R.F. IBRAGIMOV, E.E. LUPAR, I.D. KOLBIN

National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Moscow, Russia

A Library Least Square approach in the Prompt-Gamma Neutron Activation Analysis (PGNAA) process in bulk coal samples

Theoretical background

Due to each element having its own characteristic gamma ray energies, spectral intensities at these values of energy are proportional to the content of the element in the sample. The theory behind this approach supposes that any unknown sample gamma-spectrum can be derived from the sum of the products of the elemental volumetric contents and spectrum of each component for every energy channel. This can be written as:

$$f_i = \sum_{j=1}^{m} \beta_j \varphi_{ij}, i = 1, 2, 3 \dots, n,$$

where f_i is the count rate for the unknown spectrum in channel i, β_j is the amount of element j, φ_{ij} is the count rate for element j in channel i, m and $n = 1024$ are the quantity of elements in the sample and the number of energy channels respectively.

To find the most probable values of elemental contents in the sample, we should carry out the following minimization procedure:

$$\delta/_{\delta\beta_i}\left(\sum_{i=1}^{1024}\left(f_i - \sum_{j=1}^{m} \beta_j \varphi_{ij}\right)^2\right) = 0.$$

It should be assumed that φ_{ij} does not change with elemental contents in this approach. Therefore:

$$\sum_{i=1}^{1024}\left(f_i - \sum_{j=1}^{m} \beta_j \varphi_{ij}\right)\varphi_{ij} = 0.$$

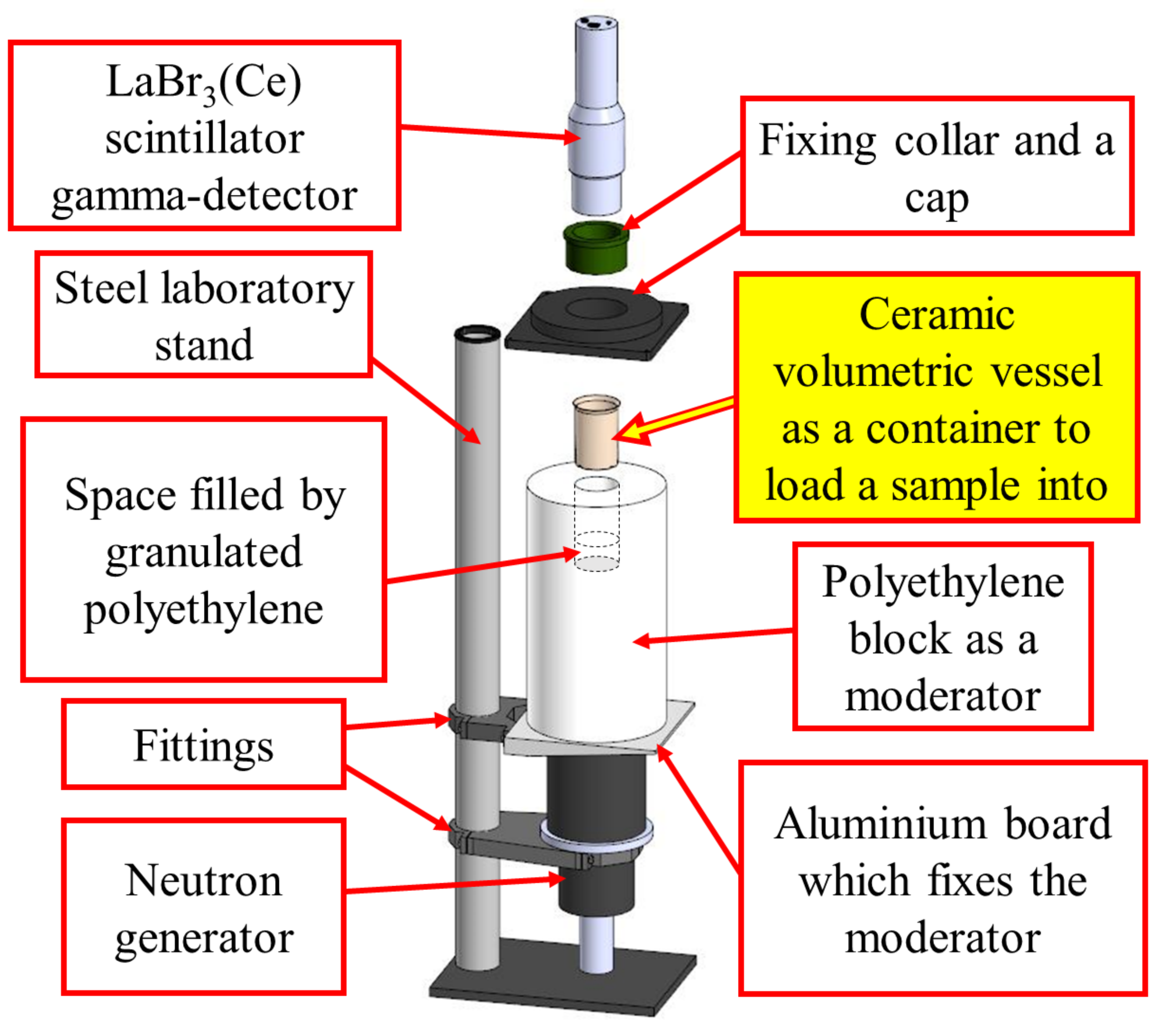


Fig. 1. Experimental facility layout.

Experimental setup

14 MeV neutrons were produced by a neutron generator via the T(d,n)[4]He reaction. A polyethylene block as a neutron moderator. The prompt gamma ray spectra from five different powders of pure compounds (namely $Ca(OH)_2$, Fe_2O_3, NaCl, SiO_2 and TiO_2) and, afterwards, several powder-mixtures of various composition were measured by a $LaBr_3(Ce)$ gamma detection system. The gamma detection system data was streamed directly to the computer, event-by-event.

Neutron source	PNG-07T	
Neutron energy, MeV	14.1	
Accelerating potential, kV	60	
Tube current, μA	98	
Current produced by Penning ion source, μA	163	
Pulse duration, μs	33	
Period, ms	100	
Neutron yield, n/s	$1.5 \cdot 10^7$	
Sample volume, sm^3	400	
Time of radiation exposure, s	7200	
γ-detector type	$LaBr_3(Ce)$	
Dead time, %	~0.4	
Reference spectroscopic gamma sources and total absorption peak energies, keV	^{137}Cs	^{60}Co
	661.7	1332.5
ADC	CAEN Vx1730	
Number of channels	4094	

Table 1. Experimental setup specification sheet.

Virtual experimental setup

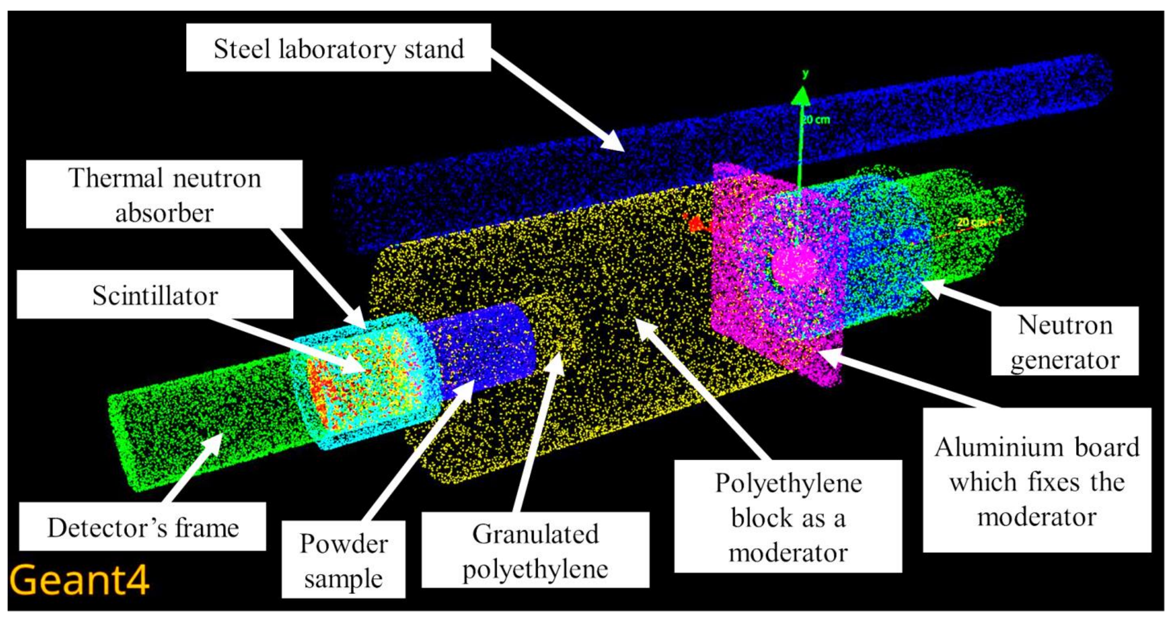


Fig. 2. GEANT4 experiment simulation.

Spectra processing algorithm

a) To carry out energetic calibration provided that gamma ray energy depends on channel number in a linear way (Fig. 3A)

b) To normalize data by the time of radiation exposure.

c) To plot an energy axis which is common to all spectra.

d) To locate spectra on the plot via piecewise-cubic interpolation taking into account that the area under the curve must be preserved.

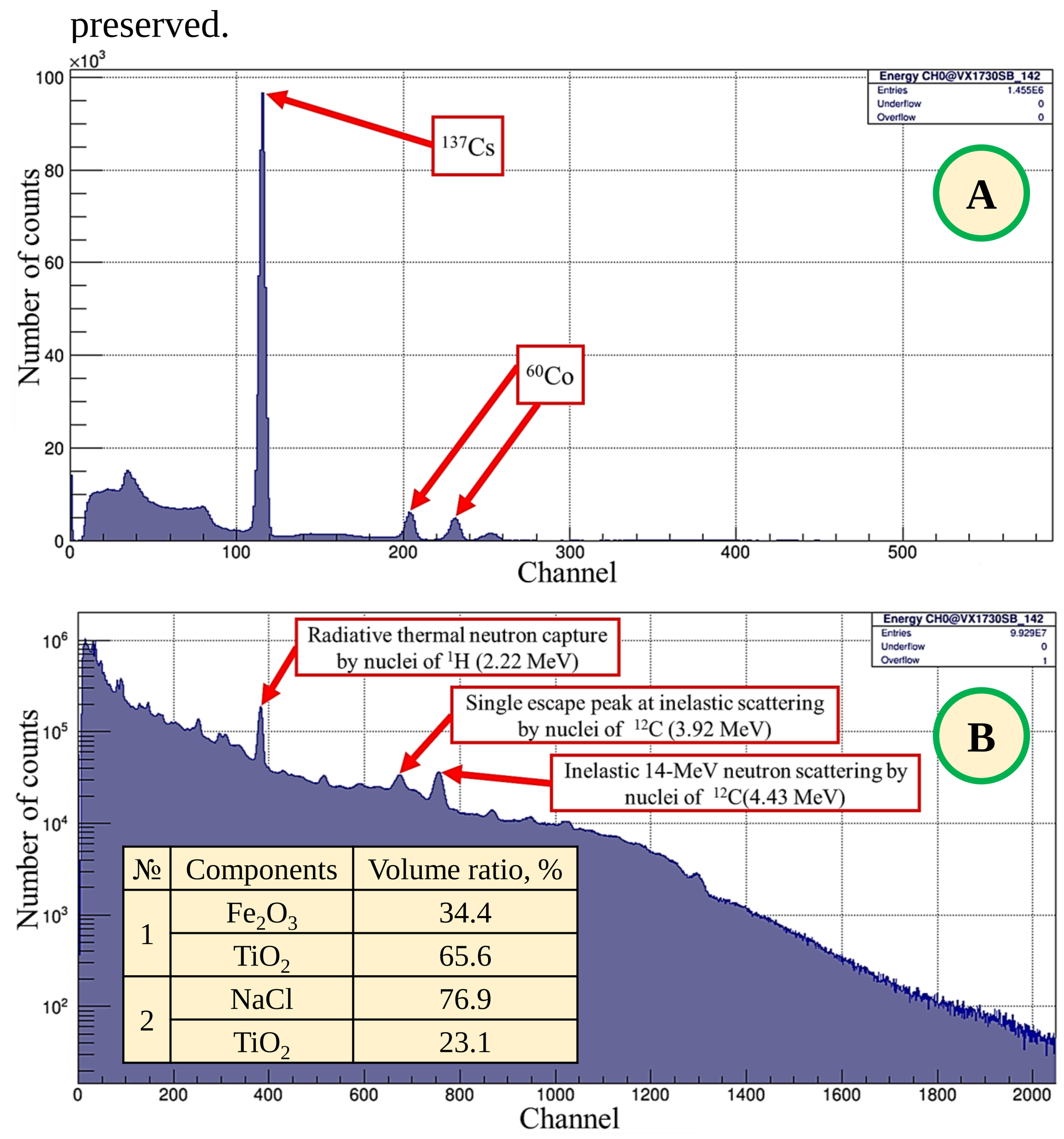


Fig. 3. Defining characteristic gamma-peaks in order to calibrate the detector (A) and a spectrum of Fe_2O_3 (34.4%) and TiO_2 (65.6%) powder-mixture (B) (*included into the graph table shows contents of experimental powder-mixtures*)

Algorithm application

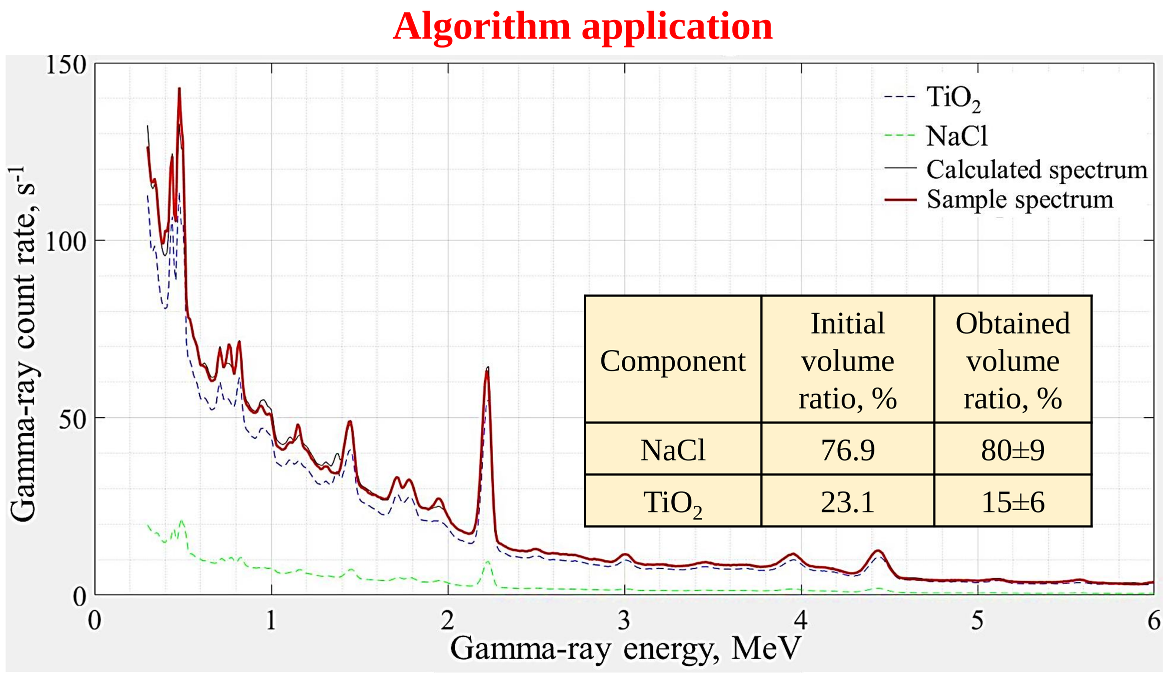


Fig. 4. Determination of the experimental mixture №2 content via PGNAA method.

Graphs like the one shown in Fig. 4 were plotted for each analyzed mixture and included:

- count rate for the sample itself;
- count rates for each of the components together with their volumetric coefficients;
- calculated spectrum defined as a sum of components' spectra.

The key criteria of algorithm's correctness is coincidence of calculated and sample spectra plots within the measurement accuracy. The graph shown in Fig. 4 represents validity of the PGNAA approach application.

The results were compared in order to optimize GEANT4 model up to the most efficient version. As an optimization final measure, the simplest decision to increase results' accuracy was to remove a steel framework and a moderator from the experimental setup simulation.

Results and discussions

As a consequence of GEANT4 model optimization, there were spectra of gamma-rays produced within the confines of the sample obtained. In addition to this, these results were compared to the detector response gamma-spectra (Fig. 5).

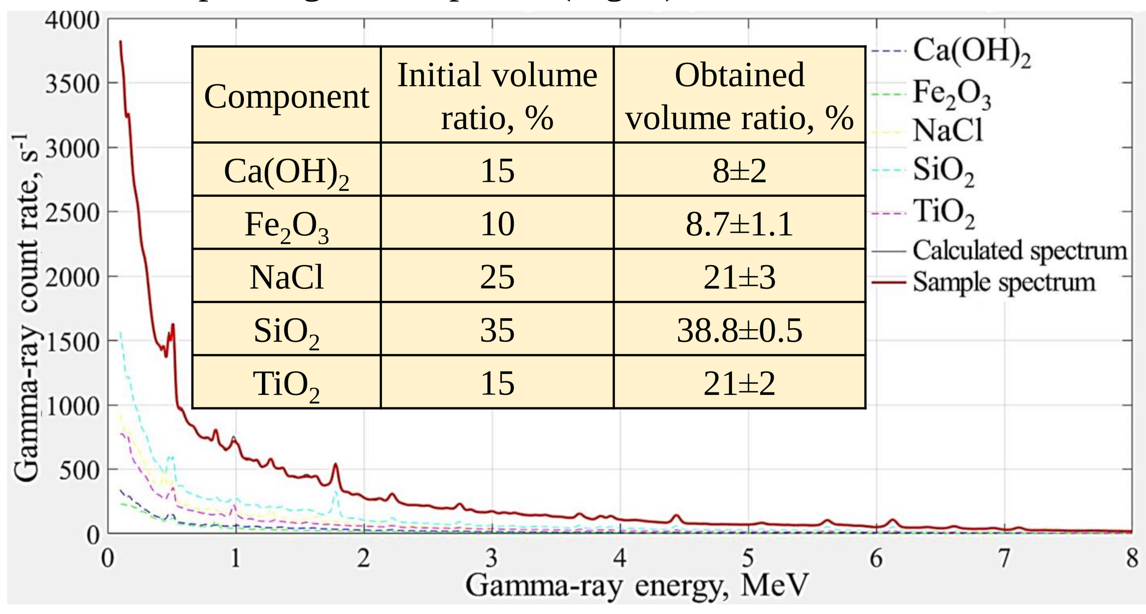


Fig. 5. PGNAA-decomposition of a model five-component mixture.

Error dependence of an initial amount of a component was derived. A graph shown in Fig. 6 represents following tendency: the larger initial volumetric coefficient of a component is, the lower is measure of inaccuracy.

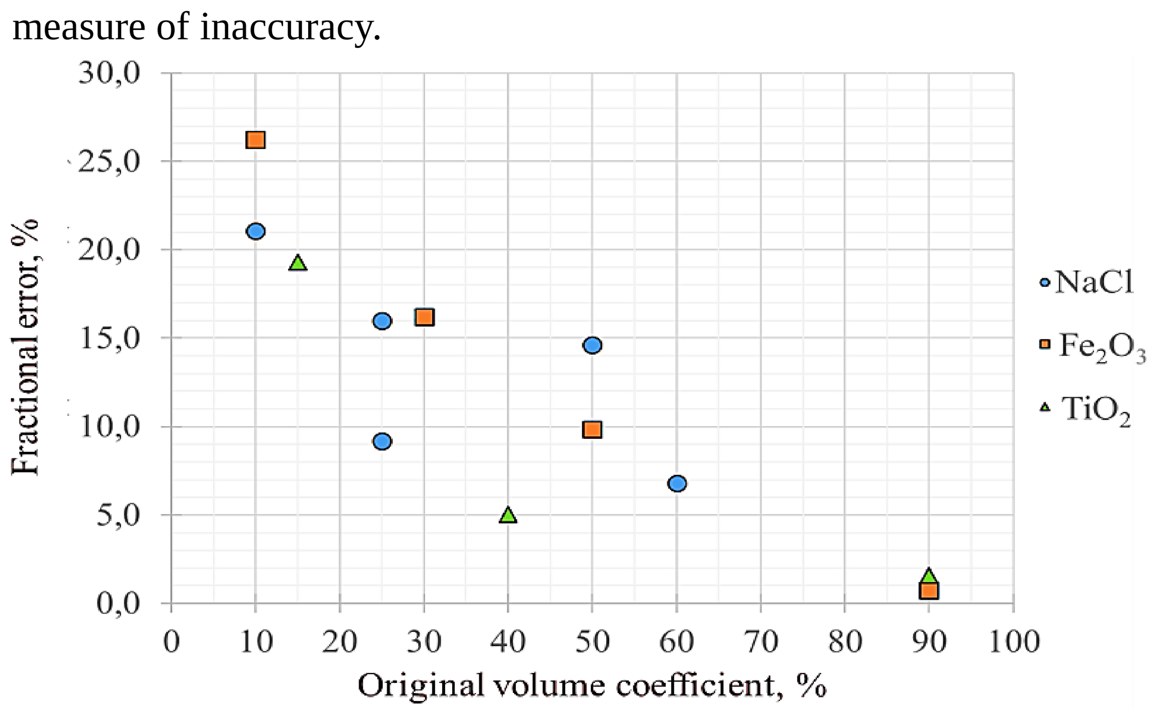
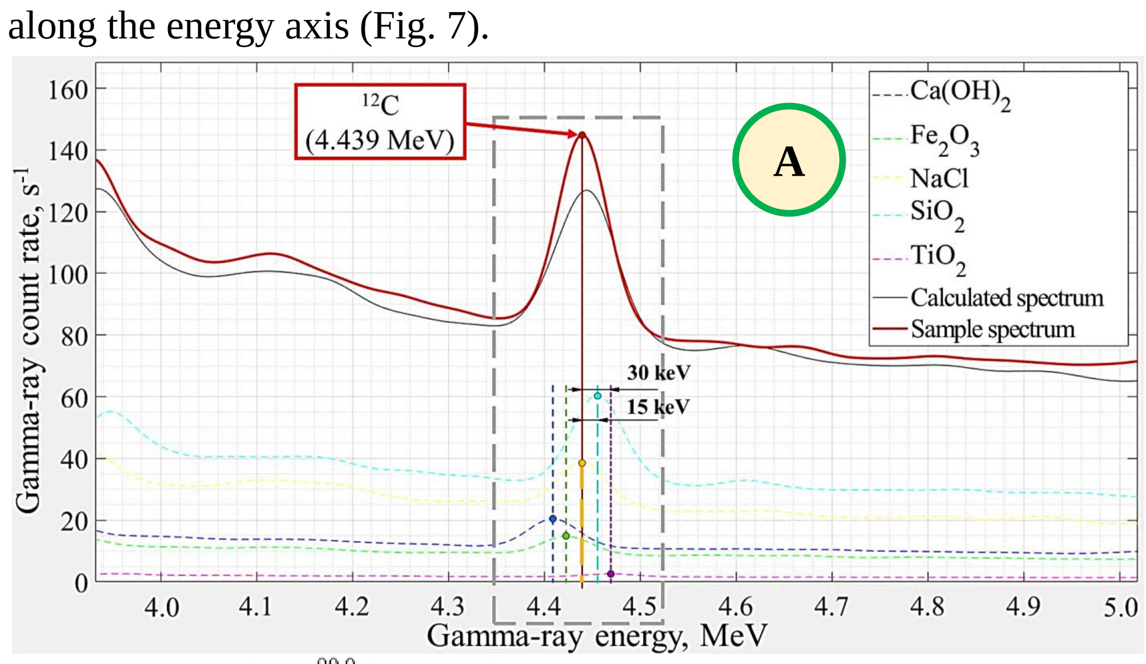


Fig. 6. Ratio error dependence of an original amount of a mixture component.

To show spectra calibration is rather important for PGNAA elemental analysis, a series of calculations were carried out. Used data contained different sets of components' volume coefficients which were obtained using various combinations of spectra displacements along the energy axis (Fig. 7).



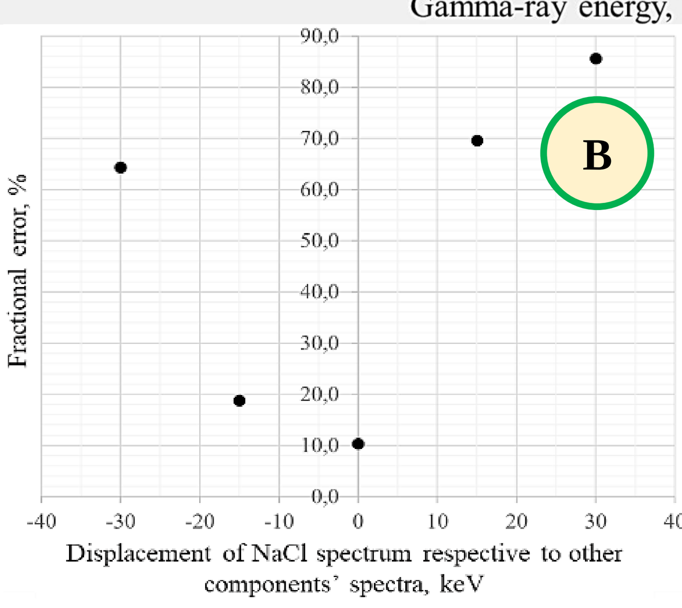


Fig. 7. Impact of spectra displacement on the analysis accuracy (A) and error dependence of NaCl spectrum displacement (B).

The graph of derived dependence (Fig. 7B) represents following trend: the bigger is displacement of a component spectrum relative to other compounds' spectra, the larger is a value of ratio error.

Conclusion. PGNAA technique recommendations

1. Provide a library of detector responses for compounds.
2. Exclude massive objects close to the sample and the detector.
3. PNG is necessary to fixate an impulse duration in order to distinguish NCGR and ISGR spectra.
4. Preferable detector counting window is (0.1; 8.0) MeV.
5. Thorough spectra calibration: corresponding γ-radiation peaks of a pair of spectra must not be further than 15-30 keV from each other.
6. The larger initial amount of a component is, the more accurate are the results of applying the LLS algorithm to the component.

1. Iskender Atilla Reyhancan, Alborz Ebrahimi, Üner Çolak, M. Nizamettin Erduran, Nergis Angin A Monte Carlo Library Least Square approach in the Neutron Inelastic-scattering and Thermal-capture Analysis (NISTA) process in bulk coal samples // Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment. – 2017. – №843. – pp. 29-33.
2. Analiz i predstavleniye rezultatov eksperimenta: uchebno-metodicheskoye posobiye / N.S. Voronova, S.G. Bezhanov, C.A. Voronov [i dr.] (In Russian). – Moscow, NRNU MEPhI, 2015. – 120 p.