

CONCENTRATION OF RADON IN SPRINGS OF KELBAJAR REGION, REGULARITIES OF ADSORPTION OF RADIONUCLIDES FROM SOLUTIONS ON ADSORBENTS

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Higher values of the total radiation and the intensity of alpha radiation were observed in soil areas around thermal and cold springs on the mountain slopes of the Kelbajar region of Azerbaijan. The concentration of radioactive radon in the water of the thermal springs of the "Istisu" sanatorium, located at an altitude (2203 m), is below the directive indicator. However, the concentration of radon in the water of springs, located at an altitude of 2385 m, exceeds the directive indicator by 100 times. Soil samples taken from areas around springs contain uranium isotopes in similar proportions found in natural uranium deposits. Regularities of sorption of uranium isotopes from aqueous solutions on some adsorbents have been studied. It has been established that weak Van der Waals forces exist between the absorbed isotopes and the adsorbents.

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INTRODUCTION

In addition to deposits of radioactive substances formed in the bowels of the planet during its formation, contamination of soil with radionuclides can occur as a result of radioactive dust settling from cosmic dust clouds on the surface of the planet or as one of the side effects of the development of nuclear technologies. Numerous works on soil's cleanup and prevention of groundwater pollution are discussed in the scientific literature [1 - 6].

It is proposed to create complex technologies for cleaning soil and water. Natural and synthetic materials with a large specific surface area and high dispersion are called adsorbents. The process of sorption of a substance or substances from a solution or gas by the surface of a solid substance is called adsorption, and similarly, the process of sorption of a substance or substances from a solution or gas by the surface of a liquid is called absorption. On the contrary, the transfer of a substance or substances from the surface of phase separation to the volume is called desorption. Adsorbents are mainly used to purify water from heavy metals and impurities, purify gases, absorb foreign gases and toxic mixtures from solutions. Adsorbents can also be used to purify aqueous solutions from radionuclides dissolved in them. The average value of the indicator characterizing the dimensions of internal pores, channels, free spaces in particles located in crushed porous bodies or dispersed system is called the specific surface. The specific surface shows the ratio of the total surface area of a porous and crushed object to its volume or mass and is directly proportional to the degree of grinding and inversely proportional to the particle size of the dispersed phase. Absorption capacity of adsorbents depends on the specific surface. The specific surface area of activated carbon is 500...1500, silica gels 800 and ion-exchange resins 70 m²/g. Physical and chemical adsorption types are distinguished depending on the form of interaction between adsorbed matter and adsorbent particles. The physical adsorption process is char-

acterized by weak intermolecular Van der Waals forces between adsorbent and particles of adsorbed substances. 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 heat of adsorption are observed. Chemical adsorption is accompanied by the formation of new chemical bonds, the process becomes irreversible, the speed of the process increases as the temperature increases, and a high thermal effect characteristic of chemical sorption are observed.

Radiometric measurements were carried out at different heights above sea level in the Kelbajar region of Azerbaijan during scientific expeditions of our team, and soil samples were taken from local areas around numerous thermal and cold springs located in mountainous areas of region. Search and identification of possible sources of nuclear fuel is important for meeting the energy needs of the country, and studying the possibility of developing efficient methods for cleaning environmental objects contaminated with radionuclides is also an urgent task for ensuring the radiation safety of the population.

1. EXPERIMENTAL PART

The measurements of the radioactive background were carried out with "InSpector-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, "ICPI-PM1401K-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 [7 - 10]. Analyzes on atomic absorption spectrometer (AA-6800, Shimadzu), X-ray fluorescence spectrometers (Expert-3L and XRF) and gamma spectrometer equipped with

high purity germanium detector and “Genie 2000” spectroscopic system (HP-Ge, Canberra Co.) were carried out to identify of radioactive elements in soil samples and in minerals. 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.

Determination of radioactive radon gas was carried out from the 100 ml sample taken in accordance with the method of radon gas determination in spring water. ^{222}Rn was ejected from the water by air bubbling at a speed of 1 l/min and 0.3 l/min for 10 min and entered into the ionization chamber of the radon measuring devices ALPHAGUARD Professional Radon Monitor (Bertin Technologies GmbH., Frankfurt) and RAD7 Electronic Radon Detector (DURRIDGE Company Inc., USA).

2. DISCUSSION OF THE RESULTS

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 $\mu\text{Sv/h}$ for the population living in this area [11]. 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 $\mu\text{Sv/h}$) does not exceed the Permissible Limit /PL = 0.12 $\mu\text{Sv/h}$) and the level of alpha radiation from ground cover is 0...0.03 $\text{Bq}_{\text{eq}}/\text{cm}^2$ in many areas of the country. But, the results of radiometric measurements on the mountains (slope of the Delidag ridge) of the Kelbajar region of Azerbaijan showed high levels (up to 3.8 $\mu\text{Sv/h}$) of ionizing radiation and the value of the alpha background is 0.05...0.35 $\text{Bq}_{\text{eq}}/\text{cm}^2$, 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 values of the dose rate of total radiation and alpha particle stream on local soil areas adjacent to springs at altitudes up to 1600 m above sea level vary in the range of 0.05...0.12 $\mu\text{Sv/h}$ and 0...0.05 $\text{Bq}_{\text{eq}}/\text{cm}^2$, from 1600 to 2200 m in the range of 0.10...0.20 $\mu\text{Sv/h}$ and 0...0.06 $\text{Bq}_{\text{eq}}/\text{cm}^2$, from 2200 to 2300 m in the range of 0.15...1.8 $\mu\text{Sv/h}$ and 0.01...0.10 $\text{Bq}_{\text{eq}}/\text{cm}^2$, from 2300 to 2500 m in the range of 0.15...3.80 $\mu\text{Sv/h}$ and 0.02...0.3 $\text{Bq}_{\text{eq}}/\text{cm}^2$, respectively.

The concentration of radioactive radon in the water samples (activity of ^{222}Rn fluctuate in the range of 0.5...9.0 Bq/l) taken from numerous thermal springs at altitudes up to 2300 m above sea level and in springs of the “Istisu” sanatorium (2203 m) in the Kelbajar region of Azerbaijan is below the directive indicator (60 Bq/l). These waters are suitable for use in medical procedures prescribed by the attending physicians.

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 on the mountain /Delidag/ slopes exceeds the directive indicator by 100 times. These waters are undrinkable.

Soil samples taken in the course of radiation measurements carried out on the territory of the Kelbajar region were extracted with nitric acid, hydrochloric acid, sodium hydroxide solutions and distilled water to remove radionuclides. Soil samples weighing 1 kg taken around thermal and cold springs in the territory of Kelbajar region at the slope of the Delidag mountain an altitude of 2386 m, where anomaly of background radiation (3.8 $\mu\text{Sv/h}$) were observed, were also treated with a similar procedure.

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 l 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 ^{238}U (99.24%), ^{234}U (0.0054%), ^{235}U (0.702%), respectively, which shows that soil samples taken from areas around springs contain uranium isotopes in similar proportions found in natural uranium 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 min, then removed and dried for 1 h. The quantity (or activity) of isotopes adsorbed by the adsorbent mass were determined using alpha and gamma spectroscopies. The degree of adsorption of a specific uranium isotope by the adsorbent was estimated from the activity of radiation of the adsorbed isotopes from the surface of the adsorbent. The activity of a specific 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 after removal the adsorbent. The regularities of adsorption of uranium isotopes on granular elastomer DOWEX HCR S/S masses from an aqueous solution are shown in Fig. 1.

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

As can be seen from Fig. 1, small amounts of ^{234}U and ^{235}U isotopes in solutions are completely absorbed by little amounts of granular elastomer DOWEX HCR S/S masses. But, only 71% of the ^{238}U isotope ($A_{\text{adsorb.}} \times 100\% / A_{\text{initial solution}} = 92 \text{ Bq} \times 100\% / 130 \text{ Bq} = 71\%$) from solution adsorbs in 100 g of granular elastomer DOWEX HCR S/S mass.

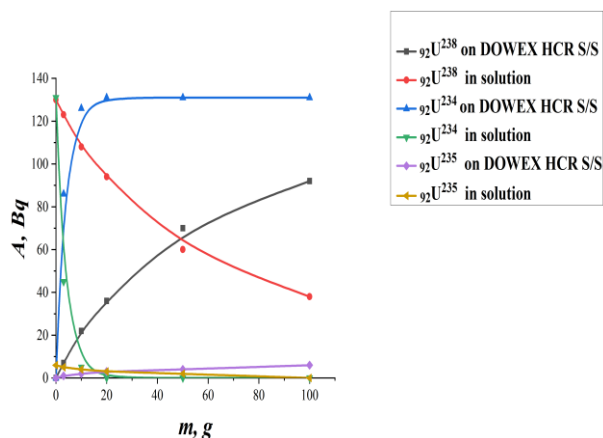


Fig. 1. Changes in the activity of isotopes in the adsorbent mass and solution as a result of the adsorption of uranium isotopes on granular elastomer DOWEX HCR S/S masses from water solution

The regularity of adsorption of uranium isotopes from an aqueous solution on activated carbon, expanded clay /keramzite/, pebble gravelly sand and granular anthracite masses are shown in Tables 1-4.

Table 1

Adsorption of ^{238}U , ^{234}U , and ^{235}U radioisotopes with specific activities of 130, 131, and 6 Bq, respectively, on activated carbon masses from water solution

Quantity of adsorbent, g	$A_{\text{adsorbent}}(^{238}\text{U})$	$A_{\text{adsorbent}}(^{235}\text{U})$	$A_{\text{adsorbent}}(^{234}\text{U})$
3	7	1	80
10	15	2	120
20	22	3	130
50	36	4	131
100	65	6	131

As can be seen from Table 1, small amounts of ^{234}U and ^{235}U isotopes in solutions are completely absorbed by little amounts of activated carbon masses and only 50% of the ^{238}U isotope ($A_{\text{ads.}} \times 100\% / A_{\text{init.solut.}} = 65 \text{ Bq} \times 100\% / 130 \text{ Bq} = 50\%$) from solution adsorbs in 100 g of activated carbon mass.

Table 2

Adsorption of ^{238}U , ^{234}U , and ^{235}U radioisotopes with specific activities of 130, 131, and 6 Bq, respectively, on expanded clay /keramzite/ masses from water solution

Quantity of adsorbent, g	$A_{\text{adsorbent}}(^{238}\text{U})$	$A_{\text{adsorbent}}(^{235}\text{U})$	$A_{\text{adsorbent}}(^{234}\text{U})$
3	6	1	76
10	12	2	116
20	18	3	130
50	33	4	131
100	60	6	131

As can be seen from Table 2, small amounts of ^{234}U and ^{235}U isotopes in solutions are completely absorbed by little amounts of expanded clay masses and only 46% of the ^{238}U isotope ($A_{\text{ads.}} \times 100\% / A_{\text{init.solut.}} = 60 \text{ Bq} \times 100\% / 130 \text{ Bq} = 46\%$) from solution adsorbs in 100 g of expanded clay /keramzite/ mass.

Table 3

Adsorption of ^{238}U , ^{234}U , and ^{235}U radioisotopes with specific activities of 130, 131, and 6 Bq, respectively, on pebble gravelly sand masses from water solution

Quantity of adsorbent, g	$A_{\text{adsorbent}}(^{238}\text{U})$	$A_{\text{adsorbent}}(^{235}\text{U})$	$A_{\text{adsorbent}}(^{234}\text{U})$
3	5	1	74
10	10	2	108
20	17	3	126
50	36	4	131
100	54	6	131

As can be seen from Table 3, small amounts of ^{234}U and ^{235}U isotopes in solutions are completely absorbed by little amounts of pebble gravelly sand masses and only 42% of the ^{238}U isotope ($A_{\text{ads.}} \times 100\% / A_{\text{init.solut.}} = 54 \text{ Bq} \times 100\% / 130 \text{ Bq} = 42\%$) from solution adsorbs in 100 g of gravelly sand mass.

Table 4

Adsorption of ^{238}U , ^{234}U , and ^{235}U radioisotopes with specific activities of 130, 131, and 6 Bq, respectively, on granular anthracite masses from water solution

Quantity of adsorbent, g	$A_{\text{adsorbent}}(^{238}\text{U})$	$A_{\text{adsorbent}}(^{235}\text{U})$	$A_{\text{adsorbent}}(^{234}\text{U})$
3	4	1	69
10	7	2	102
20	12	3	122
50	26	4	130
100	44	6	131

As can be seen from Table 4, small amounts of ^{234}U and ^{235}U isotopes in solutions are completely absorbed by little amounts of granular anthracite masses. But, only 34% of the ^{238}U isotope ($A_{\text{ads.}} \times 100\% / A_{\text{init.solut.}} = 44 \text{ Bq} \times 100\% / 130 \text{ Bq} = 34\%$) from solution adsorbs in 100 g of granular anthracite mass.

Thus, the regularities of radioisotope adsorption from aqueous solutions were studied in granular activated carbon, anthracite, pebble gravelly sand, DOWEX HCR S/S cationite, and expanded clay /keramzite/ masses. The samples of granular DOWEX HCR S/S cationite (71%), activated carbon (50%) and expanded clay /keramzite/ (46%) have the high adsorption property among the adsorbents used in experiments. Among the adsorbents used, pebble-gravel sand (42%) and granular anthracite (34%) have a relatively weak adsorption property. It was determined as a result of the conducted research, that the adsorption degree varies /decrease/ 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.

It is possible to determine in the coordinate system $\ln W_0 = f(1/T)$ the activation energy for adsorption or desorption $/\text{tg}\alpha = -E_a/R = [(\ln W_0)_1 - (\ln W_0)_2]/(1/T_1 - 1/T_2)/$.

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 (Fig. 2). The experimental desorption values in the Table 5 were used to draw up the graphical dependences in Fig. 2. 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 Fig. 2 are shown in the “activation energy” column of Table 5.

Table 5

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

Type of adsorbent	An increase in the degree of desorption of ^{238}U isotope from 100 g of adsorbents with an increase in temperature, Bq/100 g					The value of the activation energy of desorption of the ^{238}U isotope from adsorbents, E_a (kJ/mol)
	298 K	323 K		343 K		
	A_{298}	D_{323}	$\ln(D_{323}/A_{298})$	D_{343}	$\ln(D_{343}/A_{298})$	
DOWEX HCR S/S cationite	92	26	-1.264	46	-0.693	26.2
Activated carbon	65	19	-1.230	32	-0.709	23.9
Expanded clay /keramzite/	60	18	-1.204	30	-0.693	23.5
Pebble gravelly san	54	16	-1.216	26	-0.731	22.3
Anthracite	44	13	-1.219	21	-0.740	22.0

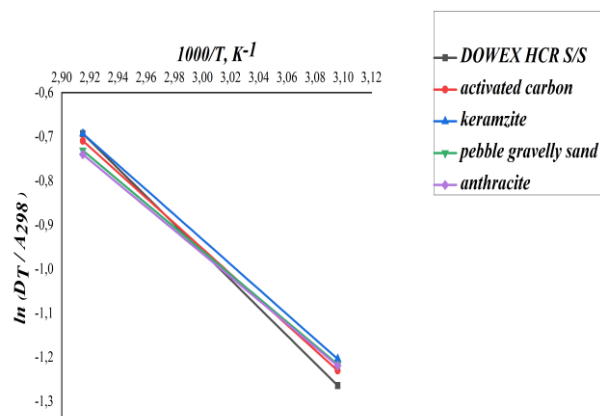


Fig. 2. Desorption of ^{238}U isotope from 100 g of adsorbents at different temperatures

The calculated value for the activation energy of the process of U^{238} desorption from DOWEX HCR S/S elastomer is 26.2 kJ/mol.

$-E_a/R = [\ln(D_{323}/A_{298}) - \ln(D_{343}/A_{298})]/[(1000/T_1) - (1000/T_2)] = [\ln(26/92) - \ln(46/92)]/[(1000/323) - (1000/343)] = -1.264 - (-0.693)/(3.096 - 2.915) = -0.571/0.181 = -3.15$; $E_a = 3.15 \times 8.3144 = 26.2$ kJ/mol.

It shows that the value of activation energy (E_a) varies in the range of 22.0...26.2 kJ/mol, after performing calculations with the same algorithm for other adsorbents (see Table 5). 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 5, U_{238} isotope adsorbed on DOWEX HCR S/S elastomer at 298 K (weak exothermic process) partially desorbs as a result of increasing the temperature to 323 and 343 K (weak endothermic process). The fact that the value of the activation energy determined for the desorption process is in the range of 8...40 kJ/mol, indicates that the adsorption

process in all 5 types of adsorbents used in the experiments, including DOWEX HCR S/S elastomer, 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.

CONCLUSIONS

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 $\mu\text{Sv/h}$) does not exceed the Permissible Limit (PL = 0.12 $\mu\text{Sv/h}$) and the level of alpha radiation from ground cover is 0...0.03 $\text{Bq}_{\text{eq}}/\text{cm}^2$ in many areas of the country. But, the results of radiometric measurements on the mountains (slope of the Delidag ridge) of the Kelbajar region of Azerbaijan showed high levels (up to 3.8 $\mu\text{Sv/h}$) of ionizing radiation and the value of the alpha background is 0.05...0.35 $\text{Bq}_{\text{eq}}/\text{cm}^2$, 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 values of the dose rate of total radiation and alpha particle stream on local soil areas adjacent to springs at altitudes up to 1600 m above sea level vary in the range of 0.05...0.12 $\mu\text{Sv/h}$ and 0...0.05 $\text{Bq}_{\text{eq}}/\text{cm}^2$, from 1600 to 2200 m in the range of 0.10...0.20 $\mu\text{Sv/h}$ and 0...0.06 $\text{Bq}_{\text{eq}}/\text{cm}^2$, from 2200 to 2300 m in the range of 0.15...1.8 $\mu\text{Sv/h}$ and 0.01...0.10 $\text{Bq}_{\text{eq}}/\text{cm}^2$, from 2300 to 2500 m in the range of 0.15...3.80 $\mu\text{Sv/h}$ and 0.02...0.35 $\text{Bq}_{\text{eq}}/\text{cm}^2$, respectively.

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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 22.0...26.2 kJ/mol and indicates that only weak Van der Waals forces exist between the adsorbed uranium isotopes and used adsorbents.

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КОНЦЕНТРАЦІЯ РАДОНУ В ДЖЕРЕЛАХ КЕЛЬБАДЖАРСЬКОГО РАЙОНУ, ЗАКОНОМІРНОСТІ АДСОРБЦІЇ РАДІОНУКЛІДІВ З РОЗЧИНІВ НА АДСОРБЕНТАХ

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Більш високі значення сумарної радіації та інтенсивності α -випромінювання спостерігалися в районах ґрунту навколо термальних і холодних джерел на схилах гір Кельбаджарського району Азербайджану. Концентрація радіоактивного радону у воді термальних джерел санаторію «Istisu», розташованого на висоті 2203 м, нижче директивного показника. Проте концентрація радону у воді джерел, розташованих на висоті 2385 м, перевищує нормативний показник у 100 разів. Зразки ґрунту, взяті з районів навколо джерел, містять ізотопи урану в таких же пропорціях, що й у природних родовищах урану. Досліджено закономірності сорбції ізоотопів урану з водних розчинів на деяких адсорбентах. Встановлено, що між поглиненими ізотопами та адсорбентами існують слабкі сили Ван-дер-Ваальса.