UDC: 544.54

METHOD FOR CLEANING SOIL CONTAMINATED WITH RADIOACTIVE WASTE

Kh.N. Shiraliyeva

Institute of Radiation Problems of Ministry of Science and Education, Azerbaijan
xagani06@mail.ru

Abstract: It was found that the amount of radionuclides 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 radionuclides. 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 manats for complete (1200-2000 times) decontamination of 1000 hectares of land contaminated with radioactive substances (nitrate uranyl). The obtained results 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. The decontamination of the soil of the adjacent territories of the nuclear reactor by the developed method will require relatively lower financial costs, taking into account the contamination of mainly the upper 5 cm soil layer even after rains.

Keywords: nuclear waste, contaminated soil, decontamination, isotopes.

1. Introduction

It is not considered appropriate to throw the waste of radiochemistry production and research containing radioactive isotopes directly into the sewage line, it is 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.

The separated radioactive substances are to be transferred to the special “Isotope” facility of MFA built for the storage of nuclear material and radioactive substances.

The development of methods for separating radioisotopes from soil samples and aqueous solutions obtained by soil extraction in order to ensure radiation safety is one of the topical topics of radiochemical research, and it is expedient to study such methods. To this end, methods have been developed for the purification of soil contaminated with uranium isotopes using chemical reagents. However, systematic research is needed to develop a new, more effective method of decontamination, because the existing methods are characterized by relatively low efficiency [1-6].

2. Methodological part

The reagents used in the experiments were “chemically pure for analysis” (Merck kGaA /Germany/, VWR PROLABO /France/, Lachema /Czech Republic/, JSC “Baza №1 Khimreaktivov" /Russia/). For the analyses, 0.2M, 0.5M, 1.0M, and 2.0M solutions of 67% nitric acid and crystalline granulated sodium hydroxide with a density of 1.4 g/cm^3 at 20°C and 0.4 M, 1.0 M, 2.0 M and 4.0 M solutions of 38% hydrochloric acid with a density 1.19 g/m^3 at 20°C were prepared in bidistilled water obtained from bidistilator GFL-2304. Uranyl nitrate salt $UO_2(NO_3)_2$, widely used in nuclear reactors, was used in experiments as nuclear material. Using gamma spectrometry with an HPGe detector, it was determined which isotopes are present in the uranyl nitrate salt and in what concentration [7-10].

Soil samples collected during the radio-monitoring conducted in the regions of the country were divided into equal portions weighing 200 grams. These soil samples were mixed with a uranyl nitrate solution to obtain model samples contaminated with nuclear waste. The activity (active concentration) of isotopes ^{238}U, ^{235}U, and $^{234,236}U$ in a uranyl nitrate salt solution impregnated into soil samples contaminated with uranyl nitrate was determined to be in a 1200:400:20 ratio.

Nitric acid, hydrochloric acid, sodium hydroxide solutions, and distilled water were used to separate radioisotopes from these soil samples. Analytical chemical and spectroscopic analyses of dry minerals obtained by mixing and evaporating the extracts were performed after extracting each sample with the listed solutions in succession [7-10].

Since radioactive substances, including the elements that form the basis of nuclear materials, mostly have metallic properties, they are assumed to react with solutions of nitric and hydrochloric acids of various concentrations, to dissolve in these acid solutions or their mixtures, just like other non-radioactive metals. In our experiments, it was used solutions of 0.2 M, 0.5 M, 1.0 M, and 2.0 M concentration of nitric acid for radionuclide extraction and also aqueous solutions of 0.2 M, 0.5 M, 1.0 M, and 2.0 M concentration of sodium hydroxide for restoring the neutral reaction property of the soil sample residues extracted at the last stage of the experiments. For more effective radionuclide extraction, aqueous solutions of nitric and hydrochloric acids in a 1:2 ratio were also used.

3. Discussion of obtained results

Considering that the radioactive substance dissolved in the reaction with chemical reagent solutions did not completely pass into the solution during filtration after extraction, and some of it remained in the residual soil after extraction with reagent the soil residue was washed with 1 liter of distilled water and gamma spectroscopic analyses of the extract with the soil residue were carried out [7-10].

The regularities of the two-step decontamination processes of soil samples (200 g) contaminated with uranyl nitrate as a model sample by a mixture of nitrogen and hydrochloric acid solutions are presented in the following figures (figure 1, / step 1/, figure 2 / step 2/).

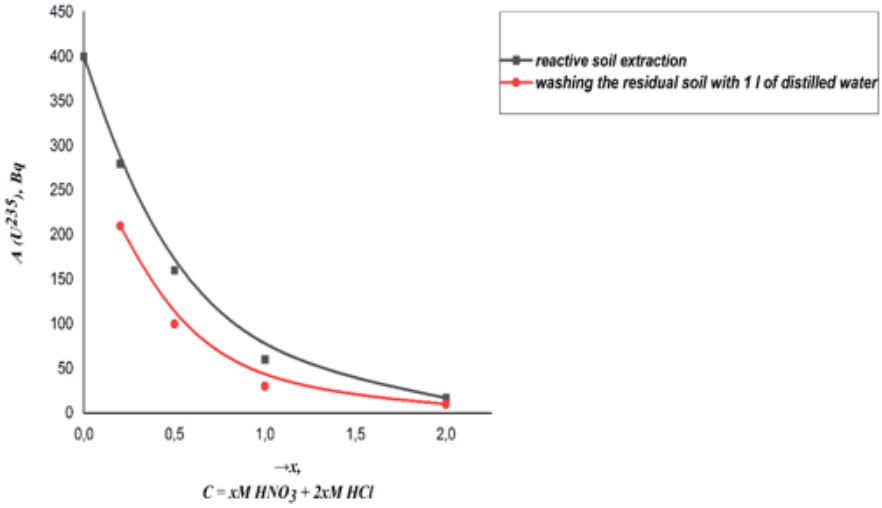

Fig. 1. Extraction of soil samples (200 g) contaminated with uranyl nitrate by a mixture of nitrogen and hydrochloric acid solutions, and reduction of ^{235}U isotope in soil due to washing with 1 liter distilled water (step 1).

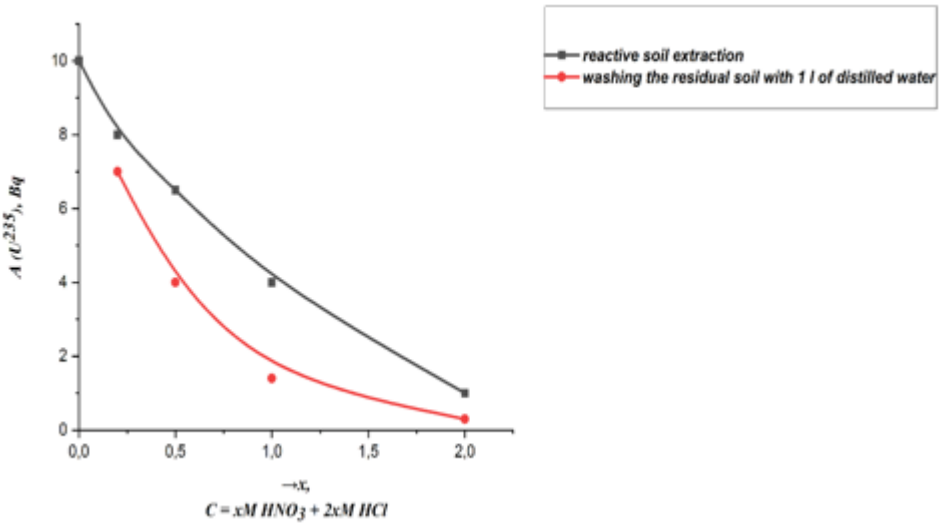

Fig. 2. Extraction of soil samples (200 g) contaminated with uranyl nitrate by a mixture of nitric and hydrochloric acid solutions, and reduction of ^{235}U isotope in soil due to washing with 1 liter distilled water (step 2).

It is possible to decrease the number of uranium isotopes in the soils up to 2000 times by two-step decontamination of soil samples containing isotopes ^{238}U, ^{235}U, $^{234,\text{ and }236}U$ with the activity in the ratio of 1200:400:20 by a mixture of nitrogen and hydrochloric acid and then by 1 liter of distilled water.

Table 1

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

n, mol ($nHNO_3$ +2n HCl in 1 liter of water)	Step 1				Step 2				Another way for Step 2			
	The soil sample is extracted with $nHNO_3$+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 $nHNO_3$ + 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 $nHNO_3$+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	DF_1	A2 ^{238}U, Bq	A2 ^{235}U, Bq	A2 $^{234,\ 236}U$, Bq	DF_2	A2′ ^{238}U, Bq	A2′ ^{235}U, Bq	A2′ $^{234,236}U$, Bq	$DF_{2'}$
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: A_i – the specific activity of Cs isotopes in the residues of the soil sample after the processing stages;

DF_i – the deactivation factor of the soil sample after the processing stages (the ratio of the specific activity of the initial soil sample to the specific activity of the soil residue after the processing stage);

d_i – reduction of the mass of the initial soil sample (in percent of the initial soil amount) after separation of the fine-dispersed radioactive soil fraction obtained after the stage of water gravity separation.

From the obtained regularities it is seen that the amount of radionuclides 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 radionuclides. The fact that two-step decontamination process of soil contaminated with uranyl nitrate wastes by a mixture of nitrogen and hydrochloric acid solutions and then with distilled water (or aqueous solution of sodium alkali) is a more effective method of treatment in comparison with decontamination with water solutions of separate reagents and also in comparison with all existing prototypes. With this method, complete decontamination (DF = 1200-2000) of soil contaminated with nuclear waste can be carried out (see Table 1).

Then a feasibility study for the new decontamination method was carried out based on the average market cost of the necessary decontamination reagents and the amount of work to be done. The necessary financial costs for decontamination of the contaminated top layer of the land plot were determined according to the following equation:

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

Where S is the total amount of funds (currency) spent on all deactivation works and necessary to ensure radiation safety;

τ – currency equivalent of working hours spent on deactivation;

E – currency equivalent of the energy spent on deactivation, soil excavation and transportation works;

R – a value of reactants used for deactivation in currency equivalent;

Δs – the value in currency equivalent of the cost of storage of the radioactive a substance released from the soil as a result of deactivation or the soil fraction enriched with radionuclides in nuclear waste storage (“cemeteries”).

The 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 manats for complete (1200-2000 times) decontamination of 1000 hectares of land contaminated with radioactive substances. The obtained results indicate the possibility of complete cleaning of adjacent territories with the application of a new method of decontamination in cases of contamination of surrounding territories with nuclear wastes as a result of accidents that may occur at nuclear reactors. The decontamination of the soil of the adjacent territories of the nuclear reactor by the developed method will require relatively lower financial costs, taking into account the contamination of mainly the upper 5 cm soil layer even after rains.

References

1. Wikipedia. Dezaktivatsiya, 2022, pp.1-2.
2. Wikipedia. Spisok radiatsionnykh avariy, February 24, 2022, pp. 1-5.
3. Y.G. Zhu. Soil contamination with radionuclides and potential remediation / Y.G. Zhu, G. Shaw //Chemosphere, 2000, v.41, pp.121-128.
4. S.A. Dmitriyev, Method for cleaning sandy soils from radionuclides / Patent RU 2388084 C1. 27.04.2010, Bull. No. 12 / Barinov A.S., Kuptsov V.M.
5. S.A. Dmitriyev. The method of reagent decontamination of soils from cesium radionuclides. Patent RU 2399975 C1. -20.09.2010, Bull. No. 26 / Barinov A.S., Kuptsov V.M.
6. S.A. Dmitriyev. The method of reagent cleaning of soils from cesium radionuclides /Patent RU 2361301 C1. 10.07.2009, Bull. No. 19 / Prozorov L., Kuptsov V.M.
7. V.M. Kondakov, A.S. Kozyrev, A.A. Zubkov, E.N.Semenov, E.B. Vashchenko. A method for monitoring the safety of radioactive waste storage sites. Patent RU 2260215 C2. pp.1-9 / A.S. Kozyrev, A.A. Zubkov, E.N. Semenov E.B. Vashchenko
8. Kh.F. Mammadov, H.N. Shiraliyeva, R.G. Allahverdiyev, G.R. Garibov. Research of radiation background and distribution of radioactive elements in soil areas of Azerbaijan // Scientific Achievements of Modern Society. XII International Scientific and Practical Conference, Liverpool, July 22-24, 2020, pp. 34-40.
9. Kh.F. Mammadov, Study of purification processes of soil contaminated with uranyl nitrate / Kh.F.Mammadov, H.N.Shiraliyeva, E.I.Mehtiyev [et al.] / Problems of Atomic Science and Technology, PAST, 2021, v.132, No.3, pp. 132-135.
10. K.F. Mammadov. Studying the treatment processes of soil samples from radionuclides / K.F. Mammadov, H.N. Shiraliyeva, E.I. Mehtiyev [et al.] // Journal of Science Rise, Estonia, – Tallinn: 2021, No.3 (74), pp.29-33.

СПОСОБ ОЧИСТКИ ПОЧВЫ, ЗАГРЯЗНЕННОЙ РАДИОАКТИВНЫМИ ОТХОДАМИ

Х.Н. Ширалиева

Резюме: При использовании реагента (HNO_3, HCl или NaOH) или смеси (HNO_3-HCl) для экстракции почвы, загрязненной радионуклидами, установлено, что количество радионуклидов 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. Учитывая загрязнение преимущественно верхнего 5-см слоя почвы даже после дождей, то дезактивация почвы прилегающих территорий ядерного реактора разработанным способом потребуют сравнительно меньшие финансовые затраты.

Ключевые слова: ядерные отходы, загрязненная почва, дезактивация, изотопы.

RADİOAKTİV TULLANTILARLA ÇİRKLƏNMİŞ TORPAĞIN TƏMİZLƏNMƏSİ ÜSULU

H.N. Şirəliyeva

Xülasə: Radionüklidlərlə 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 radionüklidlərin miqdarının azaldığı müəyyənləşdirilmişdir. Təklif edilmiş yeni dezaktivasiya üsulu mövcud ən effektiv dezaktivasiya üsulundan (prototip) 5.8 dəfə rentbellidir 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. Nüvə reaktorlarında baş vermiş qəzalardan sonra yağışlar nəticəsində ətraf ərazilərdəki torpaqların əsasən üst 5 sm qalınlığında layının çirklənməsi nəzərə alındıqda, işlənib hazırlanmış yeni dezaktivasiya prosesinə daha az maliyyə sərfiyyatı tələb ediləcəkdir.

Açar sözlər: nüvə materialı tullantıları, çirklənmiş torpaqlar, dezaktivasiyası, izotoplar.