Studding the possibility of controlling the uranium content in natural and technogenic raw materials of various compositions by Instrumental neutron activation analysis using the radionuclide neutron source based on Cf-252

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The aim of the work: Determination of the content of ^{238}U in natural and technogenic raw materials of various compositions by instrumental neutron activation analysis using a radionuclide neutron source, including in the presence of impurity elements with a high neutron absorption cross section.

Activation conditions: Radiative capture reaction ($^{238}U + n = {}^{239}U + \gamma$); neutron capture cross section σ – 2740 ± 60 mbarn.; neutron source – radioisotope, based on ^{252}Cf; суммарный neutron flux – 10^9 neutron*c^{-1}; gamma-delayed determination; activation time – 60 min; measurement time – 30 min.

Measurement conditions: Semiconductor gamma spectrometer based on Ge detector GC2018 («Canberra») and pulse signal processor SBS-75 («Green Star Technologies»). Energy resolution – 1.6 keV at energy1332 keV; relative registration efficiency at energy 1332 keV – 10 %; operating range – from 50 to 1600 keV; analytical line ^{239}U – 74,6 keV.

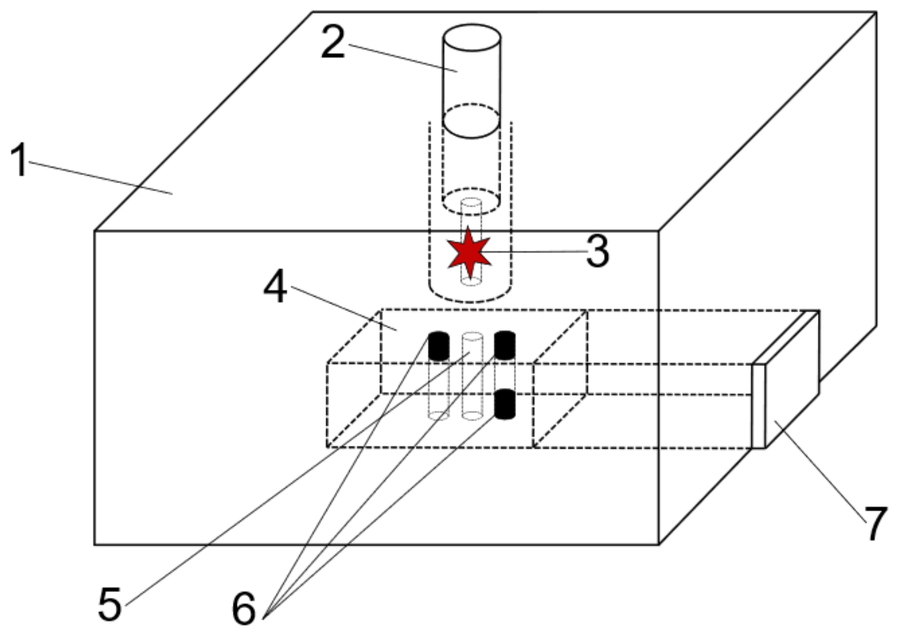

Fig.1 Activation unit: 1 – radiation protection; 2 – movable rod with a neutron source; 3 – radionuclide source ^{252}Cf; 4 – plexiglass retarder block; 5 – channel for a neutron source; 6 – activation channels for samples; 7 – sample loading unit.

Determination of uranium by the INAA method was conducted on unit developed at the Institute of Chemistry of the Far Eastern Branch of the Russian Academy of Sciences. The activation zone consists of a central channel with a radioisotope source based on Cf-252 and 6 vertical irradiators channels (Fig.1). Plexiglas was used as a moderator, providing the maximum flux of thermal neutrons in the activation zone. The measurement of the induced activity was carried out using a gamma spectrometer based on a coaxial germanium detector

The heterogeneity of the neutron flux distribution in the activation channel was calculated using gold monitors (Fig.2).

Using the additive method, a calibration dependence of the analytical signal on the concentration of uranium in aluminosilicate sand was calculated in the range of uranium concentrations from 15 to 300 ppm (Fig. 3).

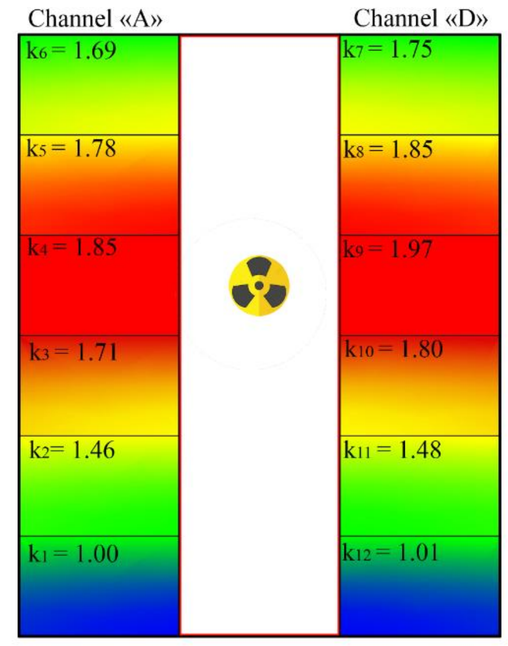

Fig.2 Neutron flux density distributions over the activation channel height

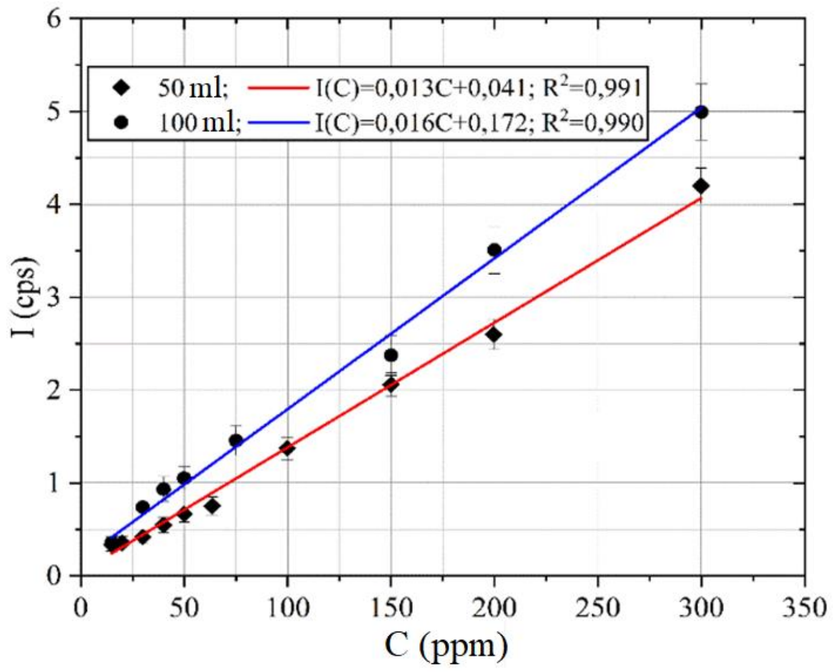

Fig. 3. Dependences of the analytical signal intensity on the concentration of uranium in aluminosilicate sand for samples of various volumes

The nonlinearity of the obtained calibration curves does not exceed 5 %. Based on the data obtained, a calculation limit of detection was made: 12.7 ± 1.5 ppm for 50 ml samples and 7.96 ± 0.45 ppm for 100 ml samples.

To assess the correctness of the results obtained by calibration curves of the INAA method, U was determined in certified standard samples of mineral raw materials. The measurements carried out show a high convergence with the data obtained for standards by the ICP-MS method (Table 1). The main part of the standards samples were aluminosilicate compounds (<85%). However, the determination of uranium in samples of various density, various macro and micromineral composition, is also of interest. An additional important task is to take into account the influence of neutron absorption by micro impurities with a high absorption cross section (for example REE), on the results of the determination of uranium by the INAA method.

Table 1: Comparison of uranium determination results in standard samples by ICP-MS and INAA methods

Standard (description of the sample)	Certified value		INAA results, ppm	Relative Bias (RB)	Z-score
	ISP-MS, ppm (Na_2O_2)	ISP-MS, ppm (4 acids)			
OREAS 100a (Uranium-bearing ore)	135 ± 11	130 ± 12	132.3 ± 4.7	- 0.015	- 96.5
OREAS 102a (Uranium-bearing ore)	662 ± 39	638 ± 43	670.6 ± 8.4	0.527	- 98.7
OREAS 147 (Pegmatitic Li-Nb-Sn ore)	15.8 ± 0.6	16.4 ± 0.7	15.8 ± 1.1	- 0.420	- 92.9

On model samples (volume of 0.75ml), doped with uranium, the effect of density, macro and micro composition on the results of INAA uranium was studied. The data obtained showed:

- Influence of the macrocomposition of the model sample: changing matrix composition from titanium oxide to zirconium oxide leads to a decrease in the analytical signal of uranium by 3.6 times was recorded (Fig. 4).
- Influence of the density of the model sample: increasing the density of the titanium oxide sample from 2.7 to 3.1 g/cm^3, leads to a decrease in the analytical signal of uranium by 30% during irradiation without cadmium shell, and by 15% during irradiation in cadmium shell (Fig. 5).
- Influence of the micro impurities with a high neutron absorption cross section: the content of 2% europium oxide in the sample, leads to a decrease in the analytical signal of uranium by 1.5 times during irradiation without cadmium shell. Decrease in the analytical during irradiation in cadmium shell was not detected (Fig.6).

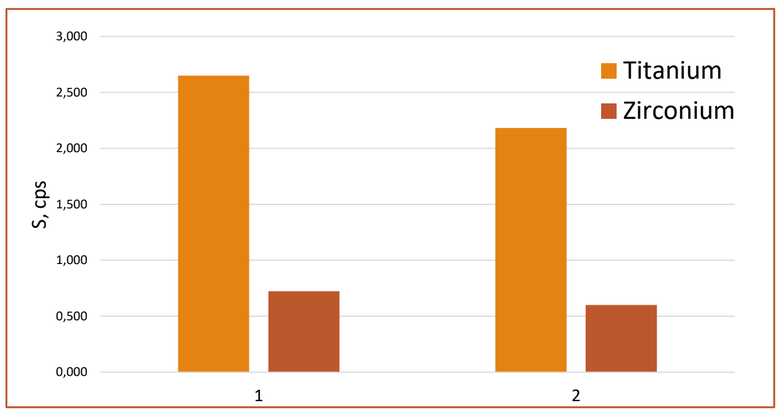

Fig.4. Effect of matrix type on the analytical signal of uranium (1 – irradiation without cadmium shell; 2 – irradiation in a cadmium shell)

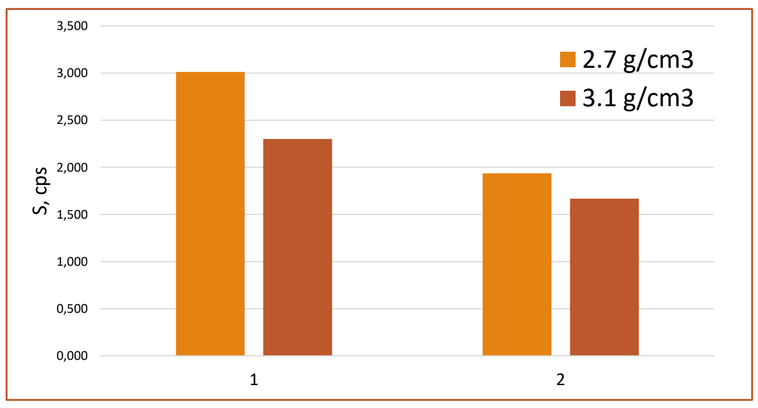

Fig.5. Effect of matrix density on the analytical signal of uranium (1 – irradiation without cadmium shell; 2 – irradiation in a cadmium shell)

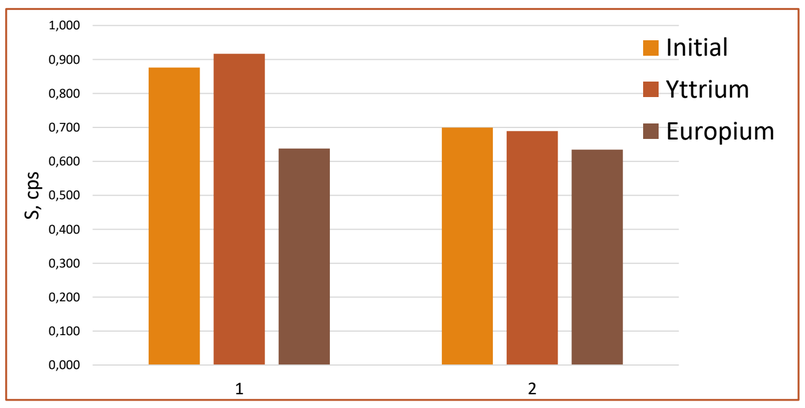

Fig.6. Effect of 6 % impurity on the analytical signal of uranium (1 – irradiation without cadmium shell; 2 – irradiation in a cadmium shell)

Conclusions: To determine the uranium content, the method of INAA with a radionuclide neutron source based on Cf-252 was used. The detection limit for was: 7.96 ± 0.45 ppm. The proposed method makes it possible to effectively determine the uranium content in samples with various macro and microelement compositions. It is necessary to take into account both the neutrons absorption by elements with a high neutron absorption cross section during activation, and the absorption of U-239 gamma radiation by the sample during the measurement. It is also possible to solve the inverse problem of determining the absorption cross section of impurity elements in uranium-containing samples.

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