Methods of Determination of Radiation Factors and Radioecological Conditions in Navoi Industrial Region

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Abstract: Radiation factors-exposure dose rate of gamma-radiation, specific effective activity (Aeff), specific activity of radionuclides, equivalent equilibrium activity of radon (EEA), activity of long-lived radionuclides (LLR), radon flux density (RFL), coefficients of radioactive equilibrium etc., and methods for definition of their significance in Navoi industrial region are actual tasks of radioecology. Values of radiation factors are regulated by normative international documents and documents of the Republic of Uzbekistan. The established norms of radiation safety are monitored in industrial and settlements as well as in the adjacent areas of the so-called observation points [1,7].

Exposure dose values are estimated by means of radiation analysis of soil, air, plants, drinking, waste and underground water in observation points of adjacent territories to sources of environmental contamination [2-6]. On the basis of the performed researches it is possible to determine -238U, 232Th, 226Ra, 40K concentrations in soil, water and technological products as well as to find the forms of elements in liquid phase using nuclear filters and to determine the fractional composition of uranium-containing solid fractions in gas emissions using nuclear filters. Based on the value of the enrichment factor of chemical elements in the soil, we can find a pattern of localization of man-made chemical elements in some areas of the studied areas of uranium production. On the basis of the studies conducted to determine the migration of chemical elements in the groundwater we can predict the possible values of groundwater contamination in the area of uranium and gold mining plants.The main component of the values of environmental exposure doses are radioactive pollutants and their compound in the soil, air, plants, drinking, waste and groundwater [8-18].

Keywords: specific total activity, radioactive equilibrium, radioactive decay chain of uranium, technological processes, liquid and solid samples, pumping wells, productive and "tailing" solutions, areas of underground leaching - (AUL) uranium, saturated and regenerated tar, bottom sediments, silt of technological maps-ponds.

1. Introduction

The purpose and objectives of the research are:

- Development of a methodology for radiometric studies, determination of gamma exposure dose rate (EDR) of technogenic objects;
- the development of methods of radiometric determination of radioisotope concentrations and their geochemical behavior in underground water samples;
- summarizing the results of studies to reveal the main factor in the mechanism of dissolution of radioisotopes;
- development of measures to reduce the radiation factor of technogenic facilities of mining and metallurgical enterprises on the environment.

To achieve the goal the following works were carried out:

- values of gamma-radiation exposure dose rate (EDR) in technogenic objects were determined and their spatial distribution was detailed;
- concentrations of various radioisotopes in water samples were determined;
- the geochemical behavior of radioisotopes during the joint storage of different types of mining and metallurgical wastes was studied;
- conclusions were made on the development of measures and reduction of the radiation factor of technogenic objects of mining and metallurgical enterprises on the environment.

Given the relevance of the above, this study presents the value of the radiation factors of uranium production and methods for their determination in the area of NSMC.

Technique and methods of physical experiment. Dosimeter DKS-96 with detector unit BDPG-96 was used for measurement of gamma-radiation exposure dose rate (EDR) of technogenic objects. The received results (more than 20 thousand instrumental measurements) on spatial distribution of EDR are compared with normative values.

Measurements of alpha-nuclide contamination of surfaces of industrial equipment, surfaces of premises, special machines and special uniforms of working personnel by DCS-96 devices.

Concentration of radioisotopes in water samples was determined by instrumental method on the device "Camera", which allows to perform express measurements faster than the traditional emanation method. The determined value of the concentration of radioisotopes allows us to assume the possibility of geochemical behavior of radioisotopes in the joint storage of different waste composition.

Obtained results and their discussion: All conducted research in the uranium production units were compared with the results obtained in other laboratories (including those accredited by the Agency "Uzstandart" for technical competence), which conduct continuous work on environmental control and with the results given in the regulatory international and Republican documents.

The following ingredients are systematically monitored in these studies:

Analyses of various soils and rocks with sampling of soil and rocks at a depth of 0-0.5 m. determined the equivalent dose rate of gamma-radiation (EDR), then in laboratory conditions measured the total specific activity of alpha-radiationAeff-soils and plants, the specific activity of natural radionuclides K40, Ra226, uranium (natural), Th232 gamma-spectrometric method.

Obtained spectra are shown in Fig. 1- 4.

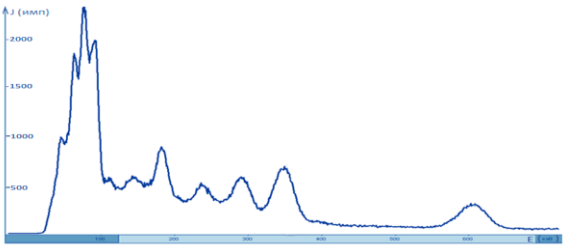

Fig.1. Spectrum of gamma-spectrometric analysis of selected rocks from uranium dumps

Fig. 2 Spectrum of gamma-spectrometric analysis of selected rocks from uranium cores

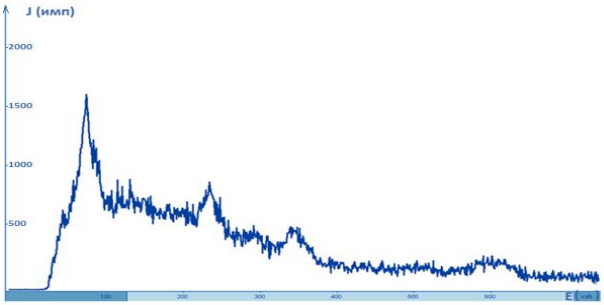

Figure 3. Spectrum of gamma-spectrometric analysis of selected soils from observation points

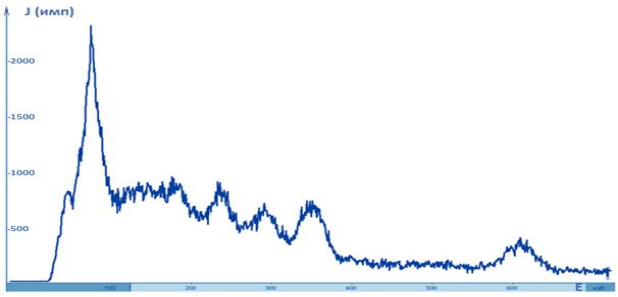

Fig. 4. Spectrum of gamma-spectrometric analysis of selected soils of settlements

From the spectra in Fig. 1-4 we can see that the activity of the samples among themselves is sharply different, that is, the sampled sample the more close to the radiation source in it the more concentrations of radionuclides and correspondingly more in their specific activity.

Correlation relation of chemical elements in soils is established on the basis of statistical methods of analysis. It is intermediate between the exact dependence given by the functional relationship and perfect independence of the variables. Thus, the dependence is found not between the values themselves, but between each of them and the corresponding mathematical expectation of the other. The correlation coefficient characterizes the degree of linear relationship between two or more variables.

To determine the correlation coefficient between chemical elements, a calculation algorithm and a special program were developed using a database management system (DBMS). The correlation coefficient between 30 elements in the samples taken in the area of the uranium object and plotted the dependence of the change of Kenr - in various uranium objects Fig. 8.

The following results were obtained:

- K = 0.7 - 0.9 between: gold - arsenic, arsenic - antimony, gold - antimony, between rare earth elements;
- K = 0.5 - 0.7 between: rubidium - cesium, mercury - silver;
- K = 0.4 - 0.5 between: gold - silver, gold - selenium, chromium - tungsten, zinc - nickel;
- K < 0,4 between: gold - tungsten, gold - iron, gold - sulfur.

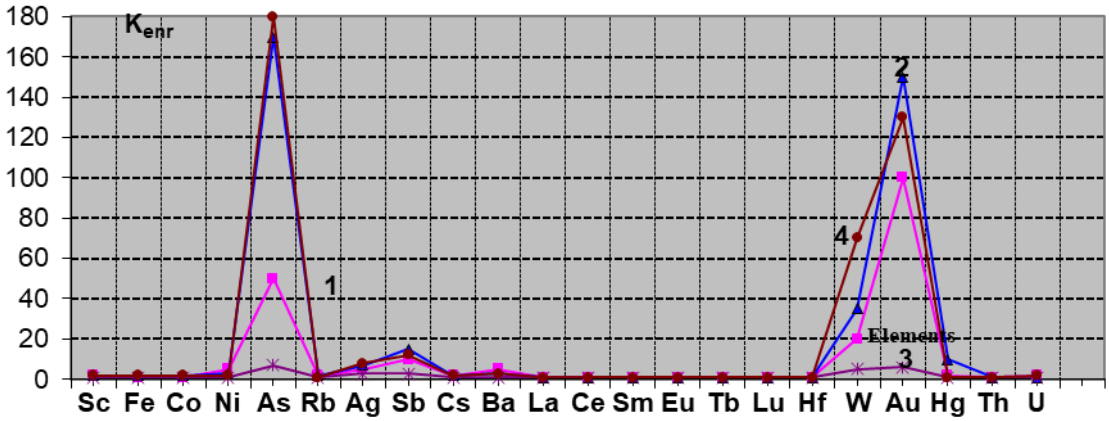

Fig. 5. Variation of Kenr - in various uranium objects

The value of the correlation coefficient between the elements in the samples taken from the studied areas differs from each other.

$$K_t = \frac{C_x * K_r}{C_r * K_x} \qquad (1)$$

The value of the enrichment factor was determined by the formula:

where Cx- content of investigated element, Cr- content of reference element, Kx- Clarke of investigated element, Kr - Clarke of reference element (scandium). Fig. 2 shows dependence (average value) of enrichment factor of some elements in soils, sampled in the area of first and second stage tailings dump, open pit and adjacent areas. The enrichment factor of 0.5 - 2.0 is observed for rubidium, barium, lanthanum, cerium, samarium, europium, terbium, lutetium, hafnium, mercury, thorium and uranium in the area of tailings 1,2, the open pit, and nearby areas (Tamdy-Bulak and Rakhmatkuduk).

The above listed elements are referred to "reference" elements, which means their natural origin.

Atmospheric air and working zone air analyses: determined radon volumetric activity, exposure or effective dose rates (EDR) in workplaces and equipment by radiometers with DCS-96 dosimeters, radon volumetric activity (VAR) in atmospheric air of settlements and in workplace air by ALPHA-GUARD instrument, equivalent equilibrium volumetric activity of radon daughter products of decay (EEVA) in atmospheric air of settlements, in the air of working area and premises by "Search" device, long-living alpha-nuclides (LAN) in the atmospheric air of settlements, in the air of working area and in premises, with sampling on aspiration filters, gamma-radiation power in continuous mode to control aerosol emission into the atmosphere.

Analyses of potable, industrial wastewater and groundwater: U, Ra, Po, Th concentrations were determined by a special method, Rn - by the emanation method, and total specific alpha and beta activity on the UMF-2000 device.

Based on the results obtained, the annual technogenic effective dose for the personnel and for the population is calculated. In addition to the above measurements, an important role is played by systematic determination of the magnitude of radioecological factors associated with the activities of uranium mining enterprises and significantly affecting the radioecological stability and purity of the environment, namely:

- the value of radioactive emissions into the atmosphere during the calcination of uranium oxide oxide; off-balance uranium-containing ore in dumps, at uranium PV sites as well as spent waste in tailing ponds; the probability of groundwater pollution by man-made compounds during uranium PV and rational options for reclamation of spent uranium PV sites.

Power of equivalent dose - (EDR) of gamma-radiation in the adjacent territories of technogenic objects is on average - 0,17-0,25 μSv/hour, with the value of natural equivalent dose rate - 0,11 - 0,15 μSv/hour. Linear dependences of DER-gamma-radiation from the point of measurement in the territories of technogenic object are shown in Fig. 8.

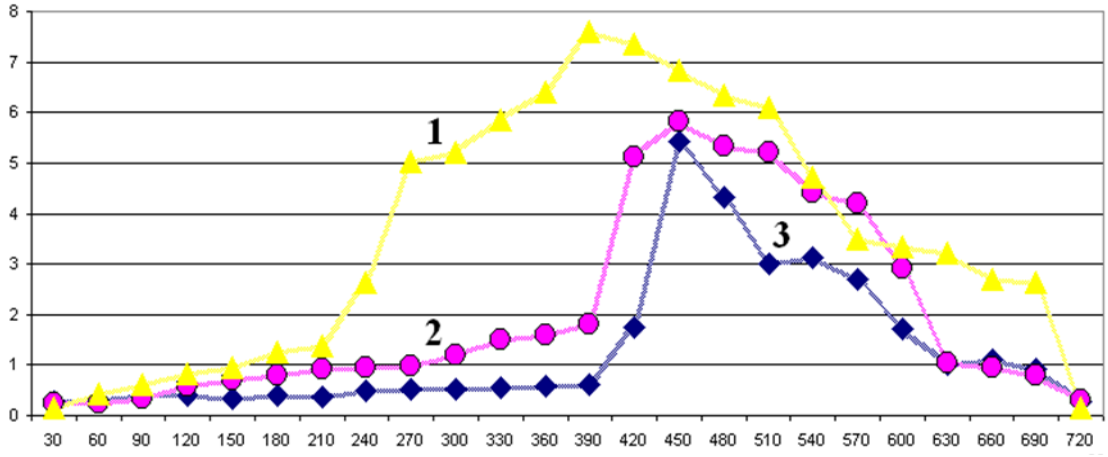

Fig. 6. Linear dependences of gamma-radiation DER from the point of measurement

Values of background and gamma-radiation DER in the junction points along the perimeter of technogenic object on average do not exceed 0.25 μSv/h.

As can be seen from Fig. 1, spatial distribution of gamma-radiation DER values in man-made objects is nonlinear. As seen from curves 1, 2, 3 of Fig. 1, values of gamma-radiation EDR vary from 0.30 μSv/hour to 7.80 μSv/hour, from 0.40 μSv/hour to 5.90 μSv/hour and from 0.28 μSv/hour to 5.30 μSv/hour respectively. This indicates that wastes of different composition have different radioisotope composition and different layer thickness.

In the investigated technogenic object the average value of gamma-radiation EDR of Fig. 1 is an order of magnitude lower than the established norm, that is, from 10 to 40 times lower. The obtained results show that all the requirements of the normative documents on the disposal of industrial wastes are fulfilled in full at this object.

Solubility coefficient of some radioisotopes in neutral media is high in comparison with sulfuric acid medium, therefore, dissolution of some radioisotopes in the liquid phase of waste of different composition, directed to obtain a thick layer, reducing the radiation impact on the environment, is possible.

When a thick layer of waste containing radioisotopes is covered, the probability of infiltration of the technogenic solution into the underground aquifer appears, as well as the probability of dissolution of radioisotope compounds from the previously placed mining and metallurgical waste and their transfer into groundwater. In order to study the probability of infiltration of these solutions, a study was conducted to determine two radioisotopes in selected samples from observation wells drilled along the perimeter of the technogenic object. Obtained results are given in Table 1.

Table 1

Results of research on the determination of two radioisotopes in selected samples from observation wells

Sample number	P_1, Bq/l	Ratio of activity P_1 to MPC	P_2, Bq/l	Ratio of activity P_1 to MPC
1	20.1	0.44	0.061	0.03
2	3.82	0.08	0.027	0.014
3	7.56	0.17	0.032	0.016
4	0.14	0.003	0.006	0.003
5	0.36	0.008	0.003	0.001
6	0.22	0.005	0.019	0.009
7	2.26	0.05	0.046	0.023
8	0.26	0.006	0.006	0.003
9	10.1	0.22	0.029	0.014
10	1.64	0.04	0.041	0.020
11	16.6	0.40	0.039	0.020

12	2.36	0.05	0.015	0.007
13	0.34	0.008	0.007	0.003
14	0.73	0.02	0.011	0.005
15	0.10	0.002	0.032	0.016
16	0.11	0.002	0.033	0.016

From the obtained experimental data Table 1 shows that the average concentration of radioisotopes P1 and P2 in waters sampled quarterly in 2018-2020 from observation wells drilled along the perimeter of the technogenic object does not exceed the regulated values of radioisotopes in water P1 (45 Bq/L) and P1 (2.0 Bq/L). From the results it is clear that the environmental protection work in this technogenic object is organized in accordance with the established requirements and fully complies with the above-mentioned norms established in the Republic of Uzbekistan.

Currently, research on the systematization of previously obtained data (for 25 years), for a more reliable clarification of the mechanism of geochemical behavior of radioisotopes in technogenic objects placed waste of different composition and solubility of radioisotopes under the influence of different compositions of liquid phases is continuing. Identification of the main factor of radioisotope solubility in aqueous samples makes it possible to promptly address the issues of assessing the magnitude of the impact of a technogenic facility on the environment.

Ways to reduce the radiation impact of uranium production on the environment During uranium mining there are three types of radioactive hazardous objects (factors affecting the environment): dumps formed during processing of uranium-bearing ores by mining methods; areas of underground uranium leaching at uranium mines; uranium waste tailings.

As our long-term (over 15 years) radiation-dosimetric surveys prove, all the waste rock and off-balance dumping sites have no radiological hazard, because most of the radionuclides they contain have dissolved and migrated to the lower layer of the heap. During acidic in-situ leaching, the vast majority of radium (99.3-99.6%) does not dissolve and remains with the solid ore residue in the Earth's interior during the in-situ leaching process. Somewhat more radium is dissolved during carbonate leaching (up to 1.5-2.2%).In this case, the concentration - 226Ra and 230Th in the discharge solutions reaches the following values: 226Ra- 35*10-6μCu/ml; 230Th-22*10-6μCu/ml, which is 10 times higher than the maximum allowable standards for discharge into open water bodies (for both isotopes). Since uranium is leached at uranium in-situ leaching sites in both acid and carbonate and hypochlorite regimes, the probability of radium getting into the uranium productive solutions, uranium chemical concentrates, tail waters and into the tailings is high.

Recommendations have been developed for reclamation of spent uranium in-situ leaching areas (in-situ leach pits) based on radioactive sludge leaching with water to decontaminate radium, and injection into the spent well.

Long-term storage of waste solid tailings is an important and complicated problem, since up to 70% of radioactive products contained in the ore are concentrated there (not counting uranium), mainly the long-lived 226Ra - an extremely dangerous potential source of radioactive contamination of the environment.

Methods to assess the degree of soil contamination with different radionuclides of uranium decay chain - U238, U234, U235, Ra226 - uranium in-situ leaching areas are controlled on the basis of the requirements of International and Republic normative documents. For this purpose the degree of soil contamination is assessed by various methods - physical-chemical, nuclear-chemical, radiochemical and radioecological methods. Most of the existing physical-chemical methods of soil remediation are based on the methods of soil purification from radionuclides and its further treatment with chemical reagents.

In order to achieve the goal the installation was created according to a new basic scheme and soil remediation of the uranium PS sites was carried out on this installation. Different radionuclides - 238U, 234U, 235U, 230Th, 226Ra and their behavior in different modes of uranium leaching were analyzed.

Among reclamation methods, the most acceptable is the physical-chemical method of decontamination (reclamation) involving a combination of the physical method and subsequent application of the chemical method with the use of various types of chemical reagents. Most of the existing physical-chemical methods of soil rehabilitation are based on methods of cleaning the soil from radionuclides and its further treatment with chemical reagents.

Recultivation process includes - bringing the disturbed lands to the initial state, revealing the local areas contaminated with radionuclides and chemical reagents, removal of these contaminated soils to the permitted landfills and industrial waste tailings, covering these areas with fertile soil.

The proposed new method of reclamation at low capital costs makes it possible to obtain in addition a certain amount of metal and to clean the contaminated soils from radionuclides at all (U238, U234, U235, Ra226, etc.).

X-ray spectral method of analysis and X-ray fluorescent analyzer APF-7 allowing quantitative determination of chemical elements in the range from Mn to U in solid and powder samples at 0.00015% (1.5 g/t) content were

used for analysis of selected solid samples. Samples weighing 1 kg were selected from soils contaminated with radionuclides and chemical reagents and prepared for analysis under laboratory conditions. For this purpose they were dried in the drying closets at 800C during 1 hour and rubbed on the laboratory tester of IDA-250 brand to a fraction of 0.074 mm.

Soil contaminated with radionuclides and chemical reagents is sampled from three parallel initial samples of 15 grams each, packed into a cuvette, placed into the measuring cell of the X-ray fluorescent analyzer ARF-7 and Uranium amount is defined in them.

The results obtained for the determination of the total uranium in the initial samples and in the samples after leaching demonstrate that the uranium concentration varies in the average range from 0.00293 % to 0.00513 %. After treatment, this limit varies from 0.00145 % to 0.00255 %. The amount of uranium in all treated samples decreases by a factor of two. Recultivated soil is considered to be cleaned from radionuclides and meets the norms established in SanPiN #0193-06.

The chemical composition of soil contaminated with radionuclides in the original samples and samples after treatment was studied by semi-quantitative X-ray fluorescence method. The results of semi-quantitative analysis of some chemical elements in the soil contaminated with radionuclides in the initial samples and samples after treatment.

From the obtained results it became clear that the concentration of most chemical elements such as - Al, Fe, Ti, Ca and S in the process of leaching approximately twice decreases in the spent samples than in the original samples. And the concentration of most chemical elements such as K, Mn, Sr etc. does not change during the leaching and their concentration is the same as in the initial samples. The concentration of SiO2 changes insignificantly.

Rational use of water resources. The expansion of processing volume of gold-containing ores and the acute deficit of technical water in Uzbekistan as an urgent task requires the search, solution and use of water-saving technology in hydrometallurgy of gold. One such opportunity is to evaluate the possibility of using ground and waste mineralized water in gold hydrometallurgy together with existing technical water.

The influence of salt composition of water on the quality of operations performed: grinding, thickening, gravitation, flotation, bio-oxidation of concentrates, sorption leaching of ores and concentrates was studied.

An acceptable - original way of using mineralized ground and waste water (with and without preliminary desalination) in a mixture with technical water from the process of gold extraction, taking into account the peculiarities of technological operations is proposed.

Assessment of the annual effective dose to the population of the region. The annual effective dose from all radiation factors for the population of the nearby towns was determined and the results showed that the established norms - 2.0 mSv/year in the Republic of Uzbekistan, according to SanPiN №0193-06 are fulfilled. In recent years, uranium mining enterprises have been monitoring the determination of individual annual dose of personnel on the ACIDC (automatic complex of individual dosimetric control) device in order to obtain reliable information, select the method of maximum reduction of the individual annual dose and timely rotation of the working personnel.

Presently the radiation situation of the personnel working at the PS uranium objects after the developed measures was improved and the annual technogenic effective dose to them is within 5-8 mSv/year under the prescribed maximum limit of 20 mSv/year in Table 2.

Table 2

Results of dosimetric survey in the points of observation of promo objects and settlements.

№ samples	**Number of sampling points, pieces**	**DER, μR/hour**		**LAN, mBq/m³**		**EEVA Bq/m³**		**Annual effective dose mSv/year**
		Value limit						
		min	**max**	**min**	**max**	**min**	**max**	
1	64 *	0.19	0.67	2.2	3.8	5	24	1.21-8.43
	20 **	0.15	0.19	1.8	5.3	1.4	2.8	0.43-0.84
2	68 *	0.18	0.64	2.1	2.8	2.1	18	2.10-7.17
	24 **	0.15	0.19	1.6	5.1	0.4	2.4	0.37-0.81
3	32 *	0.16	0.36	1.2	2.4	4.0	14	0.53-5.36
	16 **	0.15	0.19	1.1	1.5	1.0	1.8	0.24-0.53
4	58 *	0.18	0.63	3.2	4.4	4.0	23	1.43-6.34
	18 **	0.15	0.19	1.0	1.7	1.4	2.8	0.33-0.69

5	42 *	0.16	0.56	2.2	3.7	5.0	23	1.42-6.31
	28 **	0.16	0.19	1.7	1.9	2.0	2.8	0.21-0.58

Note: *-sampling points in promo objects, **-sampling points in settlements.

As can be seen from the data given in Tab. 2, the obtained annual effective doses for the personnel working at the radioactively contaminated plants do not exceed the established norm - 20 mSv/year and do not exceed the norm for the population - 2 mSv/year. On the basis of the carried out investigations and generalizations we can conclude that the technogenic influence of the radioactively contaminated objects does not go out of the territory of this object, the personnel and the environment are reliably protected from the harmful radiation influence.

2. Conclusion

From the conducted researches on determining the values of radiation factors of uranium production conducted over many years it may be concluded that the radiation situation in the region complies with the established standards, the effective annual dose to the personnel working at the uranium production facilities and the population of settlements in the adjacent areas does not exceed the values established in SanPiN-0193-06.

Based on the radiometric studies of technogenic objects, patterns of spatial distribution of gamma-radiation EDR and concentration of P1 and P1 radioisotopes in water samples were determined. From the results it can be seen that in both cases there is no excess of the indicators over the regulatory values.The main polluting area of uranium production are off-balance ores located in an extensive area, sites of underground uranium leaching and tailings for storage of spent uranium waste. To improve the radioecological state of the region it is recommended to process off-balance uranium ores using geotechnological methods (vat or heap leaching), to perform reclamation works and to cover the places of occurrence of spent uranium waste - gold ore processing wastes.Based on the studies performed the conclusion is made that the proposed new method of reclamation of soil contaminated with radionuclides is optimal. Recultivation of uranium uranium plots contaminated with radionuclides that were carried out according to the proposed method complies with all the established norms and requirements of the International and Republican norms.

On the basis of carried out researches of radiation factors it is possible in the long term to develop planned measures on systematic control of their content and behavior, and it is also possible to improve the radio-ecological condition of Navoi industrial region.

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