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Adsorption's Regularities of Radionuclides on Adsorbents from Solutions

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Abstract

In the water of numerous thermal and cold springs in the Kalbajar region of Azerbaijan, radioactive radon was determined in different concentrations, and uranium isotopes were found in the soil of the locations of these springs in the same proportions as in natural uranium deposits. Uranium isotopes were transferred from soil samples to solutions. The regularities of adsorption from solutions of uranium isotopes on the surfaces of various adsorbents are studied. The results of studies of isotope sorption from aqueous solutions on various adsorbents showed high adsorption properties of DOWEX HCR S/S granular elastomer, activated carbon and expanded clay. The activation energies of desorption of uranium isotopes from the surface of adsorbents were also determined. Relatively low values (22.1 - 26.3 kJ/mol) of the activation energy of desorption of uranium isotopes from the surface of adsorbents indicate the absence of the chemical nature of sorption and the presence of only weak Van der Waals forces between the absorbed isotopes and the surface of the adsorbents in solutions.

Keywords

Uranium Isotopes, Radon, Ionizing Radiation, Adsorption, Activation Energy

1. Introduction

Deviation from the technological regime of radiation-chemical production and small industrial accidents, as well as large-scale nuclear technological catastrophes were observed with the development of radiation-chemical and nuclear technology. Numerous works on the prevention and neutralization of the consequences of nuclear disasters and cases of environmental pollution by radioactive emissions have been published in the periodical scientific literature [1] [2] [3] [4] [5].

The results of researches on the study of mechanisms and processes for cleaning contaminated environmental objects from radioactive waste have been published. Adsorbents are mainly used to purify substances from impurities, water from gases, heavy metals and radionuclides dissolved in them, to absorb toxic mixtures and gases from solutions. The absorption capacity of adsorbents depends on their specific surface area. The specific surface area of activated carbon is 500-1500, silica gel 800, ion-exchange resins 70 m^2/g. There are physical and chemical types of adsorption depending on the form of interaction of the adsorbent with the adsorbed substance. Numerous scientific studies are devoted to the development of complex technologies for soil and water purification, in addition to purification processes using adsorbents [6]-[15].

The physical sorption process is characterized by weak intermolecular Van der Waals forces between adsorbent and particles of adsorbed substances in solutions. As a result, the presence of opposite processes of adsorption and desorption, the weakening of adsorption with increasing temperature, and characteristic small values for the energy of desorption are observed. Chemical adsorption is accompanied by a high thermal effect and the formation of new chemical bonds, the process becomes irreversible and the speed of the process increases with increasing temperature. Consequently, desorption after chemisorption, in contrast to physical sorption, is significantly hampered due to the higher activation energy of this process (more than 40 kJ/mol) [16] [17].

The areas with an increased radioactive background were identified around the thermal springs in the Kelbajar region of Azerbaijan, as a result of numerous radiometric measurements. The waters of these sources contain high concentrations of radioactive radon (100 times higher than standard values). Analysis of soil samples from these areas showed that they contain the ancestors of radon, that is, isotopes of uranium.

The search for possible natural sources of radionuclides, or the study and development of effective methods for cleaning environmental objects and soil areas contaminated with radioactive emissions are an urgent task of radiation safety.

2. Experimental Part

The express sensory assays were carried out in all possible cases on places of sampling of water samples. Pre-sterilized dishes were used for soil and water sampling, transportation and storage [10]-[15].

The determination of radioactive radon in the investigated water sources was carried out with the radon measuring devices "ALPHAGUARD, Professional Radon Monitor, Frankfurt, Germany" and RAD7 Electronic Radon Detector (DURRIDGE Company Inc., USA). The radioactive radon gas Rn^{222} was carried into the ionization chamber of the measuring device by bubbling of 3 liters of atmospheric air during 10 minutes from the water sample [18].

Analytical-chemical and physical-chemical laboratory installations and equip-

ment were used for conducting of comprehensive analyzes of soil samples, water and it's mineral. Steam sterilizers "LDZH-30FBS" and "Tengor" were used to sterilize chemical glassware and glass jars. The "GFL-2304" distiller produced double-distilled water for rinsing chemical glassware and for making solutions. Centrifuges "TDL-5M" and "TD5A-WS" were used for centrifugation and sedimentation of solid impurities in water samples in ampoules with different volumes. Membrane filters were used to isolate solid impurities from water samples and for accelerated filtration. The surfaces and structure of soil and water minerals were comprehensively examined and studied under a scanning electron microscope (SEM, Carl Zeiss) at various magnifications. Using an additional stand for the SEM equipped with an X-ray tube, X-ray spectra and the composition of the content of chemical elements in the studied water minerals were obtained.

Weighing obtained by evaporation of solutions, soil extracts, water's mineral mass was carried out on a laboratory electronic balance. Active reactions of water samples were determined by universal indicator tests and pH-meters. Evaporation of water samples and soil extracts from laboratory heat-resistant glass beakers was carried out on laboratory heaters.

The measurements of the radioactive background were carried out by "In-Spector-1000" (Canberra Co., USA) radiometer-dosimeter, "Radiagem 2000" (Canberra Co., USA) radiometer equipped with alpha, beta, gamma, neutron detectors, "Thermo Eberline R020 SI" (Thermo Electron Co., USA) dosimeter, "PRM-470CG" (Tesla Systems Ltd., USA) dosimeter with gamma ray counter, "ИСП-РМ1401К-01 IP65" (Polimaster, Belarus) dosimeter equipped with gamma and neutron counters, "IdentiFinder" (Thermo Scientific Co., AFR-USA) and "GR-135 Plus" (Exploranium Co., USA) radiometer-dosimeter for isotope determination [6]-[15].

Identification and determination of the reactive quantity, organic and inorganic (harmful chemicals, heavy metals, radioactive isotopes) emissions carried out using traditional analytical-chemical and modern research physical-chemical methods. Certified point radioactive sources and standard solutions of uranium isotopes with different activity were used for the calibration. The decrease in the activity of the point source according to the half-life of isotopes was taken into account for previously certified weak point sources. The described devices are certified according to the ISO 9001 standard on the verification of measuring instruments by the Metrology Institute of the State Agency for Antimonopoly and Consumer Market Control of the European Union and the Republic of Azerbaijan.

The value of the energy of ionizing rays or particles determined by detector of spectrometers allowed to identified the isotopes of elements (that emits these rays or particles) when carrying out radiometric measurements, alpha and gamma spectrometric researches.

Atomic absorption AA-6800 spectrometer (Shimadzu), SEM (scanning electron microscope, Carl Zeiss), XRF and Expert-3L X-ray fluorescence spectrometers were used to determine elements and their quantities in the mass of minerals, isolated by analytical-chemical procedures from soil and water samples.

Radiometer-dosimeters, equipped with "Genie 2000" spectroscopic system gamma and alpha spectrometers (Canberra) were used for radiometric measurements and analysis of soil and water samples, to determine the activity of radionuclides in soil mass, in their extracts and on adsorbents [6]-[15].

3. The Results Obtained and Their Discussion

According to the Law of the Republic of Azerbaijan "On radiation safety of the population", the allowable value of the average annual dose for the population is 1 mSv, which corresponds to a dose rate of 0.115 or approximately 0.12 μSv/h for the population living in this area [19]. The results of radiometric measurements carried out in the country's territories show that the total radiation, that is, the rate of the absorbed dose (0.03 - 0.12 μSv/h) does not exceed generally the Permissible Limit/PL = 0.12 μSv/h) and the level of alpha radiation from ground cover is 0 - 0.03 Bq/cm^2 in many areas of the country. But, the results of radiometric measurements on the mountains of the Kelbajar region showed high levels (up to 3.8 μSv/h) of ionizing radiation and the value of the alpha background is 0.05 - 0.35 Bq/cm^2, which is anomaly of background radiation. Higher values of the total and alpha radiation were observed in soil areas around thermal and cold springs.

The concentration of radioactive radon in the thermal springs in the Kalbajar region of Azerbaijan, as well as in the thermal springs of the "Istisu" sanatorium of this region is below the guideline indicator (60 Bq/l). These waters are suitable for use in medical procedures prescribed by the attending physicians.

But, the concentration of radioactive radon in the waters of cold and thermal springs located at an altitude of 2385 m above sea level in the western territories of the region exceeds the directive indicator by 100 times and these waters are undrinkable. The half-life of radioactive radon is 91.8 hours, *i.e.* approximately 4 days. Consequently, a 2-fold decrease in radon concentration is observed in water during this time.

Soil samples taken in the course of radiation measurements carried out on the territory of the Kalbajar region were extracted with nitric acid, hydrochloric acid, sodium hydroxide solutions and distilled water to remove radionuclides. Soil samples weighing 1 kg were treated with a mixture of solutions of nitric (2 M) and hydrochloric (4 M) acids, soil residues were washed with distilled water and then treated with a sodium alkali solution (1 M). The liquid fractions of all three stages of extraction of each soil sample were collected, respectively, in a separate dish. After filtration and centrifugation, the liquid fraction was evaporated. Each obtained resulting white powder mineral was dissolved in 1 liter of distilled water. Mineral powders and their aqueous solutions were analyzed on alpha and gamma spectrometers for isotope detection. The isotope activities in prepared aqueous solutions were 130, 131 and 6 Bq for U^{238} (99.24%), U^{234} (0.0054%), U^{235} (0.702%), respectively, which shows that soil samples taken from areas around springs contain uranium isotopes in similar proportions found in natural ura-

nium deposits.

Activated carbon, granular anthracite, pebble-gravel sand mass, DOWEX HCR S/S cation exchanger granular elastomer widely used at drinking water treatment stations. For the adsorption of uranium isotopes from an aqueous solution on activated carbon, anthracite, pebble-gravel sand mass, DOWEX HCR S/S granular elastomer, and expanded clay mass were used after their drying at 100˚C. Adsorbents in the amount of 3, 10, 20, 50 and 100 g were kept in solution for 30 minutes, then removed and dried for 1 hour. The quantity (or activity) of isotopes adsorbed by the adsorbent mass were determined using alpha and gamma spectroscopies. The activity of isotopes adsorbed on the adsorbent was determined both by spectroscopic measurement of its activity on the adsorbent and by the difference in the activities of radiation from a given isotope in the initial and residual solutions. The regularity of adsorption of uranium isotopes on granular elastomer DOWEX HCR S/S from an aqueous solution are shown in **Figures 1-4**.

The activity of the isotope U^{238} adsorbed on 100 g of the adsorbent DOWEX HCR S/S (within 30 min in the initial solution with an activity 130 Bq of U^{238}) was 92 Bq, on activated carbon 65 Bq, on expanded clay 60 Bq, on gravel sand 54 Bq and on granular anthracite 44 Bq, respectively. As can be seen from **Figure 1**, 3 and 4 small amounts of U^{234} and U^{235} isotopes in solutions are completely absorbed by little amounts of granular elastomer DOWEX HCR S/S masses. A similar dependence of the adsorption of such small amounts of U^{234} and U^{235} is observed for activated carbon, expanded clay, pebble gravelly sand and granular anthracite masses. But, as can be seen from **Figure 1**, **Figure 2** only 71% of the U^{238} isotope (Aads. × 100%/Ainit.solut. = 92 Bq × 100%/130 Bq = 71%) from solution adsorbs in 100 g of granular elastomer DOWEX HCR S/S mass.

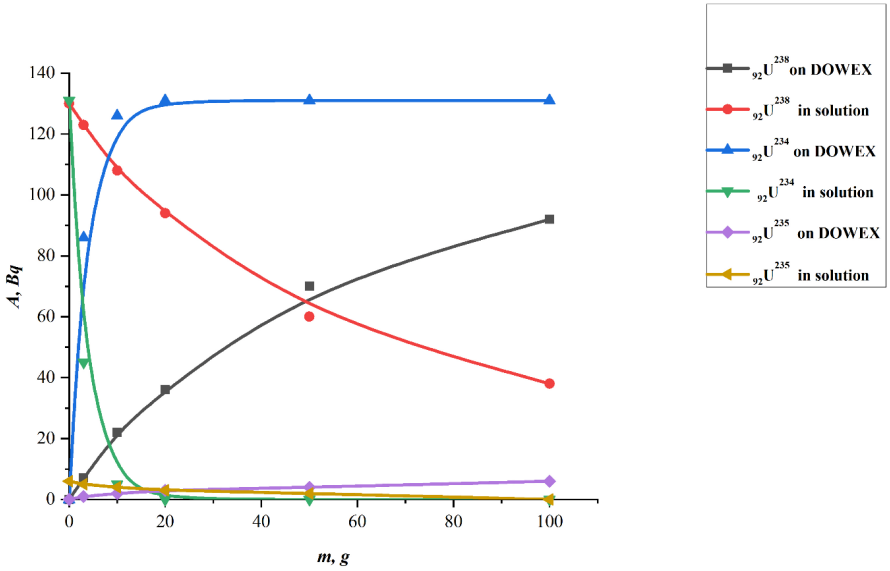

Figure 1. Changes in the activity of isotopes U^{238}, U^{234}, U^{235} in the adsorbent mass and water solution as a result of the adsorption of uranium isotopes on granular elastomer DOWEX HCR S/S masses from water solution.

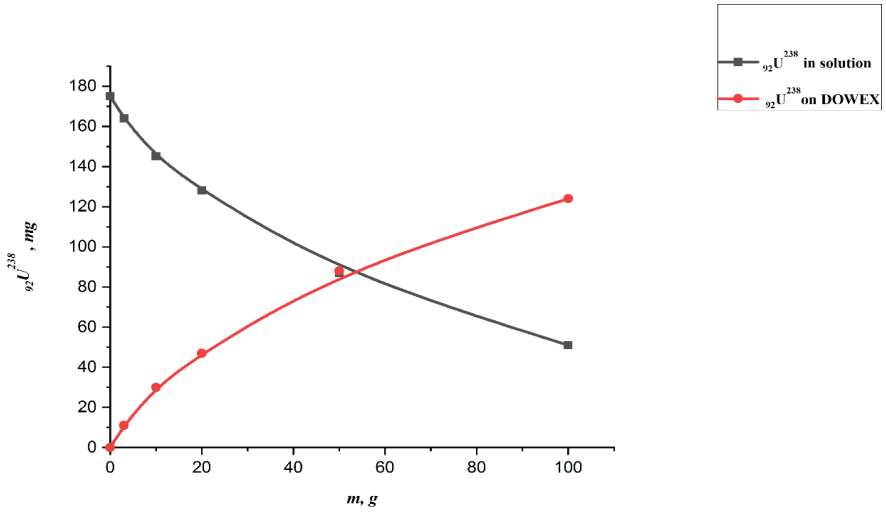

Figure 2. Changes in the amount of U^{238} isotope in the adsorbent mass and solution as a result of the adsorption of uranium isotopes (U^{238}) on granular elastomer DOWEX HCR S/S masses from water solution.

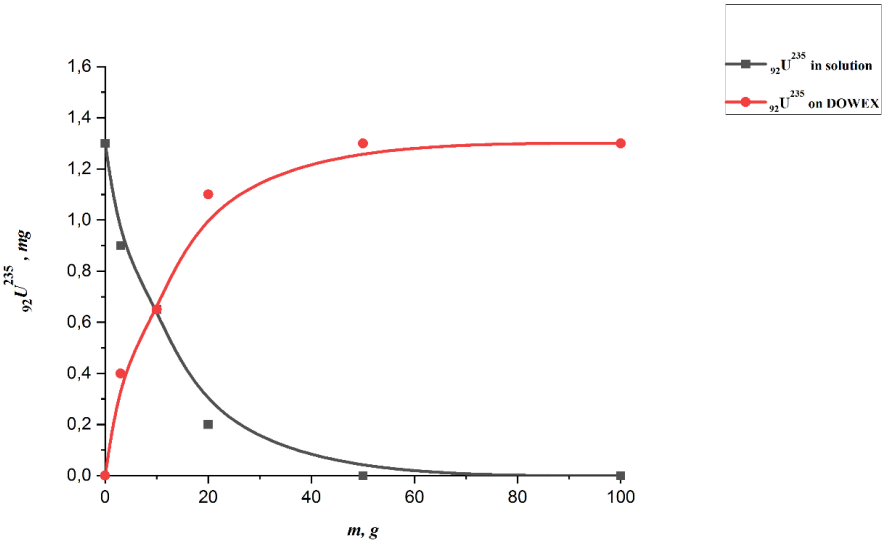

Figure 3. Changes in the amount of U^{235} isotope in the adsorbent mass and solution as a result of the adsorption of uranium isotopes (U^{235}) on granular elastomer DOWEX HCR S/S masses from water solution.

The regularity of adsorption of U^{238} from an aqueous solution on all adsorbents used in our studies (granular elastomer DOWEX HCR S/S, activated carbon, expanded clay, pebble gravelly sand and granular anthracite masses) are shown in **Table 1**.

As can be seen from **Table 1** only 71% of the U^{238} isotope from solution adsorbs on 100 g of granular elastomer DOWEX HCR S/S mass, 50% of the U^{238} isotope on activated carbon, 46% of the U^{238} isotope on expanded clay, 42% of the U^{238} isotope on pebble gravelly sand, and 34% of the U^{238} isotope on granular anthracite mass ($A_{ads.} \times 100\%/A_{initial\ solution}$ = 44 Bq × 100%/130 Bq = 34%).

Thus, the regularities of radioisotope adsorption from aqueous solutions were

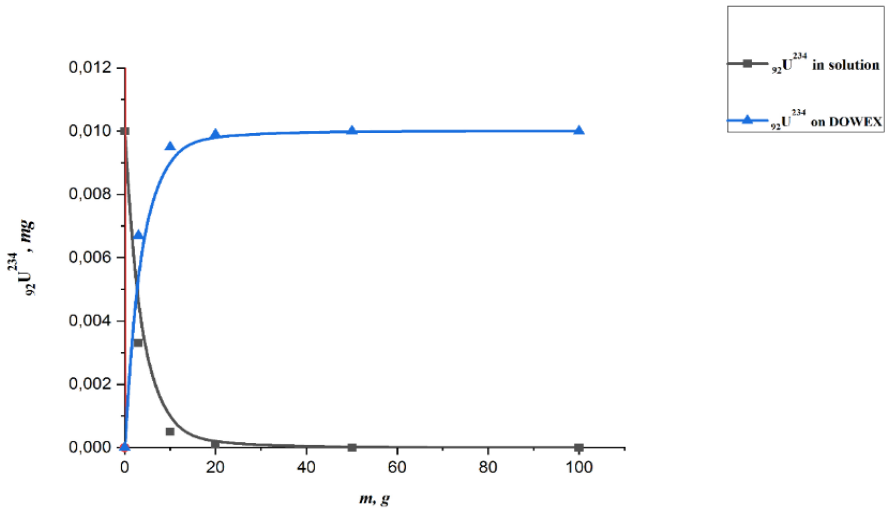

Figure 4. Changes in the amount of U^{234} isotope in the adsorbent mass and solution as a result of the adsorption of uranium isotopes (U^{234}) on granular elastomer DOWEX HCR S/S masses from water solution.

Table 1. Adsorption of U^{238} isotope with specific activities of 130 Bq on granular elastomer DOWEX HCR S/S, activated carbon, expanded clay, pebble gravelly sand and granular anthracite masses from water solution.

Quantity of adsorbent, g	Activity of U^{238} adsorbed on granular elastomer DOWEX HCR S/S, Bq	Activity of U^{238} adsorbed on activated carbon, Bq	Activity of U^{238} adsorbed on expanded clay, Bq	Activity of U^{238} adsorbed on pebble gravelly sand, Bq	Activity of U^{238} adsorbed on granular anthracite, Bq
3	8	7	6	5	4
10	22	15	12	10	7
20	35	22	18	17	12
50	65	36	33	30	26
100	92	65	60	54	44

studied in granular activated carbon, anthracite, pebble gravelly sand, DOWEX HCR S/S cationite, and expanded claymasses. The samples of granular DOWEX HCR S/S cationite (71%), activated carbon (50%) and expanded clay (46%) have the high adsorption property among the adsorbents used in experiments. The pebble gravel sand (42%) and granular anthracite (34%) have the relatively low adsorption capacity among used adsorbents. It was determined as a result of the conducted research that the degree of adsorption decreased according to the types of adsorbent in the following order: DOWEX HCR S/S cationite - activated carbon - expanded clay - pebble gravelly sand - granular anthracite.

The regularities of adsorption of uranium isotopes on adsorbent masses from aqueous solutions can be used as an original cleaning method of the drain water

of the physical stage of crude oil refining or other radiochemical processes to the norms for waste water.

İt is possible to determine the activation energy for adsorption or desorption/tgα = −Ea/R = [(lnWo)1 − (lnWo)2]/(1/T1 − 1/T2)/in the coordinate system lnWo = f(1/T).

By determining the desorption of ^{238}U isotope (adsorbed to the 100 g of adsorbent from water solutions at a temperature of 298 K) proportional to the increase in temperature a graphical interpretation of the dependence was made for the used adsorbents (**Figure 5**). The experimental values of the desorption in **Table 2** were used to draw up the graphical dependences in **Figure 5**. The values of the activation energy of the desorption process of ^{238}U isotopes from the adsorbents calculated on the basis of the graphical dependences in **Figure 5** are shown in the "activation energy" column of **Table 2**.

The calculated value for the activation energy of the process of U^{238} desorption from DOWEX HCR S/S cationite is 26.3 kJ/mol.

$$
\begin{aligned}
-E_a/R &= \left[\ln\left(D_{323}/A_{298}\right) - \ln\left(D_{343}/A_{298}\right)\right] / \left[\left(1000/T_1\right) ? \left(1000/T_2\right)\right] \\
&= \left[\ln\left(26/92\right) - \ln\left(46/92\right)\right] / \left[\left(1000/323\right) - \left(1000/343\right)\right] \\
&= -1.265 - \left(-0.693\right) / \left(3.096 - 2.915\right) \\
&= -0.572/0.181 \\
&= -3.16
\end{aligned}
$$

$$E_a = 3.16 \times 8.3144 = 26.3 \text{ kJ/mol}.$$

Table 2. Desorption of U^{238} isotopes from studied 100 g adsorbents at different temperatures.

Type of adsorbent	An increase in the degree of desorption of U^{238} isotope from 100 g of adsorbents with an increase in temperature, Bq/100 g					The value of the activation energy of desorption of the U^{238} isotope from adsorbents, E_a (kJ/mol).
	298 K	323 K		343		
	A_{298}	D_{323}	Ln (D_{323}/A_{298})	D_{343}	Ln (D_{343}/A_{298})	
DOWEX HCR S/S cationite	92	26	−1.265	46	−0.693	26.3
Activated carbon	65	19	−1.230	32	−0.709	24.6
Expanded clay	60	18	−1.204	30	−0.693	23.6
Pebble gravelly sand	54	16	−1.216	26	−0.731	22.5
Anthracite	44	13	−1.220	21	−0.740	22.1

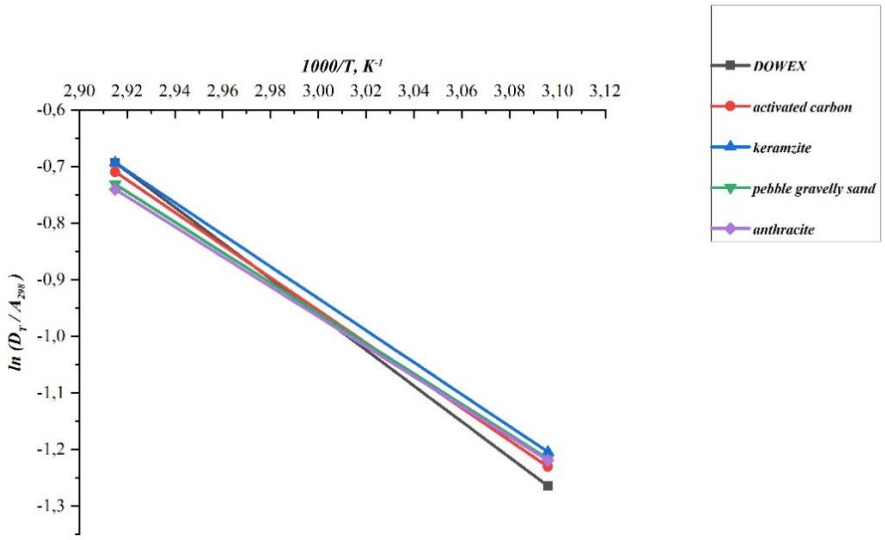

Figure 5. Desorption of U^{238} isotope from 100 g of adsorbents at different temperatures.

It shows that the value of activation energy (E_a) varies in the range of 22.1 - 26.3 kJ/mol, after performing calculations with the same algorithm for other adsorbents (see: **Table 2**). These values confirm that the adsorption process is a weak exothermic process, while the desorption process is a weak endothermic process.

As can be seen from **Table 2**, U^{238} isotope adsorbed on DOWEX HCR S/S cationite at 298 K (weak exothermic process) partially desorbs as a result of increasing the temperature to 323 K and 343 K (weak endothermic process). The fact that the value of the activation energy determined for the studied desorption processes is lower than 40 kJ/mol indicates that the adsorption process in all 5 types of adsorbents used in the experiments have a physical nature (physical sorption), and it is not accompanied by chemical sorption. This data indicates that only weak Van der Waals forces exist between the adsorbed uranium isotopes and the adsorbents in solutions.

4. Conclusions

1) The results of radiometric measurements on the mountains/on soil areas around thermal and cold springs/of the Kelbajar region of Azerbaijan showed high levels (up to 3.8 μSv/h) of ionizing radiation and the value of the alpha background 0.05 - 0.35 Bqeq/cm^2, which is anomaly of background radiation. The isotope activities in prepared aqueous solutions were 130, 131 and 6 Bq for U^{238} (99.24%), U^{234} (0.0054%), U^{235} (0.702%), respectively, which shows that soil samples taken from areas around springs contain uranium isotopes in similar proportions found in natural uranium deposits.

2) The activity of the isotope U^{238} adsorbed on 100 g of the adsorbent DOWEX HCR S/S (during 30 min in the initial solution with an activity 130 Bq of U^{238}) was 92 Bq, on activated carbon 65 Bq, on expanded clay 60 Bq, on gravel sand 54 Bq and on granular anthracite 44 Bq, respectively. It was determined,

that the adsorption degree decrease according to the types of adsorbent in the following order: DOWEX HCR S/S granular elastomer - activated carbon - expanded clay - pebble gravelly sand - granular anthracite. The adsorption's regularities of isotopes on adsorbent masses from aqueous solutions can be used as an original cleaning method of the drain water of the physical stage of crude oil refining to the norms for waste water.

3) The value for activation energy of the process of U^{238} desorption from DOWEX HCR S/S elastomer, activated carbon, expanded clay, pebble gravel sand and granular anthracite varies in the range of 22.1 - 26.3 kJ/mol. and indicates that only weak Van der Waals forces exist between the adsorbed uranium isotopes and adsorbents in solutions.

4) The concentration of radioactive radon in the water samples taken from numerous thermal springs of the "Istisu" sanatorium in the Kalbajar region of Azerbaijan is below the guideline indicator (60 Bq/l). These waters are suitable for use in medical procedures prescribed by the attending physicians. It is planned to resume (prevented more than 40 years ago due to global regional problems) the production of the original trade-mark—"Istisu" medicinal water, with the concentration of radon below the permissible value.

5) The concentration of radioactive radon in the waters of cold and thermal springs located at an altitude of 2385 meter above sea level in the western territories of the region on the mountain slopes exceeds the directive indicator by 100 times. These waters are undrinkable. But, considering the 2-fold decrease in radon concentration in water over 4 days, the waters with a 100-fold high radon content, after a certain time (several months) can be used for application in medical procedures, which allows their transportation to large distances.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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