

ALLOWANCE FOR THE SELF-ABSORPTION OF  $\gamma$  RAYS  
IN THE ACTIVATION ANALYSIS OF BULK SAMPLES

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The self-absorption coefficient is determined numerically from a family of empirical  $\gamma$ -ray absorption curves for cylindrical samples of various thicknesses, atomic weights, and densities.

The use of nuclear-physical methods of analysis to determine the elemental composition of bulk samples is complicated not only by the self-shielding effect associated with exposure of the samples to activating radiation, but also by the effect of induced radiation. This effect is particularly pronounced in the case of low-energy radiation and in the performance of high-precision measurements, where the measurement error cannot exceed 1-2%.

Previous attempts have been made to calculate the total detection efficiencies of Ge(Li) detectors [1] as well as NaI(Tl) and CsI(Tl) scintillation crystals [2-4] for point, disk, and cylindrical radiation sources. A 0.3% error of the computational results is also indicated in [3]. However, none of these calculations takes into account the real measurement conditions, multiple scattering in the sample, or scattering from the detector casing and protective shields. The special case of self-absorption in metal samples during oxygen analysis has been investigated [5]. The samples usually have various geometrical dimensions, atomic weights, and densities; in the present study, therefore, we describe a procedure for taking the self-absorption of  $\gamma$  rays into account in cylindrical samples of equal diameter, but different thicknesses, atomic weights, and densities. Samples of this type are frequently encountered in the analysis of pressed powder pellets or various finely divided objects.

We consider the sample-detector system shown in Fig. 1. The cylindrical sample is placed at a certain distance  $h_0$  from the surface of the detector and has a thickness  $h = h_1 - h_0$ . We partition the sample into plane layers parallel to the upper plane of the detector, each with a thickness  $\Delta h_i$  such that self-absorption can be neglected in the layer. The number of pulses recorded from the sample during the measurement period is

$$N = Bm\eta, \quad (1)$$

The coefficient  $B$  in Eq. (1) takes into account the activation, waiting, and measurement conditions, along with other quantities that do not depend on the geometry and absorbing properties of the sample. The efficiency  $\eta$  of  $\gamma$ -ray detection from the analyzed element depends on the geometry, atomic weight, and density of the sample. Activation analysis, as a rule, is concerned only with the photodetection efficiency of a single maximum-intensity line of the analyzed element.

When the sample is partitioned into layers, the mass of the element in the  $i$ -th layer is equal to  $m_i$ , and the  $\gamma$ -ray detection efficiency from that layer is  $\eta_i$ . The total count under the photopeak of the analytical line is

$$N = B \sum_{i=1}^n m_i \eta_i. \quad (2)$$

If the number of layers is increased and their thickness is decreased accordingly, the mass of the  $i$ -th layer can be written in the form

$$m_i = \frac{dm}{dh} \Delta h_i \equiv m_h \Delta h_i. \quad (3)$$

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The distribution of the element in the sample can be considered uniform in the first approximation, i.e.,  $m_h = \text{const}$ , whereupon the sum in Eq. (2) can be replaced by an integral:

$$N = Bm_h \int_{h_0}^{h_1} \eta(h, \mu) dh. \quad (4)$$

The detection efficiency for the given measurement geometry will depend on the thickness of the sample and the linear attenuation coefficient of the  $\gamma$ -ray analytical line. The linear density (mass per unit thickness) of the analyzed element can be found from Eq. (4):

$$m_h = \frac{N}{B \int_{h_0}^{h_1} \eta(h, \mu) dh}. \quad (5)$$

The coefficient B in Eq. (5) depends on the irradiation, waiting, and measurement conditions and, as a rule, is taken into account in activation analysis by using a standard sample with a known mass  $m_0$ . It can be made in the form of a disk with a diameter equal to the diameter of the sample and with a thickness such that the attenuation of characteristic radiation will not exceed 0.1-0.2%. The standard sample can be a foil of the analyzed element or a film containing a known quantity of it.

The standard sample is placed at a height  $h_0$  from the detector, in which case the number of pulses under the photopeak is

$$N_0 = Bm_0 \eta(h_0, \mu_0). \quad (6)$$

In Eq. (6),  $\mu_0$  can be taken equal to zero, because radiation is attenuated only in air and in the detector casing and does not depend on the sample. The value of B determined from (6) is substituted into (5), whereupon

$$m_h = m_0 \frac{N}{N_0} \frac{1}{\frac{1}{\eta(h_0, \mu_0)} \int_{h_0}^{h_1} \eta(h, \mu) dh}. \quad (7)$$

The coefficient  $\eta(h_0, \mu_0)$  does not depend on h and can be taken outside the integral sign, so that we obtain for the linear mass density of the element

$$m_h = m_0 \frac{N}{N_0} \frac{1}{I}, \quad (8)$$

where

$$I = \int_{h_0}^{h_1} \frac{\eta(h, \mu)}{\eta(h_0, \mu_0)} dh. \quad (9)$$

The integrand in Eq. (9) represents the relative detection efficiency for the individual layer. Consequently, in order to determine it we plot a family of experimental curves  $\eta(h, \mu)_{\mu=\text{const}}$ , where the parameter of the family is the linear absorption coefficient  $\mu$ . The geometry of the measurements is the same as in Fig. 1.

Curves of the detection efficiency as a function of the absorber thickness for a given  $\gamma$ -radiation line are recorded for a number of absorbers (air, plastic, aluminum, pressed  $\text{BaSO}_4$ , iron, copper, cadmium, and lead). If this line belongs to a long-lived radionuclide, a series of measurements is carried out using that radionuclide, which is prepared in the form of a thin flat source with a diameter equal to that of the sample. If the line belongs to a short-lived radionuclide, a long-lived counterpart is chosen with an energy differing 5-10 keV from the analytical. In the absence of a suitable radionuclide it is permissible to use three nuclei with energies differing a few tens of kiloelectron-volts from the analytical and then to find the detection efficiency of the given line by interpolation.

Figure 2 shows a family of curves  $\eta(h, \mu)_{\mu=\text{const}}$ , corresponding to a  $\gamma$ -radiation energy of 279 keV, which is associated with the radionuclide  $^{203}\text{Hg}$ ; this family is used to determine the gold content from the isomer  $^{197\text{m}}\text{Au}$ , which has the same  $\gamma$ -radiation energy. The diameter of the sample is 44 mm, the detector is an NaI(Tl) scintillator with a diameter of 40 mm and a thickness of 40 mm, and the distance from the sample to the detector is  $h_0 = 12$  mm.

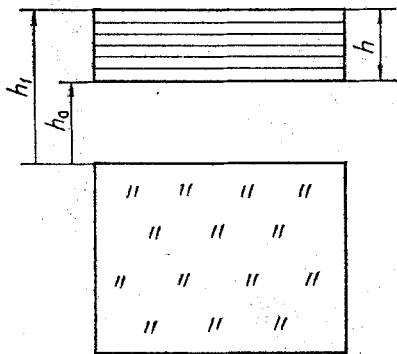


Fig. 1

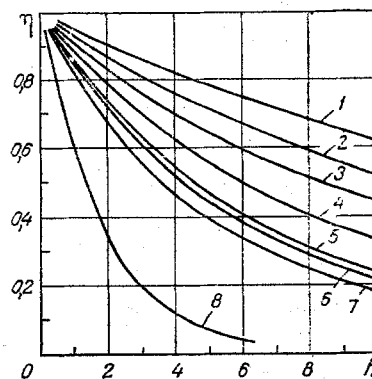


Fig. 2

Fig. 1. Geometry of the sample and detector.

Fig. 2. Family of curves of the self-absorption coefficient  $\eta$  (relative units) vs the absorber thickness  $h$  (mm) for various values of  $\mu$  ( $\text{cm}^{-1}$ ): 1)  $\mu = 0$ ; 2) 0.120; 3) 0.255; 4) 0.475; 5) 0.766; 6) 0.901; 7) 1.279; 8) 4.420.

A large statistical sample is not sufficient for obtaining high measurement precision, because the main source of error is background-subtraction error. This error is appreciable in measurements on bulk samples, in which  $\gamma$  rays suffer small-angle Compton scattering to a significant degree. A plateau is observed to the left of the photopeak, its height increasing with the thickness and density of the sample. An experiment has shown that the rms deviation of the results obtained for background subtraction with a sample density  $\rho \approx 8 \text{ g/cm}^3$  and a thickness of 10 mm attains 0.4% at a  $\gamma$ -radiation energy of 0.3 MeV. Consequently, in plotting the curves  $\eta = \eta(h, \mu)$  a fully sufficient number of pulses under the photopeak is  $N = (2-3) \cdot 10^5$ . The precision can be improved by repeating the measurement at each point or by processing the experimental curves on a computer using the least-squares method.

The resulting absorption curves can be described by fourth-degree polynomials. In particular, the curve corresponding to  $\mu = 0.901 \text{ cm}^{-1}$  and the above-indicated conditions is described by the polynomial

$$\eta(h) = 0.9999 - 1.5310 \cdot 10^{-1}h + 9.792 \cdot 10^{-3}h^2 - 1.7933 \cdot 10^{-4}h^3 - 5.0659 \cdot 10^{-6}h^4. \quad (10)$$

The rms deviation of the description of the absorption curves by fourth-degree polynomials does not exceed 0.3% for single measurements at each point with the indicated statistical sample. The measurement step is 1 mm.

To facilitate the computation of the integral I we use the data of Fig. 2 to plot a family of curves  $\eta(\mu, h)_{h=\text{const}}$ , where the parameter is now the thickness of the sample, and  $\mu$  is the independent variable. This family, processed by the least-squares method, is shown in Fig. 3. The values of  $\mu$  for the absorbers must be measured with adequate precision, because the error of handbook reference data is no better than 5% [6] and the measurement geometry in analysis is far from good. The influence of the error of determination of  $\mu$  is minimal if the measurements of  $\mu$  and the element analysis are carried out using spectrometers with close values of the energy resolution.

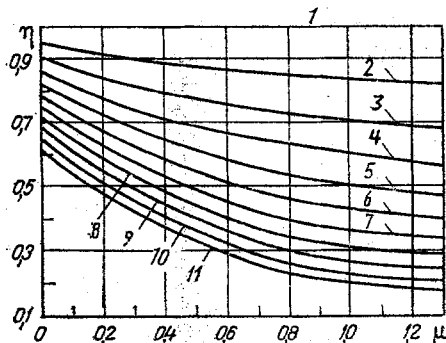


Fig. 3. Family of curves of the self-absorption coefficient  $\eta$  (relative units) vs the linear attenuation coefficient  $\mu$  ( $\text{cm}^{-1}$ ) for various values of  $h$  (mm): 1)  $h = 0$ ; 2) 1; 3) 2; 4) 3; 5) 4; 6) 5; 7) 6; 8) 7; 9) 8; 10) 9; 11) 10.

Experiments have shown that the values of  $\mu$  can be obtained with a 0.5-0.7% rms deviation during a measurement period of the order of a few minutes, which contributes on the order of 0.2-0.3% to the error of determination of the detection efficiency (for a confidence interval  $p = 0.67$ ).

The integral  $I$  is computed according to the Newton-Cotes formula with  $n = 4$ . The fourth-degree polynomial is integrated exactly in this case. The thickness  $h$  and the coefficient  $\mu$  of the sample are measured in the same geometry as in plotting the family  $\eta = \eta(h, \mu)_{\mu=\text{const}}$ . The thickness  $h$  is divided into four equal intervals  $\Delta h$ , and the values obtained for the reference points  $\Delta h \cdot i$  are used to find the values of  $\eta_i(\Delta h \cdot i, \mu)$  on the family  $\eta = \eta(\mu, h)$  that correspond to the measured value of  $\mu$  (Fig. 3). The number of curves of this family must be sufficient to ensure the prescribed interpolation accuracy. The family shown in Fig. 3 can guarantee a 3% rms reading deviation. To obtain an rms deviation of 0.2-0.3%, it is necessary to plot 10 curves between each of those shown, using empirical formulas for the polynomials  $\eta = \eta(h, \mu)_{\mu=\text{const}}$ .

The integral in (9) is computed according to the quadrature formula

$$I = \frac{h}{45} (14\eta_0 + 64\eta_1 + 24\eta_2 + 64\eta_3 + 14\eta_4), \quad (11)$$

in which  $\eta_i$  denotes the values of the efficiencies read from the family  $\eta = \eta(\mu, h)_{h=\text{const}}$  and corresponding to the partition points  $h_i = \Delta h \cdot i$ .

The procedure described here for taking self-absorption into account has made it possible to attain an rms analysis deviation of 1.5% in determining the macroscopic quantity of gold in electroplating-industrial bulk samples when all other errors of indirect measurements are equal.

#### NOTATION

$N$ , count of  $\gamma$  quanta;  $m$ ,  $m_h$ , mass and linear mass density of analyzed element, respectively;  $\eta$ , detection efficiency;  $i$ , order number of layer;  $n$ , number of layers in partition;  $h$ , thickness of sample;  $\Delta h_i$ , thickness of  $i$ -th layer;  $\mu$ , linear coefficient of  $\gamma$ -ray attenuation;  $I$ , self-absorption coefficient.

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