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Measurement of partial gamma-ray production cross-sections and k_0-factors for radionuclides with chopped-beam PGAA—Part II

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ABSTRACT

As a continuation of our previously reported experiments, the partial gamma-ray production cross-sections (σ_γ) and k_0-factors of 12 short- and medium-lived radionuclides were determined using a chopped beam of cold neutrons. This technique avoids several problems associated with epithermal activation, sample transportation and dead-time effects in neutron activation analysis (NAA) measurements. Our k_0-values were determined with internal standardization, using stoichiometric compounds or water solutions of known compositions.

1. Introduction

The standardization of prompt gamma activation analysis (PGAA) has become a well-established procedure at the Budapest PGAA laboratory [1]. A continuously evolving library comprises the partial gamma-ray production cross-sections (σ_γ) of all stable elements. These data can also be expressed as $k_{0,H}$-values, i.e. a compound nuclear constant relative to hydrogen [1]. This dataset includes several decay gamma lines from the activated radionuclides, but only those with practical use in the routine chemical analysis. Therefore a project was initiated in 2002 aimed at a more complete determination of the σ_γ - and k_0-values of those radionuclides that can be measured in beam.

In conventional PGAA, the decay peaks appear in the γ-ray spectrum along with numerous prompt peaks. The presence of prompt gammas raises the baseline in the region of interest and may produce close-lying interferences. Thus separate detections of prompt and decay radiations are preferred. The chopped-beam PGAA, a technique to realize this alternate detection, was developed and was proven to be a good method to quantify radionuclides having half-lives up to a couple of days. It has several advantages over the reactor neutron activation analysis (NAA) measurements: there is no epithermal activation, no need to transfer the sample, the dead-time is usually low and constant, and the duration of the experiment is not limited by the half-life.

The other motivation to determine decay cross-sections and k_0-values in a cold-neutron beam is to establish a methodological link between PGAA and reactor neutron activation analysis (NAA) by comparing these results to existing $k_{0,Au}$-values of NAA. The different sources of experimental errors in the two approaches can reveal discrepancies in the data.

Both fields have their own specific databases. The most complete k_0-NAA dataset is by De Corte and Simonits [2], while additional k_0 data for short-lived nuclides have been published earlier in Refs. [3, 4]. On the other hand, the Evaluated Gamma-ray Activation File (EGAF) [5], a compilation of prompt and delayed γ-ray cross-sections, also contains decay data for about 250 isotopes, but only from guided-beam experiments at present. The harmonization of the two kinds of data is a priority. As databases contain data with $\pm 1\sigma$ quoted uncertainties, this formalism will be used in the present work as well.

The first set of our k_0 data from chopped-beam PGAA measurements was published in 2006 [6]. The present work summarizes the results from a second measurement campaign, resulting in the partial gamma-ray production cross-sections and k_0-factors of 12 additional radionuclides.

2. Experimental

The main properties of the chopped-beam PGAA facility at the Budapest Neutron Centre have been published in detail in our previous papers [6–8]. The beam is periodically switched on and off using a chopper, a four-sector rotating blade with two open

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and two closed quadrants. The segments are covered with a ^{6}Li-coating to block the beam, which keeps the gamma-ray background low. In the open phase the sample is irradiated and the prompt spectrum is recorded, while in the closed phase the prompt photons are absent and only the decay radiation is detected. A dedicated unit controls the chopper and provides the timing signals to route the events [7]. An XIA Polaris digital spectrometer [9] was set up to register the events according to the status signals from the chopper electronics, i.e. to acquire the prompt and decay spectra alternately.

The γ-radiation was detected with the standard Compton-suppressed HPGe detector of the Budapest PGAA facility. The Canberra GR2027/S HPGe detector has a relative efficiency of 27%, an energy resolution of 2.2 keV at 1332 keV and is surrounded by an active BGO detector, as well as a 10-cm thick passive Pb shield [8]. The sample-to-detector distance is 235 mm and the geometrical efficiency is about 0.001, in order to keep the true-coincidence effects below 0.1%.

Since the previous measurement campaign the system has gone through important improvements in order to further enhance its performance. Thanks to the partial replacement and realignment of the neutron guides, the thermal equivalent fluence rate at the PGAA sample position is $1.5\times10^{8}\,\mathrm{cm^{-2}\,s^{-1}}$, almost five times higher than the former value of $3.3\times10^{7}\,\mathrm{cm^{-2}\,s^{-1}}$. This opened the way to measure radionuclides with weak decay peaks, which would have been too time-consuming with the previous setup. It also made it possible to use smaller samples, significantly reducing the gamma-ray self-attenuation, and to reduce the aperture of the neutron collimator to 5 mm^2, resulting in a sharper transition between open and closed phases and a better approximation of the point-source geometry.

In the previous setup the cycle time of the chopper was not sufficiently stable; it was a limiting factor in the measurement of nuclides with $T_{1/2}<0.5$ s, such as ^{24m}Na. Therefore only tentative values could be given [7] for such cases. To overcome this problem a Maxon encoder-controller unit [10] was purchased. This proved to be capable of keeping the rotation frequency constant at the level of 0.25% or better.

In the near future a new, computer-controlled beam shutter is to be installed, which will enable switching the beam on and off according to a predefined timing in the range of seconds at least. It will be a complementary tool to the existing chopper instrumentation. In combination with the list-mode capability of the XIA digital spectrometer [9], it will allow us to study the time evolution of the activation and the decay, and thus to measure half-lives or even emission probabilities.

A neutron monitor was also put into operation to follow the beam intensity in time. This, together with other instrument parameters like HPGe count rate and number of chopper cycles, was recorded with a time-resolution of one second using a NI 6601 counter/timer card.

The typical sample weight was about 50 mg in the present measurements, with the exception of $Sb(CH_3COO)_3$ (320 mg) and NH_4TcO_4 (113 mg). The powder samples were heat-sealed in Teflon bags. In a few cases water solutions were prepared, e.g. from SeO_2 (100 mg in 400 mg water) and from VF_4 (10 mg in 500 mg water). They were measured in 1-ml Teflon vials with vacuum-tight screw cap. Most of the chemicals were purchased from Aldrich, and had the purity of 99.9–99.99%. The ammonium pertechnetate sample was supplied by the Oak Ridge National Laboratory.

The irradiation times in the measurements varied between 3600 and 65,000 s, depending on the count rates of the peaks in the decay phase. The detectability of weak decay peaks in the beam-off phase has been substantially improved, as illustrated in Fig. 1.

3. Data processing

The partial gamma-ray production cross-section ((σ_γ), i.e. the product of the isotopic capture cross-section for neutrons with the velocity of 2200 m/s (σ_0), isotopic abundance (θ) and emission probability (P_γ) of the given γ-ray line, determines the count rate of a peak in the prompt gamma spectrum.

The method of internal standardization is used to determine the partial gamma-ray production cross-section for a peak of interest by direct comparison to another peak with accurately known σ_γ-value and good counting statistics. The peaks of hydrogen or chlorine served here as comparators because their nuclear data are accurately known. The comparator peaks in the prompt phase had typically a statistical precision of 0.1–0.6%, somewhat better than the peaks of interest in the decay phase (about 0.7–5%).

Formulae are given in Refs. [6,7] to calculate the σ_γ-and k_0-values of a decay peak, relative to a comparator line in the prompt gamma spectrum:

$$\sigma_\mathrm{d}=\frac{A_\mathrm{d}}{A_\mathrm{p}}\frac{N_\mathrm{p}}{N_\mathrm{d}}\frac{\varepsilon(E_\mathrm{p})}{\varepsilon(E_\mathrm{d})}\frac{\eta_\mathrm{p}}{\eta_\mathrm{d}}\frac{1}{LB}\sigma_\mathrm{p} \tag{1}$$

where A is the net peak area, N the number of given nuclide in the sample, ε the counting efficiency and η the dead-time correction factor for the two different phases, while L quantifies the proportion of the decay counting phase within a cycle. The indices p and d refer to the prompt and decay phase, respectively. Finally, the so-called *in-beam saturation factor* (B) is included to correct for growth and decay of the nuclides throughout the experiment. Following the categorization of radioactive decays in Ref. [2], it is calculated as follows:

$$B=1-\frac{1-\mathrm{e}^{-\lambda T}}{\lambda T}\quad\text{for Type I and IV/B }(\lambda\equiv\lambda_3)\text{ nuclides}$$

$$B=1-\frac{1-\mathrm{e}^{-\lambda_3 T}}{\lambda_3 T}+\frac{\sigma_\gamma^\mathrm{m}}{\sigma_\gamma^\mathrm{g}}F_2\left[1+\frac{\lambda_2^2(1-\mathrm{e}^{-\lambda_3 T})-\lambda_3^2(1-\mathrm{e}^{-\lambda_2 T})}{(\lambda_3-\lambda_2)\lambda_2\lambda_3 T}\right]$$

for Type IV/A nuclides

$$B=\frac{(1-\mathrm{e}^{-\lambda t_a})\mathrm{e}^{-\lambda t_w}(1-\mathrm{e}^{-\lambda t_c})}{\lambda(1-\mathrm{e}^{-\lambda t_r})t_a L}\quad\text{for nuclides with } T_{1/2}<0.5\text{ s},$$

where λ is the decay constant, T the true time of the experiment, m and g denote the metastable and ground states, respectively, and $(\sigma_\gamma^\mathrm{m}/\sigma_\gamma^\mathrm{g})F_2$ is a multiplication factor expressing the ratio of activation via the metastable state and the ground state. In the third formula $T_{1/2}$ is the half-life, t_a, t_w and t_c are the activation, waiting and counting times within a cycle, respectively, while t_r is the cycle repetition time.

It is to be noted that the multiplication factor $(\sigma_\gamma^\mathrm{m}/\sigma_\gamma^\mathrm{g})F_2$ was not determined in our present measurements, but it had to be taken from the literature for every IV/A nuclide. For Type IV/B cases, we are able to determine the sum of the cross-sections to the metastable state and to the ground state (σ_{m+g}), just as in NAA measurements with long cooling times. The B-factor can be derived from the Bateman–Rubinson equations for more complicated decay schemes as well.

In internal standardization only ratios of quantities are used, and hence the uncertainties related to the absolute value of the neutron fluence rate and the efficiency curve cancel. The $N_\mathrm{p}/N_\mathrm{d}$ ratio is known from the stoichiometry of the sample and is assumed to be exact.

The ratio of two partial cross-sections can be expressed as a k_0-factor if multiplied with the ratio of the molar weights:

$$k_{0,\mathrm{p}}(d)\equiv\frac{M_\mathrm{p}}{M_\mathrm{d}}\frac{\sigma_\mathrm{d}}{\sigma_\mathrm{p}} \tag{2}$$