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Analysis of Uranium Isotope Composition in Uranium Products

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Abstract *- This article presents the results of analyzes of the isotopic composition of uranium in the products of uranium production. At uranium mining enterprises, radiation and dosimetric monitoring is carried out in order to assess the degree of impact of ionizing radiation on the environment. Research carried out in recent years, in the chosen uranium production facility, in the temporary storage of raw materials (chemical concentrates of uranium and uranium oxide-oxide), an increase in the radiation background is observed 3-5 times more than the natural background. Revealing the true cause of this fact and the development of measures to reduce the radiation background in these objects is of scientific and practical interest.*

Keywords: uranium object, natural background, uranium chemical concentrates, uranium oxide-oxide, radiation background, chain of uranium radioactive decay, daughter radionuclides, uranium isotopes, radioactive equilibrium coefficient, isotope analysis.

1. INTRODUCTION

In the literature [1,2], the nuclear-physical characteristics of radionuclides in the chain of radioactive decay of uranium are given. Daughter radionuclides, such as ^{234}U, ^{226}Ra, ^{222}Rn, ^{218}Pb, ^{214}Bi, etc., are formed from the parent nuclide ^{238}U in the process of nuclear transformation. [3-7].

When determining the nuclear-physical characteristics of these radionuclides, it becomes possible to obtain scientifically substantiated information about the factors and reasons proving an increase in the radiation background at these objects. The study of literary sources shows that this issue is poorly studied for the selected objects.

Based on the above, the analysis of the isotopic composition of uranium in the products of uranium production is an urgent task in analytical chemistry, radiation chemistry, uranium geotechnology and radioecology [8-15].

The aim of this work is to analyze the isotopic composition of uranium ^{234}U, ^{235}U, ^{238}U in uranium production products. The reason for the increase in the radiation background in the warehouses for temporary storage of uranium raw materials may be an increase in the ^{234}U content in these products.

Technique and experimental technique. For analysis of the isotopic composition of uranium ^{234}U, ^{235}U. ^{238}U, the method of alpha-spectrometry and an alpha-spectrometer of the α-Analyst type (firm "Canberra", USA) are used. The advantage of the method is the simplicity of the analysis, the low cost of the analysis of the isotopic composition of uranium. The essence of the method is to measure the alpha spectrum of a counting sample containing uranium isotopes, isolated from a sample of chemical concentrates and uranium oxide-oxide, by radiochemical preparation and electrolytic deposition.

2. Results obtained and discussion.

Natural uranium consists of three isotopes: ^{234}U, ^{235}U and ^{238}U, the relative content of which in the mixture is 99.27%, 0.71% and 0.0056%, respectively.

In tab. 1. Specific activities and relative abundances of uranium isotopes are given. From tab. 1. It is seen that the specific activity of the ^{234}U isotope is four orders of magnitude higher than that of the ^{238}U and ^{235}U isotopes.

Tab1. Specific activities and relative abundances of isotopes ^{234}U, ^{235}U and ^{238}U

Isotope	Specific activity, Bq / g	Relative content,%.
^{234}U	2,31*10^{8}	0,0056
^{235}U	7,91*10^{4}	0,71
^{238}U	1,25*10^{4}	99,27

All intermediate isotopes in the uranium decay chain are extremely unstable and decay with half-lives from the first hundred thousand years (^{234}U – $T_{1/2}$=2,47·10^{5} years) to ten thousandths of a second (^{214}Po – $T_{1/2}$=1,6·10^{-4}s).

As you can see from tab. 1, the ^{234}U isotope has a high specific activity of 2,31*10^{8} Bq / g than the other two isotopes ^{235}U and ^{238}U. At that time, the specific activity of the natural mixture of uranium was only 2.5 * 10^{4} Bq / g. Consequently, such a difference in the specific activity of the isotopes ^{234}U, ^{235}U, ^{238}U can be the reason for an increase in the specific activity of the entire uranium product.

The increase in specific activity is explained by the violation of the isotopic equilibrium between the isotopes of uranium ^{234}U / ^{238}U due to the increase in the fraction of the isotope ^{234}U in the products of uranium production. To reveal the true reason for this fact, we carried out a cycle of analyzes of the isotopic composition of uranium ^{234}U, ^{235}U. ^{238}U in uranium chemical concentrates and uranium oxide, depending on the geotechnology processes, uranium leaching.

Figure 1. shows the alpha spectrum of the standard sample OSK-3, No. 02.08.2K20N, with a total activity of 7.5 Bq, prepared by the radiochemical method and measured on an alpha-spectrometer of the α-Analyst type (Canbera, USA) for 6 hours.

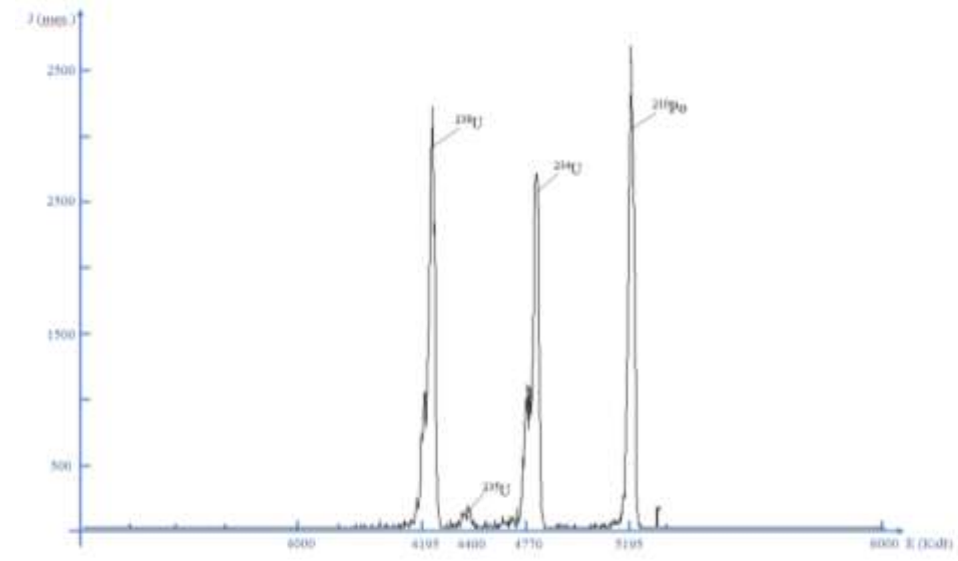

Figure: 1. Alpha spectrum of the standard sample OSK-3.

As can be seen from the spectrum, in addition to the isotopes of uranium ^{234}U, ^{235}U. The ^{238}U isotope ^{210}Po was also registered. On the basis of this spectrum, the efficiency of the alpha-spectrometer is checked, and the isotopes of uranium ^{234}U, ^{235}U are identified. ^{238}U and their number is determined. It can be seen that the energy of the ^{234}U isotope is 4770 keV, ^{235}U is 4400 keV and ^{238}U is 4195 keV, and the energy of the ^{210}Po isotope is 5195 keV.

In fig. 2 shows the alpha spectrum of a sample of a chemical concentrate taken from a geotechnological mine of underground leaching of uranium. Acid leaching of uranium is used in this area.

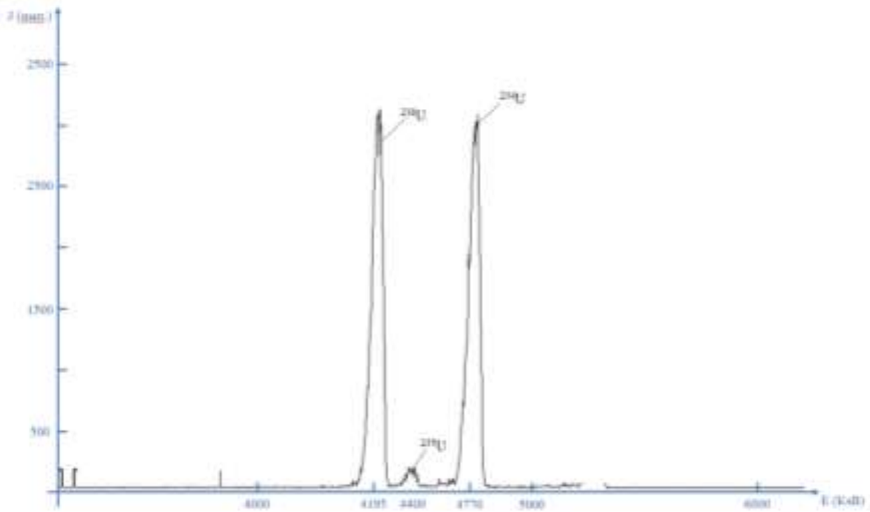

Fig2. Typical alpha spectrum of a chemical concentrate sample taken from a geotechnological mine of underground leaching of uranium.

As seen in Fig. 2the height of the ^{238}U isotope peak is higher than the peak height of the ^{234}U isotope and, in turn, the number of pulses is greater. And the number of pulses is directly proportional to the concentration of the isotope ^{234}U and ^{238}U. Based on this spectrum, the ^{234}U / ^{238}U activity ratio and the relative mass content of ^{234}U (mg / g) in the samples of chemical concentrates or uranium oxide-oxide are determined.

Radiochemical preparation of samples of chemical concentrates and uranium oxide-oxide for alpha-spectrometric analysis is carried out in the following sequence.

An aliquot of a sample of a chemical concentrate or nitrous oxide containing approximately 100 mg of uranium was placed in a beaker, 5 ml of concentrated nitric acid and 1 ml of hydrogen peroxide were added, covered with a watch glass, and boiled until the hydrogen peroxide decomposed.

The treatment with hydrogen peroxide was repeated until the solution was completely clarified. The clear solution was transferred into a 100 ml volumetric flask, brought to the mark with distilled water, closed with a stopper, and the solution was mixed well.

A 0.5 ml aliquot of the solution was evaporated to dryness in a water bath. The dry residue was dissolved in 5 ml of 1% solution of ratrilone B. To a solution containing uranium isotopes, 1 ml of a 25% solution of ammonium chloride, 2 ml of a saturated solution of ammonium oxalate, 2 drops of nitric acid (1: 4) were added and transferred to an electrolyzer. Using a universal indicator paper, the pH was checked, and if the pH was less than 5, then ammonia was added dropwise until pH = 5-7. The glass was washed with 10 ml of distilled water and the contents were added to the basic solution in the electrolytic cell.

The results of determining the average concentration of ^{234}U (mg / g) in chemical concentrates selected by quarters during 2018 are given in tab. 2.

Tab. 2 Results of determining the average concentration of 234U in chemical concentrates (mg / g) during 2018

Sampling site	I quarter	II quarter	III quarter	IV quarter	Average value
P-3	50,1	49,9	50,8	48,5	49,8

P-2	50,4	49,5	51,1	49,4	50,1
P-1	51,1	49,3	47,1	50,1	49,4
C	53,1	51,7	51,9	53,0	52,4
К	57,8	56,7	56,2	56,0	56,7
Cr	69,9	67,5	73,1	68,7	69,8

As you can see from tab. 2, during 2018 the average concentration of ^{234}U in samples of chemical concentrates P-1, P-2 and P-3 varies from 49.4 mg / g to 50.1 mg / g and does not exceed the equilibrium content, that is, 53.41 mg / g., in sample C, the concentration of ^{234}U in the 1st and 2nd quarters of 2018 approaches the equilibrium content of 53.41 µg / g and averages 52.4 mg / g, and in samples К and Cp it varies from 56.7 mg / g to 69.8 mg / g and exceeds the equilibrium content.

Based on the determined values of the radioactive equilibrium coefficients-Crr between ^{234}U / ^{238}U in the chemical concentrates of 6 mines analyzed during 2012, 2014, 2016 and 2018, linear histograms of dependence were constructed. The results are shown in Fig. 3.

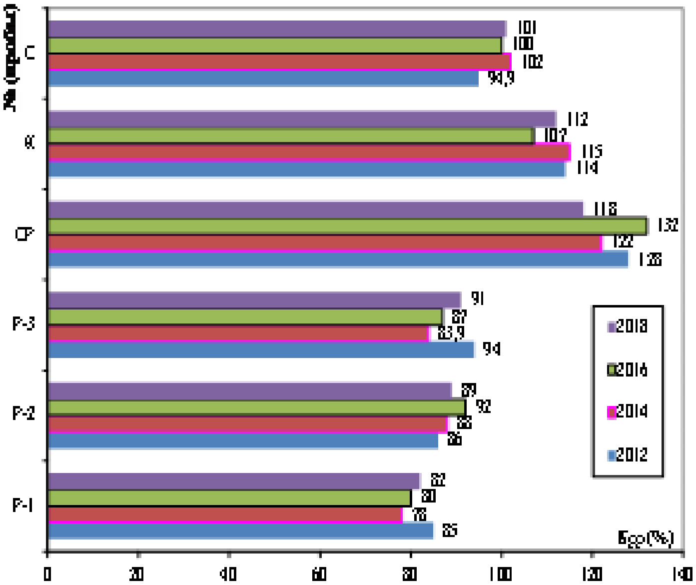

Fig. 3. Linear histograms of the dependence of the radioactive equilibrium coefficients Krr between ^{234}U / ^{238}U in chemical uranium concentrates.

It can be seen from Fig. 3 that the ^{234}U concentration in 4 samples obtained from the productive solutions of six in-situ leaching plots, UG, does not exceed the equilibrium content, that is, 53.41 mg / g, and in the K and Cp samples it exceeds the equilibrium content. It can be seen that the value of the relative coefficients of radioactive equilibrium between uranium isotopes for each deposit differs from each other by no more than 10%.

The results obtained showed that the alpha-spectrometric method with the radiochemical preparation of samples for analysis makes it possible to determine the isotopic composition of uranium and the relative content of ^{234}U in the samples of chemical concentrates and uranium oxide-oxide with a total uncertainty of the result of no more than 2.5% at a confidence level of P = 95%.

Upon receipt of a commercial uranium product that is exported to foreign countries that meets the requirements of consumers (ASTM967) for a ^{234}U content equal to or less than 56.0 µg / g, chemical concentrates from different mines with different contents of the ^{234}U isotope were mixed in the required proportions.

Measurement of ^{234}U content in more than 180 samples of exported uranium oxide-oxide was carried out during 2012-2018. The results are shown in Figure 4.

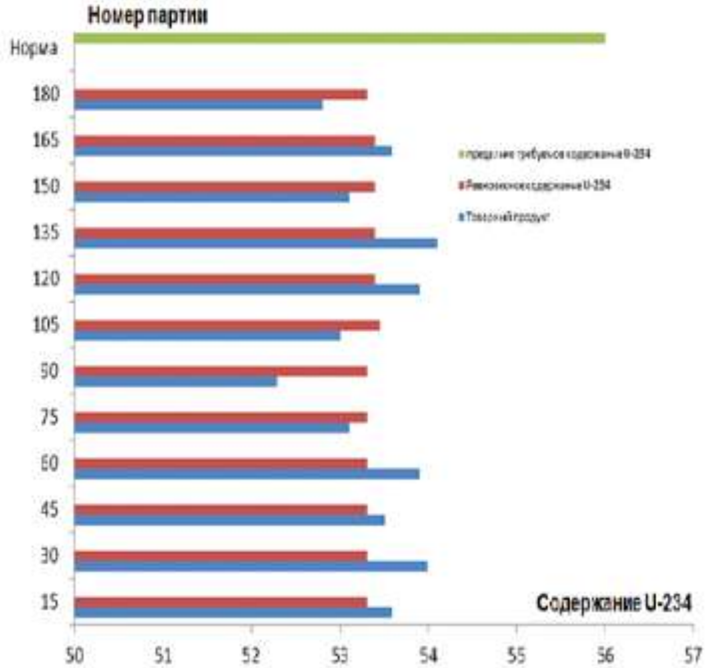

Fig 4. Change in the concentration of ^{234}U in the marketable product of uranium oxide-oxide during 2012-2018.

Fig. 4 it can be seen that the proposed batching method in the obtained batches of uranium oxide-oxide, the concentration of ^{234}U is below the threshold, i.e. set value.

Experimental data show that the concentration of the ^{234}U isotope in various sections of the UW uranium, where the mini-reagent and hypochlorite leaching technology is carried out, a violation of the radioactive equilibrium coefficient Krr between ^{234}U / ^{238}U is observed, moves towards the ^{234}U isotope and the concentration of the ^{234}U isotope in these chemical concentrates reaches values up to 70 µg / g.

This fact is probably explained as follows. As is known, the decay scheme of ^{238}U is represented as

$$^{238}U \xrightarrow[4{,}5*10^{9}\,year]{\alpha} {}^{234}Th \xrightarrow[24{,}1\,day]{\beta} {}^{234}Ra \xrightarrow[1{,}2\,\min]{\beta} {}^{234}U \xrightarrow[2{,}5*10^{5}\,year]{\alpha} {}^{230}Th... \qquad (1)$$

When an α-particle leaves the nucleus, it experiences recoil with energy,

$$E_{U-234} = \frac{M_{\alpha}}{M_{U-234}} * E_{\alpha} \qquad (2)$$

where: Mα is the atomic mass of the α-particle; MU-234 is the atomic mass of the isotope ^{234}U; Eα is the energy of the ^{234}U α-particle, when emitted, it acquires a kinetic energy of ~ 0.1 MeV.

The recoil energy is sufficient to break the bond of the daughter nucleus with the parent in the crystal lattice and overcome the energy bond field. The ^{238}U isotope remains in the nodes of the crystal lattice, while the ^{234}U isotope, leaving the crystal lattice, will be in the interstitial spaces of the crystal and easily gets into pores, cracks and other crystal defects. The same process explains the ^{234}U enrichment of pore waters near uranium mines.

3. Conclusion

Therefore, the ^{234}U isotope is geochemically more mobile than the parent ^{238}U isotope. In other words, the energy of ^{234}U α particles, equal to 4770 keV, is higher than the energy of ^{238}U α particles, which is equal to 4195 keV.

Thus, based on the obtained research results, the following conclusions can be drawn:

Based on the results obtained on the determination of the ^{234}U isotope in chemical concentrates, it is possible to propose a method of batching, when obtaining export uranium products, from equilibrium and non-equilibrium chemical uranium concentrates.

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