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CALCULATION OF THE ACTIVITIES OF GAMMA-RAY EMITTERS PRESENT IN HOMOGENEOUS CYLINDRICAL SAMPLES

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When the activities of gamma-ray emitters present in samples are calculated from gamma-ray spectra by automated spectral analysis procedures, data describing realistic counting conditions must be used in the procedures in order to obtain reliable results. Therefore parameters such as sample dimensions, composition and density must enter into the calculations as well as the probabilities for coincident detection of photons. To carry out the calculations, two basic approximations are made: the self-attenuation function is calculated supposing a simple detector model and the spatial dependence of the efficiencies for point sources is approximated by exponential functions. The approaches leading to these approximations are briefly described.

Introduction

Nowadays measurement of the activities of gamma-ray emitters is widely performed by gamma-ray spectrometers with semiconductor detectors. The spectrometers sort and register electric pulses generated by gamma-rays in the detectors according their height. The height of each pulse is proportional to the energy of the photon deposited in the sensitive volume of the detector, and since photons from nuclear or atomic decays are emitted at discrete energies, the measured energy distribution exhibits peaks at the emission energies. The count rate in a peak at the energy E is proportional to the activity of the source *a*, and the probability *p(E)* that a photon with energy *E* is emitted in the

$$\frac{N(E)}{T} = p(E)\, a \quad .$$

decay, depositing all its energy in the detector and registering in its peak:
To calculate the activity it is necessary to know the probability for registration.The factors influencing this probability are:

- the dimensions of the source,
- the material of the source,
- the relative position of the detector and source,
- the properties of the detector,
- the probability for emission
- the probabilities for detection of any radiation in coincidence with the photon.

The probability for detection of coincident radiation depends on the properties of the detector, the sample and the decaying nucleus. Therefore the probability for registration can be a complicated function of the probabilities for emission and detection of all radiations emitted simultaneously.

The probability that a photon is detected in such a way that it deposits all its energy in the sensitive volume of the detector is referred to as the peak efficiency. It is influenced by the sample properties, detector properties and the sample-detector arrangement. The influence of the detector properties as well as the influence of the sample material on the efficiency depend on the energy of the photon. Both are determined by the energy-dependent probabilities for interaction between the photon and the materials of the detector and the sample.

The efficiency at a certain energy can be measured by measuring the registration probability in the corresponding peak in cases when the losses due to coincident registration of other radiation are negligible or well known. Such measurements are performed by measuring spectra from calibrated sources emitting photons at several energies. The measured calibration curve is valid just for the specific sample-detector arrangement and sample matrix. In circumstances where samples of different materials are measured in a variety of shapes this approach is not appropriate, since much labour has to be invested in measuring and maintaining the calibration curves. In our laboratory, where samples of various materials are measured and the sample geometries are not known in advance, another approach was developed and it is the aim of this contribution to present it.

The Approach

Calculation of the efficiency

In our laboratory the counting efficiency of homogeneous cylindrical samples is calculated at the time of sample analysis. This requirement sets restrictions on the method of calculation, since the calculation time must be restricted to a few minutes. A semiempirical method is implemented, where the use of geometrical representation is avoided to the largest possible extent. Namely, the language of geometry, speaking in terms of distances and solid angles, is not particularly appropriate in those cases where simultaneous description of sample-detector arrangements with the sensitive volume of the detector much larger than the sample, and arrangements with the sample much larger than the sensitive volume, are required. Instead, the problem is formulated in terms of mathematical analysis.

The efficiency for a point source emitting photons with energy E and placed at a point $\underline{r}$

in the vicinity of the detector may be expressed as [1]:

$$\varepsilon_{PS}(\vec{r},E) = \frac{1}{N(\vec{r},E)} \int_{V_D} dn(\vec{r}, \vec{R},E) \quad ,$$

where $N(\underline{r},E)$ denotes the number of photons with energy E emitted at $\underline{r}$, $dn(\underline{r},\underline{R},E)$ the number of photons emitted at $\underline{r}$ and interacting at $\underline{R}$ within the sensitive volume of the detector V_D in such a way that they deposit all their energy there. The efficiency of an extended homogeneous source is calculated by averaging the point-source efficiencies over the sample volume taking into account the attenuation within the sample [1,2,3,4]:

$$\varepsilon_V(E,\mu) = \frac{1}{V_S} \int_{V_S}\int_{V_D} e^{-\mu(E)\, s(\vec{r},\vec{R})} \frac{dn(\vec{r},\vec{R},E)}{N(\vec{r},E)}\, dV_S(\vec{r}) \quad ,$$

where V_S denotes the sample volume, $\mu(E)$ the linear attenuation coefficient in the source and $s(\underline{r},\underline{R})$ the path in the sample of photons emitted at $\underline{r}$ and interacting at $\underline{R}$. By introducing the efficiency of the point source embedded in an absorbing medium

$$\varepsilon_{PS}(\vec{r},E,\mu) = \frac{1}{N(\vec{r},E)} \int_{V_D} e^{-\mu(E)\, s(\vec{r},\vec{R})}\, dn(\vec{r},\vec{R},E)$$

$$\varepsilon_V(E,\mu) = \frac{1}{V_S} \int_{V_S} \varepsilon_{PS}(\vec{r},E,\mu)\, dV_S(\vec{r}) \quad .$$

the efficiency for an extended source can be to expressed as an average over point sources:

These efficiencies for point sources are difficult to measure and therefore they are not appropriate as a basis for efficiency calculations. To avoid their use the self-attenuation function [1] $F(\underline{r},E,\mu)$ is introduced by rewriting the last equation to the following form:

$$\varepsilon_V(E,\mu) = \frac{1}{V_S} \int_{V_S} \frac{\varepsilon_{PS}(\vec{r},E,\mu)}{\varepsilon_{PS}(\vec{r},E)} \, \varepsilon_{PS}(\vec{r},E) \, dV_S(\vec{r}) = \frac{1}{V_S} \int_{V_S} F(\vec{r},E,\mu) \, \varepsilon_{PS}(\vec{r},E) \, dV_S(\vec{r})$$

The self-attenuation function describes the attenuation of photons within the sample. Since it is defined as a ratio of two point source efficiencies, the detector properties to a large extent cancel out. It depends mainly on the point of emission $\underline{r}$ and the attenuation coefficient μ. The second factor, $\varepsilon_{PS}(\underline{r},E)$, describes the detector properties. This efficiency is measured with point sources on a grid of points within the space near the detector occupied by the sample in order to determine its spatial dependence. It is measured with many different radionuclides in order to determine its energy dependence. The measured efficiencies characterize the detector properties in such a way that they are appropriate for further calculations.

Since the self-attenuation function only weakly depends on detector properties, it is acceptable to calculate it with a simple detector model. If the detector model describes well the attenuation of gamma-rays in the sample at some attenuation coefficient μ, it describes the attenuation as well or better at any smaller attenuation coefficient. To obtain reliable results at large self-attenuations, a detector model describing photon detection at low energies, where the attenuation coefficients are largest, is used in the calculations. In the model the sensitive volume of the detector is replaced by a sensitive surface of the same diameter, placed coaxially with the detector crystal. The surface is placed behind the front surface of the crystal at the average penetration depth of the photons. The quantity $dn(\underline{r},\underline{R},E)$ is supposed to be proportional to the illumination of the surface at the point $\underline{R}$ from a point light source positioned at $\underline{r}$. Consequently, the self-attenuation function is calculated from the expression

$$F(r,h,E,\mu) = \frac{\int_0^{R_c}\int_0^{2\pi} e^{-\mu(E)\frac{h_0}{h(E)}d(\vec{r},\vec{R},E)} \dfrac{R\,dR\,d\phi}{d^3(\vec{r},\vec{R},E)}}{\int_0^{R_c}\int_0^{2\pi} \dfrac{R\,dR\,d\phi}{d^3(\vec{r},\vec{R},E)}} \quad ,$$

where R and φ denote the coordinates on the detector surface where the illumination is observed and represent the integration variables, r is the distance of the source from the symmetry axis of the detector, $h(E)$ is the distance between the source and the detector surface, $h_0/h(E)$ is the part of that distance occupied by the sample material, R_c is the radius of the detector surface and $d(\underline{r},\underline{R},E)$ is the distance between the source and the

$$d^2(\vec{r},\vec{R},E) = r^2 + R^2 - 2\,r\,R\,\cos(\phi) + h^2(E) \quad .$$

point where the illumination is observed:

In the calculation of the counting efficiency the self-attenuation function is computed on a grid of points within the sample, thereafter multiplied by the measured efficiencies and averaged over the sample volume. In this way the material of the sample as well as the sample dimensions are taken into account when calculating the efficiency.

Calculation of the probability for registration

At low count rates the losses in the peaks due to coincident detection of photons are due to detection of two or more photons emitted in a cascade decay, following the formation of the daughter nucleus. The probability for simultaneous detection of photons depends on the details of the deexitation scheme, and therefore it is nuclide-specific and must be calculated for specified counting conditions for each emitter individually. The magnitude of the effect depends on the probabilities that two or more of the radiations emitted in a cascade are registered anywhere in the spectrum. These probabilities are referred to as the total efficiencies.

In a simple decay where just one photon with energy E is emitted in each decay, the probability for registration can be expressed by the probability for emission of the photon $b(E)$, and the efficiency:

$$p_V(E,\mu) = b(E)\ \varepsilon_V(E,\mu) \quad .$$

If the photon is emitted at the point $\underline{r}$ and another photon with energy E_1 is emitted in coincidence with it its registration probability is given as

$$p(\vec{r},E,\mu) = b(E)\ \varepsilon_{PS}(\vec{r},E,\mu)\ [1 - b(E_1)\ \varepsilon^T_{PS}(\vec{r},E,\mu)] \quad ,$$

where $b(E_1)$ denotes the probability for the emission of the photon with energy E_1 when the photon with energy E is also emitted and $\varepsilon_{PS}{}^T(\underline{r},E_1,\mu)$ denotes the total efficiency for the photon with energy E_1. The registration probability for an extended sample is given by the average over the sample volume [5,6]:

$$p_V(E,\mu) = \frac{b(E)}{V_S}\int_{V_S} \varepsilon_{PS}(\vec{r},E,\mu)\ [1 - b(E_1)\ \varepsilon^T_{PS}(\vec{r},E_1,\mu)]\ dV(\vec{r}\,) =$$

$$= b(E)\ \varepsilon_V(E,\mu) - \frac{b(E)\ b(E_1)}{V_S}\int_{V_S} \varepsilon_{PS}(\vec{r},E,\mu)\ \varepsilon^T_{PS}(\vec{r},E_1,\mu)\ dV(\vec{r}) \quad .$$

It is observed that the probability for registration in the peak is expressed by integrals of the products of peak and total efficiencies for point sources. All the integrals can in principle be calculated since all efficiencies for point sources can be obtained from the detector model and the detector characterization data. The number of integrals to be calculated in the case when n states in the daughter nucleus are populated is of the order $4^{(n-1)}/2$ [7]. Therefore approximations must be introduced in order to bring the amount of computing within acceptable limits.

The spatial dependencies of the point source efficiencies $\varepsilon_{PS}(\underline{r},E,\mu)$ and $\varepsilon_{PS}^{T}(\underline{r},E,\mu)$ are

$$\varepsilon_{PS}(\vec{r},E,\mu) = \varepsilon_{00}(E)\ e^{-\phi(E,\mu)\,r^2-\psi(E,\mu)\,z-\omega(E,\mu)\,r^2\,z}$$

approximated by exponential functions [7]:

$$\varepsilon_{PS}^{T}(\vec{r},E,\mu) = \varepsilon_{00}^{T}(E)\ e^{-\phi^T(E,\mu)\,r^2-\psi^T(E,\mu)\,z-\omega^T(E,\mu)\,r^2\,z} \quad ,$$

and

where r and z denote the coordinates of the vector $\underline{r}$. The integral over the sample volume of the exponential functions can be expressed by the exponential integral functions in closed form. Since an exponential function retains its form after multiplication by other exponential functions, the integration of products reduces to evaluation of exponential integral functions at appropriate arguments. By this approximation the probability for registration can be calculated for extended sources in the case of cascade decays, consequently enabling the calculation of activities according to Eq. (1).

Conclusion

The introduction of a simple detector model and the approximation of the spatial dependence of the efficiency for point sources by exponential functions has enabled us to use data describing realistic counting conditions in calculations of activities from peak count rates. The approach described is implemented in an automatic spectral analysis procedure. In this way human resources in the laboratory were freed from correcting results obtained with procedures supposing idealized counting conditions to results taking into account realistic counting conditions. Now the attention of the spectroscopist may be focused on the quality of the measurement, and the reliability and consistency of the results. It is our opinion that good quality analysis cannot be performed without human supervision of the entire measuring process. The main reason for this we see in the first step of the spectral analysis, the peak evaluation procedure. Here the statistical nature of spectral acquisition can introduce data sets which present day routines cannot handle reliably.

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References:

[1] M. Korun, R. Martinčič, *Appl. Radiat. Isot.* **1992**, *43*, 29-35.
[2] M. Noguchi, K. Takeda, H. Higuchi, *Int. J. Appl. Radiat. Isot.* **1981**, *32*, 17-22.
[3] R. M. W. Overwater, P. Bode, J. J. M. de Goeij, *Nucl. Instr. Methods A* **1993**, *324*, 209-218.
[4] O. Sima, C. Dovlete, *J. Radioanal. Nucl. Chem., Letters* **1995**, *200*, 191-203.
[5] K. Debertin, U. Schötzig, *Nucl. Instr. Methods* **1979**, *158*, 471-477.
[6] M. Blaauw, M. J. J. Ammerlaan, S. J. Gelsema, *Nucl. Instr. Methods A* **1997**, *385*, 330-334.
[7] M. Korun, R. Martinčič, *Nucl. Instr. Methods A* **1995**, *355*, 600-608.

Povzetek: Pri računanju aktivnosti sevalcev gama iz spektrov s pomočjo avtomatske analize je pomembno, da podatki, ki opisujejo pogoje metitve, ustrezajo dejanskim merskim pogojem. Zato mora analiza upoštevati podatke kot so dimenzije vzorca, njegova gostota in sestava, pa tudi verjetnosti za hkratno detekcijo več fotonov. Pri računanju aktivnosti upoštevamo dva osnovna približka: Funkcija lastne atenuacije je izračunana v okviru enostavnega modela detektorja, prostorska odvisnost izkoristka za točkaste izvore pa je aproksimirana z eksponentno funkcijo. V prispevku so predstavljeni pristopi, ki vodijo k tema približkom