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METHOD FOR CLEANING SOIL CONTAMINATED WITH NUCLEAR WASTE

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Abstract. İt was found that the amount of nuclear material in the soil decreases inversely with an increase in the amount or concentration of the reagent, as well as an increase in the amount of water that washes the soil when using a reagent (HNO_3, HCl or NaOH) or a mixture (HNO_3-HCl) for the extraction of soil contaminated with nuclear waste. The proposed new method of decontamination is 5.8 times more profitable than the existing most effective method of decontamination (prototype), and this method requires only 2.47 billion AZN for complete (1200-2000 times) decontamination of 1000 hectares of land contaminated with radioactive substances (nitrate uranyl). The results obtained indicate the possibility of complete soil cleanup by applying the proposed new method of decontamination in cases of contamination of adjacent territories by nuclear accidents.

İt is not considered appropriate to throw the waste of radiochemistry production and research containing radioactive isotopes directly into the sewage line, it is absolutely necessary to adapt the composition of such waste to the requirements of relevant sanitary norms and rules. The mass of solid industrial waste containing radionuclides must be collected in separate containers.

Separated radioactive substances should be handed over to the "Isotope" special plant of the Ministry of Emergency Situations, which was built for the storage of nuclear materials and radioactive substances.

The development of methods for the separation of radioisotopes from soil samples and aqueous solutions obtained by soil extraction is one of the topical topics of radiochemical research, and it is appropriate to study such methods to ensure radiation safety. The methods of cleaning soil contaminated with uranium isotopes using chemical reagents have been developed for these purposes. The existing methods are characterized by relatively low efficiency. Therefore, systematic research is needed to develop a new, more effective deactivation method [1-6].

METHODICAL PART

"Chemically pure for analysis" reagents (Merck kGaA /Germany/, VWR PROLABO /France/, Lachema /Czech Republic/, AO "Base of Chemreactives №1" /RF/) were used during the experiments. Solutions of 67% nitric acid with a density of 1.4 g/cm^3 at 20ºC, crystalline granular sodium hydroxide and 38% hydrochloric acid with a density of 1.19 g/cm^3 at 20ºC were prepared in bidistilled water obtained from the "GFL-2304" bidistillator for the analysis,. Samples of uranyl nitrate UO2(NO3)2 salt, widely used in nuclear reactors, were used as nuclear material in the experiments. Which isotopes are present in the uranyl nitrate salt and in what concentration was determined by gamma spectrometry with a HPGe detector [7-10].

The soil samples taken during the radio monitoring carried out in the regions of the country were divided into equal parts weighing 200 grams. These soil samples were mixed with uranyl nitrate solution in order to obtain model samples contaminated with nuclear material waste. The activity (activity concentration) of ^{238}U, ^{235}U, ^{234,236}U isotopes in the uranyl nitrate salt solution soaked in soil samples contaminated with uranyl nitrate was determined to be in the ratio of 1200:400:20.

Nitric acid, hydrochloric acid, sodium hydroxide solutions and distilled water were used to separate radioisotopes from these soil samples. Analytical chemical and spectroscopic analyzes of the

dry mineral obtained by mixing and evaporating the extracts were carried out after extracting each sample with the listed solutions in a row [7-10].

İt is expected that they react with solutions of nitric and hydrochloric acids of various concentrations, dissolve in these acid solutions or their mixtures, like other non-radioactive metals, since radioactive substances, including the elements that make up the basis of nuclear materials, mainly have metallic properties. 0.2 M, 0.5 M, 1.0 M and 2.0 M concentrated solutions of nitric acid were used, as well as 0.2 M, 0.5 M, 1.0 M and 2.0 M concentrated sodium hydroxide in order to restore the neutral reaction property of the soil sample residues extracted at the last stage of the experiments. aqueous solutions were used for these reason, in our experiments, for the extraction of radionuclides. We also used aqueous solutions of nitric and hydrochloric acids in a ratio of 1:2 for more effective extraction of radionuclides,.

DISCUSSION OF OBTAINED RESULTS

The radioactive substance dissolved in the reaction with chemical reagent solutions did not completely pass into the solution during filtration after extraction, and part of it remained in the residual soil, after extraction with the reagent, taking into account, the soil residue was washed with 1 liter of distilled water, and gamma-spectroscopic analyzes of the extract with the soil residue were performed [7-10] .

The regularities of the 2-stage deactivation processes of samples of soil (200 g) contaminated with uranyl nitrate as a model sample with a mixture of nitric and hydrochloric acid solutions are presented in the following pictures (figure 1, /1st stage/, figure 2 /2nd stage/).

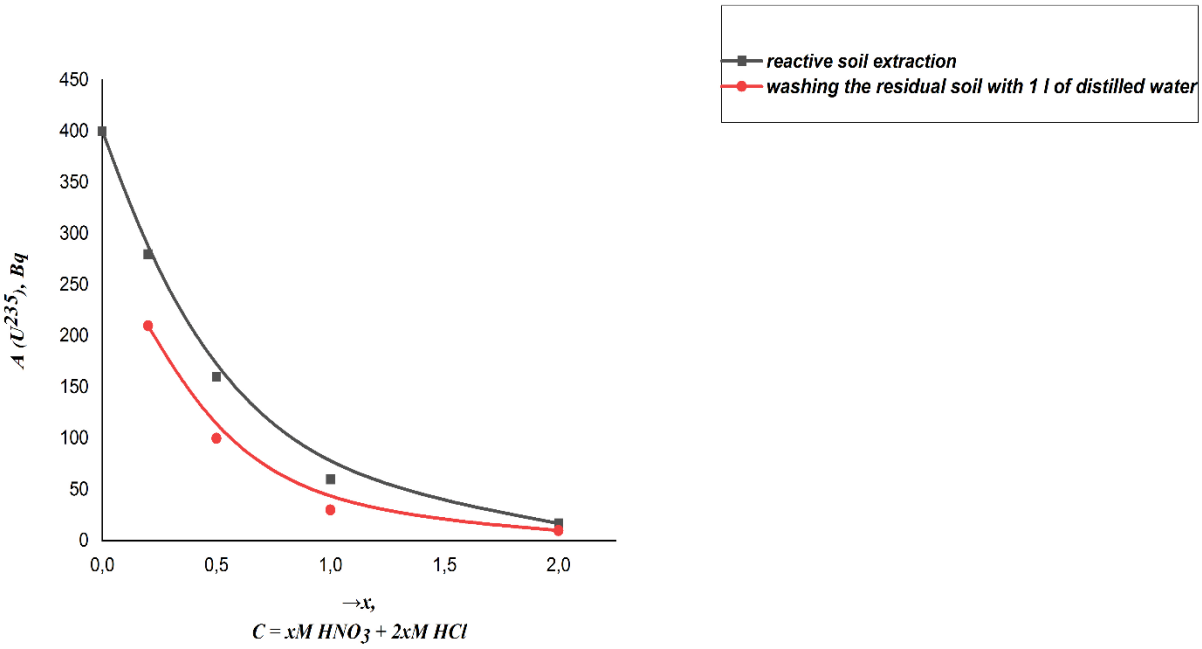

Figure 1. Reduction of ^{235}U isotope in the soil as a result of leaching of samples of soil (200 g) contaminated with uranyl nitrate by extraction with nitric and chloride acid mixture solutions and then with 1 l. distilled water (stage 1).

It is possible to reduce the amount of uranium isotopes in those soils up to 2000 times with the 2-stage deactivation processes of soil samples with the activity of ^{238}U, ^{235}U, $^{234,236}U$ isotopes in the

ratio of 1200:400:20 by extraction with a mixture of nitric and hydrochloric acid solutions and then with 1 liter of distilled water.

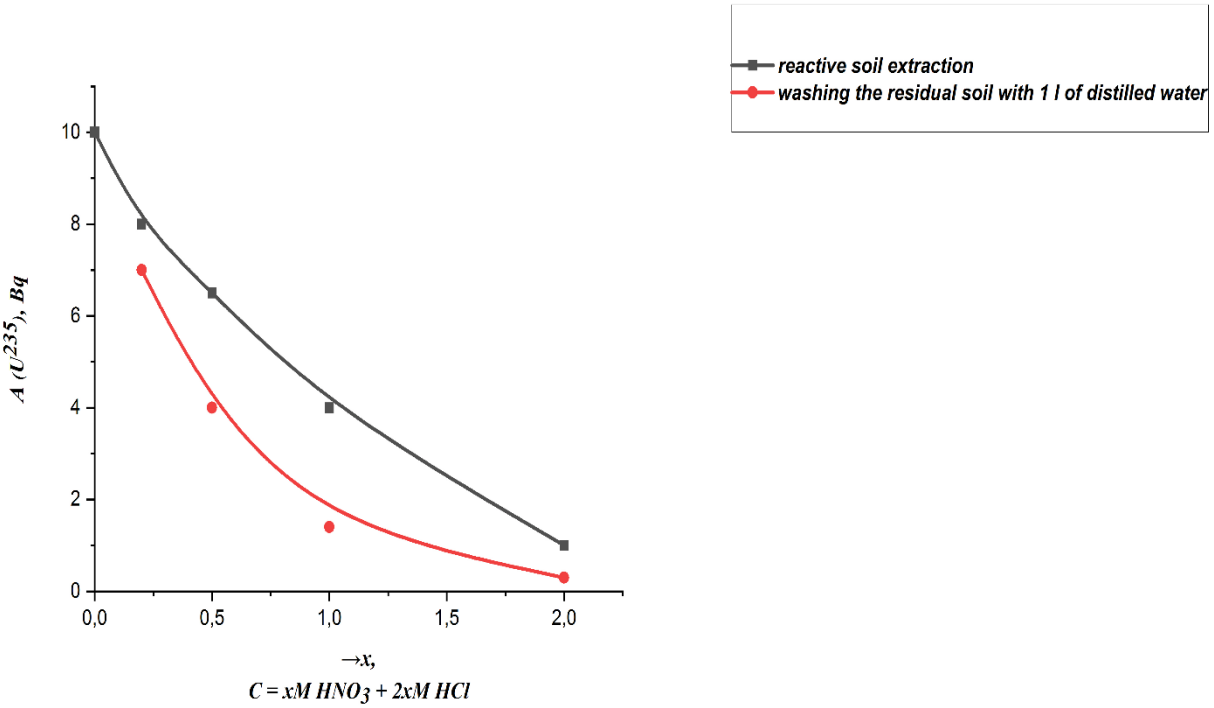

Figure 2. Reduction of ^{235}U isotope in the soil as a result of leaching of samples of soil (200 g) contaminated with uranyl nitrate by extraction with a mixture of nitric and hydrochloric acid solutions and then with 1 l. distilled water (stage 2).

It can be seen from the obtained laws, that when individual reagents (HNO3, HCl or NaOH) or their mixtures (HNO3-HCl) are used for the extraction of contaminated soils, the amount or concentration of the reagent increases, as well as inversely proportional to the increase in the amount of water with which the soil is washed, there is a decrease in the amount of radionuclides in the soil [7-10].

The 2-stage deactivation process of soil contaminated with uranyl nitrate waste with a mixture of nitric and hydrochloric acid solutions and then with distilled water (or with an aqueous solution of sodium alkali) is a more effective cleaning method compared to deactivation with aqueous solutions of separate reagents, as well as compared to all existing prototypes is assigned.

It is possible to carry out complete deactivation (DC = 1200-2000) of soil contaminated with nuclear material waste with this method (see table 1).

A technical and economic justification of the application of the new deactivation method was based on the market average price of the necessary reagents for deactivation and on the price of work to be done. The required financial expenditure for the deactivation of the contaminated upper layer of the soil area is determined by the following equation:

$$S = \tau + E + R + \Delta s \quad (1)$$

where S – is spent on all the work to be done for deactivation and funding necessary to ensure radiation safety the total amount of funds (currency);

τ – currency of working hours spent on deactivation equivalent;

E - deactivation, earth excavation and transportation works currency equivalent of consumed energy;
R – the currency of the reagents used for deactivation value with equivalent;
Δs – radioactive substance released from the soil as a result of deactivation or core of soil fraction enriched with radionuclides storage in waste storage facilities ("cemetery") cost in currency equivalent.

Table 1

Results of deactivation of 200 gram soil samples contaminated with uranyl nitrate (specific activity of ^{238}U, ^{235}U and $^{234,236}U$ radioisotopes 1200, 400 and 20 Bq, respectively) with a mixture of nitric and hydrochloric acids solution

n, mol ($nHNO_3$+2nHCl in 1 l. water)	1st stage				2nd stage				Other way for 2nd stage			
	The soil sample is extracted with nHNO3+2nHCl solution for 1 hour and the soil residue is washed with 1 liter of distilled water for 1 hour				The soil residue is extracted with nHNO3+ +2nHCl solution for 1 hour and the final soil residue is washed with 1 liter of distilled water for 1 hour				The soil residue is extracted with nHNO3+ +2nHCl solution for 1 hour and the final soil residue is extracted with nNaOH (M) solution for 1 hour			
	A1 ^{238}U, Bq	A1 ^{235}U, Bq	A1 $^{234,236}U$ Bq	DC_1	A2 ^{238}U,Bq	A2 ^{235}U, Bq	A2 $^{234,236}U$ Bq	DC_2	A2′ ^{238}U, Bq	A2′ ^{235}U, Bq	A2′ $^{234,236}U$ Bq	$DC_{2'}$
0.2	650			1.85	21			57	17			70
		210		1.9		7		57		5		80
			12	1.7			0.3	70			0.2	100
0.5	370			3.3	14			85	11			110
		100		4.0		4		100		3		133
			7	2.9			0.2	100			0.1	200
1.0	130			9.2	6			200	4			300
		30		13		1.4		333		1		400
			3	6.7			0.05	400			0.03	667
2.0	30			40	1.1			1100	1			1200
		10		40		0.3		1330		0.3		1330
			1	20			0.01	2000			0.01	2000

Note: Ai - Cs in the remains of the soil sample after the development stages specific activity of isotopes;
DC_i – the deactivation coefficient of the soil sample after the processing stages (from the development stage of the special activity of the initial soil sample the ratio of the subsequent soil residue to the specific activity);
d_i – water obtained after the stage of gravity separation of pomegranate-disperse of the original soil sample after separation of the radioactive soil fraction decrease in mass (percentage of the original soil amount).

Only 2.47 billion AZN financial expenditure is required for complete (1200-2000 times) deactivation of 1000 ha of soil contaminated with nuclear material waste with this new deactivation method. The developed new deactivation method is 5.8 times more efficient and cost-effective than the existing most effective deactivation method (prototype). The obtained results indicate the possibility of complete cleaning of the surrounding areas with the use of a new deactivation method in cases where the surrounding areas are contaminated with nuclear material waste as a result of accidents that may occur in nuclear reactors. Considering the contamination of the layer mainly in the upper 5 cm residue of the soil in the surrounding areas as a result of the rains after the nuclear reactor accidents, the newly developed deactivation process will require less financial expenditure.

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RADİOAKTİV TULLANTILARLA ÇİRKLƏNMİŞ TORPAĞIN TƏMİZLƏNMƏSİ ÜSULU

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Xülasə. Nüvə materialı tullantıları ilə çirklənmiş torpaqların ekstraksiyası üçün reaktivdən (HNO_3, HCl və ya NaOH) və ya qarışıqdan (HNO_3-HCl) istifadə etdikdə reaktivin miqdarının və ya qatılığının artmasına, həmçinin sonra həmin torpağın yuyulduğu suyun miqdarının artmasına tərs mütənasib olaraq torpaqda nüvə materialı tullantılarının miqdarının azaldığı müəyyənləşdirilmişdir. Təklif edilmiş yeni dezaktivasiya üsulu mövcud ən effektiv dezaktivasiya üsulundan (prototip) 5.8 dəfə rentabellidir və bu üsulla nüvə materialı tullantıları ilə (uranil nitratla) çirklənmiş 1000 hektar torpaqların tam (1200-2000 dəfə) dezaktivasiyasının aparılması üçün yalnız 2,47 milyard AZN maliyyə sərfiyyatı tələb olunur. Alınmış nəticələr nüvə qəzaları nəticəsində ətraf ərazilərin çirklənməsi hallarında həmin ərazilərin təklif edilmiş yeni dezaktivasiya üsulunun tətbiqi ilə tam təmizlənməsi mümkünlüyünü göstərir.

Açar sözlər: *nüvə materialı tullantıları, izotop, çirklənmiş torpaqlar, dezaktivasiyası.*

СПОСОБ ОЧИСТКИ ПОЧВЫ, ЗАГРЯЗНЕННОЙ ОТХОДАМИ ЯДЕРНОГО ТОПЛИВА

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Резюме. При использовании реагента (HNO_3, HCl или NaOH) или смеси (HNO_3-HCl) для экстракции почвы, загрязненной ядерным топливом, установлено, что количество отходов ядерного топлива в почве уменьшается обратно пропорционально увеличению количества или концентрации реагента, а также к увеличению количества воды, которой промывается почва. Предлагаемый новый метод дезактивации в 5,8 раза выгоднее существующего наиболее эффективного метода дезактивации (прототип), и при этом методе требуется всего 2,47 млрд. манатов для полной (в 1200-2000 раз) дезактивации 1000 га земель, загрязненных радиоактивными веществами (нитратом уранила) Полученные результаты свидетельствуют о возможности полной очистки почвы путем применения предложенного нового метода дезактивации в случаях загрязнения прилегающих территорий ядерных аварий.

Ключевые слова: *ядерные отходы, изотоп, загрязненная почва, дезактивация.*