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# N values estimation based on photon flux simulation with Geant4 toolkit

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## HIGHLIGHTS

- The equation of N values for computer simulation was derived and Bremsstrahlung photon flux was simulated with Geant4.
- We confirmed the feasibility of estimating N values before the irradiation starts with MC simulations of photon flux.
- N values are highly correlated with the beam parameters and the setup of the electron-photon converter.
- This practice is valuable for radiation safety concerns before the irradiation starts.

## ARTICLE INFO

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## ABSTRACT

N values are routinely introduced in photon activation analysis (PAA) as the ratio of special activities of product nuclides to compare the relative intensities of different reaction channels. They determine the individual activities of each radioisotope and the total activity of the sample, which are the primary concerns of radiation safety. Traditionally, N values are calculated from the gamma spectroscopy in real measurements by normalizing the activities of individual nuclides to the reference reaction [$^{58}$Ni(γ, n)$^{57}$Ni] of the nickel monitor simultaneously irradiated in photon activation. Is it possible to use photon flux simulated by Monte Carlo software to calculate N values even before the actual irradiation starts? This study has applied Geant4 toolkit, a popular platform of simulating the passage of particles through matter, to generate photon flux in the samples. Assisted with photonuclear cross section from IAEA database, it is feasible to predict N values in different experimental setups for simulated target material. We have validated of this method and its consistency with Geant4. Results also show that N values are highly correlated with the beam parameters of incoming electrons and the setup of the electron-photon converter.

## 1. Introduction

Photon activation analysis (PAA) is a well-established and versatile tool in nuclear and radioanalytical chemistry (Segebade et al., 1988; Segebade and Berger, 2008; Starovoitova and Segebade, 2016). After irradiating samples with high energy photons, the qualitative and quantitative information of the target nuclides can be obtained from the decay spectra of product nuclides recorded by gamma spectrometers. In recent years, this traditional technique has found new applications in agriculture, medical isotope production, archeology, cosmochemistry, environmental sciences, and nuclear physics (Sun et al., 2013, 2014; Rotsch et al., 2016; Mamtimin et al., 2013; Agar et al., 2017; Boztosun et al., 2016; Aygun et al., 2016; Tickner, 2015). In all these cases, the high-energy photon beam for activation was unanimously created by an electron LINAC via an electron-photon converter made of high Z material, such as Tungsten or Tantalum (Starovoitova and Segebade, 2016).

Using Bremsstrahlung radiation as the photon source allows the irradiation achieving photon fluxes many orders of magnitude higher than traditional isotopic sources. However, since the Bremsstrahlung beam has an energy range from zero to the maximum energy of the incoming electrons, generally it creates a series of nuclear reactions in irradiation. Typically, (γ, n), (γ, 2n), (γ, 3n), (γ, p), (γ, pn) reactions all can be produced in samples. Besides these photon-induced reactions, neutron capture reactions will exist as well, since an electron-photon converter itself is also a strong neutron source (Starovoitova and Segebade, 2016). The multichannel reactions in the sample are inevitable. This situation makes the calculation of activity of end products very difficult. In some cases, the respective activity contribution from each reaction channel is hard to estimate.

To compare the relative activities of the product nuclides of these multichannel reactions, N values are routinely introduced in nuclear

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activation. N value is the ratio of specific activity of the product nuclides normalized by the reference reaction [$^{58}$Ni(γ, n)$^{57}$Ni], which is simultaneously activated with the same experimental setup at the moment exactly one hour after irradiation. [$^{58}$Ni(γ, n)$^{57}$Ni] reaction is selected because Ni foil is widely used as the flux monitor in photon activation. N values can be obtained experimentally by the measurements of decay spectra of products nuclides and Ni-57. Experiments have shown that N values are highly related to the experimental setup of electron-gamma converter and the physical parameters of the incident electron beam (Sun et al., 2017). It is obvious that N values are very important in determining the individual activities of each radioisotope and the total activity of the sample, which are the primary concerns of radiation safety and work control documents.

However, in reality, radiation safety and work control documents usually have to be drafted and approved before the actual experiment starts, instead of being as an after effect of irradiation. Health physicists need to know what kind of radioisotopes will be produced, what is the activity of each radioisotope is, and what the total radioactivity will be generated. Experimentally determined N values are accurate and reliable, but may be too late for radiation safety and work control documents. Based on physical setup of electron-gamma converter and parameters of the incident electron beam, could we have a good estimation of N values even before the actual irradiation starts? At present, the only feasible way is to conduct calculations with the assistance from photon flux and cross sections, which accordingly are originated from Monte Carlo simulations of photon shower with high performance computers and IAEA database of photonuclear cross sections.

Monte Carlo method is a stochastic technique which uses random numbers and probability statistics to simulate physical phenomena and problems in the real world. Monte Carlo method was initially invented by the physicists at Los Alamos National Laboratory (LANL) when they investigated particle-matter interaction problems for the Manhattan project (Waters, 2002). The most popular ones in nuclear simulations are MCNPX from LNAL and Geant4 from the European Organization for Nuclear Research (CERN) (Waters, 2002; Geant4 Collaboration, 2017). As a common platform and toolkit for description of the particles passing through matter in a wide energy range, Geant4 has some irreplaceable advantage: unlike MCNPX programs completely finalized after compilation, Geant4 uses the advanced software-engineering techniques and the object-oriented programming language C++ to achieve its transparency. Users can modify the underlying codes to customize their own simulation. Beneath the Geant4 toolkit are a series of physics lists supported by repositories of nuclear data and models in contemporary nuclear and particle physics (Agostinelli et al., 2003; Allison et al., 2006; Boudreau et al., 2010).

In this paper, we employed Geant4 toolkit to simulate the photon shower of the photon activation and obtained the photon flux in the sample with its energy distributions. With the assistance of historical cross sections from IAEA databases, we can calculate the reaction density of the reaction channels of interest. With the reaction density information and initial incoming electron beam parameters, we can computer the activities of the different radioisotopes of the sample can be computed, which leads directly to the prediction of N values.

The aim of the paper is to estimate N values even before the actual irradiation starts. For this purpose, we firstly derive the modified equation of N value according to the particular situation of computer simulations, from either the original definition of N value or its equation of traditional measurements.

## 2. Theory

According to reference (Segebade et al., 1988; Segebade and Berger, 2008), the activity of product nuclides of interest immidiately after an hour of irradiation is

$$A_p(T) = \frac{m_p h_p L}{A_{r,p}}(1 - e^{-\lambda_p T}) \int_{E_{thres}}^{E_{max}} \varphi(E)_p \sigma(E)_p dE \tag{1}$$

For $^{58}$Ni(γ, n)$^{57}$Ni reaction, the activity of $^{57}$Ni is

$$A_{Ni}(T) = \frac{m_{Ni} h_{Ni} L}{A_{r,Ni}}(1 - e^{-\lambda_{Ni} T}) \int_{E_{thres}}^{E_{max}} \varphi(E)_{Ni} \sigma(E)_{Ni} dE \tag{2}$$

In the above Eqs. (1) and (2), $A$ is the activity, $T$ is the normalized irradiation time (exactly equals to one hour), $\lambda$ is the decay constant of the product, $m$ is the mass of irradiated material, $h$ is natural abundance of the nuclide under study, $L$ is Avogadro constant, $A_r$ is the relative atomic mass of irradiated nuclides, $\varphi(E)$ is the energy differential Bremsstrahlung flux density, $\sigma(E)$ is the energy-differential activation cross section within the corresponding energy range, subscript $p$ refers to the element under study, subscript $Ni$ refers to the Nickel reference monitor, $E_{thres}$ is the threshold energy, i.e. the minimum photon energy to induce a photonuclear reaction, and $E_{max}$ is the maximum energy of the Bremsstrahlung continuum, which equals the energy of the electrons incident to the converter.

The ratio of the specific activities ($SA$) after an hour of irradiation leads to N value,

$$N = \frac{SA_p(T)}{SA_{Ni}(T)} = \frac{A_p(T)/m_p}{A_{Ni}(T)/m_{Ni}} \tag{3}$$

Inserting Eqs. (1) and (2) into (3), we obtain

$$N = \frac{h_p}{h_{Ni}} \cdot \frac{A_{r,Ni}}{A_{r,p}} \cdot \frac{1-e^{-\lambda_p T}}{1-e^{-\lambda_{Ni} T}} \cdot \frac{\int_{E_{thres}}^{E_{max}} \varphi(E)_p \sigma(E)_p dE}{\int_{E_{thres}}^{E_{max}} \varphi(E)_{Ni} \sigma(E)_{Ni} dE} \tag{4}$$

Eq. (4) usually cannot be directly applied to Monte Carlo simulations, because simulations are not able to generate $\varphi(E)$ directly, but the conversion rate $Y(E)$ of the incoming electrons and emitting photons instead. Their relationship is described as

$$\varphi(E) = Y(E) I_{beam}/q_e \tag{5}$$

where $I_{beam}$ is the current of incoming electron beam and $q_e$ is the charge of an electron.

By inserting Eq. (5) back to Eq. (4), we get

$$N = \frac{h_p}{h_{Ni}} \cdot \frac{A_{r,Ni}}{A_{r,p}} \cdot \frac{1-e^{-\lambda_p T}}{1-e^{-\lambda_{Ni} T}} \cdot \frac{\int_{E_{thres}}^{E_{max}} Y(E)_p \sigma(E)_p dE}{\int_{E_{thres}}^{E_{max}} Y(E)_{Ni} \sigma(E)_{Ni} dE} \tag{6}$$

Eq. (6) is the modified N value equation for the particularity of computer simulations. We will follow this equation in the section of data analysis and result discussions. Traditionally, N value is calculated after measurements of gamma spectra (Segebade et al., 1988; Sun et al., 2017). It follows

$$N = \frac{SA_p(T)}{SA_{Ni}(T)} = \frac{P_p}{P_{Ni}} \cdot \frac{m_{Ni}}{m_p} \cdot \frac{\xi_{\gamma,Ni}}{\xi_{\gamma,p}} \cdot \frac{\eta_{\gamma,Ni}}{\eta_{\gamma,p}} \cdot \frac{\lambda_p}{\lambda_{Ni}} \cdot \frac{1-e^{-\lambda_p T}}{1-e^{-\lambda_{Ni} T}} \cdot \frac{1-e^{-\lambda_{Ni} t_i}}{1-e^{-\lambda_p t_i}} \cdot \frac{1-e^{-\lambda_{Ni} t_{c,Ni}}}{1-e^{-\lambda_p t_{c,p}}} \cdot \frac{e^{-\lambda_{Ni} t_{d,Ni}}}{e^{-\lambda_p t_{d,p}}} \tag{7}$$

where $P$ is net peak area (viz. count number), $\xi$ is absolute gamma-ray emission probability, $\eta$ is detector counting efficiency at respective gamma-ray energy, subscript $p$ refers to the gamma-ray under study, $t_i$ is the exposure time, $t_d$ is the decay time, and $t_c$ is the counting period.

At first sight, Eqs. (4) and (6) look quite different with (7). However, given the below formulas (8) and (9) of the net peak areas of product nuclides and Ni-57, one will immediately find that Eqs. (4) and (6) can be derived from Eq. (7) by inserting (8), (9) into (7). Therefore, the modified N value Eq. (6) for simulations is consistent with the