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COLLOID TRANSPORT OF PLUTONIUM IN WATER BODIES IN THE SEMIPALATINSK TEST SITE

A.S. Toropov

Scientific advisor professor L.P. Rikhvanov

National Research Tomsk Polytechnic University, Tomsk, Russia

The consequence of nuclear power facilities operation, accidents at nuclear fuel cycle enterprises, nuclear weapons tests was result of the presence in the environment of technogenic radionuclides, particularly transuranic isotopes, which are able to make a substantial contribution to the total radioactivity of natural objects [1]. It is necessary to take into account the duration of transuranic elements decay, the half-life of them reaches dozens of thousands years, and exposure to biosphere very longtime.

The actual rate of migration such elements as Pu, Am, Eu, etc. with groundwater significantly higher than the predicted using thermodynamic calculations that took into account sorption on host rocks, complexation in solution and the formation of slightly soluble compounds [1, 22]. In [4, 8] it is emphasized that the description of plutonium migration with groundwater is not possible without considering the contribution of the colloidal particles in the process. Colloidal particles are essential to mobility of radionuclides in groundwater and surface systems, but systematic research of physical and chemical forms of the actinides in natural waters quite a bit.

Underestimating the "colloidal transport" mechanism of these radionuclides has resulted to mistakes in the calculation of the plutonium migration rate in groundwater at the Nevada Test Site [3, 5]. Despite the significant role of colloids in the migration of some radionuclides, yet there is no a unified model of migration behavior of radionuclides, which takes into account the role of colloidal particles [4, 6, 10]. Scientists in this field highlights the lack of reliable experimental data (both real and for model systems), as well as the lack of understanding of the mechanisms of formation of stable colloidal particles and regularities occurring on their surface sorption reactions [4].

In the 2000s, the fragmentary studies of radionuclide content and form of their occurrence in water bodies of the Semipalatinsk test site within the various programs and projects were carried out [9, 12].

The authors studied the radionuclide content and forms of occurrence of plutonium and uranium in well water of the village "Sarzhai", located near the borders of the nuclear test site, wells of wintering in the "North" part of the Semipalatinsk test site; stream Uzynbulak, Shagan River and lakes Telkem-1 and Telkem-2 [9]. They distinguish the following forms of speciation: suspended matter, oxidized forms of uranium and plutonium (oxidation state +5, +6, total), reduced forms of plutonium uranium (oxidation state +3, +4 and total) by the method "limited iron" technique and sequential precipitation reduced and oxidized forms. According to the authors of [9, 12] in the water from funnel of explosion "Telkem-2" from 89 to 98% of plutonium was in a state of Pu (III, IV), the remaining part, they believed, was in a state +6. High proportion of reduced forms they associated with the presence in water a large amount of dissolved organic matter.

The reports of of the Radium Institute named Khlopin has unpublished research (2006) under Professor Y. Dubasov, where they studied the water of the tunnel creek an object D2 territory of the ground "Delegen". It was found that the percentage of plutonium in a fraction of the pseudo-colloids is between 9 to 20%, the proportion of plutonium in the +6 state ranged from 7 to 11%. The main plutonium fraction (70-80%) is in form Pu (III, IV).

The aim of this work is the study of the speciation of plutonium in the water bodies of the Semipalatinsk test site.

The selection of the objects for the study of the forms of radionuclides in the water is based on literature data on the content of radionuclides in the water bodies of the Semipalatinsk Test Site [1]. The objects of this study were watercourses of portal areas of tunnels 176, 177, 503, 504 and 511 at "Delegen" ground of the Semipalatinsk Test Site.

The volume of water samples ranged from 2 to 10 liters. The water sample was collected into clean polyethylene containers, avoiding stirring up of sediments and inclusion any extraneous impurity, and then filtered through a cellulose filter with a pore diameter of 5-8 microns on place or within 24 hours of sampling in the laboratory.

Then, the sample was divided in two parts, one part of the sample was acidified and concentrated with nitric acid to pH = 2 and co-precipitation of iron hydroxide was carried out. Another part was filtered through a membrane polyethylene terephthalate filter with a pore diameter of 0.2 microns, permeate was acidified whereupon added isotope tracers and concentration in a similar way was carried out. Thus, this sample preparation allowed arbitrarily distinguishing the following forms of occurrence: "suspended matter", "colloidal substances" and "true-soluble form".

Basic parameters of the tunnel water streams "Delegen" test ground composition are presented in Table 1.

According to the general chemical composition, tunnel streams are predominantly fresh and slightly mineralized waters (Tun. 504), in anion-cation composition – for tunnels 176, 177, 511 watercourses bicarbonate and calcium ions are dominant, and for tunnels 503, 504 streams dominant ions are potassium, magnesium and sulfate.

It has been established that plutonium activity in the studied water bodies ranged widely – from $n \cdot 10^{-2}$ to $n \cdot 10^0$ Bq/l (Table 2).

Table 1

Chemical composition of «Degelen» test ground water bodies

Name of the object	pH	Mineralization	Na ⁺ +K ⁺	Ca ²⁺	Mg ²⁺	Cl ⁻	HCO ³⁻	SO ₄ ²⁻
Tunnel 176	7,7	170	7,5	45	5,1	5,9	120	42
Tunnel 177	6,4	400	31	76	17	9,8	210	120
Tunnel 503	6,8	430	22	90	17	9,3	120	220
Tunnel 504	5,9	1100	54	160	100	14	4,9	870
Tunnel 511	6,4	410	23	72	22	6,5	270	37

Note: mineralization and the main measuring ions unit - mg/l.

Table 2

Activity of Pu isotopes in water bodies of Semipalatinsk test site separated by cascading filtration, Bq/l

№	Name of the object	Suspended matter	Colloidal substance	Dissolved
1	Tunnel 176	0,11±0,02	$7,3 \cdot 10^{-2} \pm 1,4 \cdot 10^{-2}$	$2,7 \cdot 10^{-2} \pm 0,9 \cdot 10^{-2}$
2	Tunnel 177	0,13±0,02	$4,3 \cdot 10^{-2} \pm 1,0 \cdot 10^{-2}$	$9,7 \cdot 10^{-2} \pm 1,8 \cdot 10^{-2}$
3	Tunnel 503	2,2± 0,7	1,1 ± 0,1	4,8 ± 0,1
4	Tunnel 504	$3 \cdot 10^{-2} \pm 1,3 \cdot 10^{-2}$	$1,8 \cdot 10^{-2} \pm 0,6 \cdot 10^{-2}$	$2,7 \cdot 10^{-2} \pm 0,9 \cdot 10^{-2}$
5	Tunnel 511	$< 6,5 \cdot 10^{-3}$	$2,2 \cdot 10^{-2} \pm 0,7 \cdot 10^{-2}$	$< 4,2 \cdot 10^{-3}$

The maximum content of plutonium isotopes in natural waters of the Semipalatinsk test site identified at the level of 8.1 Bq/l (the sum of all forms of speciation) in water stream from tunnel 503, including the amount of colloidal and dissolved forms - 5.9 Bq/l. This is more than an order of magnitude higher than the action level set by hygienic regulations [11]. In other water sources the maximum permissible level exceedance has not been found.

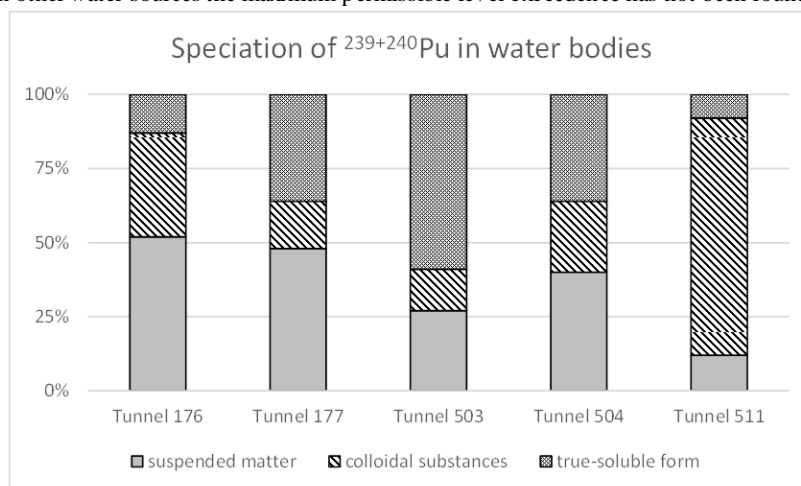


Fig. Speciation of Pu isotopes in water objects of the Semipalatinsk Test Site, %.

It is well known that plutonium has a complex chemical behavior in natural waters, tends to hydrolysis, coagulation, complexation with mineral and organic ligands, colloid formation, and change in the degree of oxidation, and accordingly, the change in the form of speciation as a result of even minor fluctuations of water composition [2, 4, 7].

The distribution of the forms of occurrence of plutonium has been uneven (Fig. 1). It is noted that the radionuclide is present in the water in all forms studied. Depending on the water source, the proportion of suspended forms ranges from 27% to 52.4%, plutonium, associated with colloidal substances - from 13 up to more than > 80%, and dissolved forms - from 13 to 59%.

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INTEGRAL ANALYSIS OF GEOLOGICAL AND FIELD DATA FOR SELECTION OF OILFIELD DEVELOPMENT STRATEGY

K.V. Tsivelev, A.A. Milke

Scientific advisors associate professor O.S. Chernova

National Research Tomsk Polytechnic University, Tomsk, Russia

The purpose of the research is selection of optimum development system and economic calculations for studied oilfield A.

The oilfield A is located in the Kaymysovky oil region next to Dvurechenskoe and Krapivenskoe oilfields. There are 9 exploration wells drilled in field A, five of which have core data. The base properties, such as porosity, permeability and water saturation, are determined on the basis of core data. As the main material the following logs data are used: gamma ray log (GR), spontaneous potential log (SP), resistivity logs, neutron logs, potential logs, and so on. There were 270 samples taken from core of different production intervals. All data were analyzed separately for two production intervals: U_{1-2} and U_{1-3-4} . The properties distribution was constructed on the basis of core data.

First of all, the intervals, which could be potential sources of hydrocarbons, were determined on the basis of SP and GR logs for all wells [3]. The shaliness should be calculated for the effective porosity calculations. Shale normally contains radioactive bearing minerals and gamma ray log could be used for shale identification. Shaliness was analyzed using different models, the best result was achieved by the Larionov model. On the basis of this method the effective porosity curve was calculated for each well. The comparison of core porosity and log porosity for U_{1-2} and U_{1-3-4} was made separately. Then on the basis of the «base well» concept, the effective porosity curves were built for wells without core data. The «base well» was determined using the following criteria: wells lithology similarity and the lateral distance between the wells (well A2 was chosen).

The permeability was measured with the use of nitrogen gas, so the core permeability data were corrected on the Klinkenberg effect, provided that there was a slippage effect of gas molecules along the grain surface. Then the relationship between porosity and permeability was calculated based on core data. The effective porosity curves were used for this reason. It was decided to use unique relationship for each development object. Despite the fact that the exponential type of correlation was obtained, the determination coefficient was high. This is due to log and core porosity similarity. The average log derived parameters for U_{1-2} : $\varphi=15\%$, $k=7.7$ mD, $S_w=0.48$; for U_{1-3-4} : $\varphi=17.2\%$, $k=176$ mD, $S_w=0.43$. The average core derived parameters for U_{1-2} : $\varphi=13.2\%$, $k=8.8$ mD, $S_w=0.56$; for U_{1-3-4} : $\varphi=18\%$, $k=162$ mD, $S_w=0.37$.

One of the main objectives of property estimation is correct calculation of Stock Tank Oil Initially in Place (STOIIIP). However, additional step should be made in order to eliminate the values which are of minor importance for reservoir field development. The criteria for this elimination include several steps: water saturation, porosity, permeability and shaliness cut-off criteria. The cut-off criteria are estimated separately for each productive formation. The cut-off criteria are defined for U_{1-2} : $S_w=0.76$, $\varphi=0.116$, $k=0.88$ mD, $V_{sh}=0.24$ and for U_{1-3-4} : $S_w=0.73$, $\varphi=0.1202$, $k=1.06$ mD, $V_{sh}=0.27$.

The interpreted parameters such as porosity, lithology logs and well picks of formation boundaries were used as an input data. The structures of geological model were constructed by offsetting Bazhen bottom structure.

The next step was to build up of 3D structural grid. Using modeling software, separate grids were constructed for U_{1-2} and U_{1-3-4} . Geological model was build using cell size of 100 m by 100 m for U_{1-2} and U_{1-3-4} . The size was chosen to optimize calculating time as well as obtain accurate model. The number of layers were selected so that the model fully describes vertical heterogeneity typical for regional depositional environment. The STOIIIP (Stock Tank Oil Initially In Place), which estimated by geological model, was $U_{1-2} = 7.92$ mln m and $U_{1-3-4} = 34.81$ mln m.

The lithology was distributed by means of indicator modeling. In terms of lateral trends variogram from similar surrounding deposit was used and vertical lithology was distributed by vertical proportional curves for each layer separately. Variogram parameters for lateral distribution were: azimuth – assumed direction of sediment deposition, long section rank – 4000 m, cross-section rank – 2000 m. Vertical variogram parameters: 1 m for U_{1-2} and up to 3 m for U_{1-3-4} . In terms of input data the pointwise interpretation of porosity log was used and then it was scaled into cells. Then the porosity parameter was distributed by kriging interpolation method. The same azimuth of variogram was used for lithology distribution.