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DELIVERABLE IRA-Particle Behaviour-D4

Hot Particles containing NORM. Characterization of HP and their matrix.

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Executive Summary

The present work focuses on the study characteristics of residues of uranium ore processing localized in the tailings of Pridneprovsky Chemical Plant (Dneprodzerginsk, Ukraine). The main tasks for the reported period within the framework of Work Package 3, task 4 were:

- Assessment of the physical-chemical characteristics (pH, moisture content, dispersal composition) of sludge sampled in the PChP tailings.
- Assessment of presence the hot particles (small, discrete, high α-active particle or cluster) in sludge sampled of the PChP tailings by method of radiography.
- Assessment of the chemical composition of the wastes stored in the PChP tailings.
- Estimation of the total contents of the uranium series radionuclides in the waste samples.

The main results achieved during the reported period are presented below.

Journal article «Protsak V., Kashparov V., Prokopchuk N. High Alpha-Active Particles in Tailings of Pridneprovsky Chemical Plant» in press.

Introduction

Pridneprovsky Chemical Plant (PChP) was the first Soviet enterprise for processing uranium ore. It was built in 1947 in the suburb of Dniprodzerjisk in Ukraine. Up to 65 % of all uranium ore of the Soviet Union was processed at PChP. The plant was built for processing uranium ore from the Zhovtorichensky ore deposit and slags from the Dniprovsky metallurgical plant (blast furnace #6), which were formed due to melting of the uranium-containing iron ore of the Pervomaysky deposit. The fluids formed as a result of the technological operations were then recycled into the ammoniac and sodium nitrates. Since 1963 the uranium salts had been extracted from the uranium ore and concentrates delivered from the former USSR's republics, Eastern Europe (DDR, Hungary, Romania), France and Spain. During the operation period seven tailing dumps were created, namely Zahidne, Centralnyy Yar, Pivdenno-Shidne, Dniprovske, Sukhachivske (first and second sections), Lanthanum fraction, tailing dump at Sergiy Lazo street, and two dumps for the uranium processing waste, DP-6 and Base-S. During the exploitation period of the enterprise (1949-1991), up to 42 millions of tons of uranium ore processing products with a total activity of $2.7.10^{15}$ Bq have been accumulated in the tailing dumps, and up to 0.2 millions of tons of uranium processing wastes with a total activity of $4.4.10^{14}$ Bq have been accumulated in the dumps DP-6 and Base-S. At the tailing dumps territories the exposure dose rate varies in the range from 0.2 to 33 µGy/hr. Table 1 presents the brief information about the tailings where the studies were carried out.

Table 1. Brief information about the PChP tailings

Name	Area, ha	Mass of waste, ton	Total activity Bq	Period of operation
Zahidne	4	770 thousand	$1.81\cdot10^{14}$	1949-1954
Centralnyy Yar	2,4	220 thousand	$1.04\cdot10^{14}$	1950-1954
Pivdenno-Shidne	3,6	330 thousand		1956-1990
Dniprovs'ke	73	12 million	$1.4\cdot10^{15}$	1954-1968
Sukhachivs'ke (I section)	150	19 million		1968-1983
Sukhachivs'ke (II section)	70	150 thousand		Since 1983

Analysis of the literature sources shows the absence of data which would satisfactory characterize the physical-chemical parameters of the PCP's tailings' media and could be used for the comprehensive prognosis evaluation of the dumps' impacts on the ecological situation. The available information in this field was obtained mainly in the course of the works aimed in passportization of the tailings, and in some Governmental ecological programs and international projects.

Brief information about tailings where the samples were taken within the framework of Work Package 3 (task 4) is presented below.

Zahidne tailing. The tailing's area is approx 60,000 m^2. The operational period lasted from 1949 to 1954. The dump contains 0.77 millions of tons of radioactive waste (tails) with a total activity of $1.81 \cdot 10^{14}$ Bq. In 2005 at its surface, measures were taken to renew the coats and the drainage system and to reinforce the slopes. The γ-radiation dose rate at the tailing dump varies within the range of 0.1 - 9.8 μGy/hr. The tailing dump's territory is covered with a 1-m thick layer of loam and soil, and is partially asphalted. The dump's slopes are covered with crushed stone and fertile soil, which is fixed with grass. For this dump, a retaining wall and drainage channel for discharge of surface waters were created. However, the drainage system is not efficient enough, and its capacity is limited due to design defects and because of obstacles formed by the debris brought from the adjacent territories.

Centralnyy Yar tailing. The tailing's area is approx 24,000 m^2. The operational period lasted from 1950 to 1954. According to the tailing dump passports' data (Sanitary passports which were created in 1983-1989), the dump contains 0.22 millions of tons of radioactive waste (tails) from uranium processing (mainly of the radium fraction) with a total activity of $1.04 \cdot 10^{14}$ Bq. The tailing dump is located at the territory of a former ravine, which was divided into two parts by the dam. The tailing is almost completely covered with wild trees, shrubs and grasses. At the studied sites, the exposure γ-radiation dose rate varied within the range of 0.12 - 6.1 μGy/hr. Analytical studies of soil contamination showed textremely high levels of radium-226.

Dniprovske tailing. The operational period lasted from 1954 to 1968. According to literature data on the actual and potential environmental impacts, the tailing dump is one of the most dangerous radiation objects in this region (Korovin et al. 2004; Voitsekhovich et al. 2009). It is located in the direct vicinity of the PChP territory at the Konoplyanka river's bank, in the Dnieper river's floodland. The tailing was created by the hydraulic fill method, but filtration barriers were not formed either in the dam or in the basement of the bed. In total, it contains 5.6 millions of tons of uranium ore processing with residual contents of uranium up to 0.023%. In 1976 – 1980, the tailing dump was covered with a 0.5 – 13.5 m thick phosphate-gypsum layer (3.04 millions of m^3). The total activity of radioactive waste (RAW) in the tailing dump reaches $1.4 \cdot 10^{15}$ Bq. The radionuclide composition of RAW is determined by uranium and its decay products. The exposure γ-radiation dose rate in the RAW volume reaches 100 μGy/hr. A very dangerous fact is that the radioactive waste is saturated with water. The level of saturation of the pores with water reaches 99%. The main for the ground water table rising is the banking of the tailing dump's ground water flux by sludge storages and gold storages of coke-chemical and metallurgic manufactures. Besides, the ground water table rising was supported by the additional infiltration from the PChP's and Dniprovsky metallurgical complex's industrial sites, and the general rising of the water level in Dnieper (1958). Currently, these conditions remain actual and determine the high level of the ground water and flooding the territory. As a result of the water migration of the chemical substances and radionuclides, a plume of aquifer contamination has formed. It reaches the Konoplyanka and Dnieper rivers, which serve as ground water discharge.

Methods

1.1 Sampling

Samples of sludge were collected at various depths in the three tailings (Zahidne, Centralnyy Yar and Dniprovske) during the “dry” auger drilling of the boreholes (Fig. 1). These samples were packed into plastic bags and placed into a refrigerator where they were stored until the beginning of the analyses. After their delivery to the laboratory, the sample was taken from the central part of each bag for further analysis (Fig. 2).

a) Tailing “Dniprovske”

b) Tailing “Zahidne”

c) Tailing “Centralnyy Yar”

Fig. 1. Sampling on tailings of the Pridneprovsky Chemical Plant.

Fig. 2. Sludges of the Pridneprovsky Chemical Plant.

1.2 Analyses

Values of pH of the water extracts from the samples were determined according to ISO 10390-2001 at the solid : liquid ratio of 1:5.

The sample humidity was measured according to the standard method. The samples material was placed into the weighing bottles and covered with the caps. The total mass of the weighting bottle, cap and material, A, was measured with the precision of 0.001 g. Then the caps were removed, the samples were dried at 105 ^{0}C during 12 hr, covered with the caps again, cooled in an exsiccator and their masses, B, were measured again. Humidity (in %) was calculated as

$$humidity,\% = \frac{A-B}{B-m} \cdot 100 ,$$

where m is a total mass of the weighting bottle and the cap.

Dispersal composition of sludge was determined using the laser analyzer of the particle size Analysette 22 COMFORT (Fritsch GmbH, Germany) after ultrasonic dispersion of the samples.

The mass contents of chemical elements in the sludge samples were measured using the energy-dispersion roentgen-fluorescent analyzer EXPERTMobile. The analyzer provides the mass fraction measurements for elements from ^{12}Mg to ^{92}U in the range of 0.01 – 99.9 %.

Gamma-spectrometry of the samples was performed using the HpGe detector 7229P (Canberra) and multichannel analyzer. GammaVision-32 software was applied for the spectra treatment. For γ-spectrometry the samples were encapsulated and kept for three weeks in order to reach equilibrium between ^{226}Ra and ^{222}Rn. ^{235}U was identified using the peak of 143.8 keV, and the peak of 63.3 keV was used to determine ^{234}Th and ^{238}U in the equilibrium state. ^{214}Pb and ^{226}Ra were identified at 351.9 keV, and ^{212}Pb and ^{232}Th at 238.6 keV.

Alpha-spectrometric measurements were performed at EG&G ORTEC OCTETE PC with the silicon detectors of BU-017-450-100 ULTRA series.

Autoradiography and α-track radiographic studies were carried out with a goal of visualization of high α-active and β-active particle in samples. LR 115 film (Kodak) and X-ray film RP-U “Onico” (Ukraine) were used as detectors. Processing of the films was done according to the producer’s recommendations. For comparison of dispersed composition of high α-active particles (or clusters) in sludge with dispersed composition of chernobyl HP, both types of samples (sludge and bottom sediments of the cooling pond the Chernobyl Nuclear Power Plant (ChNPP)) were exposed in the same conditions. Chernobyl fuel particles (FPs) are α and β emitters and a method of evaluation of the dispersed composition has been developed for them. Therefore, if we expose different samples in the same conditions, we can do a rough estimate of size of α-emitting particles by comparing the size of the spots α-tracks (Fig. 3).

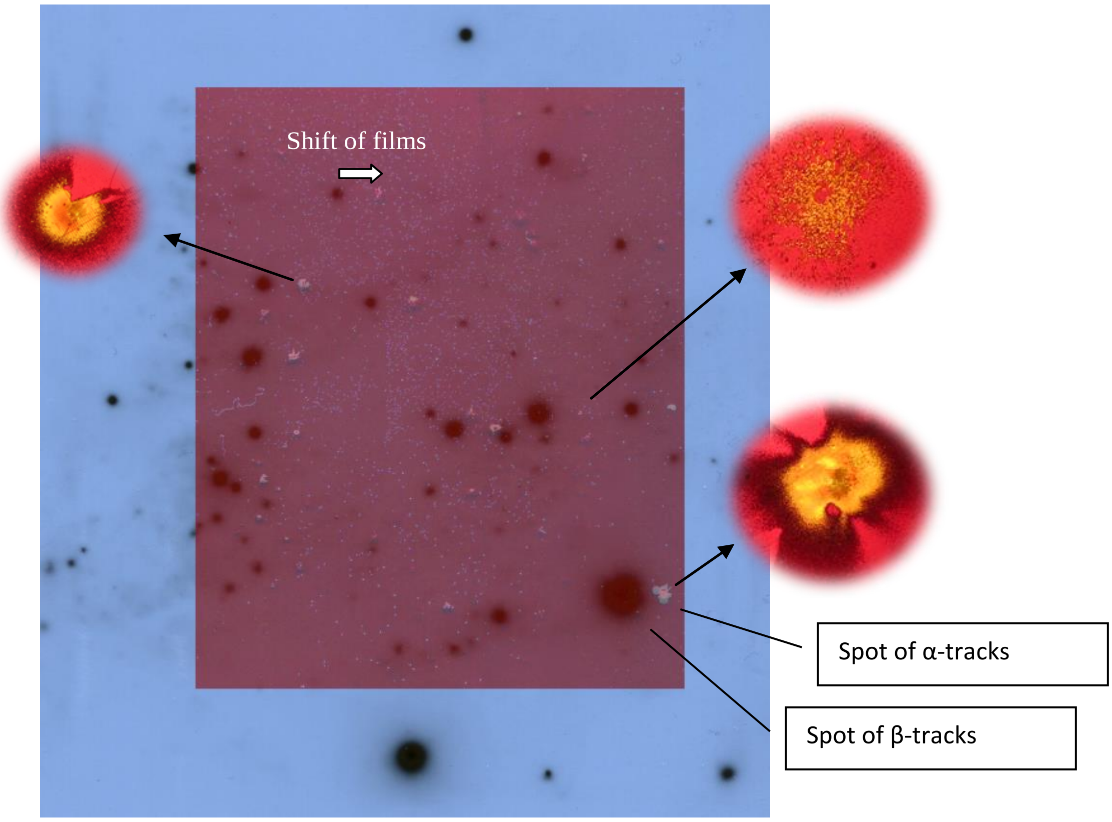

Fig. 3. X-ray film and film LR 115 after joint exposure on the sample of bottom sediments of cooling pond ChNPP.

Weighed and air-dried samples of the sludge and bottom sediments were thin layer distributed (extremely close to monolayer) on a hard surface (Fig. 4). The scheme of the exposure process is presented in Fig. 5.

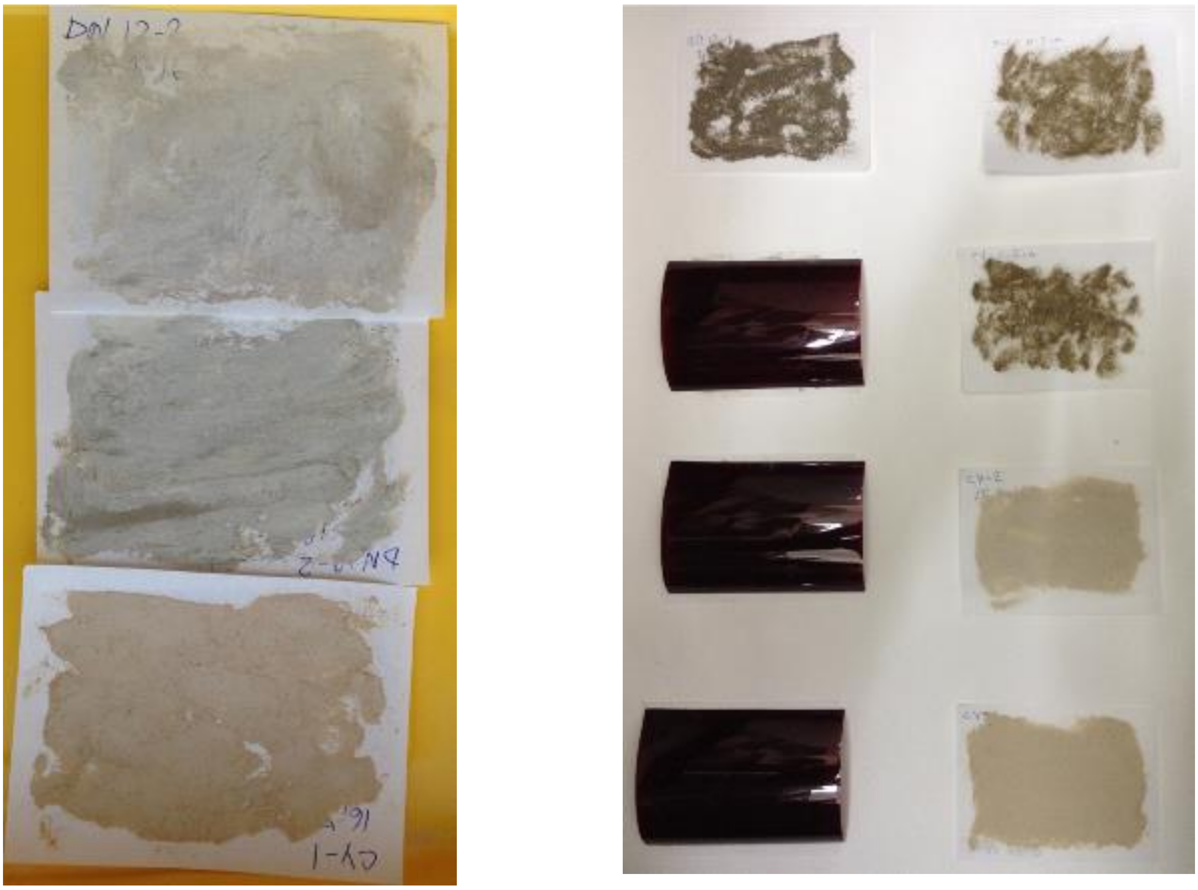

Fig. 4. Samples of sludge and bottom sediments for radiographic studies.

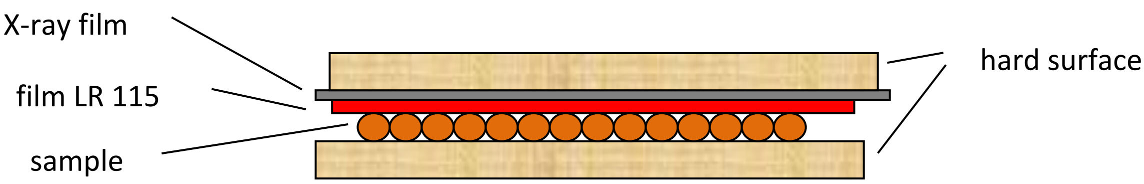

Fig. 5. Scheme of exposure samples.

Results

1.3 Actual acidity and humidity of the sludge samples

Results of the laboratory measurements of the actual acidity and humidity of the samples collected in various boreholes at various depths are presented in Table 2.

Table 2. Coordinates of the sludge sampling (# of borehole and depth from the soil surface) and values of the actual acidity (pH) and humidity (W, %).

Tailing dump								
Centralnyy Yar			Zahidne			Dniprovske		
CY 12-1			**ZP 12-1**			**DN 12-2**		
H, m	**pH**	**W, %**	**H, m**	**pH**	**W, %**	**H, m**	**pH**	**W, %**
2-2.5	7.8	12	1.0-1.5	9.2	21	13.8-14.3	7.4	156
3.5-4		55	3.0-3.5	8.8	23	15.7-16.2	7.3	67
6-6.5	4.1		4-4.5	9.5	25	17.7-18.2	6.8	68
9-9.5	4.4	65	7.0-7.5	9.8	22	18.2-18.4	6.7	26
13.5-14	4.1	67	8.5-9	9.8	26			
16-16.5	4.2	77	10-10.5	9.7	32			
17.5-18	4.3	28						

Obtained results show acid media in the Centralnyy Yar tailing (pH 3.6 - 4.4), alkaline media in the Zahidne tailing (pH 8.8 – 9.8) and close to the neutral media in the Dniprovske tailing (pH 6.7 – 7.4). Different acidity may determine the difference between the chemical forms and mobility of the chemical and radioactive contaminants stored in these dumps.

The sludge humidity in the tailings differs too. For the Zahidne tailing it makes 23-32 % while for the Centralnyy Yar tailing it is 55-77 %. The high saturation was observed at the horizon 13.8-14.3 m in the borehole DN 12-2 (Dniprovske) where the sludge humidity reached 156 %.

1.4 Dispersal composition of the sludge samples

Enlarged image of sludge particles of tailings of Pridneprovsky Chemical Plant is shown in Fig. 6.

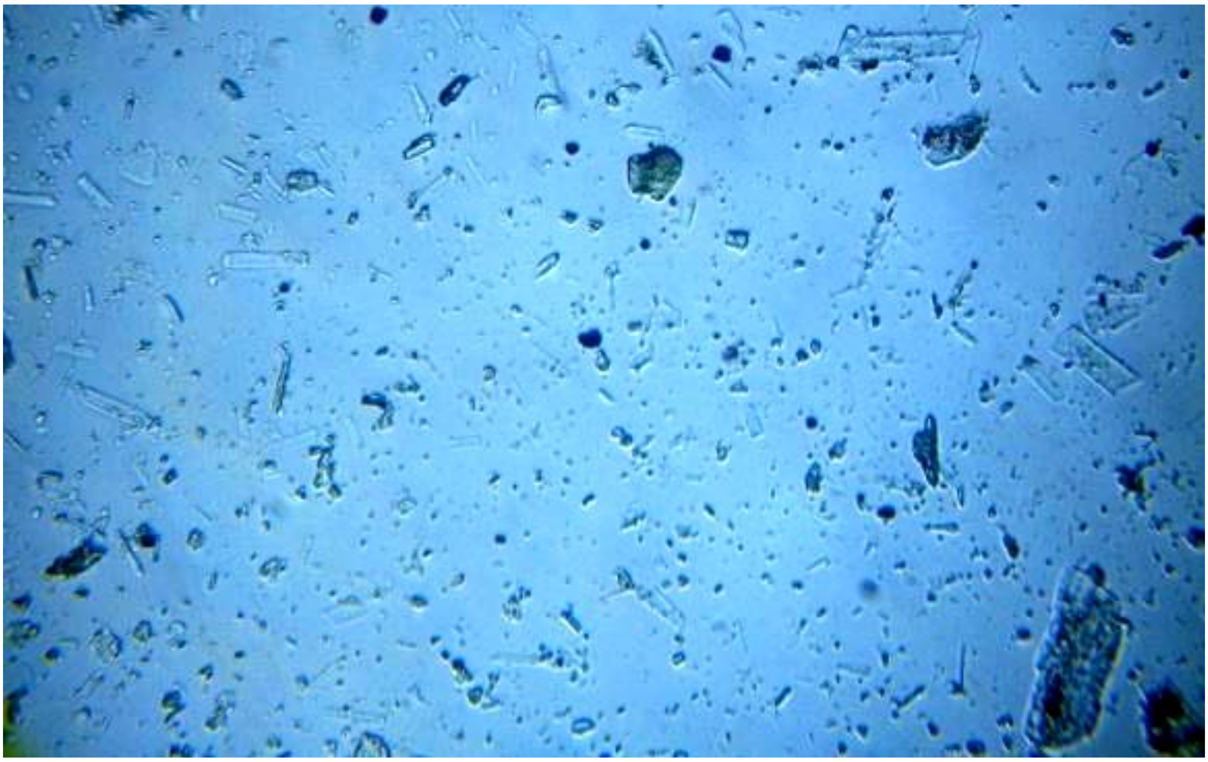

Fig. 6. Enlarged image of sludge particles PChP.

Obtained cumulative and differential frequency distributions of the mixture particle sizes from the PChP tailings are presented at Fig. 7 – 14.

Zahidne Tailing

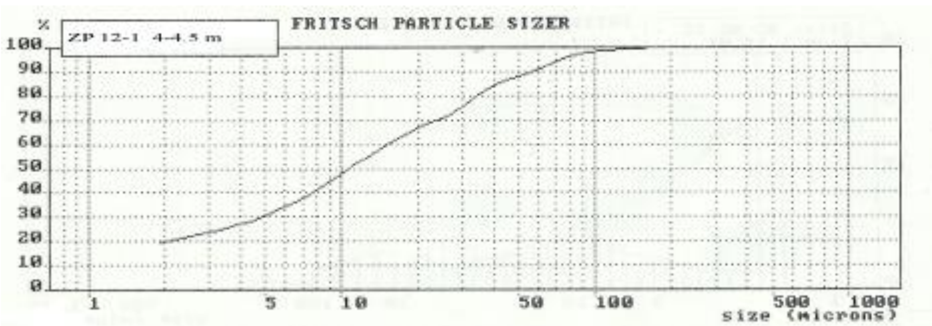

Fig. 7. Cumulative distribution of the sludge particle granulometric sizes (borehole ZP 12-1, horizon 4-4.5 m).

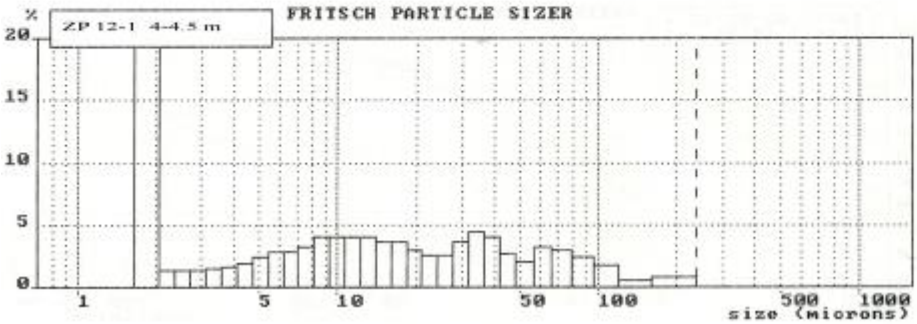

Fig. 8. Differential distribution of the sludge particle granulometric sizes (borehole ZP 12-1, horizon 4-4.5 m).

Centralnyy Yar Tailing

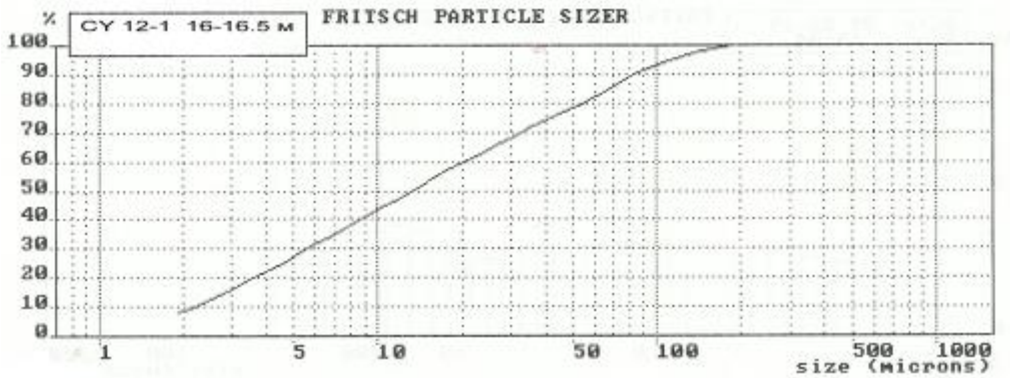

Fig. 9. Cumulative distribution of the sludge particle granulometric sizes (borehole CY 12-1, horizon 16-16.5 m).

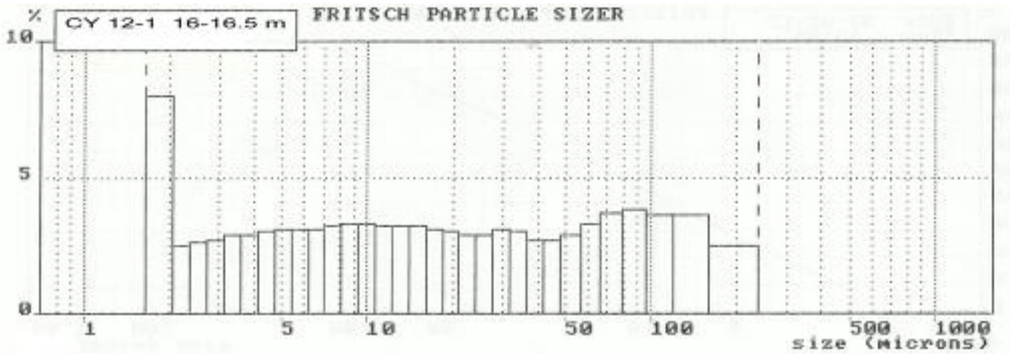

Fig. 10. Differential distribution of the sludge particle granulometric sizes (borehole CY 12-1, horizon 16-16.5 m).

Dniprovske Tailing

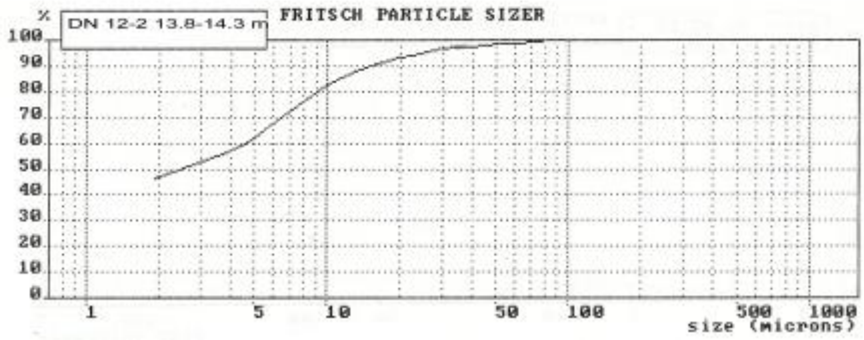

Fig. 11. Cumulative distribution of the sludge particle granulometric sizes (borehole DN 12-2, horizon 13.8-14.3 m).

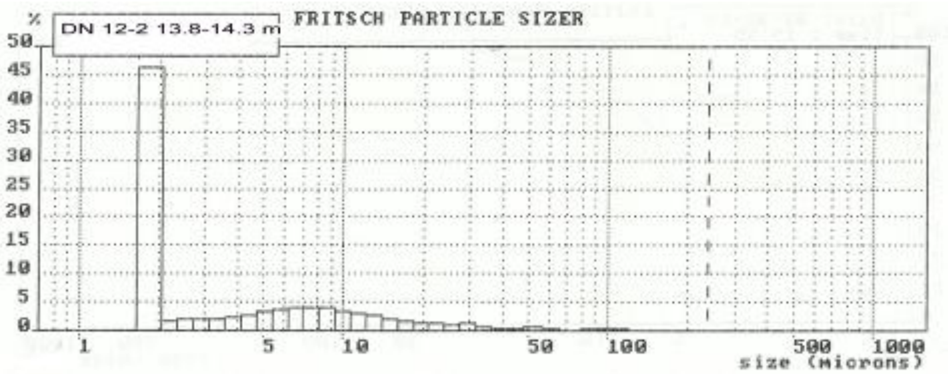

Fig. 12. Differential distribution of the sludge particle granulometric sizes (borehole DN 12-2, horizon 13.8-14.3 m).

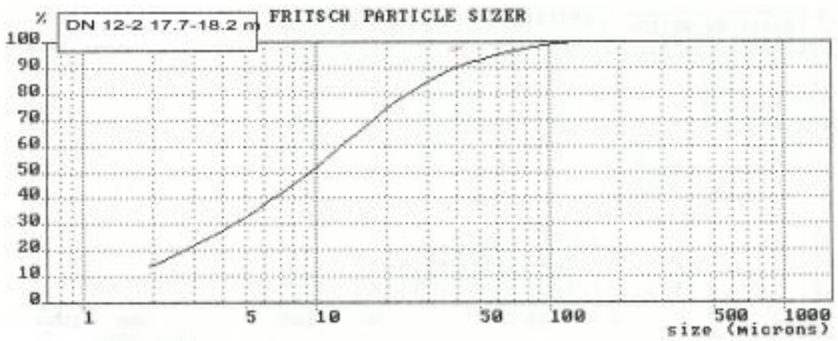

Fig. 13. Cumulative distribution of the sludge particle granulometric sizes (borehole DN 12-2, horizon 17.7-18.2 m).

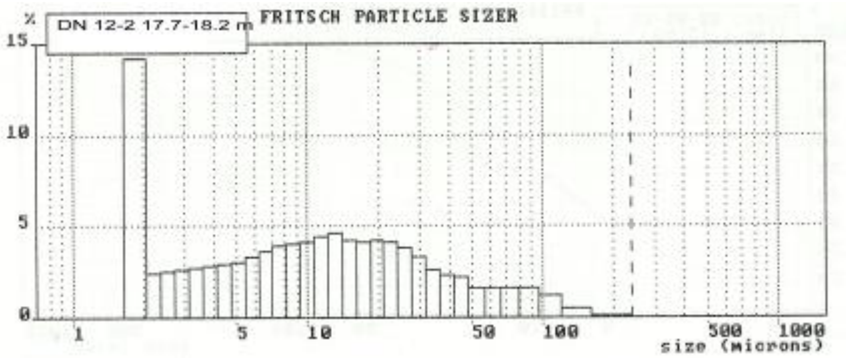

Fig. 14. Differential distribution of the sludge particle granulometric sizes (borehole DN 12-2, horizon 17.7-18.2 m).

According to the measurement results the median diameter value of the sludge particles in the Zahidne tailing is approx 10 µm (borehole ZP 12-1) and in the Centralnyy Yar tailing it is 14-22 µm (borehole CY 12-1). At the horizon of 13.8-14.3 m in the borehole DN 12-2 (Dniprovske) the particles are finer (median diameter is equal to 2.5 µm), which explains the high water retention capacity value for this horizon (sludge humidity of 156 %, Table 2). At the horizon of 17.7-18.2 m in the borehole DN 12-2 the sludge particles median diameter is about of 9 µm.

1.5 Alpha-track radiographic of the sludge samples

Alpha-track radiographic confirmed the existence high α-active particle in the sludge of tailings PChP. The images of α-track radiography of samples are shown in Figure 15-17.

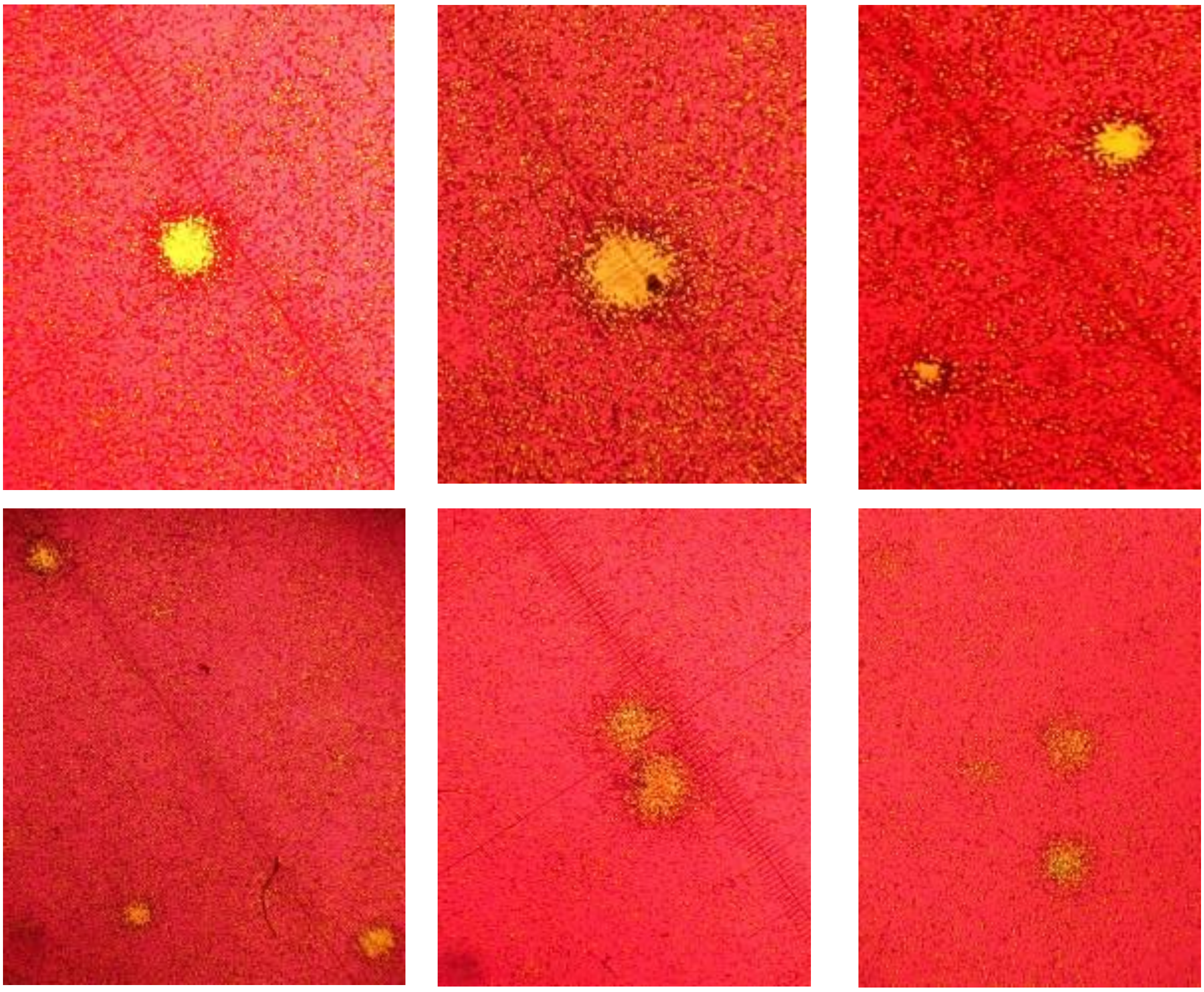

Fig. 15. Images of α-track radiography of sludge samples from tailing “Centralnyy Yar”.

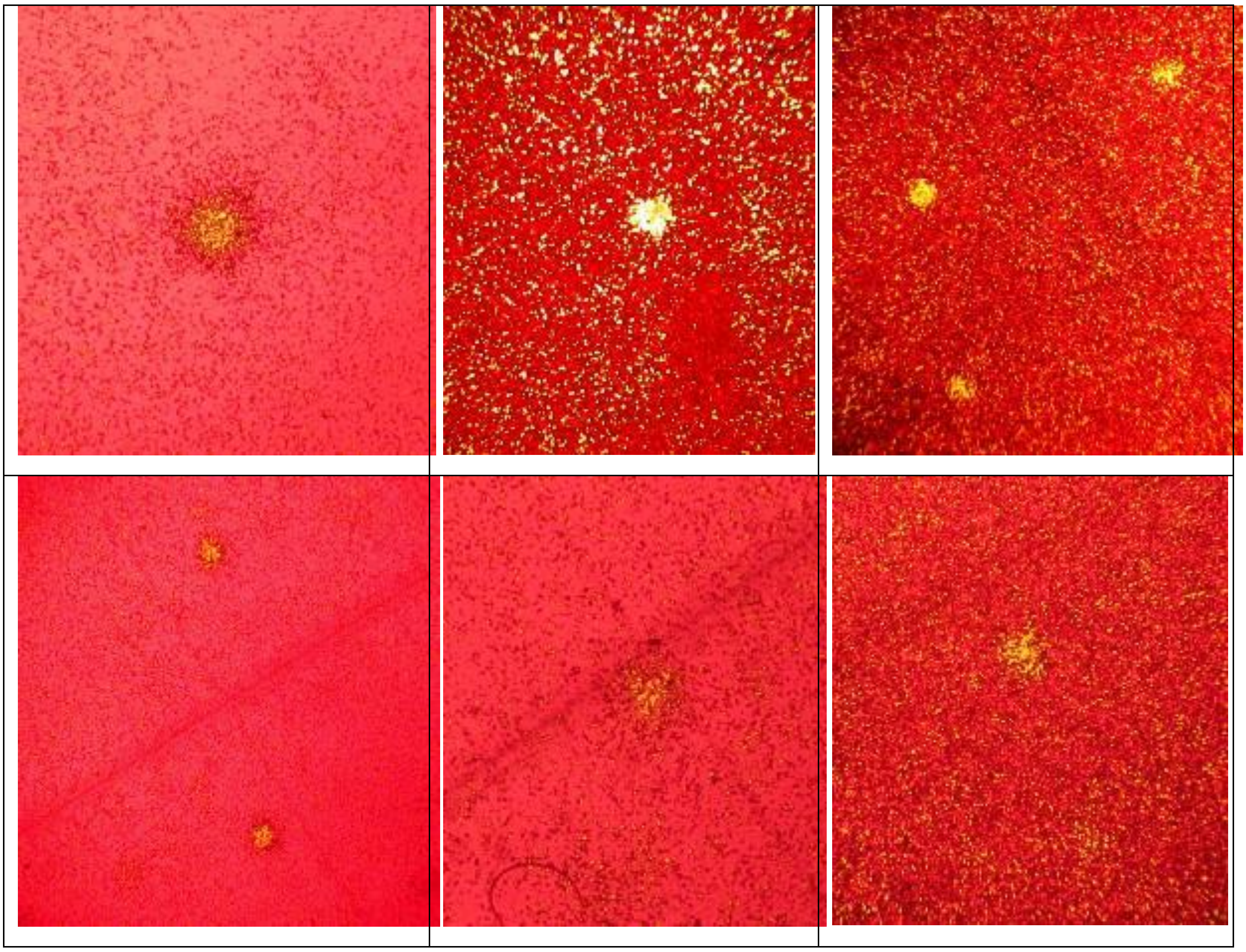

Fig. 16. Images of α-track radiography of sludge samples from tailing “Dniprovske”.

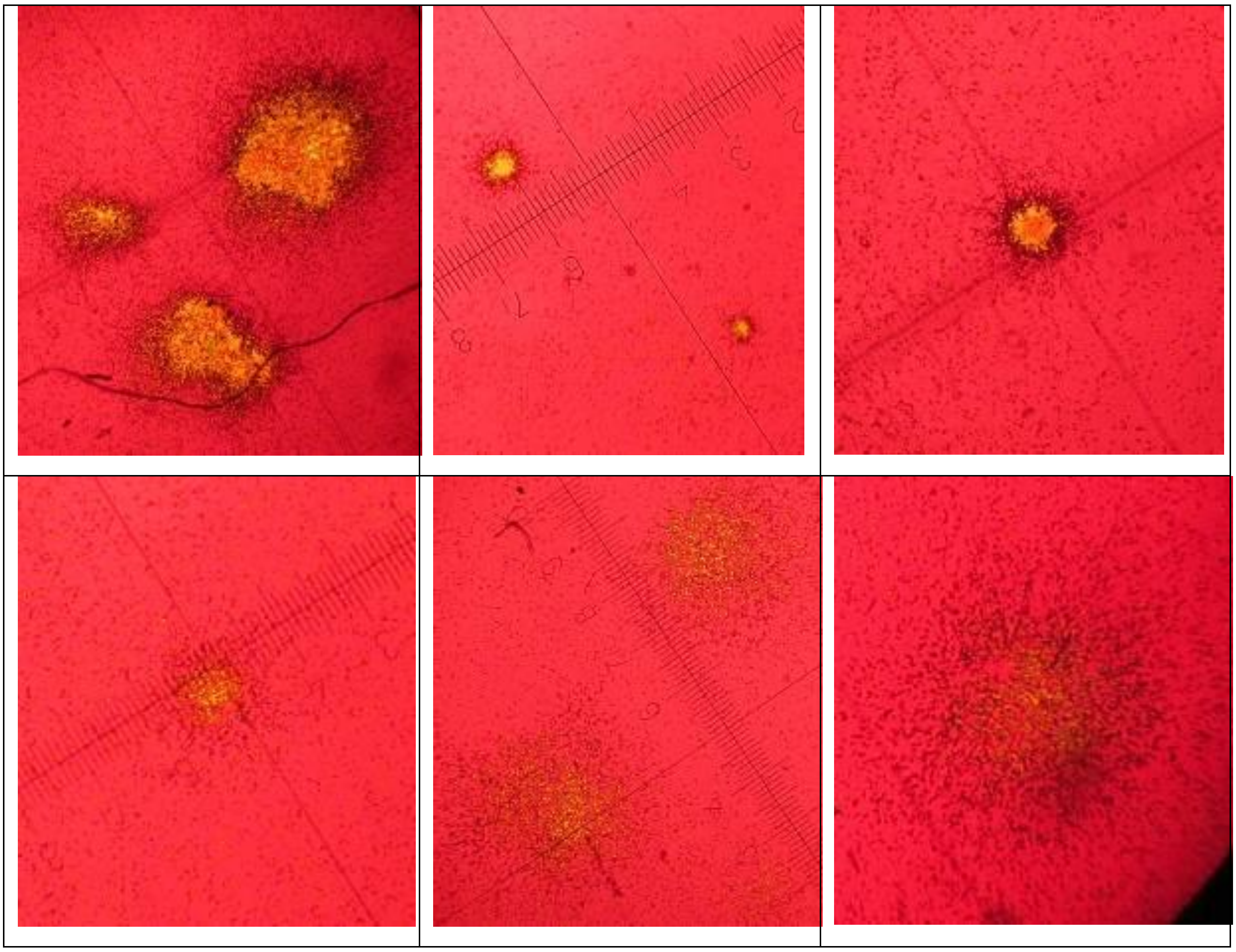

Fig. 17. Images of α-track radiography of sludge samples from tailing "Zahidne".

For comparison spots of α-tracs Chernobyl FP exposed in the same conditions are shown in Figure 18.

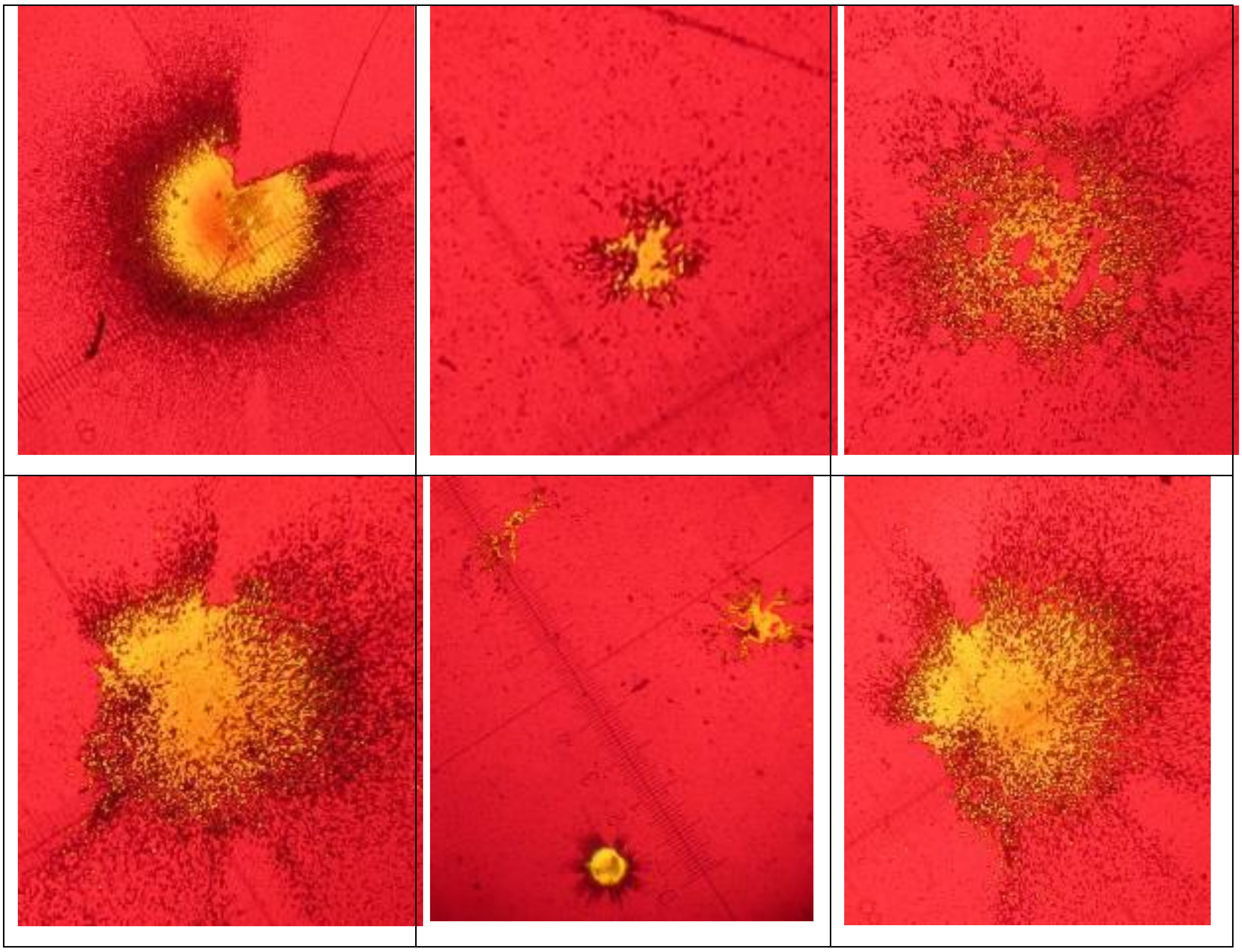

Fig. 18. Images of α-track radiography of samples of bottom sediments of cooling pond ChNPP.

Analysis of the received information indicates the presence of small, discret and high α-activity particles (or cluster) in sludge of the tailing PChP. Interval of size detected particle is 2 to 20 μm. Their α-activity is commensurate with the α-activity of Chernobyl FP. Differences in dispersed composition of α-emitting particles for tailings are observed. The largest particles are mainly observed in the tailing “Zahidne”. The contribution of fine particles dominated in the tailing “Dniprovske”. Evaluation of the number of particles per unit mass of sludge for different tailings PChP is shown in Table 3.

Table 3. Specific number (N) α-activity particles in sludge (dry weight) of PChP tailing

Tailing dump PChP	N, pcs/g
"Central Yar"	420
"Dneprovskoe"	280
"Zapadnoe"	160

Currently, works on the selection of individual fuel particles from samples of sludge are being carried out.

1.6 Element concentrations in the sludge samples

The accompanying chemical elements which are not extracted from the ores accumulate in the tailing dumps of the ore-processing enterprises. For reason the tailing dumps are often considered as prospective anthropogenic deposits. At the same time the tailing dumps are intensive sources of the Environment contamination because they contain among others a significant part of toxic chemical elements.

According to the concentration of the chemical admixtures the uranium ores are classified as uranium, uranium-molybdenum, uranium-vanadium, uranium-nickel-cobalt-bismuth-silver and others. According to the chemical composition of the non-metallic compounds among the uranium ores the following ore types are distinguished: silicate (composed mainly from the silicate minerals), carbonate (contain more than 10-15 % of carbonate minerals); iron-acid (iron-uranium); sulfide (contain more than 8-10 % of sulfide minerals), and caustobiolith (composed mainly of organic matter). Chemical composition often plays a determining role in the selection of the ore procession method. For example, from the silicate ore, uranium is extracted by acids, and from the carbonate ore, by soda solutions, while the iron-acid ore is melted and uranium is concentrated in slag. Caustobiolith ores sometimes are enriched by means of their annealing.

The composition and total contents of the chemical elements and oxides in the PChP tailings were determined by means of X-ray fluorescence analysis of the collected samples. The results are presented in Tables 4 – 7.

Table 4. Concentrations of chemical elements in the sludge samples at various depths in the PChP tailings

Element	Borehole, depth (m), mass fraction (%)											
	Zahidne (ZP 12-1)			Centralnyy Yar (CY 12-1)						Dniprovske (DN 12-2)		
	4.25	*8.75*	*10.25*	*2.25*	*3.75*	*6.25*	*9.25*	*16.25*	*17.75*	*14*	*18*	*18.6*
^{8}O	46.093	48.221	43.474	47.492	49.679	49.859	49.598	50.788	50.294	41.466	46.914	49.574
^{12}Mg			3.213							4.429		
^{13}Al	6.67	8.776	7.163	4.854	1.417	1.332	2.715	1.538	6.356	5.122	2.82	5.631
^{14}Si	27.839	32.46	22.547	30.9	32.047	32.541	30.874	34.638	35.929	16.308	25.099	35.521
^{16}S	0.529	0.197	0.306	2.372	8.497	8.327	8.711	7.646	1.692	4.233	9.632	0.725
^{19}K	0.804	5.028	3.649	2.034	0.311	0.299	0.414		2.16	2.92	0.801	1.638
^{20}Ca	2.931	2.167	7.583	7.722	6.133	5.346	5.65	3.059	0.878	13.072	10.331	0.989
^{22}Ti	0.408	0.16	0.487	0.514	0.424	0.502	0.474	0.514	0.48	0.126	0.334	0.384
^{23}V	0.059		0.046		0.058	0.09	0.082	0.055		0.091	0.053	
^{24}Cr	0.019		0.015							0.016		
^{25}Mn	0.128	0.107	0.705	0.174	0.038	0.039	0.046	0.064	0.037	0.51	0.098	0.167
^{26}Fe	14.159	2.616	9.852	3.747	1.215	1.425	1.213	1.467	1.902	11.219	3.472	5.257
^{28}Ni			0.041	0.002						0.011		
^{29}Cu	0.018	0.002	0.064	0.007	0.001				0.004	0.017	0.11	0.003
^{30}Zn	0.018	0.013	0.212	0.024	0.001				0.004	0.022	0.005	0.005
^{31}Ga		0.004	0.003						0.001			
^{33}As	0.007	0.01	0.135	0.002	0.002	0.002	0.002		0.001	0.042	0.013	
^{37}Rb	0.003	0.047	0.027	0.007					0.004	0.012		0.008

Table 4 (continued)

Element	Borehole, depth (m), mass fraction (%)											
	Zahidne (ZP 12-1)			Centralnyy Yar (CY 12-1)						Dniprovske (DN 12-2)		
	4.25	*8.75*	*10.25*	*2.25*	*3.75*	*6.25*	*9.25*	*16.25*	*17.75*	*14*	*18*	*18.6*
^{38}Sr	0.025	0.059	0.02	0.022	0.013	0.016	0.019	0.016	0.008	0.122	0.144	0.011
^{39}Y	0.018	0.002	0.006	0.005	0.002	0.003	0.002	0.002	0.004	0.008	0.005	0.003
^{40}Zr	0.138	0.022	0.045	0.065	0.071	0.093	0.066	0.06	0.07	0.018	0.053	0.044
^{41}Nb		0.004		0.002	0.002							0.003
^{42}Mo									0.005	0.008		
^{45}Rh				0.003		0.003						
^{47}Ag							0.003					
^{50}Sn	0.021	0.012	0.023	0.013	0.01	0.01	0.012	0.008	0.01	0.022	0.01	0.015
^{51}Sb								0.008				
^{52}Te	0.028	0.019						0.013				
^{53}I		0.017										
^{56}Ba			0.087		0.061	0.091	0.093	0.063	0.034		0.039	
^{72}Hf								0.004				
^{74}W		0.003										
^{82}Pb	0.059	0.055	0.259	0.013				0.004		0.189	0.027	0.007
^{90}Th	0.009					0.003						0.005
^{92}U	0.019		0.039	0.027	0.017	0.021	0.026	0.051	0.127	0.018	0.041	0.009

Table 5. Chemical composition of sludge recalculated to oxides (mass fraction, %) at various depths in the Centralnyy Yar tailing

Oxide	Centralnyy Yar , borehole CY 12-1					
	2.25 m	3.75 m	6.25 m	9.25 m	16.25 m	17.75m
Ag_2O				0.003		
Al_2O_3	9.171	2.677	2.517	5.129	2.906	12.011
As_2O_3	0.003	0.002	0.002	0.002		0.002
BaO		0.068	0.101	0.103	0.07	0.038
CaO	10.805	8.581	7.48	7.905	4.28	1.229
CuO	0.009	0.002				0.004
Fe_2O_3	5.357	1.738	2.037	1.734	2.097	2.719
Ga_2O_3						0.002
HfO_2					0.005	
K_2O	2.451	0.375	0.36	0.499		2.602
MnO_2	0.275	0.060	0.061	0.072	0.102	0.058
MoO_3						0.008
Nb_2O_5	0.003	0.002				
Ni_2O_3	0.003					
PbO					0.004	
Pb_2O_3	0.014					
Rb_2O	0.007					0.005
Rh_2O_3	0.004		0.004			
SO_2	4.739	16.975	16.636	17.403	15.275	3.38
Sb_2O_3					0.01	
SeO_2				0.002	0.002	
SiO_2	66.104	68.560	69.614	66.049	74.102	76.862
SnO_2	0.016	0.012	0.012	0.015	0.01	0.012
SrO	0.025	0.015	0.019	0.022	0.02	0.01
TeO_2					0.016	
ThO_2			0.003			
TiO_2	0.858	0.708	0.837	0.792	0.857	0.801
UO_3	0.033	0.021	0.026	0.032	0.061	0.153
V_2O_5		0.104	0.161	0.146	0.099	
Y_2O_3	0.006	0.002	0.003	0.003	0.002	0.004
ZnO	0.030	0.002				0.005
ZrO_2	0.087	0.096	0.126	0.089	0.082	0.094

Table 6. Chemical composition of sludge recalculated to oxides (mass fraction, %) at various depths in the Zahidne tailing

Oxide	Zahidne, borehole ZP 12-1		
	4.25 m	8.75 m	10.25 m
Al_2O_3	12.602	16.582	13.534
As_2O_3	0.009	0.014	0.178
BaO			0.097
CaO	4.101	3.032	10.61
Cr_2O_3	0.027		0.022
CuO	0.022	0.003	0.08
Fe_2O_3	20.244	3.74	14.086
Ga_2O_3		0.006	0.004
I		0.017	
K_2O	0.969	6.056	4.396
MgO			5.328
MnO_2	0.203	0.169	1.115
Ni_2O_3			0.058
Nb_2O_5		0.005	
Pb_2O_3	0.065	0.061	0.288
Rb_2O	0.003	0.051	0.03
Rh_2O_3			
SO_2	1.056	0.394	0.612
SiO_2	59.557	69.442	48.236
SnO_2	0.027	0.015	0.029
SrO	0.03	0.069	0.024
TeO_2	0.035	0.024	
ThO_2	0.01		
TiO_2	0.68	0.267	0.813
WO_3		0.004	
UO_3	0.023		0.047
V_2O_5	0.105		0.082
Y_2O_3	0.022	0.003	0.008
ZnO	0.022	0.016	0.264
ZrO_2	0.186	0.03	0.06

Table 7. Chemical composition of sludge recalculated to oxides (mass fraction, %) at various depths in the Dniprovske tailing

Oxide	Dniprovske, borehole DN 12-2		
	14 m	18 m	18.5 m
Al_2O_3	9.677	5.328	10.64
As_2O_3	0.055	0.018	
BaO		0.043	
CaO	18.289	14.455	1.384
Cr_2O_3	0.023		
CuO	0.021	0.138	0.004
Fe_2O_3	16.04	4.964	7.516
K_2O	3.517	0.964	1.974
MgO	7.345		
MnO_2	0.808	0.155	0.264
MoO_3	0.012		
Nb_2O_5			0.004
Ni_2O_3	0.016		
Pb_2O_3	0.21	0.03	0.008
Rb_2O	0.013		0.008
SO_2	8.457	19.242	1.448
SiO_2	34.887	53.696	75.991
SnO_2	0.028	0.013	0.02
SrO	0.144	0.171	0.013
ThO_2			0.006
TiO_2	0.21	0.557	0.64
UO_3	0.022	0.049	0.011
V_2O_5	0.163	0.094	
Y_2O_3	0.01	0.006	0.004
ZnO	0.028	0.006	0.006
ZrO_2	0.025	0.071	0.06

Analysis of the obtained results reveals the differences between the chemical composition of sludge in the tailings. In the Centralnyy Yar tailing prevail oxides SiO_2 (mass fraction of 74 - 66 %), SO_2 (17 - 15 %), CaO (8 - 4 %), Al_2O_3 (5 - 2.5 %) and Fe_2O_3 (2 - 1.7 %). The corresponding sequences for the tailings Zahidne and Dniprovske are SiO_2 (69 - 48 %), Fe_2O_3 (20 - 4 %), Al_2O_3 (17 - 13 %), CaO (10 - 3 %), K_2O (6 - 1 %) and SO_2 (1 - 0.3 %), and SiO_2 (53 - 34 %), SO_2 (19 - 8 %), CaO (18 - 14 %), Fe_2O_3 (16 - 5 %), Al_2O_3 (10 - 5 %) and K_2O (3.5 - 1 %), respectively.

The highest mass concentrations of arsenic (^{33}As) were found in the profiles of the Zahidne tailing (0.007 - 0.135 %) and Dniprovske (0.013 - 0.042 %).

1.7 Radionuclide composition

At the first stage of determination of specific activities in the PChP tailings the collected samples were analyzed by means of γ-spectrometry. Results of the γ-spectrometric measurements are presented in Fig. 19-21. Confidence probability of the presented uncertainties is 0.95.

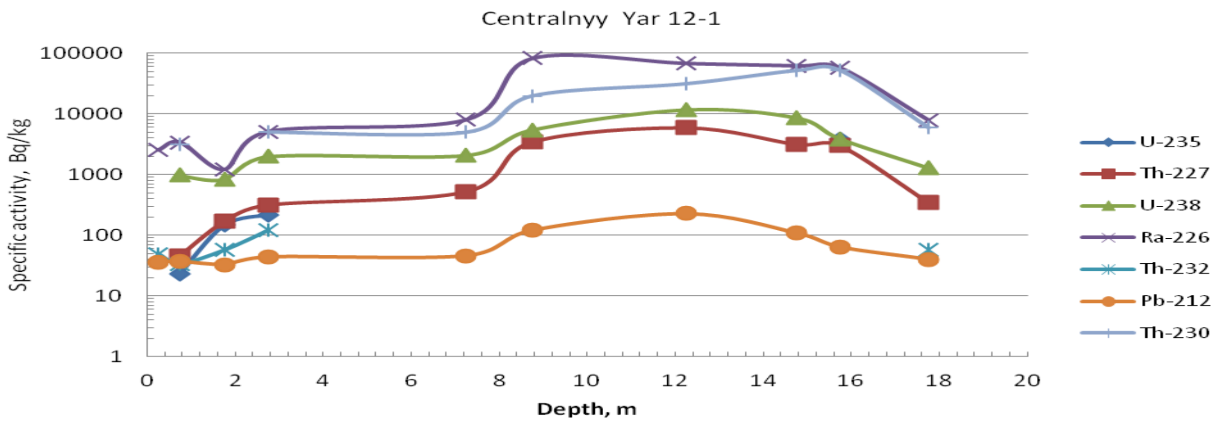

Fig. 19. Distribution of specific activities of the uranium-thorium series radionuclides in the vertical profile of the Centralnyy Yar tailing (borehole 12-1).

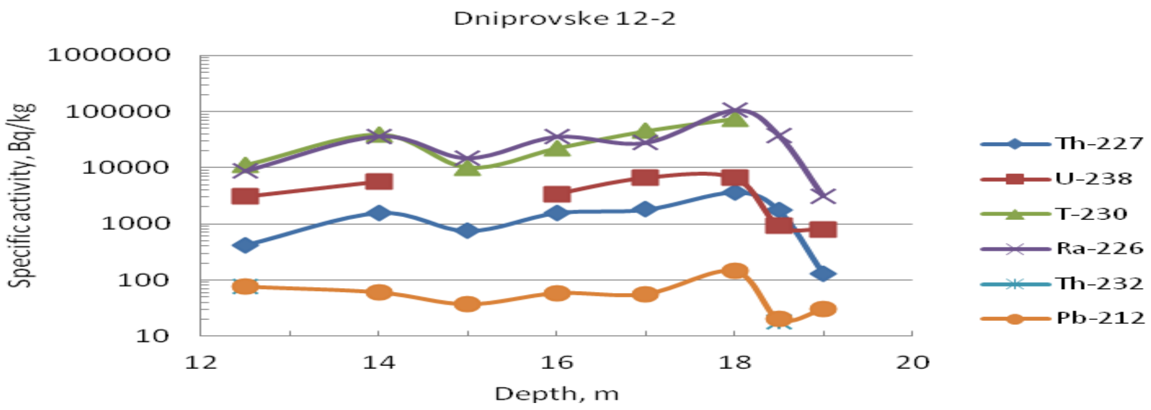

Fig. 20. Distribution of specific activities of the uranium-thorium series radionuclides in the vertical profile of the Dniprovske tailing (borehole 12-2).

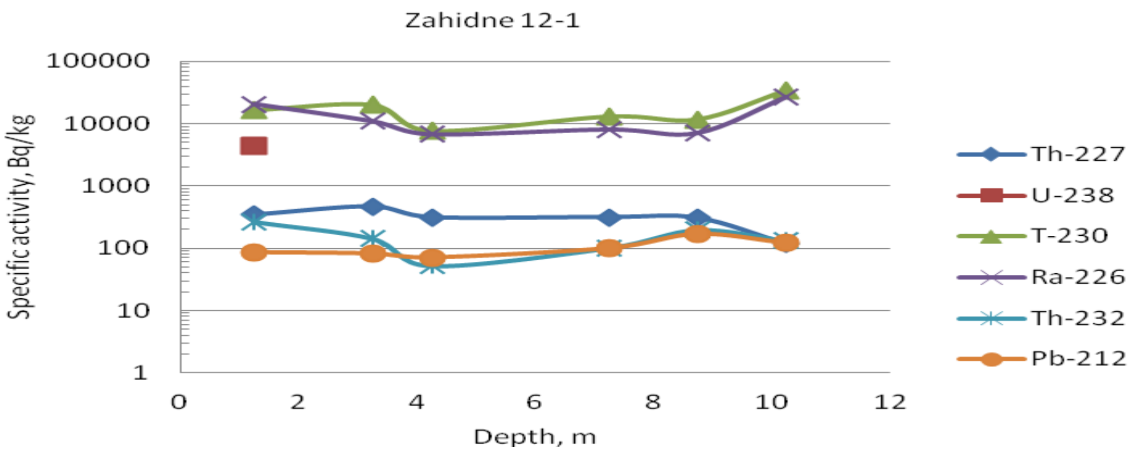

Fig. 21. Distribution of specific activities of the uranium-thorium series radionuclides in the vertical profile of the Zahidne tailing (borehole 12-1).

According to the obtained results, the highest specific activities of the uranium-thorium series radionuclides are found in the Centralnyy Yar tailing. Among other radionuclides, the highest specific activities in this tailing were measured for ^{226}Ra (up to 100000 Bq/kg). Absence of radioactive equilibrium between ^{230}Th and ^{226}Ra may indicate that solutions enriched with radium were stored in this tailing dump.

Discussion and conclusions

Obtained results allowed estimation of the main physical-chemical parameters and elemental composition of core material of PChP tailings. The highest differences between the element concentrations in the tailing dumps are found for such elements as Al, S, K, Ca, Mn, Fe, As, Sr. Specific activities of the uranium-thorium series radionuclides decrease in the PChP tailings sequence: Central Yar > Dneprovskoe > Zapadnoe.

By autoradiography using track detectors the homogeneity of the distribution of α-emitting radionuclides in sludge of PChP tailings has been investigated. It has been found that beside a homogeneous background distribution of the α-activity, single spots with significant higher activity concentration exist in form of α-active particles. Their concentration in sludge from different tailings has been determined. If radioactivity is much higher than background level, handling TENORM may cause problems in dry processes, where NORM scales and dust become air borne and have a significant chance to enter the body. The main hazards associated with α-active particles are inhalation and ingestion routes of entry. Hence, the study of the characteristics of the α-active particles in tailing dumps should be continued.

References

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