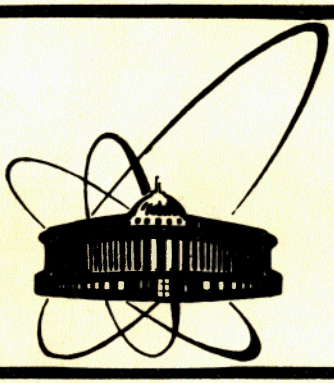

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E18-83-200

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APPLICATION OF A NEW STANDARDIZATION METHOD IN ACTIVATION ANALYSIS WITH REGISTRATION OF SOFT GAMMA RADIATION

Submitted to "Journal of Radioanalytical Chemistry"

1983

INTRODUCTION

During the several last years it was shown that the detection of low-energy radiation (several tens of KeV) following the sample activation provided an attractive approach for analysis of a wide range of elements [1-4]. Nevertheless, the strong self-absorption of such radiation in the matter of samples makes it necessary to use the time-consuming and laborious method of Internal-Standard Addition. This situation limited the application of the above-mentioned procedure of activation analysis for the rapid determination of the element content in numerous samples with different matrices. In the present work we report the application of the new standardization method [5] that makes it possible to avoid these difficulties.

THEORETICAL

The new standardization method developed in ref. [5] can be briefly outlined as follows. Let us consider a cylindrical sample (with radius R and height H) and point detector situated at the extension of the cylinder axis at a distance of x_0 from its bottom. Let ρ, z, ϕ be the cylindric coordinates of the samples. Then the radiation intensity of the radioactive nuclide dispersed in the sample homogeneously is [6]:

$$P_c = 2\pi\Gamma_\gamma qR \int_0^1 \int_0^k \frac{m\,dm\,dn}{(n+p)^2+m^2} \cdot \exp\left\{-\mu R \frac{n}{n+p}\sqrt{(n+p)^2+m^2}\right\} = \quad (1)$$

$$= 2\pi\Gamma_\gamma qR\, G_2(k, p, \mu R),$$

where $m = \rho/R$, $n = z/H$, $p = x_0/R$, $k = H/R$, q is the volumetric density of the radionuclide, μ, Γ_γ are, respectively, the linear attenuation coefficient and the dimension-constant for the considered radiation with a given energy.

Let us consider the standard form of a disc with the radius r. Then the registered radiation intensity of the radionuclide, distributed uniformly on the disc surface is given by

$$P_d = \pi\sigma\Gamma_\gamma \ln[1 + (r/y_0)^2], \quad (2)$$

where σ is the surface density of the radionuclide, y_0 is the distance from the disc centre to the detector situated at the

extension of the disc axis. We assume that the attenuation of radiation by the disc matter is negligible. As the disc standard differs from the internal one that has the same shape and absorption as a real sample, so corrections for these factors were made by replacing the experimental value of P_d by

$$kP_d = \frac{2qRG_2(k, p, \mu R)}{\sigma \ln[1 + (r/y_0)^2]}, \qquad (3)$$

where k is the correction factor, q and σ satisfy the relation $r^2\sigma = R^2 qH$.
Thus

$$k = \frac{2r^2 G_2(k, p, \mu R)}{RH \ln[1 + r/y_0)^2]}, \qquad (4)$$

the content x of the considered nuclide is calculated from the following equation

$$x = \frac{mN_0 RH \ln[1 + (r/y_0)^2]}{2MN_1 r^2 G_2(k, p, \mu R)}, \qquad (5)$$

where M is the sample weight, m is the amount of the element distributed on the disc standard, N_0 and N_1 are, respectively, the measured intensities of the activated sample and the disc-standard for the analytical peak used. Eqs. (1)-(5) are valid for a point detector. As semiconductor detectors widely used for radiation detection are rather large, it is necessary to use the concept of the effective centre. In other words, we consider the detector as a point one situated at its effective centre. The distance from it to the disc or a cylindrical-shape sample can be obtained by measuring the intensity of the activated disc with the radius R (for a cylinder) or r (for a disc), located at different distances from the detector surface. Using the least-squares method the values of y_0 and x_0 can be obtained from Eq. (2). It is however more convenient to choose $R = r$ and, therefore, $x_0 = y_0$. In order to use Eq. (5) for practical calculations one must know the value of μ. It can be determined in the following manner. Let the irradiated disc be located on an inactivated sample. We can consider such a geometry as a barrier one. So, the radiation intensity registered by the detector can be expressed as:

$$P_s = 2\pi\Gamma_\gamma \sigma [Ei(-\mu H\epsilon) - Ei(-\mu H)], \qquad (6)$$

where $Ei(x) = \int_{-\infty}^{x} \frac{e^u}{u} du$ is an exponential integral function, ϵ is a calibration constant. The latter can be determined in an experiment with a substance for which μ is well known, for example, aluminium or water. From Eqs. (2) and (6) we have

$$P_s/P_d = 2[Ei(-\mu H\epsilon) - Ei(-\mu H)]/\ln[1 + (r/y_0)^2]. \qquad (7)$$

From this equation the values of ϵ and then μ for samples can be easily determined.

EXPERIMENTAL

To test the advanced method the activation analysis of man-made uranium samples has been performed. In order to have a wide range of μ samples were prepared by mixing H_3BO_3 powder with $BiCl_3$ in different proportions. The stock solution of uranium was prepared by weighing a definite amount of $UO_2(NO_3)_2 \cdot 2H_2O$ and dissolving it in distilled water. Different aliquout amounts of the stock solution were added to the samples with a micropipet. Elemental concentrations were chosen to give good counting statistics, but without excessive activity. After evaporation to dryness by gentle heating, the samples were carefully mixed and put into cylindrical plexiglass containers with the radius R = 10 mm and height H = 6 mm. To minimize radiation absorption the container ends were closed by mylar film 10 μm thick. The standards were prepared by accurately micropipetting a definite amount of a uranyl nitrate solution on to a filter paper disc with the same radius as that of the sample container. For irradiation the discs are placed at both ends of the container to take into account the gradient of the irradiation flux.

IRRADIATION

In one run of irradiation ten containers filled with samples each weighing about 1.5-3 g, as well as disc-standards, were placed into an irradiation canister in the "Sandwich" manner: disc - sample - disc - sample, etc., so that their axis coincided. The irradiation was carried out along the axis by bremsstrahlung beam from the microtron MT - 22 under the following conditions: the electron beam energy - 20.5 MeV, mean electron current - 10-12 μA, bremsstrahlung intensity at a distance of 1 m from target - 6000 roentgens per minute, the irradiation time - 3 hours.

GAMMA-RAY SPECTROMETRY

The uranium content was determined according to the 59.54 keV photo-peaks of the isotope ^{237}U,, obtained in the reaction $^{238}U(\gamma,n)^{237}U \xrightarrow[6.75d]{\beta^-} {}^{237}Np$. The spectra were registered by a Princeton Gamma-Tech intrinsic Ge detector of 2.1 cm^3 active volume and 7 mm thick with a resolution of about 500 eV (FWHM) at 122 keV. Information was accumulated in a LP - 4900 multichannel pulse-height analyzer (NOKIA ELECTRONICS). The spectra were treated using the program PROBA /7/ designed to calculate the energy, area, FWHM and error for each peak found.

MEASUREMENTS AND CALCULATION OF THE URANIUM CONTENT

To reduce the measuring time for the effective centre x_0, the calibration constant ϵ and linear attenuation coefficient a disc with a large amount of uranium (10 mg) was irradiated. At first the disc intensity dependence on the distance from the detectors surface was measured for the 59.54 keV line (fig.1) and x_0 was determined. Then the disc intensity was measured for the cases in which it was situated on the detector and then on a water-filled container. By using the above obtained value of x_0 the constant ϵ was found from Eq. (7). Finally the disc was placed on each sample, the 59.54 keV photo-peak intensity was measured and the coefficient μ was obtained from Eq. (7). Note that all these manipulations were carried out before the sample irradiation. After that the areas of the 59.54 keV photo-peaks for the samples and standard-discs were determined. All the calculations to determine the uranium content were performed using a computer FORTRAN program consisting of a subroutine for

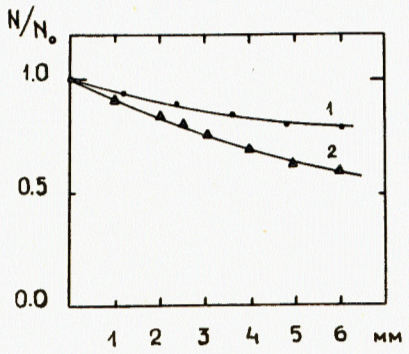

Fig.1. The relative intensity of the disc standard (with radius r = 10 mm) as a function of the distance from the detector surface. 1 - 208.00 keV, Ge(Li) detector, 2 - 59.54 keV, Ge - in detector, N_0 - the disc intensity on the detector surface.

x_0 evalution by the least-squares method, a subroutine for obtaining the ϵ and μ values, a subroutine for calculating the double integral G_2 and, finally, for determining the uranium content from Eq. (5). As we used two discs placed on the container ends as a standard for a given sample, in Eq. (5) N_1 is the sum of their peaks areas while m is the total amount of uranium on the discs.

RESULTS AND DISCUSSION

The analyses of eighteen samples were carried out using the 59.54 keV photo peak and, for comparison, of five samples, using the 208 keV photopeak due to ^{237}U. The spectra of these five samples were registered by a 25 cm^3 Ge(Li) detector having 2.5 keV resolution for the ^{60}Co 1332 keV peak. The linear attenuation coefficient varied in a wide range from 0.01 cm^{-1} to 5.01 cm^{-1}. Because the $BiCl_3$ and H_3BO_3 powders were mixed with uranyl nitrate in a mortar and only part of this mixture was put into the sample container for irradiation, the uranium content of the sample differs from the calculated one due to unavoidable inhomogeneity. So, the linear regression analysis was chosen to test the reliability of this advanced method. The correlation between the activation analysis and the calculated content is shown in Fig.2. The correlation coefficient is equal to 0.98 and a slope in the regression equation differs from unity by 3.6%. Figure 2 illustrates the inaccuracy of the external standard method too. As the sample μ becomes larger, an error in the content value determined by this method increases. For example, for a sample with μ = 0.06 cm^{-1} an error was 9.8% while for μ = 5.01 cm^{-1} it amounts to 78.8%.

From the results obtained in this work it can be concluded that the standardization technique proposed in ref. /5/ can be

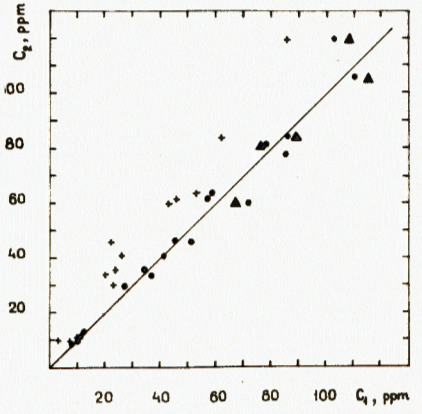

Fig.2. The correlation of the uranium content. C_1 - the content calculated from sample preparation manipulations, C_2 - the content obtained using activation analysis with application of the new standardization method (● - 59.54 keV, ▲ - 208.00 keV) and the method of external standard (+ - 59.54 keV).

used for mass activation analysis involving the registration of low-energy radiation instead of the laborious internal - standard method. Further efforts must be devoted to its simplification, especially in calculation procedures to meet the needs of ordinary analytical laboratories.

REFERENCES

1. Weise H.-P., Segebade Chr. J.Radioanal. Chem., 1977, 37, p. 195.
2. Amiel S., Mantel M., Alfassi Z.B. J.Radioanal.Chem., 1977, 37, p. 189.
3. Vo Dac Bang, Zamiatnin Ju.S., Truong Thi An, JINR, 18-80-407, Dubna, 1980.
4. Henderson P., Williams C.T. J.Radioanal. Chem., 1981, 67, p. 445-452.
5. Vo Dac Bang, USSR authors's certificate No.973249, Bull., OIPOTZ, 1982, 41, p. 196.
6. Gusev N.G. et al.Zashchita ot Izlucheniya Protyazhennykh Istochnikov, Atomizdat, M., 1961, p. 25.
7. Phillippot J.Ck. IEEE, Trans. NS-17, 3, 1970, p. 446.

Received by Publishing Department
on March 31, 1983.

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E18-83-200

Применение нового метода эталонирования при радиоактивационном анализе по мягкому гамма излучению

Описывается применение нового метода эталонирования при радиоактивационном анализе по мягкому гамма излучению. Метод позволяет проводить массовый анализ без применения трудоемкой процедуры приготовления внутренних стандартов.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1983

Vo Dac Bang, Phan Thu Huong
E18-83-200

Application of a New Standardization Method in Activation Analysis with Registration of Soft Gamma Radiation

An application of the new standardization method for rapid activation mass analysis with the registration of the strongly absorbed low-energy gamma radiation is described. This method makes it possible to avoid the use of the time-consuming and laborious method of Internal Standard.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1983