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STUDYING REGULARITIES OF RADIONUCLIDE ADSORPTION ON ADSORBENTS

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Abstract: The adsorption regularities of radioisotopes from aqueous solutions of nuclear material waste in granular activated coal, anthracite, gravel sand, DOWEX HCR S/S resin, and expanded clay masses have been studied. Among these adsorbents, granular DOWEX HCR S/S resin (72-75%), activated carbon (50-65%), and expanded clay (47-50%) have the highest adsorption property, and gravel sand (41%) and granular anthracite (33%) the lowest. Despite the fact that the values of the deactivation coefficient (1.05 - 3.75) of the purification processes carried out with the studied adsorbents are lower than those given in the scientific literature for the existing purification methods (DC > 4). However, expanded clay masses, which have large reserves as natural raw materials in our country, may be used for wastewater treatment to safety standards at the stage of physical oil refining. It was found that the activation energy (E_a) of the uranium isotope desorption process from the studied adsorbents varies in the range of 20.3-26.5 kCl/mol. These values indicate that the desorption process is a weak endothermic process in contrast to the exothermic adsorption process. The activation energies determined for the desorption process show that the adsorption process is of a physical nature (physical sorption) in all 5 types of adsorbents used in the experiments, and only weak van der Waals forces exist between the absorbed uranium isotopes and the adsorbent.

Keywords: adsorbents, nuclear material waste and solutions, isotopes, desorption.

1. Introduction

Natural and synthetic materials with a large specific surface area and high dispersion are called adsorbents. The absorption of gaseous substances or liquids in adsorbents upon contact with their surfaces is called adsorption. The average value of the indicator characterizing the dimensions of internal pores, channels, and free spaces in particles located in crushed porous bodies or a dispersed system is called the specific surface. The specific surface indicates 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. The absorption capacity of adsorbents, the efficiency of solid catalysts, and the filtering capacity of materials depend on the specific surface area. The specific surface values range from 500 to 1500 m² per gram for activated carbon, up to 800 m² per gram for silica gels, up to 70 m² per gram for macroporous ion exchange resins, and up to 10 m² per gram for diatomaceous gas-liquid chromatographs. Adsorbents are mainly used to purify water from heavy metals and impurities, as a catalytic carrier mass in gas masks, purify gases and alcohols, separate alcohols, and absorb foreign gases and toxic mixtures from solutions [1–4].

Natural and synthetic materials with a high specific surface area and high dispersion are called adsorbents. The absorption of substances from gases or liquids in adsorbents, where they come into contact with their surfaces, is called adsorption. The average value of an indicator characterizing the size of internal pores, channels, and spaces of particles in the main phase of a porous body or disperse system is called the specific surface area.

2. Methodical part

Isolation of radionuclides from samples taken from environmental objects is carried out under the conditions of dosimetric and mass spectroscopic control in a stationary laboratory using traditional analytical and chemical methods, grinding and preparing porridge samples in blenders, diluting and dissolving in distilled water, heat treatment and processing with aggressive substances, decantation, settling, filtration, sedimentation on a centrifuge, extraction, and evaporation.

A conclusion on the effectiveness of the decontamination process was made on the basis of gamma spectroscopic analyses of minerals obtained after sequential treatment of soil samples contaminated with radionuclides using weak acidic and alkaline solutions.

Model soil samples contaminated with nuclear waste used for analysis were prepared by adding a solution of uranyl nitrate with a given activity to 200 g samples of soil taken from the regions of the country.

To ensure radiation safety, the development of methods for isolating radioisotopes from soil samples and aqueous solutions obtained by soil extraction is one of the topical aims of radiochemical research, and there is a need to study such methods. In the experiments carried out for this purpose water extracts of soils contaminated with uranium isotopes were used adsorbents used in water treatment technology (granular activated carbon, anthracite, gravel-sand mass, DOWEX HCR-S / S cation exchanger) and raw materials with large reserves - expanded clay (brick production waste).

Samples of uranyl nitrate salt (UO₂(NO₃)₂), which is widely used in nuclear reactors, were used as the nuclear material in the experiments. The types of isotopes (238 U, 235 U, 234,236,237 U) and their concentration in the uranyl nitrate salt were determined by analyses performed on a gamma spectrometer with an HPGe detector.

3. Discussion of the obtained results

Taken during the radio monitoring carried out in the regions of the country soil samples were divided into parts weighing 200 grams. In order to obtain model soil samples contaminated with nuclear material waste from these soil samples, they were mixed with uranyl nitrate solution. The soil samples contaminated with nuclear waste were extracted with nitric acid, hydrochloric acid, sodium hydroxide solutions, and distilled water for cleaning them from nuclear waste. It was determined by the study of the regularities of uranium's adsorption in various adsorbents that the activity (activity concentration) of ²³⁸U, ²³⁵U, ^{234,236,237}U isotopes in aqueous solutions of a uranyl nitrate salt is in the ratio of 90:60:3.

The soil extracts were analyzed by HPGe gamma spectrometer with a high-purity germanium detector, provided with the Genie 2000 spectrometric system of Canberra Company in order to detect uranium isotopes and separate them with natural adsorbents. The activities of ²³⁸U and ²³⁵U isotopes were 90 Bq and 60 Bq, respectively, in extracts of model soil samples contaminated with nuclear material and the total activity of ^{234,236,237}U isotopes was 3 Bq. Analytical chemical analyses showed that investigated model soil samples were contaminated with contained nitrate salt /UO₂(NO₃)₂/ of isotopes ₉₂U²³⁸ and ₉₂U²³⁵ with trace amounts of U²³⁴, U²³⁶, and U²³⁷. Activated carbon /charcoal/ widely used in drinking water treatment plants, anthracite, activated pebble gravel sand mass, DOWEX HCR S/S cationic, and expanded clay /keramzite/ mass was used for adsorption of uranium isotopes from aqueous solution after drying at 100°C. Adsorbents in the amounts of 3 g, 10 g, 20 g, 50 g, and 100 g were kept in the solution

for 30 minutes, then removed and dried for 1 hour. Amounts of isotopes captured by adsorbent mass (activity determined by gamma spectroscopy) were determined.

Figure 1 and Table 1 show the adsorption of uranium isotopes from uranyl nitrate solution on activated carbon/charcoal masses and Figure 2 shows the regularity of β -radiation activity on the activated coal surface



Fig. 1. Changes in the activity of isotopes in the adsorbent mass and solution as a result of the adsorption of uranium isotopes from uranyl nitrate solution on activated carbon /charcoal/masses.



Fig. 2. The intensity of β -radiation generated on the surface of the adsorbent as a result of the adsorption of uranium isotopes from the aqueous solution of uranyl nitrate on activated carbon /charcoal/masses.

Table 1

Adsorption of ²³⁸U and ²³⁵U radioisotopes with specific activities of 90 and 60 Bq, respectively, on activated carbon /charcoal/ mass from uranyl nitrate solution and deactivation coefficient of the process ($DC = A_{initially solution}/A_{solution residue}$).

Quantity of adsorbent, g	Adsorption of uranium isotopes from an aqueous solution of uranyl nitrate to the activated carbon /charcoal/ mass during 1 hour and the change in the deactivation coefficient of the corresponding process				
	$A_{dsorbent}(^{238}U)$ or /Ainit. solution – A solution residue/	DC(²³⁸ U), (A _{init. solut.} / A _{solut.residue})	Adsorbent(²³⁵ U) (Ainit.solutAsol. residue)	DC(²³⁵), (A _{init.solut} ./ A _{sol. residue})	
3	5	1.06	2	1.03	
10	10	1.13	7	1.13	
20	15	1.20	12	1.25	
50	25	1.38	22	1.58	
100	45	2.00	38	2.73	

As can be seen from Figure 1, trace amounts of U^{234} , U^{236} , and U^{237} isotopes in uranyl nitrate solution are completely absorbed by small amounts of activated carbon /charcoal/ mass.

As can be seen from Table 1, 50% of the U^{238} isotope (A_{ads}.×100% / A_{init.solut}.= 45 Bq×100% / 90 Bq = 50%), and 63% of the U^{235} isotope (A_{ads}.×100% / A_{init.solut}.= 38 Bq×100% / 60 Bq= 63%) in the uranyl nitrate solution adsorbs in 100 g of activated carbon.

Tables 2, 3, 4, and 5 show the regularity of adsorption of uranium isotopes from uranyl nitrate solution in granular anthracite, activated pebble gravel sand, ion-exchange resin, and expanded clay /keramzite/ masses, respectively.

Table 2

Adsorption of ²³⁸U and ²³⁵U radioisotopes with specific activities of 90 and 60 Bq, respectively, from uranyl nitrate solution on granular anthracite masses and deactivation coefficient of the process (DC = $A_{initially solution}/A_{solution residue}$).

Quantity of adsorbent, g	Variation of the deactivation coefficient corresponding to the adsorption of uranium isotopes from an aqueous solution of uranyl nitrate to granular anthracite mass for 1 hour			
	A(²³⁸ U)	DC(²³⁸ U)	A(²³⁵ U)	DC(²³⁵ U)
3	2	1.02	2	1.02
10	5	1.06	5	1.06
20	8	1.10	8	1.10
50	18	1.15	13	1.17
100	30	1.5	19	1.27

As can be seen from Table 2, 33% of U^{238} and U^{235} isotopes in uranyl nitrate solution are adsorbed in 100 g of anthracite mass.

Table 3

Adsorption of ²³⁸U and ²³⁵U radioisotopes with specific activities of 90 and 60 Bq, respectively, from uranyl nitrate solution on pebble gravelly sand masses and deactivation coefficient of the process (DC = $A_{initially solution}/A_{solution residue}$).

Quantity of adsorbent, g	Variation of the deactivation coefficient corresponding to the adsorption of uranium isotopes from an aqueous solution of uranyl nitrate to a pebble gravelly sand mass within 1 hour			
	A(²³⁸ U)	DC(²³⁸ U)	A(²³⁵ U)	DC(²³⁵)
3	5	1.06	3	1.05
10	7	1.13	7	1.13
20	13	1.20	12	1.25
50	25	1.38	20	1.50
100	37	1.70	24	1.70

As can be seen from Table 3, 41% of U^{238} and U^{235} isotopes in uranyl nitrate solution are adsorbed in 100 g of pebble gravel sand mass.

Table 4

Adsorption of radioisotopes ²³⁸U and ²³⁵U with specific activities of 90 and 60 Bq, respectively, from uranyl nitrate solution on DOWEX HCR S/S cationite and deactivation factor of the process (DC = $A_{initially solution}/A_{solution residue}$).

Quantity of adsorbent, g	Variation of the deactivation coefficient corresponding to the adsorption of uranium isotopes from an aqueous solution of uranyl nitrate onto granular DOWEX HCR S/S cationite mass over 1 h			
	A(²³⁸ U)	DC(²³⁸ U)	A(²³⁵ U)	DC(²³⁵)
3	5	1.06	2	1.03
10	15	1.20	7	1.13
20	25	1.38	15	1.33
50	48	2.14	30	2.00
100	64	3.46	44	3.75

As can be seen from Table 4, 100 g of DOWEX HCR S/S cationite mass adsorbs 72% of the U^{238} isotope and 75% of the U^{235} isotope of the uranyl nitrate solution.

Large-scale production areas operating in the country include bricks, glass, glass containers, cement, bentonite, aluminum, saw stones, travertine, marble and granite facings, etc. production, ferrous and non-ferrous metallurgy, electrical engineering, chemistry, petrochemical, etc. industries have a special weight. Among the products and wastes of these production areas, the substance with the highest adsorption properties is "high-temperature baked porous clay residue scraps" - expanded clay /keramzite/ mass, accumulated in the surrounding areas as a waste of brick production plants operating in the Surakhani region of Baku and in most regions of the country. Since the porous granular baked clay waste (keramzite mass) processed at high temperatures typical of the brick "baking" industry with a large supply of raw materials is

suitable for use as an adsorbent, uranyl nitrate solutions were used in experiments to study the regularities of uranium isotope adsorption.

Table 5

Adsorption of ²³⁸U and ²³⁵U radioisotopes with specific activities of 90 and 60 Bq, respectively, from uranyl nitrate solution on expanded clay /keramzite/ masses and deactivation coefficient of the process ($DC = A_{initially solution}/A_{solution residue}$).

Quantity of adsorbent, g	Variation of the deactivation coefficient corresponding to the adsorption of uranium isotopes from an aqueous solution of uranyl nitrate to the expanded clay /keramzite/ mass within 1 hour			
	A (²³⁸ U)	DC (²³⁸ U)	A (²³⁵ U)	DC (²³⁵ U)
3	4	1.05	2	1.03
10	8	1.10	6	1.10
20	12	1.15	10	1.20
50	23	1.34	20	1.50
100	41	1.84	30	2.00

As can be seen from Figure 1, trace amounts of U^{234} , U^{236} , and U^{237} isotopes in the uranyl nitrate solution are completely absorbed by small amounts of adsorbent mass.

As can be seen from Table 5, 47% of the U^{238} isotope and 50% of the U^{235} isotope in the uranyl nitrate solution are adsorbed in 100 g of expanded clay /keramzite/.

Thus, the regularities of radioisotope adsorption from aqueous solutions were studied in granular activated charcoal, anthracite, pebble gravelly sand, DOWEX HCR S/S cationite, and expanded clay /keramzite/ masses. Among the mentioned adsorbents, the samples of granular DOWEX HCR S/S cationite (72-75%), activated carbon/ charcoal/ (50-65%), and expanded clay/ keramzite/ (47-50%) have the highest adsorption property, and gravel sand (41%). and granular anthracite (33%) is the weakest.

The regularities of adsorption of nuclear material from aqueous solutions on adsorbent masses of isotopes can be used as an original cleaning method to adapt the physical purification stage of oil or other radiochemical processes to the safety norms of wastewater.

It was determined as a result of the conducted research that depending on the amount of adsorbent used, the deactivation coefficient of the process varies according to the types of adsorbent in the following order: DOWEX HCR S/S cationite - activated charcoal /carbon/-expanded clay /keramzite/- pebble gravelly sand - granular anthracite.

The process of absorption 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 absorption of a substance or substances from a solution or gas by the surface of a liquid is called absorption, and on the contrary, the transfer of a substance or substances from the surface of phase separation to the volume is called desorption. Physical and chemical adsorption types are distinguished depending on the form of interaction between adsorbate and adsorbent particles.

The physical adsorption process is characterized by weak intermolecular Van der Waals forces between adsorbent and adsorbate particles. 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 is observed.

It is possible to determine the activation energy from the graph constructed for adsorption or desorption in $\ln W_o = f(1/T)$ coordinate $/tg\alpha = -Ea/R = [(\ln W_o)_1 - (\ln Wo)_2]/(1/T_1 - 1/T_2) / system.$

By determining the desorption of 238 U isotope atoms adsorbed to the adsorbent (100 g) from the uranyl nitrate solution at a temperature of 298 K, proportional to the increase in temperature (238 U activity in the adsorbent at each temperature), a graphical interpretation of the dependence was made for the mentioned adsorbents (see: Figure 3). The experimental values of the desorption rate in Table 6 were used to draw up the graphical dependencies in Figure 3. In the "activation energy" column of Table 6, the values of the activation energy of the desorption process of 238 U isotopes from the adsorbents used in the research, calculated on the basis of the graphical dependences in Figure 3, are shown.

Table 6

Adsorption of ²³⁸U isotopes from uranyl nitrate solutions on studied 100 g adsorbents at different temperatures.

Type of adsorbent	Adsorption on adsorbent at room temperature partial desorption of the ²³⁸ U isotope proportional to the temperature increase, Bq/100 g.				The value of the activation energy of desorption of the ²³⁸ U isotope	
	298 K	323 K 343 K		$\frac{110111}{E} \frac{ausorberns}{mal}$		
	A ₂₉₈	D ₃₂₃	ln(D ₃₂₃ /A ₂₉₈)	D ₃₄₃	ln(D ₃₄₃ /A ₂₉₈)	E_a (KJ/mol).
DOWEX HCR S/S cationite	64	18	-1.269	32	-0.693	26.5
Activated charcoal	45	13	-1.242	22	-0.716	24.2
Keramzite	41	12	-1.229	20	-0.718	23.5
Pebble gravelly sand	37	11	-1.213	18	-0.721	22.6
Anthracite	30	9	-1.204	14	-0.762	20.3



Fig. 3. Desorption of adsorbed from uranyl nitrate solutions to adsorbents $(100 g)^{238}U$ isotopes are proportional to temperature.

$$\begin{split} -E_a/R &= [\ln(D_{343}/A_{298}) - \ln(D_{343}/A_{298})]/[(1000/T_1) - (1000/T_2)] = \\ & [\ln(18/64) - \ln(32/64)]/[(1000/323) - (1000/343)] = \\ -1.269 - (-0.693)/(3.096 - 2.915) = -0.576/0.181 = -3.18; \\ & -E_a/R = -3.18; E_a = 3.18 \times 8.3144 = 26.5 \text{ kJ/mol.} \end{split}$$

It shows that the value of a quantity (E_a/R) is 2.44-3.19 for all adsorbents used in the experiments and the value of activation energy (E_a) varies in the range of 20.3-26.5 kC/mol, after performing calculations with the same algorithm for other adsorbents (see: Table 6).

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 6, ²³⁸U isotope adsorbed on DOWEX HCR S/S cationite at room temperature (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 desorption process is in the range of 8-40 kC/mol, indicates that the adsorption process in all 5 types of adsorbents used in the experiments, including DOWEX HCR S/S cationite, have a physical nature (physical sorption), and it is not accompanied by chemical sorption, indicates that only weak Van der Waals forces exist between the adsorbed uranium isotopes and the adsorbent.

It can be seen from the obtained regularities that when using the same type of adsorbent, the reduction of uranyl nitrate from the solution occurs inversely proportional to the increase in the amount of adsorbent.

4. Conclusions

The regularities of adsorption of radioisotopes from aqueous solutions of nuclear material waste in granular activated carbon /charcoal/, anthracite, pebble gravel sand, DOWEX HCR S/S cationite, expanded clay masses have been studied. The samples of granular DOWEX HCR S/S

cationite (72-75%), activated carbon /charcoal/ (50-65%), and expanded clay /keramzite/ (47-50%) have the highest adsorption property among the mentioned adsorbents, and the weakest adsorption property has pebble gravel sand (41%) and granular anthracite (33%).

The values of the deactivation coefficient (1.05 - 3.75) of the cleaning processes carried out with the application of the studied adsorbents are smaller than the deactivation coefficients of the existing cleaning methods (DC > 4) shown in the scientific literature. However, expanded clay /keramzite/, which has large natural raw material reserves in our country, can be used as a cleaning component for adaptation of the physical purification stage of oil to the safety standards of wastewater.

It was determined that the activation energy (E_a) of the desorption process of uranium isotopes from the studied adsorbents varies in the range of 20.3-26.5 kJ/mol. These values indicate that the desorption process is a weak endothermic process, in contrast to the exothermic adsorption process. The values of the activation energy determined for the desorption process show that the adsorption process has a physical nature (physical sorption) in all 5 types of adsorbents used in the experiments, and only weak Van der Waals forces exist between the absorbed uranium isotopes and the adsorbents.

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ИЗУЧЕНИЕ ЗАКОНОМЕРНОСТЕЙ АДСОРБЦИИ РАДИОНУКЛИДОВ НА АДСОРБЕНТАХ

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Резюме: Изучены закономерности адсорбции радиоизотопов из водных растворов отходов ядерных материалов гранулированным активированным углем, антрацитом, гравийным песком,

катионитом DOWEX HCR S/S, керамзитовыми массами. Среди указанных адсорбентов наибольшей адсорбционной способностью обладают образцы гранулированного катионита DOWEX HCR S/S (72-75%), активированного угля (50-65%) и керамзита (47-50%), а наименьшей адсорбционной способностью гравийный песок (41%) и гранулированный антрацит (33%). Значения коэффициента дезактивации (1,05 - 3,75) процессов очистки, проводимых с применением исследованных адсорбентов, меньше приведенных в научной литературе коэффициентов дезактивации существующих методов очистки (DE > 4). Однако, керамзитовые массы, имеющие большие запасы в качестве природного сырья в нашей стране, может быть использован для очистки сточных вод стадии физической очистки нефти до норм безопасности. Установлено, что энергия активации (Еа) процесса десорбции изотопов урана из исследуемых адсорбентов колеблется в пределах 20,3-26,5 кКл/моль. Эти значения указывают на то, что процесс десорбции является слабым эндотермическим процессом, в отличие от экзотермического процесса адсорбции. Значения энергии активации, определенные для процесса десорбции, показывают, что процесс адсорбции имеет физическую природу (физическая сорбция) во всех 5 типах адсорбентов, используемых в экспериментах, и между поглощаемыми изотопами урана и адсорбента существуют лишь слабые Ван-дер-Ваальсовые силы.

Ключевые слова: адсорбенты, отходы и растворы ядерных материалов, изотопы, десорбция.

ADSORBENTLƏRDƏ RADİONÜKLİDLƏRİN ADSORBSİYASI QANUNAUYĞUNLUQLARININ ÖYRƏNİLMƏSİ

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Xülasə: Nüvə materialı tullantılarının sulu məhlullarından radioizotopların dənəvər aktivləşdirilmiş kömürdə, antrasitdə, çınqıllı qumda, DOWEX HCR S/S kationitində, keramzit kütlələrində adsorbsiyası qanunauyğunluqları öyrənilmişdir. Qeyd edilmiş adsorbentlərdən ən yüksək adsorbsiya xassəsini dənəvər DOWEX HCR S/S kationit (72-75%), aktivləşdirilmiş kömür (50-65%) və keramzit (47-50%) nümunələri, ən zəif adsorbsiya xassəsini isə çınqıllı qum (41%) və dənəvər antrasit (33%) nümunələri göstərmişdir. Tədqiq edilmiş adsorbentlərin tətbiqi ilə aparılmış təmizləmə proseslərinin dezaktivasiya əmsalının (1.05 - 3.75) qiymətlərinin elmi ədəbiyyatda göstərilmiş mövcud təmizləmə üsullarının dezaktivasiya əmsallarından (D ∂ > 4) kiçik olmasına baxmayaraq ölkəmizdə təbii xammal ehtivatları böyük olan keramzitdən istifadə etməklə neftin fiziki təmizlənməsi mərhələsinin tullantı sularının təhlükəsizlik normalarına uyğunlaşdırılması üçün təmizləmə üsulu qismində istifadə edilə bilər. Tədqiq edilmiş adsorbentlərdən temperatur yüksəlməsi ilə baş verən uran izotoplarının desorbsiya prosesinin aktivləşmə enerjisinin (E_a) qiymətinin 20.3-26.5 kC/mol intervalında dəyişdiyi təyin edilmişdir. Bu qiymətlər ekzotermik adsorbsiya prosesinin əksinə olaraq desorbsiya prosesinin zəif endotermik proses olduğunu göstərir. Desorbsiya prosesi ücün təyin edilmiş aktivləşmə enerjisinin qiymətləri təcrübələrdə istifadə edilən bütün 5 növ adsorbentlərdə adsorbsiya prosesinin fiziki xarakter daşıdığını (fiziki sorbsiya), udulan uran izotopları ilə adsorbent arasında yalnız zəif Van-der-Vaals qüvvələrinin mövcud olduğunu göstərir.

Açar sözlər: adsorbentlər, nüvə materialı tullantıları və məhlulları, izotoplar, desorbsiya.