Determining concentrations of elements with different reaction channels in photon activation

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HIGHLIGHTS

- Concentrations of selected elements are calculated via different reaction channels in photon activation.
- N-values, which indicate the relative intensity of reaction channels, were calculated and compared.
- The design of electron-gamma converter may play a dominant role in determining N values of reaction channels.

ARTICLE INFO

ABSTRACT

In photon activation, same element may be activated by the bremsstrahlung beam through different nuclear reaction channels and produce different radioisotopes. These radioisotopes follow their own decay schemes and generate characteristic gamma rays. This phenomenon usually is an interference in spectra analysis, but it also offers a theoretical feasibility to determine the concentration of one element through different reaction channels.

To realize this theoretical feasibility, we conducted series of photon activation experiments with sample and reference of known concentrations. Irradiation of the samples and the references were conducted with electronic LINAC with different photon converters at a peak energy around 30 MeV. Several elements and their corresponding reaction channels were chosen to validate this procedure. Calculations of PAA were based on the internal monitor method. Our results have confirmed the advantages of current PAA reaction channel selection, and show that it might be beneficial to calculate the concentration of same elements with different reaction channels in some certain occasions. N-values, which indicate the relative intensity of reaction channels, were calculated and compared with those values generated from the photon activation at the Federal Institute for Materials Research and Testing in Germany (BAM). Results suggested that N values are impacted by several parameters of electron beam, and the design of electron-gamma converter may play a dominant role in determining N values.

1. Introduction

Photon activation analysis (PAA) is one of the most sensitive radioanalytical techniques used for the quantitative multi-element analysis. It dates back to 1934 when Chadwick and Goldhaber studied the disintegration of deuterium [^{2}H(γ,n)^{1}H] with photons produced by radioactive nuclides ^{208}Tl (Chadwick and Goldhaber, 1934). In 1954, Basile introduced bremsstrahlung radiation as high energy photon source for the first time. He analyzed some light elements with photonuclear reactions caused by bremsstrahlung photons induced by a Betatron (Basile et al., 1954). In 1986, Segebade published his encyclopedia book on PAA (Segebade et al., 1988). It summarizes almost

all the theoretical and experimental aspects of this technique, which indicated standardization, maturity, and wide acceptance of this nuclear tool. In recent years, PAA has found new applications in agriculture, medical isotope production, archeology, cosmochemistry, environmental sciences, and nuclear physics (Sun et al., 2013; Rotsch et al., 2016; Maschner et al., 2007; Mamtimin et al., 2013; Agar et al., 2017; Boztosun et al., 2016; Aygun et al., 2016; Tickner). In these cases, the high-energy photon beam of irradiation was unanimously created by an electron LINAC via an electron-photon converter made of high Z material, such as tungsten or tantalum. A recent review by Dr. Starovoitova addressed several issues of this high intensity photon source, such as converting efficiency, neutron production, and heat deposition
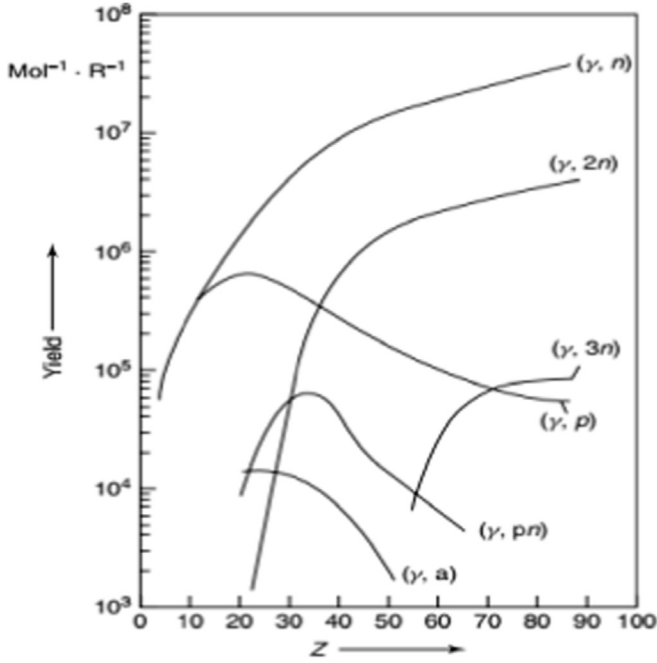

Fig. 1. Yields of photonuclear reactions for 30 MeV bremsstrahlung as a function of the atomic number (Segebade and Berger, 2008).

(Starovoitova and Segebade, 2016).

Using bremsstrahlung radiation as the means for photon activation, such as electron accelerators equipped with a bremsstrahlung converter, allows the irradiation achieving photon fluxes many orders of magnitude higher than isotopic sources. However, since the bremsstrahlung beam is not monochromatic, but spread from zero to the maximum energy of the incoming electrons, it created a complicate situation in nuclear reactions. Typically, (γ, n), $(\gamma, 2n)$, $(\gamma, 3n)$, (γ, p), (γ, pn) all can be generated in the target samples (see Fig. 1). Besides these photon-induced reactions, neutron capture reactions will exist as well, since an electron-photon converter itself is also a strong neutron source. The fast neutrons produced via photon capture in the sample can be slowed down to thermal or epithermal range and induce further neutron capture reactions, e.g. (n, γ), (n, p), (n, α), etc. The multichannel reactions are inevitable with bremsstrahlung photon source.

How to select spectra peaks for PAA analysis for the element of interests? This is not a simple task as we thought. As mentioned, using bremsstrahlung photon source may induce a bunch of photon capture and neutron capture reactions and generated different product isotopes. These product isotopes will create different decay spectra recorded by gamma spectrometers. Even for a single product isotope, it usually has several gamma lines with different energy. Selection of the characteristic peak needs to consider the interference of nearby peaks from other reaction channels.

The phenomenon of multichannel reactions accompanied by bremsstrahlung photon source may usually be an interference in gamma spectra analysis, but it also offers a theoretical possibility to determine the concentration of a certain element through different reaction channels. In this paper, we try to analyze the possible origins of the interferences in gamma spectra for selected elements, compare pros and cons for each reaction channel, and explain the selection rules why certain reaction channel is the most suitable choice for PAA.

2. Theory

Concentration of element of interest is related to the net peak area of characteristic energy line by the equation

$$c_m = \frac{PA_r\lambda}{\eta\theta\xi mhL(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})e^{-\lambda t_d}\varphi\sigma_{eff}} \tag{1}$$

where c_m is the concentration of the nuclides of interest, P is the net peak area (or counts) of the characteristic gamma line, A_r is the atomic mass of the nuclide, λ is the decay constant of radioactive nuclide, η is the detector efficiency, θ is the branching ratio, ξ is the intensity of the gamma line, m is the mass of the whole sample, h is the natural abundance of the target nuclide, L is the Avogadro constant, t_i is the irradiation time, t_c is the counting time, t_d is the decay time, φ is the integral photon flux, σ_{eff} is the effective cross section of photonuclear reaction of interest (Sun et al., 2014).

For relative methods which irradiating the sample (S) with reference material (R), one can use Eq. (1) twice respectably on the same element in the sample and reference. Dividing these two equations on both sides, one gets:

$$\frac{c_S}{c_R} = \frac{P_S}{P_R} \cdot \frac{m_R}{m_S} \cdot \frac{\varphi_R}{\varphi_S} \cdot \frac{1-e^{-\lambda t_{cR}}}{1-e^{-\lambda t_{cS}}} \cdot \frac{e^{-\lambda t_{dR}}}{e^{-\lambda t_{dS}}} \tag{2}$$

Since we do not know the flux ratio, we have to introduce an internal monitor Sc, which has known concentration in both the sample and reference material. We choose internal monitor method since it has the best accuracy and sensitivity in relative methods (Sun et al., 2014). Following Eq. (2) of the internal monitor, one gets:

$$\frac{c_{iS}}{c_{iR}} = \frac{P_{iS}}{P_{iR}} \cdot \frac{m_R}{m_S} \cdot \frac{\varphi_R}{\varphi_S} \cdot \frac{1-e^{-\lambda_i t_{cR}}}{1-e^{-\lambda_i t_{cS}}} \cdot \frac{e^{-\lambda_i t_{dR}}}{e^{-\lambda_i t_{dS}}} \tag{3}$$

$$c_S = c_R \cdot \frac{c_{iS}}{c_{iR}} \frac{P_S}{P_R} \frac{P_{iR}}{P_{iS}} \frac{e^{-\lambda t_{dR}}}{e^{-\lambda t_{dS}}} \cdot \frac{e^{-\lambda_i t_{dS}}}{e^{-\lambda_i t_{dR}}} \cdot \frac{1-e^{-\lambda t_{cR}}}{1-e^{-\lambda t_{cS}}} \cdot \frac{1-e^{-\lambda_i t_{cS}}}{1-e^{-\lambda_i t_{cR}}} \tag{4}$$

It should be noted that all the mass data are eliminated in Eq. (4). Therefore, there is no need to measure the mass of the sample and reference in the experiments. This is an inherent advantage of the internal monitor method.

The uncertainty in concentrations of unknown elements is primarily propagated from the statistically uncertainties of peak areas and that of concentrations of known elements. It follows the equation below.

$$\Delta c_S = c_S \sqrt{\left(\frac{\Delta c_R}{c_R}\right)^2 + \left(\frac{\Delta P_S}{P_S}\right)^2 + \left(\frac{\Delta P_R}{P_R}\right)^2 + \left(\frac{\Delta P_{iS}}{P_{iS}}\right)^2 + \left(\frac{\Delta P_{iR}}{P_{iR}}\right)^2} \tag{5}$$

As discussed in Section 1, bremsstrahlung photon source will induce a bunch of photon capture and neutron capture reactions. Different reaction channels produce different product isotopes. To compare the relative intensity of these reaction channels, we introduce N value to indicate the ratio of special activities. The activities of the individual nuclides produced during activation were normalized to the reference reaction of the nickel monitor simultaneously irradiated. All the reaction channels will be normalized by the relative intensity corresponding to $^{58}Ni(\gamma, n)^{57}Ni$ reaction channel after exactly an hour of irradiation. According to Section 5.3 of reference (Segebade et al., 1988), the N value of a certain reaction channel is

$$N = \frac{A_{0,el}(T)}{A_{0,Ni}(T)} = \frac{P_{el}}{P_{Ni}} \cdot \frac{m_{Ni}}{m_{el}} \cdot \frac{\xi_{\gamma,Ni}}{\xi_{\gamma,el}} \cdot \frac{\eta_{\gamma,Ni}}{\eta_{\gamma,el}} \cdot \frac{\lambda_{el}}{\lambda_{Ni}} \cdot \frac{1-e^{-\lambda_{el}T}}{1-e^{-\lambda_{el}t_i}} \cdot \frac{1-e^{-\lambda_{Ni}t_i}}{1-e^{-\lambda_{Ni}T}} \cdot \frac{1-e^{-\lambda_{Ni}t_{c,Ni}}}{1-e^{-\lambda_{Ni}t_{c,el}}} \cdot \frac{e^{-\lambda_{Ni}t_{d,Ni}}}{e^{-\lambda_{Ni}t_{d,el}}} \tag{6}$$

In Eq. (6), A is the special activity, T is normalization time (exactly an hour), subscript 0 means it is immediately after the irradiation, subscript el is the element of interest, subscript Ni is about the element Nickel, subscript γ is of the gamma line of interest, subscript d is of decay, subscript i is of irradiation, subscript c is of counting.