Method for Conducting of Uranium Isotopic Analysis in Various Natural Waters of Uranium-Bearing Regions of Uzbekistan

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Abstract: The paper presents the results of a uranium isotopic analysis by alpha spectrometric method in various natural waters of uranium-bearing regions of Uzbekistan. To perform isotopic analysis of uranium in various natural waters, preliminary radiochemical preparation of water samples is carried out, which includes concentrating of uranium isotopes from water samples, removing of the interfering radionuclides and iron, preparation of a counting sample by electrical method, electrolytic deposition of uranium, etc. Based on the obtained data on the isotopic composition of uranium in the studied water samples, disruption in the radioactive equilibrium between uranium isotopes and a directly proportional relationship between concentration of the ^{234}U isotope and the total volume alpha activity of the water was established.

Keywords — Alpha-spectrometric technique; uranium isotopic analysis; concentrating of uranium isotopes; water samples; radioactive equilibrium coefficient; extraction separation; electrolytic deposition of uranium; volumetric alpha activity; concentration of the ^{234}U isotope.

1. INTRODUCTION

Performing an isotopic analysis of uranium in various natural waters of uranium-bearing regions is of scientific and methodological interest from the point of view of assessing the level of radiation pollution of these waters by various natural and technogenic radionuclides, where geotechnological leaching of uranium is carried out. At the geotechnological method of uranium mining, the probability of contamination of natural waters by various radionuclides due to their migration in ecosystem objects remains. In addition to these, various foreign and domestic sources have information about the disruption between the uranium isotopes of which affects the total volume alpha activity of water samples.

The study of the chemical composition of natural waters, to obtain complete data on the isotopic composition of uranium and the radioactive equilibrium between uranium isotopes are a vital task of analytical chemistry, applied nuclear physics and radioecology [1-4,8].

There are mass spectrometric, alpha spectrometric and other methods of analyzing the isotopic composition of uranium. Of these, the most accessible method for determining the isotopic composition of uranium in various natural waters is the alpha spectrometry method [5-7]. This method differs from other methods in its simplicity, the availability of radiochemical preparation of water samples, the singular amounts of the selected peaks in the obtained spectra, and the simplicity of identifying these peaks.

Carrying out isotopic analysis of uranium includes - preliminary radiochemical preparation of water samples, which includes the concentration of uranium isotopes from water samples, extraction separation from interfering radionuclides and iron, and the preparation of an electrically counted sample.

The electrolytic deposition of uranium is carried out on a corrosion-resistant stainless steel substrate. This "counting" sample - a stainless steel substrate is supplied to the measurement.

The concentration of radon in the ground layer in this area is 3.52-7.64 Bq / m^3. However, a number of finishing materials have high rates of effective specific activity of natural radionuclides (for example, ceramic tiles), so the use of such products can significantly increase the gamma background of the premises.

2. EXPERIMENTAL METHOD

Salt composition, hardness, the sum of cations and anions in water samples are determined by the chemical method on a photo colorimeter - KFK-03, pH-value of water on a pH meter I-160, uranium isotopic composition was determined by alpha spectrometry on an alpha spectrometer of the type ALPHA ANALYST "CANBERRA".

3. RESULTS AND DISCUSSION

In the selected water samples from various reservoirs of the Kyzylkum region, a complete chemical analysis was first carried out to assess the change in the quality of these waters due to geotechnological processes of uranium mining. Salt composition, hardness, pH value of water, the sum of cations and anions are determined. The results are shown in table 1.

Table 1. The results of a complete chemical analysis of samples taken from various reservoirs of the Kyzylkum region

№ samples or sampling sites	CATIONS mg/dm³						pH
	Ca^{2+}	Mg^{2+}	$Na^{+}+K^{+}$	Fe^{3+}	NH_4^{+}	$\sum$	
21	7,50	222,72	511,62	237,52	<0,02	427,35	8,03
3	7,50	228,82	505,85	244,61	<0,02	428,95	7,97
M	4,50	228,83	510,38	173,71	<0,02	372,33	8,05
B	16,50	161,12	1159,07	1003,24	0,00	1180,87	7,67
	ANIONS mg/dm³						
Dry residue, mg/dm³	CO_3^{2-}	HCO_3^{-}	SO_4^{2-}	Cl^{-}	NO_2^{-}	NO_3^{-}	$\sum$
1236,00	7,50	222,72	511,62	237,52	<0,02	1,31	980,67
1242,00	7,50	228,82	505,85	244,61	<0,02	0,60	987,38
1140,00	4,50	228,83	510,38	173,71	<0,02	13,03	930,45
3614,00	16,50	161,12	1159,07	1003,24	0,00	0,00	2339,93

The results show that in samples 21, 3 and M, the cationic composition, anionic composition, pH, dry residue and the sum of cations and anions are very close to each other. The reason for this similarity is the close location of these water samples. The results of samples B sharply differ in cationic, anionic composition, pH, dry solids and the sum of cations and anions.

Specific alpha-activity of the presented samples was determined on a radiometer-UMF-2000. Obtained in four samples, the value of specific alpha activity is from 0.12 Bq / L to 0.28 Bq / L. Based on these data Fig.1, the dependence of the specific alpha activity on the concentration of ^{234}U was constructed.
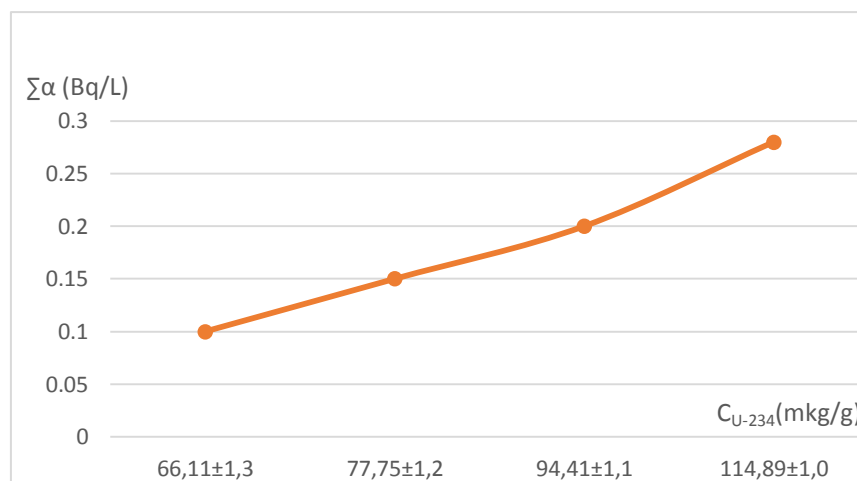

Fig 1. The dependence of specific alpha activity on the concentration of ^{234}U.

To reveal the reason for the increase in specific alpha activity in natural waters, depending on the concentration of radioisotopes in the decay chain of uranium, a preliminary assessment of natural waters for radioactivity was carried out by determining the specific alpha activity, since if the values of the annual effective dose are lower than 0.2 mSv /yr, then a preliminary assessment of natural waters is not required (this value of the dose with water consumption of 2 kg per day corresponds to the average values of specific activity Bq / kg - (^{238}U-6,2; ^{235}U-6,0; ^{234}U-5,8; ^{230}Th-1,32; ^{226}Ra-1,0; ^{222}Rn-60; ^{210}Bi-220; ^{210}Po-0,24; ^{210}Pb-0,4 etc.) and if the effective dose is in the range from 0.2 to 1.0 mSv / yr, isotope analysis is required to determine the specific activity of each radioisotope.

To identify the main cause of the increase in the total volume alpha activity of these waters and to reveal the true cause of the increase, an isotopic analysis was performed.

The reason for the isotope analysis was that the ^{234}U isotope is one of the decay products - ^{238}U and it has a specific activity of 2.3 * 108 Bq/g. And the total specific activity of all three (^{234}U, ^{235}U, ^{238}U) isotopes of uranium is - 2.5 * 10^4 Bq / g. Therefore, such a difference in specific activity can be explained by a disruption of the isotopic equilibrium of uranium due to an increase in the proportion of the ^{234}U isotope in the studying samples. The activity of the ^{238}U isotope, taking into account its mass fraction in natural uranium, having a weight of 1 g, is (Bq/g in a mixture of isotopes of natural uranium):

$$A_{U-238} = \frac{0,992745 * \ln 2 * 6,022136 * 10^{23}}{T_{U-238} * 238} = 1,2347 * 10^4 \, Bq \qquad (1)$$

where 0.992745 is the relative mass fraction of the isotope ^{238}U, g; 6.022136 *10^{23}- Avogadro constant; T$_{U-238}$.- half-life decay period of ^{238}U, s; 238 - atomic mass of ^{238}U.

The mass (equilibrium) content having the same activity of the ^{234}U isotope in 1 g of equilibrium uranium will be equal to (g):

$$M_{U-234} = \frac{1,2347 * 10^4 * T_{U-234} * 234}{\ln 2 * 6,02213 * 10^{23}} = 53,41 * 10^{-6} \, g \qquad (2)$$

where T$_{U-234}$ is the half-life decay period of the isotope ^{234}U, s; 234 — atomic mass of the isotope ^{234}U.

Based on 1 and 2, the conversion factor can be introduced- K, that is, in 1 g of a mixture of natural uranium isotopes in equilibrium uranium samples, the content of the ^{234}U isotope is:

$$K = \left(\frac{M_{U-234}}{M_U}\right) \qquad (3)$$

where - MU-234 – is the mass (equilibrium) content of the isotope ^{234}U, g; M$_U$ – is mass of a mixture of isotopes of natural uranium, g.

The ratio of the activities of A$_{U-234}$/A$_{U-238}$ in the studied samples is:

$$\frac{A_{U-234}}{A_{U-238}} = \frac{S_{U-234}}{S_{U-238}} \qquad (4)$$

where S$_{U-234}$ is the peak area of ^{234}U; S$_{U-238}$ is the peak area of ^{238}U.

By taking into account 1, 2, 3, and 4, the determination of the mass content of the ^{234}U isotope in the studying samples (mkg/g of uranium) can be expressed by the formula:

$$M_{U-234} = \left(\frac{A_{U-234}}{A_{U-238}}\right) * K \qquad (5)$$

where A$_{U-234}$ – is the measured activity - ^{234}U (Bq); A$_{U-238}$ – is the measured activity - ^{238}U (Bq); K - conversion factor equal to 53.41 (mkg / g of natural uranium).

Based on the above analytical formulas, selected from various water reservoirs of the Kyzylkum region, using alpha spectrometry the isotopic composition of uranium in natural waters was analyzed (Table 2).

Table 2. The results of determining the isotopic composition of uranium in various natural waters of the Kyzylkum region.

№ samples	Number of pulses - ^{234}U/^{238}U	Mass content - ^{234}U(mkg/g)	
		Parallel samples	Average result
21	16500/13200	66,76±1,2	66,11±1,3
	12400/10200	65,45±1,4	
3	15300/10600	77,10±1,2	77,75±1,2
	18200/12400	78,39±1,1	
M	18900/10600	95,23±1,2	94,41±1,1

	21200/12100	93,58±1,0	
B	23900/11200	113,97±1,0	114,89±1,0
	21900/10100	115,81±1,0	

The results showed that there is indeed a disruption in the radioactive equilibrium between the uranium isotopes (^{234}U/^{238}U) in the investigated groundwater. At a radioactive equilibrium of uranium isotopes, ^{234}U content should be at the level of 53.41 ± 1.5 mkg / g. As can be seen from the results of tab. 2, the content of ^{234}U in all cases is greater than 53.41 ± 1.5 mkg / g.

Uranium isotopes in natural objects, including rocks, pass from them into water due to leaching. Uranium isotopes has various solubility, migratory abilities, valence states, ion radii, recoil energy and α-particle exit energy. It is difficult to determine which of these factors is the main and predetermining in disruption of the radioactive equilibrium. Nevertheless, conducting a study on the basis of a method of radiochemical extraction separation of uranium from associated radioactive isotopes, preparing a "counting sample", identifying each isotope using the alpha spectrometric method, and studying the influence of geotechnological processes takes place in the scientific substantiation of this fact.

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