beta = \frac{v}{c}$, the relative phase velocity of the particle, and $I=[(9.76+58.8Z^{-1.19})Z, \textit{when } Z>12]$ is the mean excitation

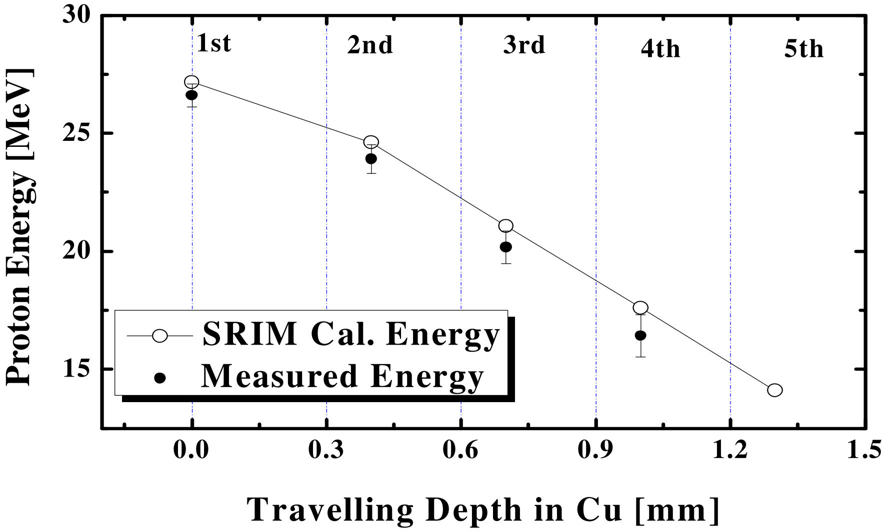

Figure 5. Comparison between the measured proton energy with SRIM simulated ones.

potential of the absorber in units of eV. With the help of a very convenient stopping-range calculation program SRIM-2003 (Ziegler et al., 2003), we found that the 35 MeV proton beam reaches the first copper foil of the stack with an energy of 27 MeV, due to the energy loss by initial Al metal window. The proton energy along the stack was calculated by using the computer program SRIM-2003 (Ziegler et al., 2003), and is shown in Figure 5. Since the stack was made by a total of 15 foils (0.1 mm thickness of each), therefore, the actual traveling path of a proton through the stack is 1.5 mm. Reduced proton energy (shown in Figure 5) indicates that a proton losses its energy due to the traveling through the Cu, Mo and Al foils in the stack.

RESULTS AND DISCUSSION

The measured proton energy was compared with the theoretical ones, and presented in the Figure 5. This measurement showed a general good agreement to the calculated ones within the experimental error. Figure 5 indicates that the proton losses kinetic energy continuously during its travel through the copper foils in the stack. Therefore, it was assumed that the activity of the Cu foils varies linearly as a function of the traveling depth. Moreover, the theoretical calculations by SRIM-2003 program also represent a similar trend of traveling path of protons to the measured ones. Therefore, data of activation analysis from a stacked target thin metallic Cu foils with natural isotopic compositions can be used to calibrate the low energy (<30 MeV) proton beam with an overall uncertainties of about 7%.

CONCLUSIONS

The proton beam energy of MC-50 cyclotron at the KIRAMS was investigated using the stacked target technique with an activation analysis. Two different proton-induced reactions of natural copper foils were used to develop a diagnostic technique to measure the beam energy. Since, the accuracy of cross-sections largely depends on the beam energy; therefore, accurate measurements of proton energy along the stacked samples plays an important role to measure a reliable cross sections and/or excitation functions of important medical radionuclides through an activation experiment. As the present results showed generally a good agreement with the theoretical ones, therefore, this may be considered as a useful technique for beam monitoring purposes in activation experiment.

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