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DELIVERABLE IRA-Particle Behaviour-D3

Set of parameters for FP dissolution in the drained bottom sediments, characterization of FP matrix.

Journal article submitted.

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Table of Contents

Executive Summary

Assessments of the occurrence of chemical forms of radionuclides ^{90}Sr, ^{137}Cs, ^{238}Pu, $^{239,240}Pu$, ^{241}Am and the dispersal composition of fuel particles in bottom sediments of the cooling pond of the Chernobyl nuclear power plant (CP ChNPP) were carried out. According to the obtained results, in slimy bottom sediments more than 98 % of ^{90}Sr, ^{137}Cs, ^{238}Pu, $^{239,240}Pu$, ^{241}Am are in non-exchangeable forms. However, in sandy bottom sediments about 10 % of ^{90}Sr and ^{137}Cs are in exchangeable forms. At the same time, about 70 % of total ^{90}Sr and more than 80 % of total ^{241}Am and isotopes of plutonium in CP ChNPP may be located in the matrix of the fuel particles. The obtained results also indicate selective leaching of radionuclides from the matrix of fuel particles and the presence of spatial heterogeneity of the chemical forms of radionuclides in sediments of the ChNPP cooling pond.

Journal article has been published with reference to the COMET project (http://jnpae.kinr.kiev.ua/15.3.html**):**

Protsak V. P., Odintsov O. O. Assessment of forms finding of Chernobyl radionuclides in bottom sediments of cooling pond of the ChNPP //Nuclear Physics and Atomic Energy, 2014, V.15, N.3, p.259-268 (Ukr.) (**Annex : Journal article submitted - pdf file).**

Introduction

The cooling pond of the Chernobyl Nuclear Power Plant (CP ChNPP) is a manmade water reservoir created for cooling the heat exchangers of the four nuclear reactors of the ChNPP. CP ChNPP is located in the northern part of Ukraine, at the right-bank floodplain of the Pripyat river. The CP ChNPP shore is formed both as the headrace of the river and as the 25-km long levee. The area surrounded by the levee includes the riverbed, former riverbeds and lakes. The CP area is approximately 22 km², its volume is 151.2 million m³, its length 11.5 km, and its mean width 2.2 km. Average and maximum depths are 6.6 m and about 18 m, respectively, and the water elevation of the CP is about 5-7 m over the Prypiat river. In order to maintain this level of water in CP ChNPP (i.e. to compensate the water loss due to filtration and evaporation), water is permanently pumped from the river into CP, which implies significant financial expenditures.

As a result of the accident in 1986, large amounts of radioactive aerosols and particles of the dispersed fuel from the ruined reactor deposited to the CP surface, and the pond was highly contaminated. Besides, about 5000 m³ of highly contaminated water came into the pond from the main circuit of the accidental reactor and from the systems of the technical maintenance of the unit (Gudkov et al., 2007). Also, parts of the water used for the fire suppression at the 4th unit during the accident, as well as a part of water which was collected in the drainage system after decontamination of the ChNPP industrial site, penetrated the pond.

According to previously published results (Buckley M.J. et al., 2002; Kashparov et al., 2004), the main mass of radionuclides of bottom sediments in the cooling pond is included in matrix of fuel particles (FP). Certain radionuclides such as ^{95}Zr, ^{95}Nb, ^{99}Mo, ^{141,144}Ce, ^{154,155}Eu, ^{237,239}Np, $^{238\text{-}242}$Pu, ^{241,243}Am, ^{242,244}Cm were released from the accidental unit in fuel particles matrix only. More than 90% of ^{89,90}Sr and ^{103,106}Ru activity also was released in FP form. All these radionuclides were deposited on the various surfaces in the matrix of FP in various degree of transformation and were initially characterized by a low mobility in the environment. Fuel particles (separate grains, crystallites of uranium oxide) with a median

diameter of about 4-6 μm and their aggregates can be divided into 3 groups according to their dissolution rates under natural conditions (Kashparov ey al., 2004, 2012):

1. chemically extra-stable particles (hypothetically U-Zr-O), formed as a result of high-temperature annealing of UO_2 in the presence of zirconium in building materials). These particles formed at the first moment of the accident on 04/26/86 and fell within the narrow western trace;
2. non-oxidized chemically stable fuel particles (UO_2) of the first release (04/26/86), formed as a result of the mechanical destruction of nuclear fuel. These particles were also created along the narrow western trace of fallout. Leaching of the fission products from particles of this type during the accident was minimal. This fact is confirmed by a constant ratio in the fission products;
3. particles of low chemical stability (UO_{2+x}), formed as a result of oxidization of the nuclear fuel in the period 04/26/86-05/05/86. These particles were predominantly in the northern and southern traces of fuel fallout.

A superposition of all three types of fuel particles was observed in the initial fallout at various points in the 30-km zone. Their partial contribution of these three groups of FP varies depending on the direction and distance from the ChNPP. This division of fuel particles into three types is hypothetical because there are no clear boundaries between the various types of particles. In addition, the extent of nuclear fuel oxidation and its zirconium content, as well the volumetric distribution, can vary over a wide range. An analysis of soil physical and chemical properties affecting FP dissolution shows that the rate of dissolution for particles of the same group can correlate, to the largest extent, with the pH of the water extract. Most of particles remained unaffected in neutral soils (pH>6) independently of their direction and distance from the ChNPP.

If the decision of turning off pumps is made, the water level in CP ChNPP will decrease by 6-7 m in 5-7 years. Hence, 13-18 km^2 of highly radioactive bottom sediments will be uncovered (Petrov and Kireev, 2006). Intensive processes of soil formation due to aeration, oxidizing of organic residues, and changes of agrochemical characteristics, might begin in the drained sites. Therefore, dissolution of FP, leaching of radionuclides from their matrix, and

increased mobility and migration ability of radionuclides should be expected. In order to obtain predictive estimates for dynamics of this process, the data on radionuclides species presented in bottom sediments are required.

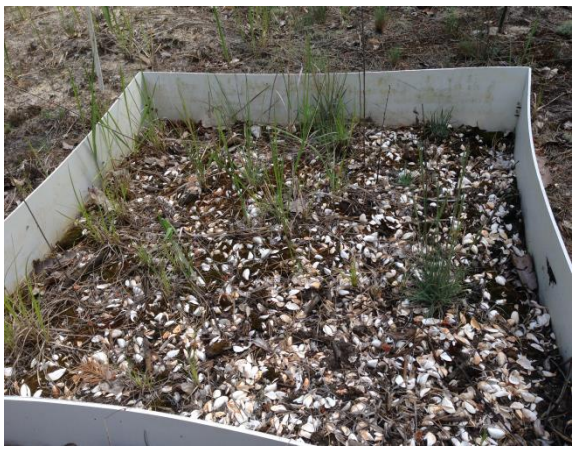
Methods

1.1 Sampling

An experimental field plot for the simulation of drainage of sediment and its exposure to natural conditions was created. For this purpose, samples of the bottom sediments were collected by boat in the northern-western part of cooling pond of ChNPP at a depth of 5–7 m by a bottom sampler . The water area for sampling was about 3000 m^2. Nearly 120 liters of the bottom sediments were collected during the sampling. Cuvettes with a dimension of 1x1 m (Fig. 1), without bottom, made beforehand and placed directly on the bank of CP ChNPP, were filled with collected bottom sediments. These samples were then used in model experiments on dynamics of radionuclides species in the bottom sediments of CP ChNPP to imitate draining and exposure to natural conditions (Protsak and Odintsov, 2014).

Periodic sampling of the bottom sediments from the filled cuvettes (trays) in the experimental site (Fig. 2), was also carried out in order to control main agrochemical characteristics, and estimate temporal dynamics for the radionuclide species presented in the bottom sediments and modified due to the imitation of their draining.

a) b)



c)

Fig. 1 – Preparation of the experimental site (a,b). A view of the bottom sediments of the CP ChNPP at the experimental site after 3 years of the exposure to the natural conditions (c) .

Decrease of the water level in the ChNPP cooling pond started from Feb 2015 (Fig.2), and the water level has now decreased by 3.5 m (Fig.3 - 5). Thus, keeping conditions of radionuclides have started to change. In this regard, study the dynamics of chemical forms of radionuclides on drained sites was started.

Fig. 2. Water level in the ChNPP cooling pond near the pumping station (BPS-3) in February 2015.

Fig. 3. Water level in CP ChNPP near the pumping station (BPS-3) in May 2015.

Fig. 4. Water level in the northern part of CP ChNPP in June 2015.

Fig. 5. Water level of the ChNPP cooling pond in September 2015.

On the drained sites of the ChNPP cooling pond, 6 research points were chosen (Fig. 6). At these points, samples of the dried sediments were collected from different soil layers at the depths of 0-25 cm, and samples of the first vegetation (leaves of young shoots of willow) were also collected.

Fig. 6. Location of the research points for study of the dynamics of radionuclide forms.

1.2 X-ray radiography of the samples of bottom sediments

Radiographic studies were carried out with thea goal of visualizating Chernobyl FP in samples of the bottom sediments from the CP ChNPP, and estimation of their dispersed composition. Weighed and air-dried samples of the bottom sediments were thin layer distributed (extremely close to monolayer) on a hard surface and sealed with polyethylene film. Then these preparations were exposed during 750 hours on X-ray film RP-U "Onico" (Ukraine). Development and fixation of the film were done according to the producer's recommendations. Patterns of black spots, shown up on the films due to radioactive emission of hot particles, were digitalized by an Epson V330 Photo scanner in indirect lighting, with resolution 600 dpi (dots per inch), and analyzed consequently in the software environment «ImageJ». In order to estimate dispersal composition of FP, the experimentally found relationship between the diameter (pixels) of black spots and dose rate (result of multiplication of ^{90}Sr (Bq) activity in FP on exposure time (c)) was used. The relationship was obtained through exposure of single fuel particles line on X-ray film during various periods of time (Fig. 7). These particles were isolated from the soil samples and air filters. The ^{90}Sr activity was determined previously by the instrumental method (Yoschenko et al., 2003) for every sample. Detection sensitivity of the method at 750-hour exposure was 0,0004 Bq of ^{90}Sr per particle, that corresponded to FP diameter of 0,5 μm.

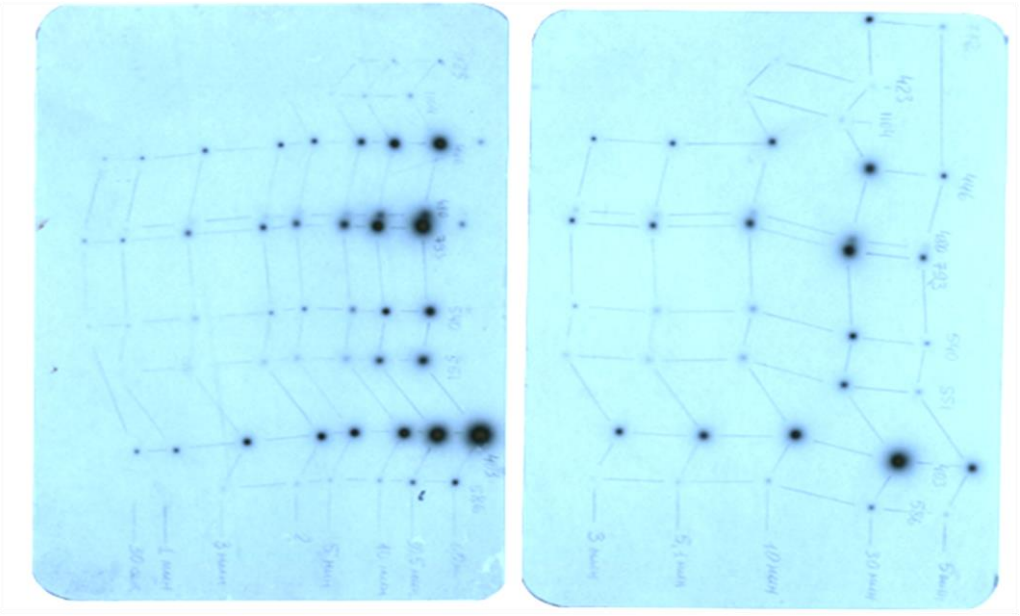


Fig.7. Radiography of the individual fuel particles with various exposure periods used in order to obtain the relationship between the diameter of black spots and irradiation rate. Film size was 13x18 cm.

1.3 Method of sequential extraction

Physical - and – chemical species of ^{90}Sr, ^{137}Cs, $^{238,239,240}Pu$ and ^{241}Am presented in the bottom sediments of CP ChNPP were determined by the method of sequential extraction.

Air-dried samples with mass of 10 g were selected from the general sample with three replications. By the gamma-spectrometry measurements it was indicated that the difference of the ^{137}Cs and ^{241}Am specific activity in the integrated samples and in their aliquots didn't exceed 15 %.

Leaching of radionuclides was carried out with three replications for each sampling. The ratio of solid phase to liquid phase was 1:5. Leaching conditions, reagents, and radionuclides species are represented in Table 1.

Table 1. Conditions for sequential leaching of radionuclides from samples of the bottom sediments of CP ChNPP

Stage №	Reagent and conditions of leaching	Radionuclide species
I	Distilled water; 24 hours at room temperature and periodic shaking	water soluble
II	1 M CH_3COONH_4 (NH_4Ac); pH 7; 24 hours at room temperature	exchangeable
III	1 M HCl; 24 hours at room temperature	mobile
IV	0.2 M $(NH_4)_2C_2O_4$ + 0.1 M $H_2C_2O_4$ (Tamm solution); pH 3,2; 2 hours at room temperature	Bound with amorphous oxides and hydroxides of Fe and Al in the form of organic-mineral complexes
V	8 M HNO_3; 24 hours at room temperature	Sparingly soluble and associated with fuel particles (U_xO_y)
VI	Residue after stage 5 was washed at 550 °C during 6 hours, treated by the acid mix of 8 M HNO_3 + 10 M HCl for 2 hours at 95° C.	Bound with organic components of the bottom sediments and fuel particles. (UO_2)
VII	Residue after stage 6 was leached by the acid mix: 8 M HNO_3 + 4 M HF for 2 hours at 95° C.	Strongly fixed in mineral components of the bottom sediments and in matrix of the constructive fuel particles ($U_xZ_yO_z$)
VIII		Insoluble rests.

Extracts after leaching were separated from the solid residue by centrifugation at a velocity of 6000 rpm. Decanted solutions were filtrated through the paper filter (“blue band”). After the II-VII leaching stages, solid residues were washed with distilled water, and supernatants obtained after the centrifugation and filtration were combined with the corresponding extracts of leaching. The filters used in these procedures were combined with the solid residues.

1.4 Measurement of specific activity of radionuclides in samples.

The ^{137}Cs and ^{241}Am content in the extracts after leaching was determined by gamma-spectrometry, ^{90}Sr was determined by beta-radiometry after radiochemical extraction, and the content of ^{238}Pu, $^{239+240}Pu$ and ^{241}Am was determined by alfa-spectrometry after radiochemical extraction.

The activity of ^{137}Cs, $^{154,155}Eu$ and ^{241}Am was measured on a γ-spectrometric complex equipped with semi-conductive detector GL2020R (CANBERRA, USA) of ultra-pure germanium. Minimum detectable activity for our geometry of measurements for γ-line 59 keV (^{241}Am) reached 0, 1 Bq/sample and 0,4 Bq/sample for $^{137}Ba^{m}$ (661,6 keV). Treatment of the spectra was carried out in the software environment GENIE-2000.

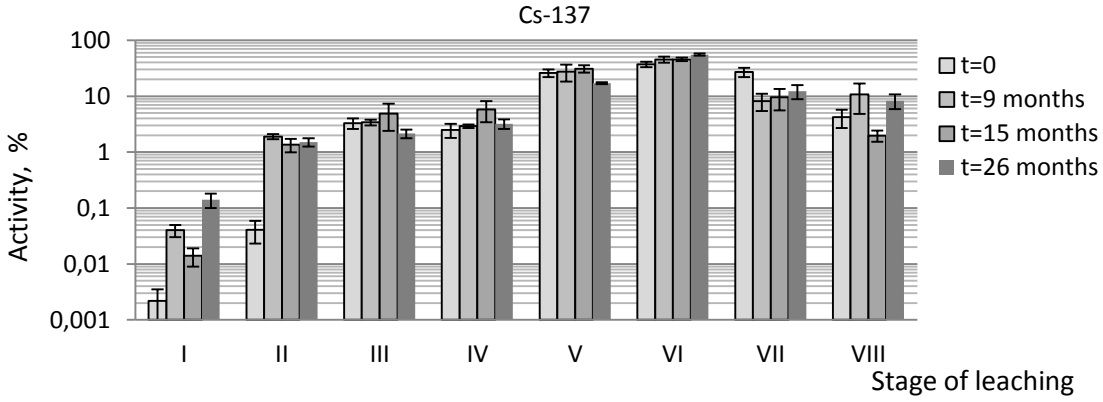
Beta-radiometric measurements of ^{90}Sr ($SrCO_3$) were carried out on a beta-radiometer with a low background UMF-1500 (in original УМФ-1500), immediately after its radiochemical extraction by precipitation.

Radiochemical extraction of Plutonium and Americium after leaching was carried out by the ion-exchangeable method (Ageyev et al., 2005). In order to determine chemical yield of Plutonium and Americium, tracers (^{242}Pu i ^{243}Am) with known volume activity have been used.

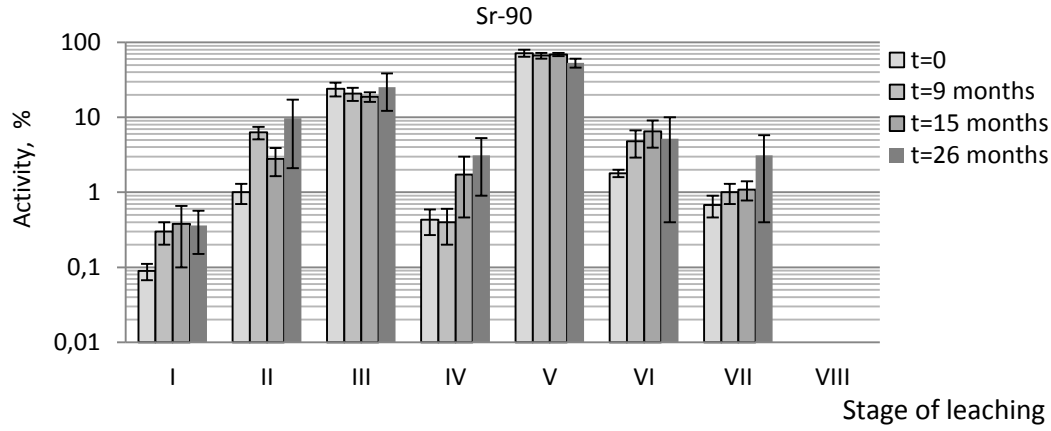
Alfa-spectrometric measurements were carried out on an eight-channel α-spectrometer EG&G ORTEC OCTETE PC with semi-conductive siliceous detectors of series BU-017-450-100 ULTRA, at efficacy of registration 25% at the distance 12 mm from source. The background of the spectrometer for energies exceeding 3MeV was not more than 1 impulse per hour, and the separating ability of the spectrometer was 19 keV for 5486 keV line (^{241}Am).

Results

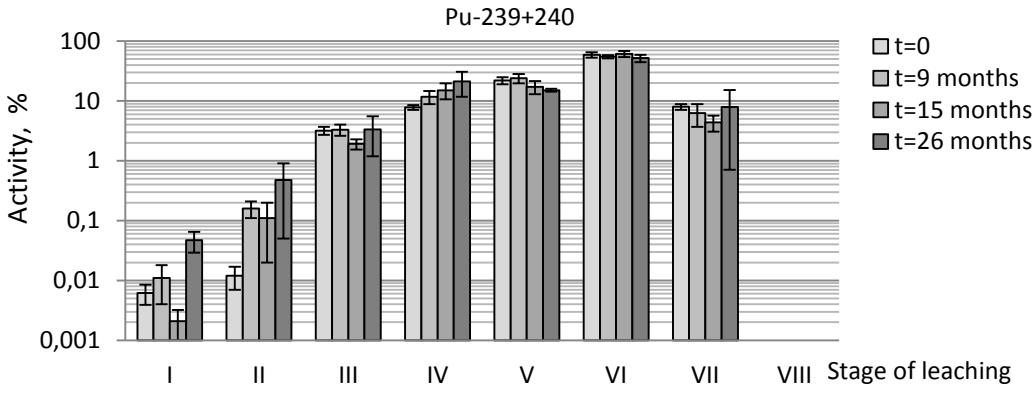
The dynamics of ^{137}Cs , ^{90}Sr, $^{239+240}Pu$ and ^{241}Am activity among fractions leached from the bottom sediments of CP ChNPP before their draining (t=0), after draining and exposure during 9, 14 and 26 months to natural conditions at the experimental site are presented in Fig. 8.


Cs-137
100
10
1
0,1
0,01
0,001
Activity, %
t=0
t=9 months
t=15 months
t=26 months
I
II
III
IV
V
VI
VII
VIII
Stage of leaching

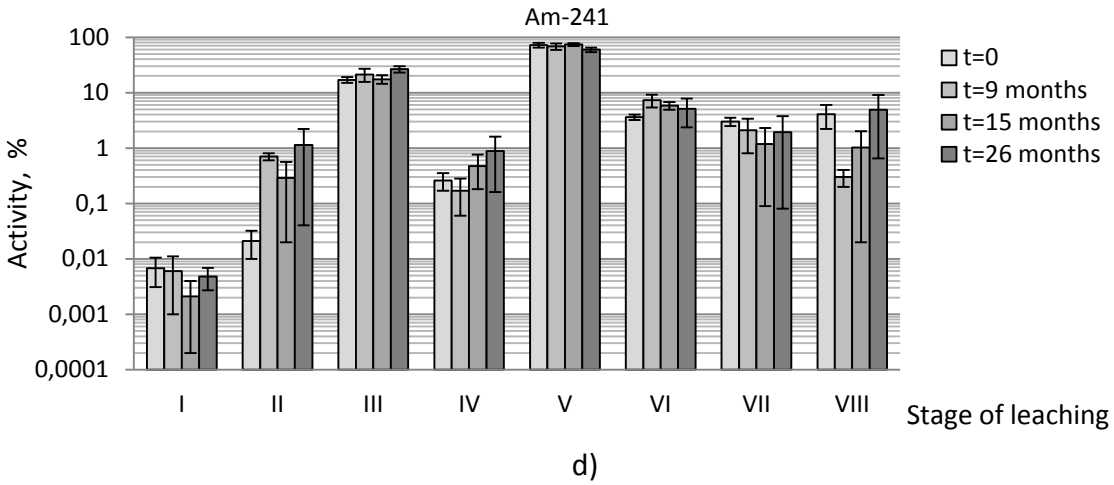
a)


Sr-90
100
10
1
0,1
0,01
Activity, %
t=0
t=9 months
t=15 months
t=26 months
I
II
III
IV
V
VI
VII
VIII
Stage of leaching

b)


Pu-239+240
100
10
1
0,1
0,01
0,001
Activity, %
t=0
t=9 months
t=15 months
t=26 months
I
II
III
IV
V
VI
VII
VIII
Stage of leaching

c)



d)

Fig. 8. The dynamics of ^{137}Cs (a), ^{90}Sr (b), $^{239+240}$Pu (c) and ^{241}Am (d) activity among fractions leached from the bottom sediments of CP ChNPP after their draining (t=0) and exposure during 9 months (t=9), 14 months (t=14) and 26 months (t=26) in the natural conditions on the experimental site.

Analysis of the results indicates that total content of ^{137}Cs, $^{239 + 240}$Pu and ^{241}Am exchangeable species is quite small (less than 2% of the total content) despite the increase of radionuclides exchangeable species amount by a factor of 10 during the 26 months of exposure to natural conditions at the experimental site (fig. 8).

A slightly higher content of ^{90}Sr exchangeable species (10 % of the total content) can be caused by its leaching from shells. During our study, the content of these species of radionuclides in the samples of sediments amounted to approximately 15 % of dry weight. Special work is planned to test this assumption.

The previous experimental results indicated that the rate of Chernobyl fuel particles destruction was minimal in a neutral medium, and that transfer of FP increased in alkaline or acidic medium. Hence, pH is one of the key factors affecting the amount of radionuclides leached from the fuel particles. Dynamics of pH values in the experimental site is shown in Fig. 9. As it is shown in the figure, pH of the medium in the experimental site remained slightly alkaline for a long time. Probably, it is a main reason for the low coefficient of the radionuclide leaching from modified bottom sediments.

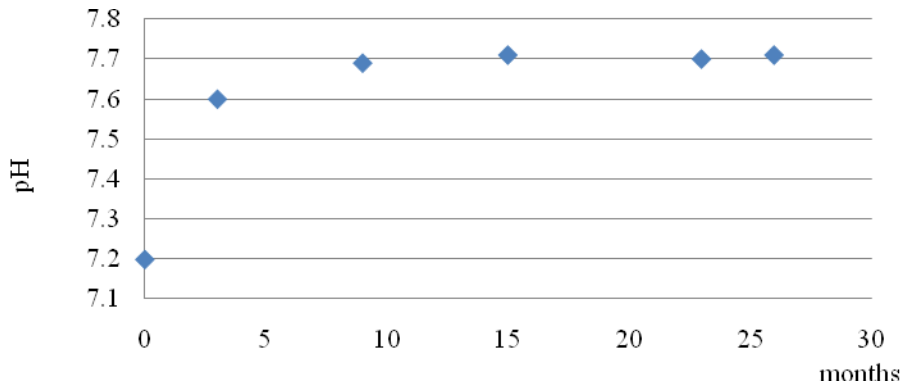


Fig. 9. – Dynamics of pH in medium of modified bottom sediment at the experimental site.

In our opinion, the sediments at the experimental site have remained slightly alkaline for a long time is due to the large number of shells found there. A large number of shells in the bottom sediments is a characteristic of CP ChNPP. According to published data, up to 20 kg per 1 m^2 of zebra mussel can be found in some areas of CP ChNPP. This factor may affect directly the dynamics of radionuclide leaching from the fuel particles, and as a result of it this factor may influence mobility and migration properties of radionuclides on the drained areas of bottom, especially in the early years.

The results of the sequential leaching of ^{137}Cs and ^{90}Sr from the samples of bottom sediments from the ChNPP cooling pond, selected after drainage at point T1 (Fig.6) are shown in Figure 10.

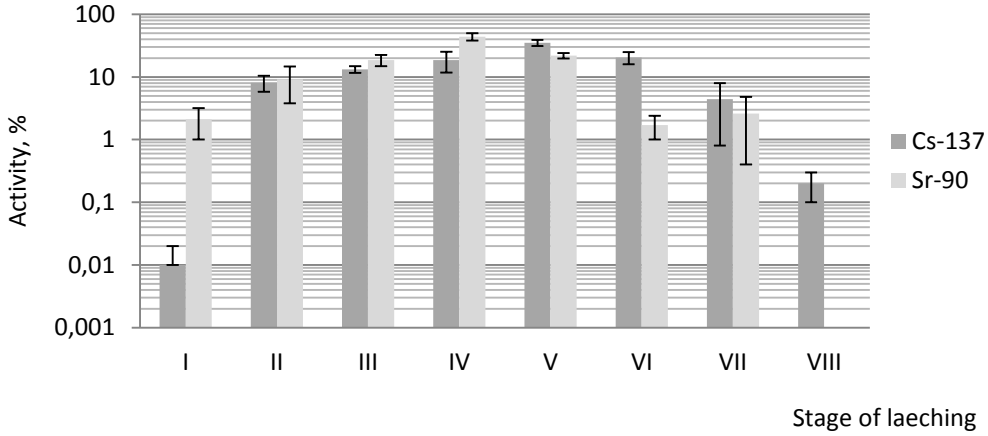


Fig. 10. Activity of ^{137}Cs and ^{90}Sr in leaching fractions of bottom sediments of the ChNPP cooling pond after the draining of it (point T1).

About 10 % of the activity of ^{137}Cs and ^{90}Sr was transferred in acetate extract (stage II of leaching, table 1). These results differ from the results obtained for samples of bottom sediments collected in the deep part of the ChNPP cooling pond (fig. 8 for t=0). It can be caused by the different content of organic matter in the samples. In the samples from T1 organic matter is almost absent (pure sand), but in samples from experimental site the content of organics reached 30 % of sample weight. Perhaps, this fact indicates the spatial heterogeneity of radionuclide forms in sediments of the ChNPP cooling pond.

X-ray autoradiographs of samples of bottom sediment core from different soil layers which was sampled in the point T1 (depth of 0-25 cm) are shown in Figure 11. As it is shown in Figure 11, the main part of the fuel contamination is concentrated in the 9 - 20 cm layer. The dispersed distribution and the distribution of ^{90}Sr activities depending on the size of the fuel particles for layered samples are shown in Figure 12 (according to the results of x-ray autoradiography).

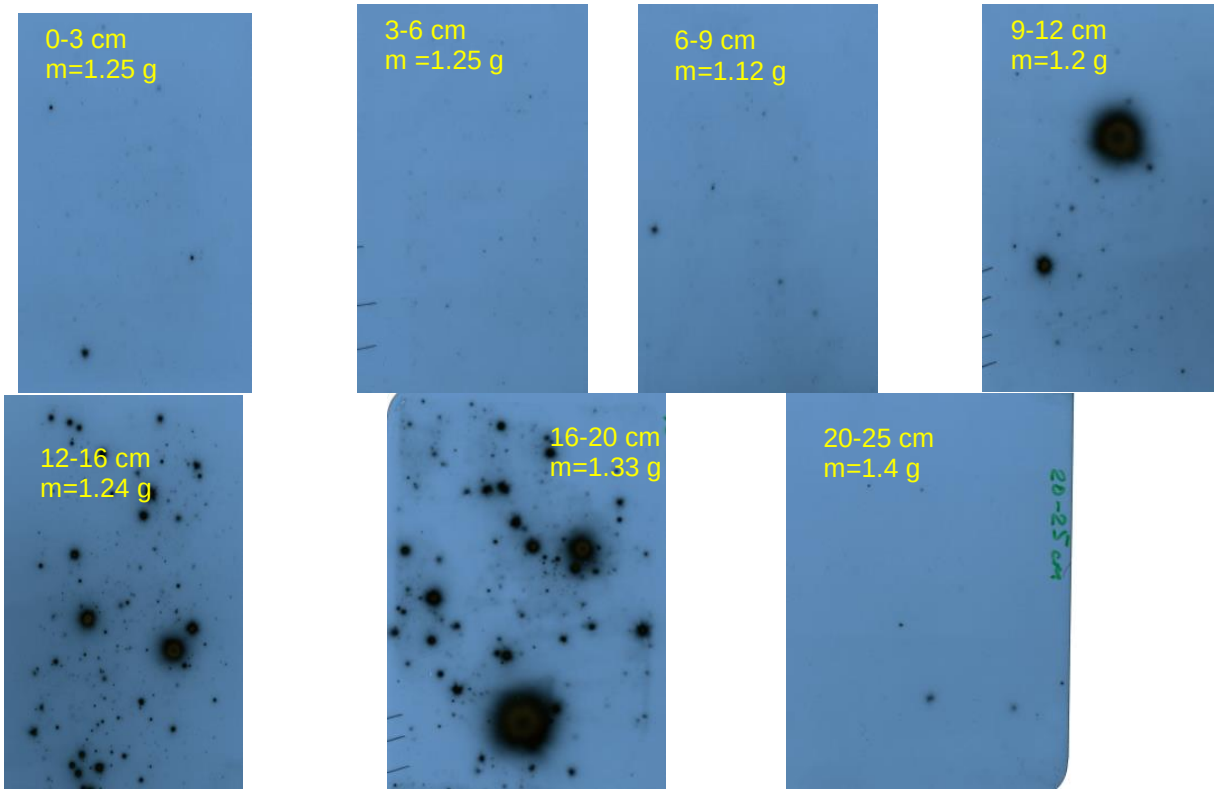


Fig. 11. X-ray autoradiography of the vertical profile of bottom sediments (0-25 cm) at the drained bottom of the CnNPP CP (point T1).

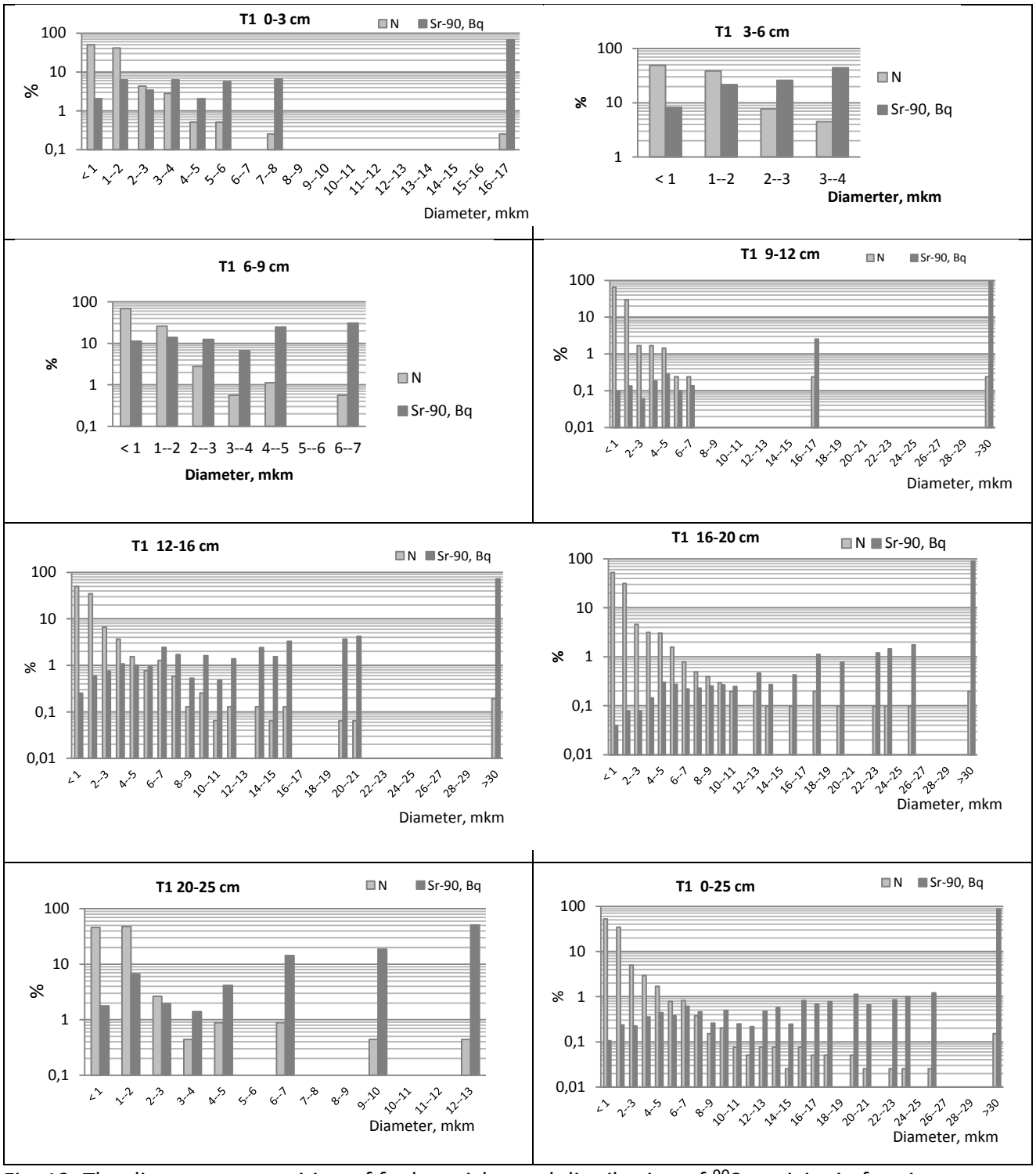


Fig. 12. The disperse composition of fuel particles and distribution of ^{90}Sr activity in fractions depending on size of fuel particles (according to the results of x-ray radiography).

Analysis of the received information indicated that the main part of the ^{90}Sr activity (> 80%) is concentrated in relatively large fuel particles (diameter > 20 μm), although the amount of large fuel particles is about 1% of total amount.

Currently, works on the selection of individual fuel particles for morphological studies and sequential leaching are being carried out.

Discussion and conclusions

According our results, the predominant part (> 98 %) of ^{137}Cs, $^{239+240}Pu$ and ^{241}Am in sediments of CP ChNPP, after drying and 26 month exposure to natural conditions at the experimental site, is still in a non-exchangeable forms (stage of leaching III-VIII). The content of ^{90}Sr exchangeable species may reach 10 %. Probably, the low rate of radionuclides leaching from modified sediment of CP ChNPP is caused by a prolonged slightly alkaline pH in the medium. In turn, this is caused by the presence of a large number of remnant shells of zebra mussels in bottom sediments of CP ChNPP. It can be stated that a sharp increase of radionuclide mobility should not be expected on the newly exposed sediments. Thus, a significant increase of radionuclide mobility in the sediments, associated with beginning of the destruction of the fuel particles after drying and 26 month exposure to natural conditions, was not observed. It contradicts the existing forecasts. According to data [*A. Bulgakov et al., 2009*], 3 years after the sediment exposure about 10% of the fuel particles would dissolve, and pH of exposed sediments would be 6.5. But this was not observed.

A significant part of the radionuclide activity was not removed from the solid phase of the bottom sediment in the CP ChNPP, even under "extra strong" extraction conditions (stage of leaching V-VII, table 1). Probably, this fact indicate that part of radionuclides are kept in the chemically very stable form of particles, and for this reason these radionuclides will not be transferred into the environment for many years. A significant part of ^{137}Cs was deposited to the ChNPP CP in the form of volatile compounds after its release from the nuclear fuel during the accident, due to the high temperatures. Because of the low energy of hydration, part of

radiocesium could be firmly bound in the lattice of clay minerals of the bottom sediments. This part of radiocesium was not leached in the sequential extractions.

It is possible to estimate radionuclide content of FP in the bottom sediments of CP ChNPP, if it is assumed that radionuclides leached during the sequential extraction procedure on I + II + III + IV stages are associated with the various components of soil absorptive complex, and, therefore they represent a part of FP dissolved in the bottom sediments. Summarized content of ^{90}Sr, $^{239+240}Pu$ and ^{241}Am in extracts of I + II + III + IV stage of leaching (fig. 8, for t=0) is presented in Table 2.

Table 2. Relative content of radionuclides in I + II + III + IV fractions of leaching, percentage of the content of the certain radionuclide of a total amount of radionuclides in a sample of bottom sediments of CP ChNPP (fig. 8)

^{90}Sr	$^{239+240}Pu$	^{241}Am
% activity in fractions I + II + III + IV of leaching		
25 ± 5	10 ± 2	17 ± 2

Thus, it can be supposed, that at the present moment, from 70 to 90 % of radionuclide activity in the bottom sediments of CP ChNPP are still associated with fuel particles.

Taking into account that ^{90}Sr, ^{241}Am and plutonium isotopes, in contrast to ^{137}Cs, were not fractionated scarcely in nuclear fuel during the accident, and during fallout they were associated with fuel matrix, dynamics of their leaching at the sequential extractions must be similar. The obtained results revealed that behavior of ^{90}Sr and ^{241}Am during leaching are similar but differ from $^{238,239,240}Pu$. Perhaps, it indicates heterogeneous distribution of radionuclides in FP or selectivity of their leaching from the whole volume of FP. For a certain stage of leaching (I + II + III + IV) similar behavior of ^{90}Sr and ^{241}Am is explained by the following statements: firstly, strontium and americium are presented in the form of hardly soluble salts of oxalic acid, and, secondly, strontium and americium are less disposed to form complex compounds with components of the soil absorptive complex, than plutonium.

The obtained results indicate the spatial heterogeneity of forms of radionuclides in sediments of the ChNPP cooling pond. More detailed studies of this phenomenon should therefore be performed.

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Annex

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