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SPATIAL NONUNIFORM DISTRIBUTION OF ^{235}U ISOTOPE AT SUPERCRITICAL FLUID EXTRACTION WITH CARBON DIOXIDE IN A GRADIENT TEMPERATURE FIELD

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The spatial redistribution of the ^{235}U isotope of natural uranium in a gradient temperature field along the height of the reactor in supercritical carbon dioxide has been experimentally investigated. The scheme of the reactor is given and the principle of operation of the reactor is described. The method of preparation of initial samples from granite samples containing natural uranium and the procedure of extraction are described. The conclusion about the spatial redistribution of ^{235}U isotopes in supercritical carbon dioxide is based on the analysis of gamma spectra of extracts. It is shown that the concentration of the ^{235}U isotope in a supercritical fluid is maximal near the lower heated flange of the reactor, and decreases with approaching the upper, cooled flange. It was concluded that the separation factor of the ^{235}U isotope in supercritical carbon dioxide can be about 1.2 ± 0.12.

ENERGY URANIUM ISOTOPE. METHODS FOR URANIUM EXTRACTION FROM NUCLEAR MATERIALS

The main fuel isotope in open-cycle uranium-fueled thermal reactors is ^{235}U. Since the world nuclear energy uses this uranium isotope, then, naturally, its production and depletion rate in the world are growing [1].

Generation of ^{235}U isotope is based on its separation from the bulk of natural uranium, which mainly consists of ^{238}U isotope in an approximate ratio 99.2745% ^{238}U:0.7200% ^{235}U [2].

Production of uranium compounds for the needs of nuclear power is carried out by direct mining and refining of uranium from uranium deposits [1, 3], as well as by spent nuclear fuel reprocessing (SNF) using well-developed methods (bismuth-phosphate process, liquid extraction in the form of redox and purex processes) [4–9].

The disadvantage of uranium production by processing of uranium ores is its incomplete (up to 95…97%) extraction from the feedstock. Therefore, a large amount of waste uranium has accumulated in Ukraine in the form of tailing dumps, which form technogenic deposits of a total area of about 2.5 mln m^2 and a mass of about 40 mln t. Waste heaps of coal mines of a total volume of coal rocks of the order of 1056.52 mln m^3 can be also counted as technogenic deposits containing heavy metals, uranium and thorium.

Therefore, search for new alternative, low-waste technologies for processing technogenic deposits should be considered as one of the most important tasks in the development of nuclear energy.

One of these technologies satisfied the selective, deep and low-waste processing of feedstock is the physical method of supercritical fluid extraction by carbon dioxide (SFE-CO$_2$) [10] of uranyl nitrate complexes with organophosphorus compounds. According to this method a supercritical fluid with an uranium complex dissolved in it is generated in a reactor at a high pressure (more than 7.38 MPa) and a relatively low temperature (more than 31.06 ºC). Experiments proved that up to 62% of uranium complexes are extracted at a pressure of 9 MPa and a temperature of 38 ºC [11]. Relatively low indicators of extraction efficiency in [11] were obtained at low pressures, low temperatures and using tributyl phosphate (TBP) as an organophosphorus reagent. The use of higher pressures (up to 30 MPa) and additional complexing agents make it possible to increase the extraction efficiency up to 100% [12].

A theoretical estimate has shown that the maximum extraction efficiency of the uranium complex is about 96% [13].

Since, uranium complexes are in the volume of the fluid in the process of SFE-CO2, there is a certain possibility to carry out local changes in the ratio of the amount of isotopes by the effect of some external factors. An example is the Rank-Hilsch vortex tubes [14] used to separate ^{36}Ar/^{40}Ar isotopes in the fossil gases H$_2$ or CF$_3$Cl, which can be considered as an external factor. In the case of SFE-CO$_2$ for uranium complexes a change in the ratio of the amount of isotopes can be implemented in the process of fluid extraction from the reaction volume on condition that the flow and temperature scenarios similar to the Zippe centrifuge are realized.

In the case of SFE-CO$_2$ the supercritical fluid containing uranium complexes is under high pressure. Such a medium is quite sensitive to the temperature differential at its boundaries: even small temperature gradients (at the level of 0.1…0.2 °C/cm) can start the convective mechanism of mass transfer, which has been experimentally confirmed in [15]. Therefore, it is of great interest to study the effect of the temperature gradient on the spatial distribution of a supercritical fluid containing complexes of uranium isotopes, which may arise even due to a small temperature difference in the probabilities of their transitions between energy levels [13].

As one of the examples of the possible redistribution of uranium isotopes under natural conditions, in our

opinion, is a natural nuclear reactor that operated for more than tens of thousands of years about two billion years ago in Gabon, West Africa [16]. To initiate chain fission reaction in such a natural reactor it is necessary to have ^{235}U enrichment at least 3%, as it is provided in modern commercial nuclear reactors. For example, light water reactors operate at pressures of about 15.0 MPa and temperatures of about 300 ℃ [17]. At such parameters the water is in a liquid state ($T_{sc} = 374$ ℃, $P_{sc} = 21.77$ MPa) and carbon dioxide, as noted above, goes into a supercritical state and turns into a good organic solvent [10, 11]. Under such conditions, at low water content, when there is no moderator and fission reactions cease, the dissolution of uranium complexes in supercritical carbon dioxide is possible. The aforementioned uranium enrichment to a level of 3% can be observed in the field of a spatial temperature gradient due to the temperature difference in probabilities of transitions of uranium isotope complexes between energy levels [13].

This paper presents the results of an experimental study of the change in the spatial distribution of ^{235}U isotope in a volume of supercritical carbon dioxide heated from below and cooled from above.

DESCRIPTION OF THE TRIAL SETUP REACTOR

Experiments for investigation of spatial distribution of uranium isotopes ^{235}U and ^{238}U in the reactor were carried out on the upgraded laboratory setup for supercritical fluid extraction "SFE-U" [11, 18]. The setup allows raising the pressure of supercritical carbon dioxide up to 20.0 MPa and maintaining the temperature from room temperature to 50 ℃. Control of pressure and temperature at different points of the reactor is provided by digital barostats and thermostats, which allow such control in automatic, semi-automatic and manual modes. When carrying out the extractions, Carbon dioxide meeting the requirements of GOST 8050-85 with a volumetric CO_2 content of 99.8% was used for extractions.

The updating of the laboratory setup was carried out only for the reactor, which in its original form was a cylinder 9 cm high and 2.6 cm in diameter. In the upgraded reactor its height was increased to 21.8 cm. Its lower and upper flanges were maintained at specified temperatures by means of adjustable heaters and the temperature of the lower flange was set higher than the temperature of the upper one.

The internal arrangement of the upgraded reactor is presented in Fig 1.

A specific feature of this upgraded reactor is a sealed axial mounting of a stainless steel tube of a diameter of 6.1 mm and a wall thickness of 1 mm. At the end of the tube there is a intake device 10 (see the fragment of section A in Fig. 1) in the form of an umbrella, which doses removing a part of the extract from the volume of the reactor by lowering the pressure in it. Decreasing the pressure is carried out stepwise, starting from the initial pressure release by about 4.0 MPa and subsequent pressure release in total by an amount within the range of 9.6…10.0 MPa from the initial one. As shown in [11], the bulk of the uranium complex containing ^{235}U isotope

is recovered during the first pressure release. The fraction of the primary recovered extract volume was estimated by the same ratio as the primary pressure release, 0.25 of the total volume, and basically corresponded to the area located below the intake device.
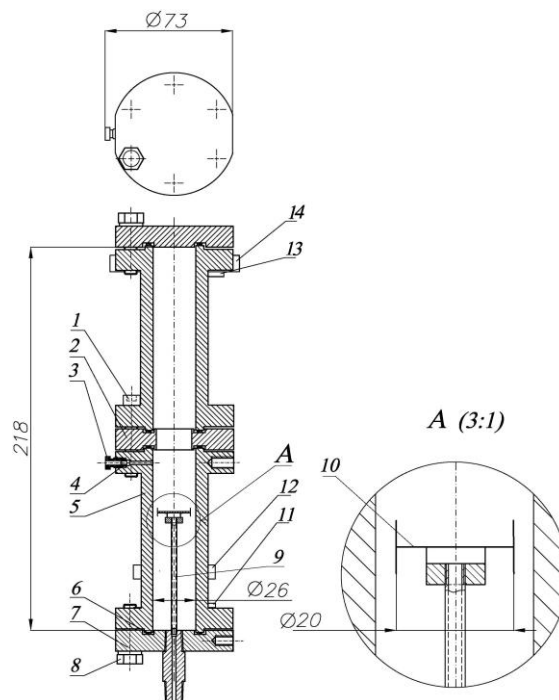

Fig. 1. Internal arrangement of the elements of the upgraded reactor:
1 – bolt; 2 – middle flange; 3 – fitting;
4 – sealer; 5 – case; 6 – insert;
7 – bottom flange; 8 – bolt; 9 – tube; 10 – intake device;
11 – lower flange temperature sensor;
12 – lower flange heating element;
13 – upper flange temperature sensor;
14 – upper flange heating element

The intake device was located at a given distance from the lower flange of the reactor in each series of experiments.

SAMPLE PREPARATION

Sample preparation is necessary to impart to the initial samples the properties of solubility in supercritical carbon dioxide. Sample preparation was carried out similarly to that described in [19], where samples of granites with uranium content from 0.13 to 667 µg/g were used as the initial material containing natural uranium. The uranium content in the initial samples was determined by gamma spectrometry using SEG-50 (P) spectrometer based on a semiconductor germanium-lithium diffusion-drift detector DGDK-60V.

A granite sample with the highest natural uranium content was used for the experiments. The original sample was crushed and grinded to powder of a grain size of less than 50 µm. Then 10 g of powder was poured with 20 ml of 40% nitric acid and kept for a day. The resulting solution was filtered. After filtration the solution was mixed with 6 ml of 30% TBP solution in kerosene and was shaken. After stratification the organic phase was filtered through a paper filter. Thus, under the assumption that the efficiency of all sample preparation

procedures is close to 100%, the uranium content in 1 ml of solution was estimated to be no more than 1.1 mg.

EXTRACTION PROCEDURE

The extraction on the upgraded SFE-U unit was carried out in the following order:

– a glass container (short test tube) containing 2 ml of the obtained organic extract was placed in the reactor;

– the upper flange of the reactor was heated up to 35 ºC, the lower flange – up to 40 ºC;

– injection of carbon dioxide by means of a compressor up to a pressure of 16.0 MPa;

– exposition of the extract for 30 min;

– discharge of the extract (~ 1 ml) into a glass jar for 5 min while releasing pressure from 16 to 12 MPa;

– discharge of the extract (~ 1 ml) into a glass beaker for 5 min while releasing pressure from 12 to 6 MPa;

– after releasing the pressure to atmospheric, the extract residue was removed from the reactor.

The extraction procedures described above were carried out three times at three different points along the height of the reactor: I, II, and III. At point I the distance from the lower flange of the reactor to the intake device (see item 10 in Fig. 1) was – 5 cm; at point II – 10 cm; at point III – 15 cm.

PROCESSING OF THE OBTAINED EXPERIMENTAL RESULTS

The initial samples of the solution (before CFE-CO_2 carrying out) and the samples obtained as a result of CFE-CO_2 were analyzed for the content of uranium ^{235}U isotope according to the spectra obtained on the gamma-ray detector.

A detector based on high-purity germanium was used as a gamma-ray detector – a coaxial germanium detector GC 1818 (Germanium Coaxial) with a relative detection efficiency $\varepsilon_{отн} = 18\%$, an energy resolution $\Delta E = 180$ keV at an energy of gamma quanta $E_\gamma = 1.33$ MeV.

Table 1

The results of the analysis of ^{235}U isotope content in natural uranium using the FRAM program

Sample No. (Volume)	Content ^{235}U, %	Error; +/-, %
0 (2 ml) initial	0.69	0.012; (1.75)
0 (6 ml) initial	0.64	0.007; (1.03)
I.1 (2 ml)	0.91	0.018; (1.95)
I.2 (2 ml)	1.35	0.101; (7.5)
I.3 (2 ml)	1.4	0.062; (4.44)
II.1 (2 ml)	1.57	0.352; (2.44)
II.2 (2 ml)	1.00	0.056; (5.62)
II.3 (2 ml)	1.07	0.055; (5.17)
III.1 (2 ml)	0.98	0.061; (6.19)
III.2 (2 ml)	0.97	0.030; (3.16)
III.3 (2 ml)	0.96	0.063; (6.27)
The residue (2 ml) in point III	0.63	0.017; (2.70)

When using the detector, gamma spectra were measured with their subsequent analysis by the FRAM program [20] which was developed at the Los Alamos National Laboratory. The sample was placed directly in front of the detector window.

The standard exposure time was about 2 days.

A special feature of the FRAM software is that results are obtained using only spectral data and known nuclear constants. In this case, calibration of the spectrometric system using standard samples is not required.

The results of the analysis of ^{235}U isotope content in natural uranium using the FRAM program are presented in Table1.

RESULTS AND DISCUSSION

Based on the results presented in Table 1 the following conclusions can be drawn.

The content of the uranium isotope ^{235}U in the initial samples does not exceed the values corresponding to natural uranium – 0.711…0.7200% [2].

As a result of SFE-CO_2 complexes of uranium from the region located at a distance of 5 cm from the lower flange (point I) ^{235}U isotope content varied from 0.91 % to 1.4 %. ^{235}U isotope content decreased to a level of 1.0 % with increasing the distance from the lower flange of the reactor (point II) up to 10 cm. ^{235}U content decreases to an average value of 0.97 %, but remains above the natural level with a further increase in the distance from the lower flange of the reactor up to 15 cm.

The excess of ^{235}U isotope concentration over the natural level at points I–III, in our opinion, was due to the experimental conditions: at each point of extract separation the ^{235}U concentration corresponded to its local average content in the removed volume, the value of which was estimated to be about 0.45 of the total volume.

The location of the experimental points of ^{235}U isotope concentration over the height of the reactor (black squares) is shown in Fig. 2. The X-axis direction is chosen from the cold reactor flange to the warm flange. The experimental points are well described by the linear dependence of the uranium isotope concentration $y_{exp}(x)$ on the normalized length x (the straight line in Fig. 2 passing through the experimental points), which was plotted using the least squares method:

$$y_{exp}(x) = 0.67 + 0.85 \cdot x. \qquad (1)$$

The real distribution of the uranium isotope concentration is described by the lower straight line in Fig. 2, which is obtained from expression (1) after its normalization to the entire volume of the reactor:

$$y_{real}(x) = 0.3 + 0.85 \cdot x. \qquad (2)$$

The validity of the expression (2) is confirmed by calculating the average percentage of ^{235}U isotope over the height of the reactor $\overline{y}_{real}(x) = \int_0^1 y_{real}(x')dx' = 0.725$, which approximately corresponds to the natural.
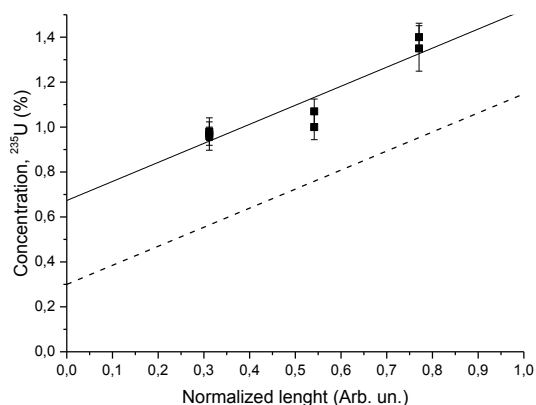
Fig. 2. Dependence of ^{235}U isotope concentration on the reactor normalized length. Solid line – formula (1), dashed line – (2)

Both straight lines in Fig. 2 characterize the uneven distribution of ^{235}U isotope over the reactor height.

To check the correspondence between the content of ^{235}U isotope in the initial sample and its total content in the extract and in the residue after extraction we measured its content in the reactor after sample acquisition at point III. The measurements showed that the content of this isotope was below the natural level and corresponded to a value of 0.63% (see Table 1). The balance equation for the uranium isotope content in the extract and in the residue in the reactor is as follows:

$$0.97 \cdot x + 0.63 \cdot (1-x) = 0.72 , \qquad (3)$$

where x – part of the volume of the supercritical fluid extracted from the reactor; the numerical coefficient for the first term is the amount of isotope extracted from the reactor at point III; the numerical coefficient for the second term is the amount of isotope remaining in the reactor; on the right side – the value of the natural amount of the isotope.

It follows from (3) that part of the volume of the supercritical fluid extracted from the reactor is equal to $x = 0.264$, which corresponds to the value of the primary pressure release – 4.22 MPa.

Thus, as a result of the experiments it was shown that at certain parameters of the supercritical fluid in a spatially inhomogeneous temperature field the ^{235}U isotope content is unevenly distributed over the reactor height: maximum at a distance of 5 cm from the lower flange (hotter) and decreases with a distance from it.

COMPARATIVE ASSESMENT OF THE EFFICIENCY OF URANIUM ISOTOPE REDISTRIBUTION

The spatial redistribution of the isotopic composition of natural uranium obtained in this work as a result of supercritical fluid extraction with carbon dioxide has an analogy with the problems of separating isotopes of various chemical elements. A series of such elements start from light elements Ar, Li [14, 21] and end with heavy ones – U, Pu [22, 23]. One of the parameters determined the efficiency of isotope separation is the separation factor [22, 23].

The separation factor α for a mixture of two isotopes is determined by the expression:

$$\alpha = \frac{c'}{1-c'}\left(\frac{c''}{1-c''}\right)^{-1} , \qquad (4)$$

where c' and $1-c'$ – concentration of light and heavy isotopes in the enriched mixture, respectively; c'' and $1-c''$ – the same in the original mixture.

From expression (2) it follows that at the height of the reactor $x = 0.67$ the content of ^{235}U is equals to 0.861%. This uranium content corresponds to the separation factor with taking into account the measurement error (see Fig. 2): $\alpha = 0.8695/0.72 \approx 1.2 \pm 0.12$.

Separation factors for uranium isotopes when using different separation methods are presented in Table 2.

Table 2
Separation factors ^{235}U and ^{238}U in different separation methods [23]

Separation method	Separation factor, α
Chemical enrichment [24]	1.0013…1.0030
Gas–diffusion enrichment	1.00429
Centrifugation (250 m/s)	1.026
Centrifugation (600 m/s)	1.233
Isotope separation in a system of opposite, axially symmetric magnetic fields [21]	30–40
CFE-CO$_2$	1.2 ± 0.12

Analyzing the results of the experiments it can be concluded that the separation factor of ^{235}U isotope is about 1.2 ± 0.12 in the SFE-U setup.

CONCLUSIONS

Redistribution of ^{235}U isotope of natural uranium in a supercritical dioxide medium with a spatial temperature gradient is shown by experiments in the paper. The diagram of the reactor of the experimental setup is presented and the principle of its operation is described. The method of sample preparation and the procedure for the extraction of natural uranium from granite samples are described. The isotopic composition of uranium in the obtained extracts is determined using the method of gamma spectrometry. It is shown that at certain parameters of the supercritical fluid the concentration of ^{235}U isotope is nonuniformly distributed over the height of the reactor: its concentration is maximum near the lower heated flange of the reactor and decreases with approaching the upper, colder flange of the reactor. The separation factor of ^{235}U isotope in supercritical carbon dioxide is determined; it is equal to 1.2 ± 0.12.

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ПРОСТРАНСТВЕННО-НЕОДНОРОДНОЕ РАСПРЕДЕЛЕНИЕ ИЗОТОПА ^{235}U ПРИ СВЕРХКРИТИЧЕСКОЙ ФЛЮИДНОЙ ЭКСТРАКЦИИ ДИОКСИДОМ УГЛЕРОДА В ГРАДИЕНТНОМ ПОЛЕ ТЕМПЕРАТУР

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Экспериментально исследовано пространственное перераспределение изотопа ^{235}U природного урана в градиентном поле температуры по высоте реактора в сверхкритическом диоксиде углерода. Приведена схема и описан принцип работы реактора. Описаны метод подготовки исходных образцов из образцов гранитов, содержащих природный уран, и порядок проведения экстракции. Вывод о пространственном перераспределении изотопов ^{235}U в сверхкритическом диоксиде углерода основан на анализе гамма-спектров

экстрактов. Показано, что в сверхкритическом флюиде концентрация изотопа ^{235}U максимальна вблизи нижнего подогреваемого фланца реактора, и уменьшается с приближением к верхнему, охлаждаемому фланцу. Сделан вывод о том, что коэффициент разделения изотопа ^{235}U в сверхкритическом диоксиде углерода может составлять величину около 1.2 ± 0.12.

ПРОСТОРОВО-НЕОДНОРІДНИЙ РОЗПОДІЛ ІЗОТОПІВ ^{235}U ПРИ НАДКРИТИЧНІЙ ФЛЮЇДНІЙ ЕКСТРАКЦІЇ ДІОКСИДОМ ВУГЛЕЦЮ В ГРАДІЄНТНОМУ ПОЛІ ТЕМПЕРАТУР

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Експериментально досліджено просторовий перерозподіл ізотопу ^{235}U природного урану в градієнтному полі температури по висоті реактора в надкритичному діоксиді вуглецю. Наведено схему і описаний принцип роботи реактора. Описані метод підготовки вихідних зразків із зразків гранітів, що містять природний уран, і порядок проведення екстракції. Висновок про просторовий перерозподілі ізотопів ^{235}U в надкритичному діоксиді вуглецю заснований на аналізі гамма-спектрів екстрактів. Показано, що в надкритичному флюїді концентрація ізотопу ^{235}U максимальна поблизу нижнього фланця реактора, що підігрівається, і зменшується з наближенням до верхнього охолоджуваного фланця. Зроблено висновок про те, що коефіцієнт поділу ізотопу ^{235}U в надкритичному діоксиді вуглецю може становити величину близько 1.2 ± 0.12.