COMPUTATIONAL METHODS FOR NEUTRON-ACTIVATION ANALYSIS OF LARGE OIL SAMPLES


An absolute method for performing neutron-activation analysis of large oil samples is presented. Software has been developed for performing a Monte Carlo calculation (based on nuclear-physical data in the ENDF and ENSDF format) of the distribution of the neutron flux and activation of the analyte nuclides in large samples. A method and software have been developed for calculating the detection efficiency for $\gamma$-rays from a large sample by a planar detector. The coefficient $\alpha$ of deviation of the epithermal neutron flux from the $1/E$ law is determined to be $\alpha = 0.023$. The content of 38 elements in samples of raw oil samples from a deposit in Eastern Siberia in the concentration range from $10^{-9}$ to 0.5% is determined.

Natural and artificial hydrocarbons are widely used in the power-generation and chemical industries, which results in significant use of raw oil and oil products. Chemical catalysts such as Fe, Co, Ni, V, W, Mo, Pt, and Pd on the one hand and the elements N, P, As, Sb, Bi, O, S, Se, and Te on the other are used to refine the oil. The elemental content of raw oil varies from $10^{-9}$ to 0.5%. For this reason, a determination of the content of elements in raw oil will make it possible to adjust the technological process of oil refining and to conserve expensive materials, such as, for example, platinum and palladium.

Ordinarily, samples with point geometry (small volume or mass) are used in instrumented neutron-activation analysis to reduce self-screening of the neutron flux inside a sample. However, the intensity of $\gamma$-lines of the analytical nuclides in the spectrum of a sample and, therefore, the sensitivity of their determination can be increased by increasing the mass of the sample to definite limits. This technique was used in the present work to improve the limit of detection and increase the number of elements determined.

The Monte Carlo method is a probabilistic one in which the path of a neutron is followed from creation to absorption or escape from the sample. All interaction processes involving the neutron are modeled. This makes it possible to model processes in complicated multi-element systems. For example, this method has been used to model the interaction of a neutron with the sample when measuring chlorine by means of neutron-radiation analysis [1] and estimating the neutron and $\gamma$-fluxes in soil wetted by pure and salt water as well as raw oil [2]. In the present work, this method was used to determine the neutron flux and specific activity of elements in large volumes of oil.

Description of the Software. The program developed was used to calculate the distribution of the thermal and epithermal neutrons in the sample and the energy dependence of the cross sections of the nuclear reactions $(n, \gamma)$, $(n, n)$, and $(n, f)$. The ENDF/B VII.0 and ENSDF (BNL-NCS-51655-01/02-Rev) nuclear-physical data are used in the program. The cross sections of the following nuclear reactions are calculated using the single-level Breit–Wigner formalism [3, 4]:

$$\sigma_\gamma = \sum_l \sum_r \sigma_{mr} \frac{\Gamma_r}{\Gamma_n} \Phi(\theta, x),$$

for radiative capture.

for the fission process,

\[ \sigma_f = \sum \sum \sigma_{\text{nr}} \frac{\Gamma_f}{\Gamma_r} \Psi(\theta, x), \]

for elastic scattering,

\[ \sigma_n = \sigma_p + \sum \sum \sigma_{\text{nr}} \left[ \cos 2\varphi_f - \left( 1 - \frac{\Gamma_{\text{nr}}}{\Gamma_r} \right) \right] \Psi(\theta, x) + \sin 2\varphi_f \chi(\theta, x), \]

where \( \sigma_{\text{nr}} = 4\pi/k^2 g_j \Gamma_{\text{nr}}/\Gamma_r \) is the cross section at the peak of the resonance; \( k = 2.196771 \times 10^{-3} A/(A + 1)E^{1/2} \) is the wave number of the incident neutron as a function of its energy \( E \) and the ratio of the atomic mass \( A \) to the neutron mass for the isotope under study; \( g_j \) is the statistical weight; \( \Gamma_{\text{nr}}, \Gamma_{\gamma r}, \Gamma_{f r}, \) and \( \Gamma_r \) are the neutron, radiation, fission and total width of the resonance, respectively; \( \sigma_p \) is the cross section for potential scattering of neutrons; \( l \) is the orbital angular momentum; and \( \varphi_f \) is the phase shift. The temperature dependence of the shape of a resonance line is determined by the functions \( \Psi \) and \( \chi \), which have the form

\[ \Psi = \frac{\sqrt{\pi}}{2} \theta \Re W \left( \frac{\theta x}{2}, \frac{\theta}{2} \right); \quad \chi = \frac{\sqrt{\pi}}{2} \theta \Im W \left( \frac{\theta x}{2}, \frac{\theta}{2} \right), \]

where \( \theta = \Gamma_r/(4kTE/A)^{1/2}; \quad x = (E - E_r')/\Gamma_r; \) \( T \) is the absolute temperature; \( k \) is Boltzmann’s constant; and \( E_r' \) is the reduced resonance energy.

The multilevel Breit–Wigner formalism is used to take account of resonance interference. In this case, the resonances are described by the same relations except that the term describing the interference of resonances is included in the relation for the elastic scattering cross section:

\[ \sigma_n^1(E) = \frac{\pi}{k^2} \sum g_j \sum \sum \frac{2\Gamma_{\text{nr}} \Gamma_{\text{ns}} ((E - E_r')(E - E_r') + (\Gamma_r / 2)^2)}{[(E - E_r')^2 + (\Gamma_r / 2)^2][(E - E_r')^2 + (\Gamma_r / 2)^2]}, \]

where \( \sigma_n^1(E) \) is the cross section of the process calculated as the sum of the resonance component and the pedestal \( \sigma(E) \), calculated from the data in section 3 of the ENDF file.

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The parameters of the angular distribution of the elastically scattered neutrons and inelastically scattered neutrons with excitation of the discrete levels of a nucleus are described in the form \( f(\mu, E) \) of the probability that the incident neutron with energy \( E \) will be scattered in the interval \( d\mu \) with an angle whose cone equals \( \mu \) in the laboratory coordinate system or in the center-of-mass system. The following nonrelativistic kinematic relations were used in the program to describe the neutron scattering in the center-of-mass system (Fig. 1) and in the laboratory coordinate system (Fig. 2):

\[ A = \frac{m_2}{m_1}; \quad A' = \frac{m_3}{m_1}; \quad \gamma = \frac{A'}{A + 1 - A'}; \quad \beta = \left( \frac{A(A + 1 - A')}{A'} \right)^{1/2} \left( 1 + (1 + A)Q \right)^{1/2}; \quad \]

\[ \varepsilon_1 = \frac{A}{A + 1} E_1; \quad \varepsilon_3 = \frac{A'}{A^2} \beta^2; \quad \varepsilon_4 = \frac{A'}{A + 1 - A'} \varepsilon_3; \quad \]

\[ \mu_3 = \mu = -\mu_4; \quad \frac{E_3}{E_1} = \frac{A'}{(1 + A)^2} (\beta^2 + 1 + 2\beta \mu); \quad \frac{E_4}{E_1} = \frac{A + 1 - A'}{(1 + A)^2} (\gamma^2 + 1 - 2\gamma \mu); \]

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