Neutron Activation Analysis at IREN and IBR-2 Facilities

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Abstract

The installation for neutron activation analysis (NAA) at the IREN facility, which consists of HPGe detector, a sample changer, chemical equipment, and control software, is described. Some results obtained by the NAA method, which have been carried out at IREN and IBR-2 facilities are presented. The neutron spectrum obtained at these facilities has big part of resonance neutrons. A program was developed for the calculation of effective resonance integrals, which takes into account the real spectrum of resonance neutrons, as well as the thermal motion of atoms and the absorption of neutrons in the sample. The flux densities for thermal and resonance neutrons have been determined.

Introduction

Neutron activation analysis (NAA) is a method for determining the elemental composition of a substance, based on measuring the characteristics of radiation emitted by radioactive nuclei that appear after neutron irradiation. The first activation of various elements by neutrons was observed by E. Fermi in 1934. G. de Heversey and H. Levi in 1936 first applied this phenomenon to determine the content of elements in the samples. At present, neutron activation analysis is widely used in geology, ecology, archeology and other fields of research [1]. The main process of interaction of neutrons with nuclei, leading to the formation of unstable nuclei, is radiation capture, that is, the capture of neutrons followed by the emission of gamma quanta. NAA allows us to determine the content of more than 40 elements with high sensitivity.

Neutron sources in FLNP and installation for activation analysis

IREN (Intensive Source of Resonance Neutrons) consists of linear electron accelerator and a tungsten target (4 cm diameter and 10 cm height), surrounded by a moderator. Electrons enter the target, lose energy and emit bremsstrahlung quanta in a wide range of energies. The number of emitted gamma rays decreases with increasing energy. The maximum energy of gamma rays is equal to the kinetic energy of the electrons. The neutrons are created as a result of the reactions (γ, n), (γ, 2n), etc. The average energy of the emitted neutrons is 1 MeV. To obtain neutrons of lower energies, the target is surrounded by a moderator. The water is used as moderator, which also cools the target. The moderator is a cylinder with a diameter of 15 cm. Neutrons pass in it a distance of the order of 5 cm [2].

At present, the maximal electron energy is equal to 50 MeV, average current approximately 5 μA. A total number of neutrons is equal to $2 \cdot 10^{11}$ 1/sec approximately.

The installation for automation of measurement of the spectra of induced activity during neutron activation analysis at IREN facility have been created. The installation consists of HPGe detector (Canberra type, 40% relative efficiency), chemical equipment, sample changer, NAA databases and software (see fig. 1).

Figure 1. Automatic system for measuring the spectra of induced activity.

The capacity of the sample changer is 45 containers. Control software allows you to simultaneously control four sample changers. The database provides storage of all necessary information about NAA. At different stages of the NAA, information is exchanged between the database and the various programs used for automation of NAA, as well as employees participating in the NAA. We use the GENIE-2000 processing program [3].

At FLNP the irradiation unit on the 3rd channel of the IBR-2 has been used for investigations also [4].

Determination of the neutron flux density

The cross section for the capture of neutrons at low energies follows the law 1/v, where v is the neutron velocity, that is $\sigma_{n\gamma} \propto \frac{1}{\sqrt{E_n}}$. At higher energies, neutron resonances are formed. Neutrons which have different energy are divided into several groups. There are thermal neutrons (mean energy 0.025 eV), resonance (from 0.5 to $5 \cdot 10^5$ eV) and fast ones. The spectrum of thermal neutrons is Maxwellian, and the flux density of resonance neutrons decreases in inverse proportion to the neutron energy in the first approximation. Experimentally, thermal and resonance neutrons are separated by cadmium, since the 113Cd isotope has a huge thermal neutron capture cross-section (of the order of 20,000 b) and a small capture cross section for resonant neutrons. The cadmium boundary is 0.5 - 0.55 eV (depending on cadmium thickness). The main contribution to the formation of radioactive nuclei is provided by thermal and resonance neutrons.

The neutron flux density in the energy range from 10^{-3} to 10^{5} eV can be represented:

$$\varphi(E_n) = \Phi_{th} f_M(E_n, T) + \frac{\Phi_{res}}{E_n^{1-\alpha}}. \quad (1)$$

Here the contributions of thermal and resonance neutrons are separated. Φ_{th} is the thermal neutron flux, the function ***f_M (E, T)*** describes the Maxwell distribution. The second term describes the resonance neutrons. In the first approximation, $\alpha = 0$ (Fermi spectrum). Thus, Φ_{res} is the neutron flux density at 1 eV. Usually, the parameter α is equal to zero. For IREN and IBR-2 facilities this parameter is equal to the value of $\alpha = 0.1$ approximately.

It is necessary to determine the fluxes of thermal and resonance neutrons. We use the method of cadmium difference. To do this, together with the sample irradiated indicators, that is, elements with well-known cross-sections. Indicators of the same type are irradiated in a shell from Cd and without it under the same conditions. From measurements of the activity of an indicator of mass m2 irradiated in cadmium protection, one can determine Φ_{res}:

$$\Phi_{res} = \frac{N_\gamma \cdot M \cdot \lambda \cdot e^{\lambda t_{d2}}}{m_2 \cdot N_A \cdot \gamma \cdot \varepsilon \cdot \theta \cdot I_{res} \cdot [1-\exp(-\lambda t_{irr})] \cdot [1-\exp(-\lambda t_{meas})]}. \quad (2)$$

Measurements of the activity of the indicator irradiated without cadmium gives a value proportional to the value of $X = \Phi_{th}\sigma_{th} + \Phi_{res}\sigma_{res}$, since the activation is produced by both thermal and resonant neutrons. Measurement of the activity of indicators irradiated in Cd protection and without it is carried out at the same time. This means that it is necessary to subtract the contribution of resonant neutrons. As a result, we get:

$$\Phi_{th} = \frac{X - \Phi_{res} \cdot I_{res}}{\sigma_{th}} = \frac{r}{\sigma_{th}} \left(N_{\gamma 1} \frac{\exp(\lambda t_{d2})}{m_1} - N_{\gamma 2} \frac{\exp(\lambda t_{d2})}{m_2} \right), \quad (3a)$$

where the common factor is:

$$r = \frac{M \cdot \lambda}{N_A \cdot \gamma \cdot \varepsilon \cdot \theta \cdot [1-\exp(-\lambda t_{irr})] \cdot [1-\exp(-\lambda t_{meas})]}. \quad (3b)$$

As indicators, gold, copper, zirconium, and others are usually used. Data on interaction cross sections, half-lives, and yields of individual gamma lines can be found in [5,6].

Calculations of the effective resonance integrals

Because of the spectrum of resonance neutrons at our facilities differ from Fermi spectrum we calculate the effective resonance integrals using experimental data for resonances. The effective resonance integral is equal to:

$$I_{res}(\alpha) = \frac{1}{n} \int_{E_{Cd}}^{\infty} \frac{\sigma_{n\gamma}}{\sigma_t} [1 - \exp(-\sigma_t \cdot) n] \frac{dE}{E^{1-\alpha}}. \quad (4)$$

Here n –sample thickness, $\sigma_{n\gamma}$, and σ_t are the radiative capture and total cross-sections, which are the sum of a number of resonances. Every resonance is described by Breit-Wigner formula.

It is possible to present the resonance integral in the next form:

$$I_{eff.res} = I_v(\alpha) + \frac{\pi}{2} \sum \frac{\sigma_{0i} G_i}{E_{ri}^{1-\alpha}} \Gamma_{\gamma i}. \quad (5)$$

Table 1. The calculated values of the effective resonance integrals

Nuclide	Number of resonances	Resonance integral, b [5]	Effective resonance integral, b	
			α = 0	**α = 0.1**
^{197}Au	24	1550 ± 28	1577	1849
^{94}Zr	18	0.23 ± 0.01	0.30	0.78
^{97}Zr	18	5.3 ± 0.3	6.43	19.5
^{63}Cu	24	4.97 ± 0.08	4.95	9.52

The first term describes contribution from thermal cross-section tail (according to *1/v* low). The second term describes the contribution of resonances. $\sigma_0 = \frac{4\pi}{k^2} g_J \frac{\Gamma_n}{\Gamma}$ is the total capture cross-section where $E = E_{ri}$, E_{ri} – resonance energy, Γ_n – neutron width, Γ_γ – radiative width, $\Gamma = \Gamma_n + \Gamma_\gamma$ is total width, g_J is the statistical factor and k is neutron impulse. The factors G_i depend on sample thickness and temperature. $\sigma_{n\gamma} = \sigma_0 \cdot \frac{\Gamma_\gamma}{\Gamma} \psi(\theta, x)$, where function $\psi(\theta,x)$ describes the thermal motion of atoms, $x = \frac{2(E-E_r)}{\Gamma}$, $\theta = \frac{\Gamma}{\Delta}$. $\Delta = \sqrt{\frac{4E_r k_B T}{A}}$ is the Doppler width. The Padé approximation has been used to calculate the $\psi(\theta,x)$ function [7]. Resonance parameters were taken from [8].

The calculations show the big difference (up to tens of percent) between calculated values and known from literature ones. The calculated values have been used for accurate determination of the neutron flux density and for calculations of concentrations of elements by the absolute method.

Conclusions

As a result of the measurements, the following neutron flux density values were obtained at the IREN facility:

$$\Phi_{th}=1{,}0\cdot10^{8}\ \text{n/(cm}^2\cdot\text{s)}\ \text{and}\ \Phi_{res}=1{,}1\cdot10^{7}\ \text{n/(cm}^2\cdot\text{s)}.$$

In our laboratory, the method of activation analysis was used to study ecological samples from Egypt [9] and for investigations of archeological samples – bracelets and fragments of human remains from the burials of the Moscow Kremlin [10].

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