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Measurement of partial γ-ray production cross-sections and k_0 factors for radionuclides with chopped-beam PGAA

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Abstract

The partial γ-ray production cross-sections (σ_γ) and k_0 factors for 16 short- and medium-lived radionuclides were determined with prompt gamma activation analysis (PGAA) in a chopped beam of cold neutrons. This technique avoids some problems of usual standardization measurements done in neutron activation analysis (NAA), e.g. epithermal activation, counting at varying count rates, and also offers better selectivities than continuous-beam PGAA. The k_0 values were determined with internal standardization using stoichiometric compounds or water solutions of known compositions. Calculation of the uncertainties is also discussed in detail.

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1. Introduction

In prompt gamma activation analysis (PGAA), radionuclides are also produced during the irradiation. Their decay γ-rays appear in the γ-spectrum along with the numerous prompt peaks. Since many of the decay peaks are among the most intense peaks of the spectrum, they are widely utilized in the routine analysis. Partial γ-ray production cross-sections (σ_γ) and k_0 values of the strongest decay lines have already been measured with high precision at our laboratory [1] using stoichiometric compounds. The determination of the cross-sections and k_0 values of these decay lines is an important methodological link between PGAA and conventional neutron activation analysis (NAA), i.e. the results can be compared to $k_{0,\mathrm{Au}}$ values measured with NAA. The most up-to-date k_0 NAA recommendation is from 2003 [2]. Additionally, k_0 data of short-lived nuclides were measured at the Atominstitut, Vienna [3], and at the DSM Research-Mol [4].

However, to investigate the short- and medium-lived nuclides, the use of a guided, cold-neutron-beam PGAA facility has advantages over standard NAA, especially in combination with beam chopping. In particular, at the PGAA facility of the Budapest Neutron Center [5],

(1) the epithermal activation is completely avoided,
(2) the longer sample–detector distance (235 mm) and the collimation (see also Fig. 2) result in a small solid angle, which, in turn, keeps the true coincidence effects low and also helps to reduce the dead-time effects,
(3) the sample is in a fixed position throughout the experiment, which allows even the investigation of nuclides with ms-long half-lives and eliminates uncertainties related to the sample repositioning,
(4) the half-life does not limit the precision, since the on-line counting can be prolonged as long as needed to attain suitable statistical accuracy,
(5) the use of a beam chopper avoids the spectral interferences and the high Compton-baseline due to prompt peaks, by separating the decay and the prompt events on the basis of their different time behaviour.

A recent experiment by Révay et al. [6] revealed that the decay spectra recorded during the closed chopper phase

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have excellent peak-to-background ratios; therefore, they are suitable to lower the detection limits of some elements in PGAA and to obtain decay cross-section data.

The first results of an ongoing project to determine partial γ-ray production cross-sections and k_0 factors of decay lines using the chopped-beam PGAA technique are reported in this paper.

2. Theoretical

2.1. Standardization of the decay lines

The intensity of a peak in PGAA is proportional to the partial γ-ray production cross-section (σ_γ), that is the product of the isotopic capture cross-section for neutrons of 2200 m/s velocity (σ_0), the isotopic abundance (θ) and the emission probability (P_γ) of the given γ-line,

$$\sigma_\gamma = \sigma_0 \ \theta \ P_\gamma. \tag{1}$$

Using the method of internal standardization, we determine the partial γ-ray production cross-section of a peak of interest from direct comparison to another peak with accurately known σ_γ value and good counting statistics. The best suitable comparator lines in PGAA have proved to be the 2223-keV peak of hydrogen, the 1951-keV line of chlorine, the 1884-keV line of nitrogen and the 841-keV line of sulphur. Occasionally, the 4945-keV line of carbon can also be used. The target sample is chosen to contain the element to be measured and one of the above-mentioned comparator elements in an accurately known molar ratio, preferably in the form of a stable, stoichiometric compound or a water solution. The sample is measured until counts for good statistical precision are accumulated for both the comparator peak and the peak of interest. This procedure has been applied for many years at our laboratory for the continuous-beam PGAA. Partial γ-ray production cross-sections of decay peaks can also be determined this way, after correcting for the time dependence of their count rate.

In chopped-beam PGAA, however, the beam is periodically opened and closed, and two spectra are recorded alternately. The decay peak is determined in the beam-off phase, while the comparator prompt peak is detected during the beam-on phase. This technique therefore requires an additional normalization of the two spectra.

In our previous publication [7], simplified formulae have been derived for the standardization with chopped-beam PGAA, as a limiting case of the cyclic activation analysis equation by Givens et al. [8]. It has been shown that the cumulative peak area in the closed-phase spectrum, from a radionuclide with $\lambda T \ll 1$ can be calculated as follows:

$$A_\mathrm{d} = \sigma_\mathrm{d} \Phi \ K \ N_\mathrm{d} \varepsilon(E_\mathrm{d}) L \ \ t_\mathrm{m} \eta_\mathrm{d} \ \ B. \tag{2}$$

Similarly, the area of the prompt peak from the comparator collected in the open-phase spectrum is given as:

$$A_\mathrm{p} = \sigma_\mathrm{p} \Phi \ K \ N_\mathrm{p} \varepsilon\left(E_\mathrm{p}\right) t_\mathrm{m} \eta_\mathrm{p}. \tag{3}$$

In the above expressions, T is the cycle time of the chopper (about 80 ms in this work), A is the net peak area, N is the number of atoms of the given nuclide in the sample, Φ is the neutron flux, K is the proportion of the irradiation time ($K = 0.5$), while L is the proportion of the decay counting time to the length of a complete cycle. A delay has to be used in the beam-off phase to allow even the slowest neutrons to reach the sample before the counting of the decay activity is started [7] and consequently, L has to be less than 0.5. σ_d and σ_p are the partial γ-ray production cross-sections for a decay and a prompt peak, ε is the detection efficiency, λ is the decay constant, B is the so-called in-beam saturation factor (see below), t_m is the measurement time, while the dead-time losses are taken into account with the factor η. This was determined from the peak areas of an auxiliary radioactive source detected in the two phases. The subscripts p and d indicate quantities associated with the prompt and decay phases, respectively.

If the ratio of a decay and a prompt peak is determined from the same measurement, then by combining Eqs. (2) and (3), some factors, e.g. the neutron flux, cancel out, eliminating possible sources of uncertainty. Thus, the k_0 value of a decay peak, relative to a comparator in the prompt spectrum is given by

$$k_{0,\mathrm{p}}(\mathrm{d}) \equiv \frac{M_\mathrm{p}}{M_\mathrm{d}} \frac{\sigma_\mathrm{d}}{\sigma_\mathrm{p}} = \frac{M_\mathrm{p}}{M_\mathrm{d}} \frac{A_\mathrm{d}}{A_\mathrm{p}} \frac{N_\mathrm{p}}{N_\mathrm{d}} \frac{\varepsilon\left(E_\mathrm{p}\right)}{\varepsilon(E_\mathrm{d})} \frac{\eta_\mathrm{p}}{\eta_\mathrm{d}} \frac{1}{LB}, \tag{4}$$

where M is the relative atomic mass of the element. Only the relative efficiency curve has to be used here, whose uncertainty is always smaller than that calculated for the absolute efficiencies. The $k_{0,\mathrm{p}}$ factors can be readily converted to those relative to the ultimate (u) comparator ($k_{0,\mathrm{H}}$ or to $k_{0,\mathrm{Au}}$) by multiplying them with the $k_{0,\mathrm{u}}$ value of the p comparator as

$$k_{0,\mathrm{u}}(\mathrm{d}) = k_{0,\mathrm{p}}(\mathrm{d}) k_{0,\mathrm{u}}(\mathrm{p}) = \frac{M_\mathrm{p}}{M_\mathrm{d}} \frac{\sigma_\mathrm{d}}{\sigma_\mathrm{p}} \frac{M_\mathrm{u}}{M_\mathrm{p}} \frac{\sigma_\mathrm{p}}{\sigma_\mathrm{u}} \equiv \frac{M_\mathrm{u}}{M_\mathrm{d}} \frac{\sigma_\mathrm{d}}{\sigma_\mathrm{u}}. \tag{5}$$

The k_0 factors of these key analytical lines were accurately determined in previous experiments at the Budapest PGAA laboratory [1,9].

2.2. Determination of the in-beam saturation factor for Type IV nuclides

Most of the radionuclides produced during the in-beam activation follow the simplest activation–decay scheme, i.e. Type I, according to the classification used in NAA [2]. An in-beam saturation factor [1],

$$B = 1 - \frac{1 - \mathrm{e}^{-\lambda t_\mathrm{m}}}{\lambda t_\mathrm{m}}$$

has been derived to correct for the growth in these cases (e.g. ^{20}F, ^{28}Al, ^{56}Mn). However, other nuclides appearing in the present work belong to Type IV, where the activation can follow different branches and all of them