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A comparison of various procedures in photon activation analysis with the same irradiation setup

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ABSTRACT

A sample of known elemental concentrations was activated in the bremsstrahlung photon beam which was created by a pulsed electron LINAC. Several procedures of photon activation analysis, including those applied with/without reference material and with/without photon flux monitor, were conducted to make a comparison of their precision and accuracy in practice. Experimental results have indicated that: (1) relative procedures usually produce better outcome despite that the absolute measurement is straightforward and eliminate the assistance of reference materials; (2) among relative procedures, the method with internal flux monitor yields higher quality of the analytical results. In the article, the pros and cons of each procedure are discussed as well.

1. Introduction

Photon activation analysis (PAA) has been a versatile nuclear technique in elemental analysis for more than half a century. The first published article of PAA dated back to 1951 when Gaudin and Pannell of MIT tried to determine the amount of beryllium in low grade beryl ores by photodisintegration of Beryllium [1]. They claimed the advantage of this method is “rapid, simple, and nondestructive”. In 1954, Basile suggested to analyze some of the light elements using photonuclear reaction induced by bremsstrahlung radiation from a betatron [2]. His work led to the wide use of the bremsstrahlung radiation as the high energy photon source of PAA. In 2008, Segebade gave a brief but complete overview of PAA research in a chapter of Encyclopedia of Analytical Chemistry [3]. In the recent few years, PAA has expanded its applications to radiotherapy, meteorology, geochemistry, archeology, industrial material, and environmental studies, etc [4–10].

Traditionally, PAA has been conducted with the relative procedures which need the assistance of reference material. In irradiation, the sample and the reference are activated with the same high energy photon beam. By comparison of the decay spectra created by the same isotope in sample and in reference, scientists can deduce/calculate the unknown amount of the isotope in the sample according to the known concentration of the isotope in the reference. Usually, the photon fluxes in both the sample and the reference are unknown. To track the discrepancy/ratio of photon fluxes, researchers apply flux monitors (e.g. Ni foils) in activation and the relative procedures are divided into three categories accordingly: mass-based (analysis without monitor), external monitor method, and internal monitor method. Relative procedures, especially the internal monitor methods, are also well developed and widely used in neutron activation analysis (NAA) [11–14]. Recently, the developments of computer technology allowed scientists to make Monte Carlo simulation of photon flux in the sample based on experimental setup, which raised a new absolute method in PAA – quasi-absolute method: with simulated photon flux and database of experimental cross sections, it is possible to calculate the concentration of target isotopes in the sample directly without any reference material [15,18]. How is the outcome of this absolute method compared with traditional relative procedures? What is the accuracy difference among the different flux monitoring methods in relative procedures? Although there is scattered study on flux monitoring in PAA [16], no one has compared the absolute measurement with relative ones in details before. A study of comparison of the results of various procedures in PAA, whether it is relative or absolute, within the same sample of the same irradiation setup, will be the key to answer those questions.

2. Calculations in PAA

2.1. Quasi-absolute method

According to Refs. [15,17–19], the net peak area (or net counts) P of the characteristic gamma line of a particular isotope in the spectrum is

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$$P = \eta\theta\zeta\int_{t_c} A(t)dt = \frac{\eta\theta\zeta m c_m hL}{A_r\lambda}(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})e^{-\lambda t_d}\int_{E_{thres}}^{E_{\max}}\varphi(E)\sigma(E)dE \qquad (1)$$

where η is the spectrometer efficiency, θ is the branching ratio of the reaction channel, ζ is the absolute intensity of the corresponding gamma line, A is the activity of radioactive nuclide, m is the mass of the whole sample, c_m is the concentration of the isotope of interest, h is the natural abundance of the target isotope, L is the Avogadro number, λ is the decay constant of product radioactive isotope, A_r is the atomic mass of the target isotope, $\varphi(E)$ is the energy differential photon flux in the sample, $\sigma(E)$ is the cross section of corresponding photonuclear reaction, t_i is the irradiation time, t_c is the counting time, and t_d is the decay time from the end of irradiation to the start of spectrum measurement.

Rearranged Eq. (1), one obtains

$$c_m = \frac{PA_r\lambda}{\eta\theta\zeta mhL(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})e^{-\lambda t_d}\int_{E_{thres}}^{E_{\max}}\varphi(E)\sigma(E)dE} \qquad (2)$$

Normally, energy differential photon flux $\varphi(E)$ cannot be measured directly. However, assisted by Monte Carlo simulations from computational nuclear physics, one can derive photon flux by simulated photon yield $Y(E)$, average beam current I_{beam} of the LINAC, and single electron charge q_e as below:

$$\varphi(E) = Y(E)I_{beam}/q_e \qquad (3)$$

With $\varphi(E)$ and cross section data $\sigma(E)$, one can compute the integral in the denominator of Eq. (2). Therefore, it is possible to calculate the concentration of target isotopes without any reference material based on Eq. (2). This procedure is regarded as quasi-absolute method (QAM) because photon flux is not assessed from any direct experimental measurement, but from Monte Carlo simulation.

2.2. Relative methods

Both the sample and the reference are irradiated with the same photon beam in the relative methods. Their decay spectra are measured with the same gamma ray detector. According to Eq. (2), for the same characteristic energy line of the same isotope in the sample and in the reference, one has

$$c_S = \frac{P_S A_r\lambda}{\eta\theta\zeta m_S hL\varphi_S\sigma(1-e^{-\lambda t_i})(1-e^{-\lambda t_{cS}})e^{-\lambda t_{dS}}} \qquad (4)$$

$$c_R = \frac{P_R A_r\lambda}{\eta\theta\zeta m_R hL\varphi_R\sigma(1-e^{-\lambda t_i})(1-e^{-\lambda t_{cR}})e^{-\lambda t_{dR}}} \qquad (5)$$

where the subscript S means the parameters are of the sample, R means the parameters are of the reference. In addition, the integral of energy differential photon flux $\varphi(E)$ and cross section $\sigma(E)$ is replaced by the product of integral photon flux φ and effective cross section σ as

$$\int_{E_{thres}}^{E_{\max}}\varphi(E)\sigma(E)dE = \varphi\sigma \qquad (6)$$

Let us divide Eq. (4) with Eq. (5) to obtain the general equation of the relative method:

$$c_S = c_R\cdot\frac{P_S}{P_R}\cdot\frac{m_R}{m_S}\cdot\frac{\varphi_R}{\varphi_S}\cdot\frac{(1-e^{-\lambda t_{cR}})}{(1-e^{-\lambda t_{cS}})}\cdot\frac{e^{-\lambda t_{dR}}}{e^{-\lambda t_{dS}}} \qquad (7)$$

On the right side of Eq. (6), the only unknown parameter is the photon flux ratio φ_R/φ_S. As mentioned, relative methods are fallen into three categories according to the way of getting the flux ratio with different positioning of the monitors: mass-based (MB), external monitor method (EM) and internal monitor method (IM). Here we discuss the internal monitor method in details; calculations of other procedures can be found in Ref [20].

The internal monitor (e.g. Sc solution) is added and mixes evenly in the sample and reference in order to represent photon fluxes in the sample and the reference. From Eq. (7), for internal monitor i, one has

$$\frac{c_{iS}}{c_{iR}} = \frac{P_{iS}}{P_{iR}}\cdot\frac{m_R}{m_S}\cdot\frac{\varphi_R}{\varphi_S}\cdot\frac{(1-e^{-\lambda_i t_{cR}})}{(1-e^{-\lambda_i t_{cS}})}\cdot\frac{e^{-\lambda_i t_{dR}}}{e^{-\lambda_i t_{dS}}} \qquad (8)$$

Rearrange Eq. (8), we can get the photon flux ratio f as

$$f = \frac{\varphi_R}{\varphi_S} = \frac{c_{iS}}{c_{iR}}\cdot\frac{P_{iR}}{P_{iS}}\cdot\frac{m_S}{m_R}\cdot\frac{(1-e^{-\lambda_i t_{cS}})}{(1-e^{-\lambda_i t_{cR}})}\cdot\frac{e^{-\lambda_i t_{dS}}}{e^{-\lambda_i t_{dR}}} \qquad (9)$$

By plugging the flux ratio of Eq. (9) back into Eq. (7), one can reach the final formula for internal monitor method:

$$c_S = c_R\cdot\frac{c_{iS}}{c_{iR}}\cdot\frac{P_S}{P_R}\cdot\frac{P_{iR}}{P_{iS}}\cdot\frac{e^{-\lambda t_{dR}}}{e^{-\lambda t_{dS}}}\cdot\frac{e^{-\lambda_i t_{dS}}}{e^{-\lambda_i t_{dR}}}\cdot\frac{(1-e^{-\lambda t_{cR}})}{(1-e^{-\lambda t_{cS}})}\cdot\frac{(1-e^{-\lambda_i t_{cS}})}{(1-e^{-\lambda_i t_{cR}})} \qquad (10)$$

One remarkable thing in Eq. (10) is: all the mass data are eliminated. Therefore, people do not need to measure the mass of the sample and reference in the experiments. This is an inherent advantage of the internal monitor method.

3. Experiments and simulations

3.1. Sample, reference and monitors

The irradiation target included the sample, the reference, and the photon flux monitors. The sample was the Standard reference material 1648a (urban particulate matter) from NIST [21]. The reference material was the fly ash (certification campaign BCR "City Waste Incineration Ash") from the Federal Institute for Materials Research and Testing in Germany (BAM) [22]. Both of them have well-known certified elements and are widely used in instrumental analytical research. High purity nickel foils used as external photon flux monitor were bought from Alfa Aesar Company. 1% scandium solution ($Sc(NO_3)_3$) was added to both the sample and the reference as the internal photon flux monitor. Initially, the sample and the reference were in powder form. Then we mixed them with cellulose evenly and employed a mechanical compressor to press them into two dime-shaped pellets. Each pellet is in 1 cm diameter and 1 mm thickness. Pellets and Ni foils were wrapped together by Al foils to serve as the irradiation target.

3.2. Experimental setup for irradiation

Fig. 1 shows the experimental setup for photon activation. The electrons are initially produced by the hot cathode and then accelerated by a series of alternating RF electric fields in the LINAC. After focused by magnetic fields, they created an electron beam about 3 mm radius at the water-cooling beam window. After the window, electrons hit a custom designed electron-photon converter (or radiator). The converter was made by 3 mm thickness of tungsten which induced the bremsstrahlung radiation of incoming electrons and generated a cone of high energy X-rays after the converter. A 3-inch-thick aluminum absorber (or hardener) is positioned directly after the converter to absorb the residual electrons and filter the low energy photons. Most of residual high energy photons hit directly with the target behind the hardener and cause photonuclear reactions inside the sample, the reference, and the monitors.