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THE SUMMARY OF RESEARCHES OF CLAYS AND CRYSTALLINE ROCKS AS GEOLOGICAL ENVIRONMENTS FOR RW REPOSITORIES

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Engineering-geological and hydrogeological problems of the safe RW disposal in clays and gneiss host-rocks are analyzed considering similar investigations at several foreign facilities. Varieties of the fundamental physical parameters describing the transport of radionuclides by groundwater ("far field") during and after-operation period are presented. The results of the geomigration modeling for radionuclides and hydrogen (as corrosion product of the isolating steel containers) in the aqueous and gas phases are discussed assuming elevated temperatures in rock massif.

Keywords: *radioactive waste, physical properties of rocks, filtration of underground waters, migration of radionuclides and gas phase.*

Introduction

The significant economic costs and the increase of environmental risks related with the storage of "legacy" RW and its accumulation at the nuclear facilities have led to approval in 2011 of a new concept of RW management, formulated as a federal law. According to this concept, the temporary storage of RW should be superseded by the final disposal of RW.

For the intermediate and low-level waste of 3 and 4 classes, the most economically reasonable option is a construction of near-surface disposal facilities. For the high- and intermediate level waste (1^{st} and 2^{nd} classes), the most promising option being considered is a construction of RW disposal facilities in clay and crystalline rocks located in deep geological environment.

The national research programs of radioactive waste management in Belgium, France and Switzerland are focused mainly on clay rock formations (Rupelian Boom Clay, Callovo-Oxfordian clayey siltstone/silty clay and Aalenian Opalinus Clay), which are planned to be used as host-rocks for RW repositories (Table 1).

It is obvious that the clay formations are represented by consolidated and overconsolidated (up to argillites) clays that can be used for disposal at a depth of several hundred meters. These intervals have been mined to construct shafts where underground research laboratories (URL) have been organized [1—4]. Table 1 shows general geological features of Vendian (Kotlin) clays, which are widespread in northwest of Russia. The intensive investigations of these formations have started in 2007—2014 and included drilling a net of research wells.

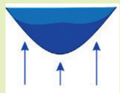
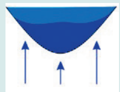
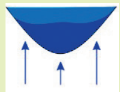
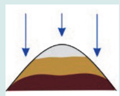
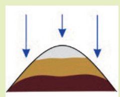
The most significant results were obtained from the study of crystalline granite-gneiss rocks in Sweden (Stripa Mine, Äspö Hard Rock Laboratory), Finland (Olkiluoto Research Tunnel) and Switzerland (Grimsel Test Site), where URLs were also constructed at the depth intervals supposed for long-term disposal facilities for RW and SNF (Table 2) [4—7].

These facilities are mainly situated in areas of the groundwater discharge to the sea. In this case the engineered barriers play the main role for the protection of repository, while the host-rocks

Table 1. Geological features of sites proposed for disposal of RW and construction of URLs in clay formations

Country	Belgium	France	Switzerland	Russia
Name of formation	Boom Clay (BC)	Callovo–Oxfordian (COx)	Opalinus Clay, Mont Terri (OPA)	Kotlin clays (Vkt)
Location	NRC Mol/Dessel	Meuse/Haute-Marne	Western Switzerland	Sosnovy Bor, North-West of Russia
RW type	ILW, HLW, LLW	HLW	HLW, LLW	LLW, ILW
Depth, m	> 200	450—500	400—800	70—100
Conception URL, wells	URL HADES, since 1984 (-223 m)	URL Bure, since 2004 (-490 m)	URL Mont Terri since 1996	Wells, since 2007
Age, mln years	29—33	152—158	180	530—650
Geological period	Lower Oligocene (Rupelian)	Medium Callovian — lower Oxfordian	Lower and medium Aalenian	Vendian
Lithological description	Consolidated gray clay	Consolidated argillites	Overconsolidated clays, argillites	Clays, argillites

Table 2. Geological features of sites proposed for disposal of RW and construction of URLs in crystalline rocks

Site/Contry	Host rock	Rock age	Depth, m	Purpose of the section	Hydrodynamic zone
Forsmark Sweden	Metamorphized biotite granites	1.8—1.9 bln years	500	HLW, SNF	
Äspö Sweden	Granitoids, low crustalline granites	1.6—1.7 bln years	200—460	URL only	
Olkiluoto Finland	Migmatized gneiss, pegmatite granites	1.8—1.9 bln years	450	URL, HLW, SNF	
Grimsel Test Site Switzerland	Granites crossed by minor intrusions of aplites and lamprophyres	300 mln years	450	URL only	
Yeniseysky Site, Russia	Gneiss crossed by minor intrusions of dolerites	1.8—2.5 bln years	400—500	URL, HLW, MLW	

serve only as the surrounding environment; the dilution of radioactive components by seawater is the additional factor for the environmental safety.

The Russian analogue of European projects of deep underground repositories is the project of a deep RW disposal facility (DRWDF) in Nizhne-Kansky gneiss rock massif (Table 2).

The location of the facility at the watershed area gives a certain advantage, because the host-rock will contribute to the protective function of contamination retention in addition to the engineered barriers, as described below.

During the development of such underground repository one of the key area is the long-term safety case that is largely determined by geomigration processes in the underground hydrosphere. Therefore, a special attention in international practice is paid to development of hydrogeological models. Moreover, the reliability of the predictive models is largely dependent on the completeness and quality of the input information: the structure of the flow, the spatial distribution and the character of faults, flow and migration (with respect to radioactive solutions and gas phase) properties of rocks, etc. [8].

Implementation of the RW management concept (adopted in Russia in 2011) takes into account the modern conditions. Moreover, it is possible to declare that there are some advances of researches of clay and crystalline formations and implementation of theoretical and experimental approaches to develop long-term safety cases for such engineering facilities. This conclusion is based on the analysis of survey, design and researches: 1) in the North-West Region of Russia with widespread

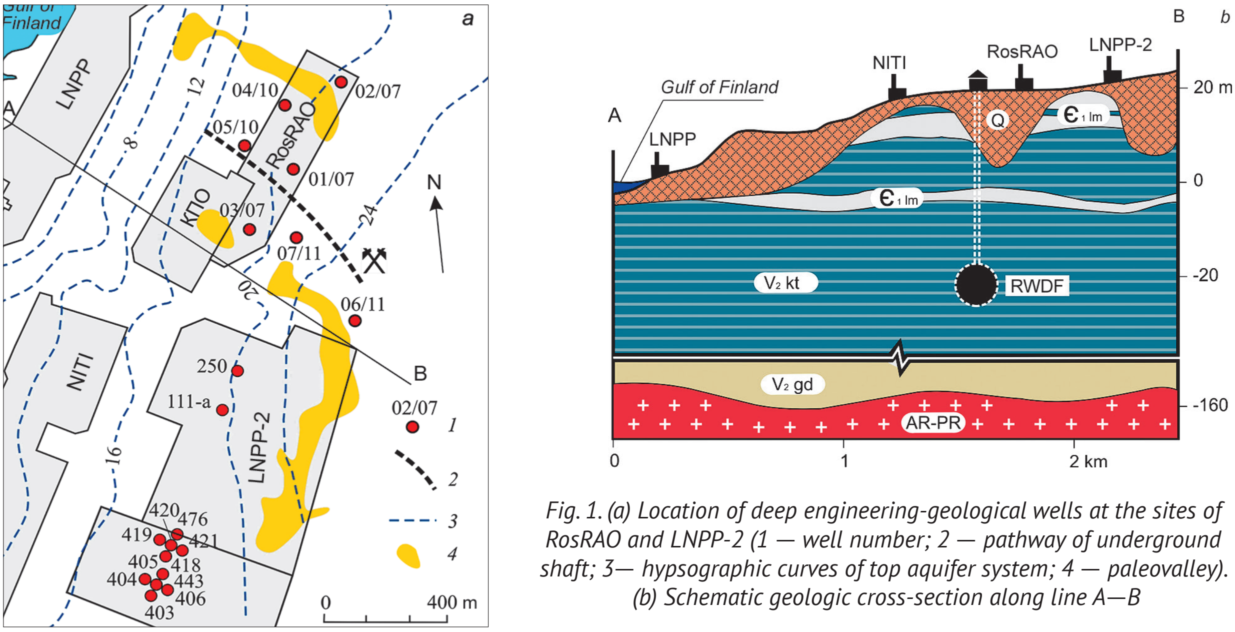


Fig. 1. (a) Location of deep engineering-geological wells at the sites of RosRAO and LNPP-2 (1 — well number; 2 — pathway of underground shaft; 3— hypsographic curves of top aquifer system; 4 — paleovalley). (b) Schematic geologic cross-section along line A—B

Vendian and Cambrian clay formations, 2) in the Krasnoyarsk region, where Achaean and Proterozoic granite-gneiss rocks are observed. The present article illustrates only the small part of problems related to parametric and model simulation of long-term safety of RW disposal in two types of environments — clays and crystalline rocks representing the experience of the Institute of Geoecology of RAS working in collaboration with Russian and foreign organizations supported by enterprises of State Corporation "Rosatom" — FSUE "NO RW" and FSUE "RosRAO".

1 RWDF in Vendian clays (Sosnovy Bor Distrit of the Leningrad Region)

Lithified clays are widespread in the North-West of the Russian platform. They include Kotlin clays (V_2 kt) of the Vendian system crossed by blue lower Caembrian clays ($\epsilon_1$ln).

Clays have been discovered south of the Gulf of Finland, within the Karelian Isthmus and in the vicinity of St. Petersburg. In particular, clays are underlying such facilities as LNPP and LNPP-2 as well as near-surface SRW storage facility of RosRAO in the vicinity of Sosnovy Bor (Fig. 1) [9].

At the pre-design stage of the construction of a near-surface (up to 100 m) RWDF the following rock properties have been investigated [9]: (1) properties affecting the geotechnical conditions of construction and operation of the facility; and (2) properties controlling the safety of the facility from the point of view of radiation impact on groundwater and neighboring environment. Most of the data was acquired from research well-drilling (up to 180 m) (Fig 1a). In order to identify ancient buried valleys additional geophysical studies were performed (Fig 1b). Moreover, considerable laboratory studies were performed.

1.1 Hydrophysical and physical-mechanical properties

Results of core analyses demonstrated the clear regularity in the spatial distribution of physical and mechanical properties of clay formation [10]. Correlations shown in Fig. 2a demonstrate that the upper zone (I) of the cross-section (up to depths of 40—50 m) is composed by moist and deconsolidated rocks.

The lower zone (II) is represented by less dense and less moist rocks.

Stabilometrical tests demonstrated a clear trend: the hardness of rock increases with the depth (Fig. 2b). Deformation properties of rocks also change with depth. Thus, the increase of the total stress-strain modulus (Fig. 2c) indicates the decrease of the compressibility of rocks at large depths (zone II) compared to near-surface zone (I). These data confirm the trends identified earlier.

The identified spatial variability of parameters allowed selection of the reasonable depths for underground construction.

1.2 Flow and solute transport properties of clays

The hydraulic conductivity of Kotlin clays was investigated by using triaxial compression meter WF-50 at monoliths oriented in two perpendicular directions.

The interval studies (Table 3) demonstrated that Kotlin clays have fairly low permeability properties. The hydraulic conductivity across the bedding is in the range from $5\cdot10^{-7}$ to $2\cdot10^{-5}$ m/d, while the hydraulic conductivity along the bedding is from $4\cdot10^{-6}$ to $2\cdot10^{-4}$ m/day. Thus, the studied parameter has a strong anisotropy (up to 16), which is explained by the fine banding of clay sediments.

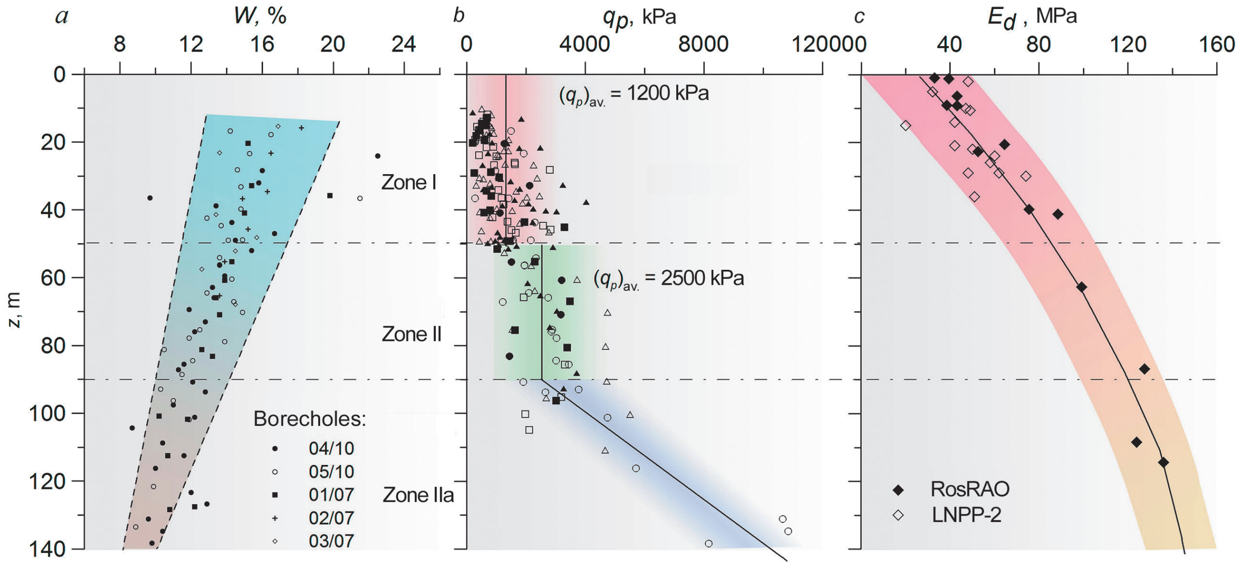


Fig. 2. Change of mositure (a), deviatoric tension (b) and total stress-strain modulus (c) with depth

Diffusion experiments with radionuclides for three various models (single chamber, two chamber, 3D) provided estimates for diffusion coefficients of Kotlin clays (Table 4) at the proposed location of RWDF. Obtained values are in good agreement with the results of foreign studies for conditions of similar facilities [3].

Anisotropy in diffusion coefficient of the clay has been confirmed experimentally. The species diffuse up to 2—6 times faster along the strata, than across it, due to fine banding structure of Kotlin clays [13].

The obtained effective diffusion coefficients (De) for various radionuclides were arranged in the order: tritium > Sr-90 > Cs-137 > Co-60. The corresponding values of De are as follows: $3.6{\cdot}10^{-10} > 2.3{\cdot}10^{-10} > 6.6{\cdot}10^{-11} > 3.0{\cdot}10^{-11}$ m²/s.

Clay samples collected from various depths have been used for sorption experiments with Sr-90, Cs-137, Am-241, Pu-239, -240 [14]. The obtained ranges for the sorption distribution coefficient Kd differ substantially for various radionuclides (Fig. 3). Also, it was clearly seen that K_d values were significantly lower for samples from the lower zone of the cross-section (deeper than 100 m), represented by Vendian sandstone. The sorption is the lowest for Sr-90, and the highest for Am-241 and Pu-239, -240; Cs-137 has intermediate values.

The performed studies allowed developing a system of numerical models. Simulation of the migration processes in the framework of safety case development has allowed assessing the spatial-time scales of diffusion in clay mass in the post-operation period (regular operation of RWDF), and the map of contamination within the Vendian layer (emergency scenario connected with loss of integrity of rock mass).

In both cases, the impact is within the acceptable limits [15].

1.3 Comparison of vendian clays (Vkt) with clay formations in western Europe (BC, COx and OPA)

Studies performed by several organizations in Western Europe and studies in Sosnovy Bor allow

Table 3. SHydraulic conductivities of Kotlin clays

Item	Well No.	Depth, m	kII, m/day	k, m/day	Anisotropy
1	5/10	48.6–49.0	$6.00{\times}10^{-6}$	$1.30{\times}10^{-6}$	4.6
2	4/10	51.8–52.1	$4.00{\times}10^{-6}$	$6.00{\times}10^{-7}$	6.7
3	1/07	55.0–55.25	–	$5.00{\times}10^{-7}$	–
4	1/07	65.0–65.25	–	$5.00{\times}10^{-7}$	–
5	4/10	69.2–69.5	$1.90{\times}10^{-5}$	$1.40{\times}10^{-6}$	13.6
6	7/11	70.3–70.6	$2.17{\times}10^{-5}$	$1.30{\times}10^{-6}$	16.7
7	7/11	94.0–94.25	$2.20{\times}10^{-4}$	$1.96{\times}10^{-5}$	11.0

Table 4. Average diffusion coefficients (D_e), м²/s

Type of experiment	3D experiment	Single chamber cell	Two chamber cell
Cl-36	$5.91{\times}10^{-10}$	–	$1.40{\times}10^{-10}$
H-3	–	$3.05{\times}10^{-10}$	–
Sr-90	$4.54{\times}10^{-10}$	$1.51{\times}10^{-10}$	–
Co-60	$4.60{\times}10^{-11}$	$3.65{\times}10^{-11}$	–
Cs-137	–	$6.56{\times}10^{-11}$	–

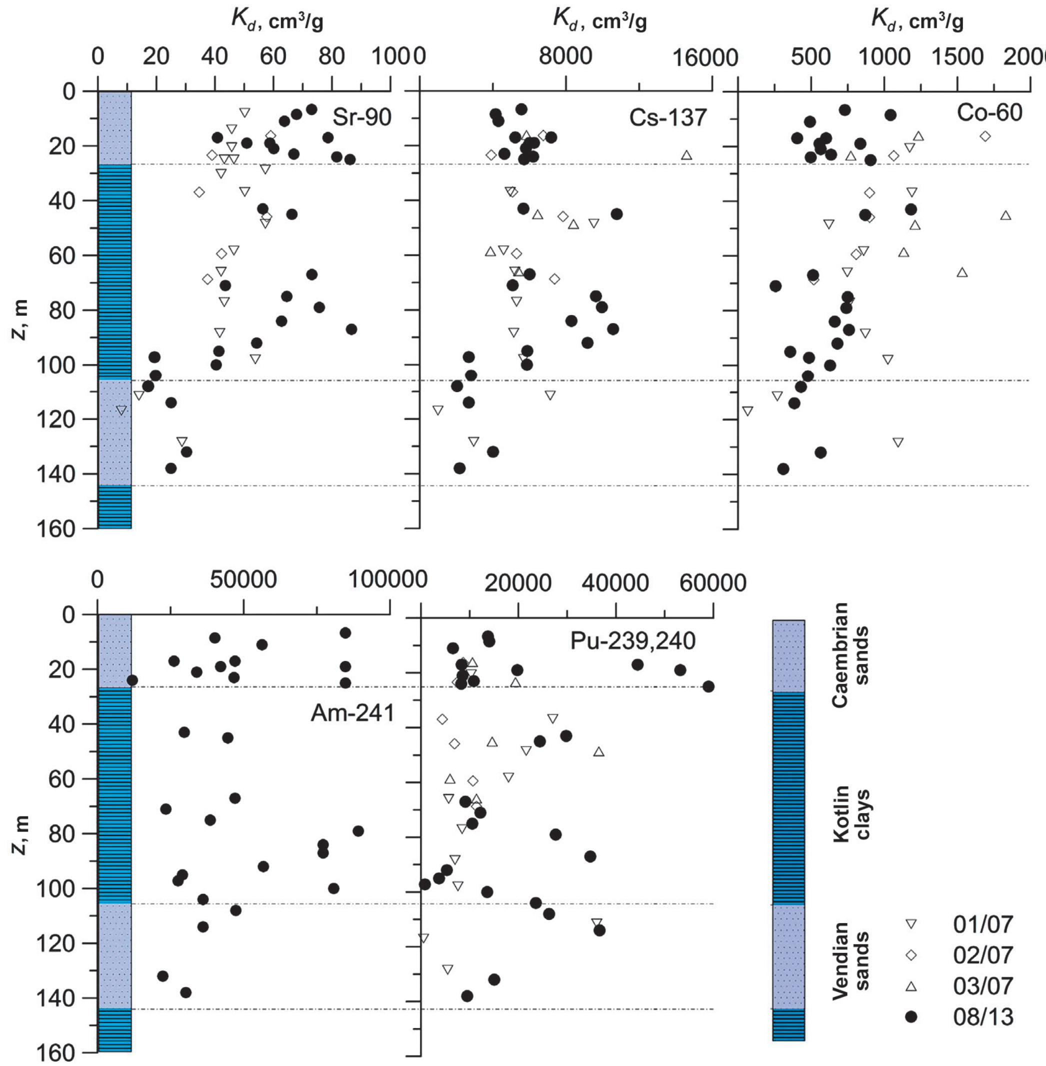


Fig. 3. Variations of K_d (cm³/g) along the depth, Z, m, of clay mass. Dotted line shows the zone of sandy clays

concluding that clays (Table 1) are suitable for the disposal of radioactive waste due to:

- small diffusion coefficients;
- the absence of water flows around the waste is determining slow leaching rate;
- the predominant radionuclide transport mechanism is a molecular diffusion;
- favorable physico-chemical conditions for radionuclide retention (low solubility);
- high sorption/retention capacity;
- “self-sealing” features, i.e. closure of fractures and cavities (swelling of clay materials).

The following common properties have been identified in the experiments and comparative analysis of published materials for the four formations Vkt, BC, COx and OPA:

- similar mineral composition;
- high sorption capability and exchange capacity;
- neutral and weakly alkaline chemical medium;
- predominantly diffusion transport.

Physical, chemical and sorption properties of clay are favorable for retention of radionuclides. Slow migration of radionuclides into the biosphere during long time period is typical for clays.

The following differences have been identified in experiments and comparative analysis for the four formations Vkt, BC, COx and OPA:

- porosity and moisture of clays reduce depending on density in the row Vkt >> BC >> OPA > COx;
- carbonate content reduces in the row COx > OPA >> BC, Vkt;
- total salt content reduces in the row OPA > COx > Vkt > BC;
- geomechanical properties (hardness and resistivity): COx (solidified) > OPA >>> BC (plastic) = Vkt (solidified, non-plastic). Strength of Vkt is lower than that of COx and OPA;
- small hydraulic conductivity (decimal order in m/s given in brackets): Vkt ~ BC (−12) > OPA (−13) > COx (−14).

Therefore, it is safe to say that most of the parameters of Kotlin clays correspond to the “standard” clay rocks considered in the Western Europe as the environment for RW repository.

2 DRWDF in granite-gneiss host-rocks (Yeniseisky site, Krasnoyarsk region)

Since the beginning of the 1990-s Nizhne-Kansky granitic-gneiss massif has been considered as a promising site due to its isolation properties. This massif is located at the edge of two major Asian geological structures — Western Siberian and Eastern Siberian plates. Various geological investigations were performed at several sites between 1992 and 2005. The Yeniseisky site was considered as promising for the future research. Comprehensive research at this site was started in 2008 by JSC "Krasnoyarskgeologiya" [16].

The overall distance of research wells (600—700 m each) has exceeded 7.5 thousands meters; water was withdrawn from hydrogeological wells (every 50 m) — more than 180 experiments; borehole drilling included a complex of such studies as flow measurements and resistivity measurements. Site-specific (surface) geophysical works included magnetometeric, resistivity measurements using VES and seismic analyses.

The site is located on the watershed of two rivers — Yenisei and Kan.

DRWDF is located at the depth of 400—450 m within the Nizhne-Kansky hydrogeological unit of fracture and vein groundwater (Fig. 4), where two types of subsurface flows are distinguished: regional flow of groundwater, guided by discharge to regional drains — the Yenisei and the Kan; local flows of groundwater controlled by local forms of landscape and discharged to the rivers of the third and the second order. The proof of separation of flows in the upper and lower zones of water mass would be one of the tasks of the model analysis to be carried out in the framework of safety case studies.

On the other hand, the acceptability of RWDF construction is determined by the permeability of the rock massif. Interval hydrogeological tests show that the hydraulic conductivity reduces with depth [16]. At the depths of 0—100 m, the most frequent values of hydraulic conductivity are $5{\cdot}10^{-3}$ m/day, while in the range of 100—200 m the average values are 5 times lower. Below 200 m, the values of hydraulic conductivity stabilizing at $(1...5){\cdot}10^{-4}$ m/day.

Data of hydrogeological studies were used to develop a 3D-mathematical model of the site (Fig. 5) based on the assumption of equivalent porous medium using MT3DMS software [17]. The model calibration was carried out by comparison of the model calculations with the data of hydrogeological testing of the massif, monitoring of pressure distributions, and hydrographs of rivers.

The modeling demonstrates that the Shumikha River, as well as the Bezymyanny and Studyony streams have a draining effect on the water-bearing rocks only in the upper part of the cross-section (Fig. 6). At the lower absolute intervals (including the supposed location of the underground facility (DRWDF)) there is no such effect: the underground waters transits this section and are unloading directly to the Yenisei River.

It is planned that the 2nd class waste in DRWDF will be located in horizontal mines (drifts) while the 1st class wastes (heat-generating waste) — in vertical wells. List of radionuclides that are potentially hazardous from the contamination point of view in the post-operation period is fairly large (I-129, Sr-90, H-3, Tc-99, Se-79, U-238, Cs-137, Cs-135, Am-243 and Pu-239). In addition to heat, gas generation is possible due to corrosion of materials.

Two types of experiments were performed to study the interaction of dissolved radionuclides and rock materials: (1) sorption of radionuclides by the dispersed material in the fracture interspace (determination of K_D, cm^3/g); (2) sorption

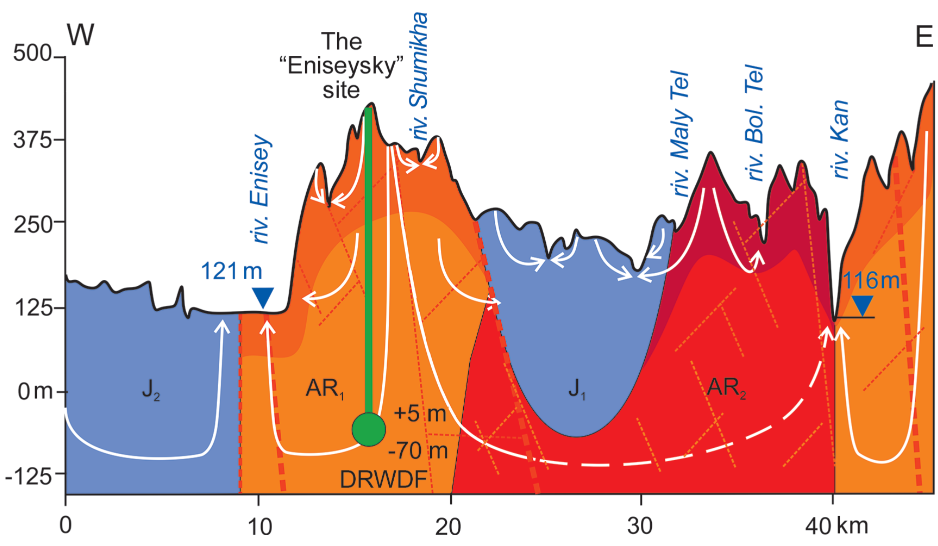


Fig. 4. Schematic geological profile (W-E) of the "Eniseyskiy" site (DRWDF). Arrows show flow directions in the zones of active and slowed water exchange

of radionuclides on the surfaces of fractures (in monolithic fragments of rocks) with estimation of the surface distribution coefficient (K_A, cm) (Fig. 7).

The modeling demonstrates that sorption and radioactive decay are the limiting factors for radioactive contamination of groundwater (Fig. 8). For example, a short half-life period and a considerable sorption lead to the situation when almost all Cs-137 remain within the boundaries of underground repository. Pu-239 with long half-life has a considerable plume in spite of its high sorption rate. Taking into account previous considerations the most hazardous are the low sorbing and long-lived radionuclides, such as I-129, Se-79 and Tc-99. This is illustrated by modeling of Se-79 contamination (Fig. 8).

Consequences of gas and heat generation have been investigated at the final stage by solving a conjugate problem of flow and heat transport.

The need to use two-phase models is explained by: (1) the formation of the gas emersion as a less dense phase, (2) the difference of phase permeability of the rock massif (relative to gas and water); (3) dissolution of gas in water and its fast transport in dissolved form. Flow structure of the model has been developed by applying the software code TOUGH2 [18] and has inherited the main features of the previously considered model. Intensity of hydrogen generation, as the most probable gas, has been estimated using the specialized software. Heat is generating due to the radioactive decay, the generation capacity was calculated based on overall activity of various radionuclides.

The calculations showed that the heat generation in the repository is represented as a decreasing exponential function. Maximum temperature

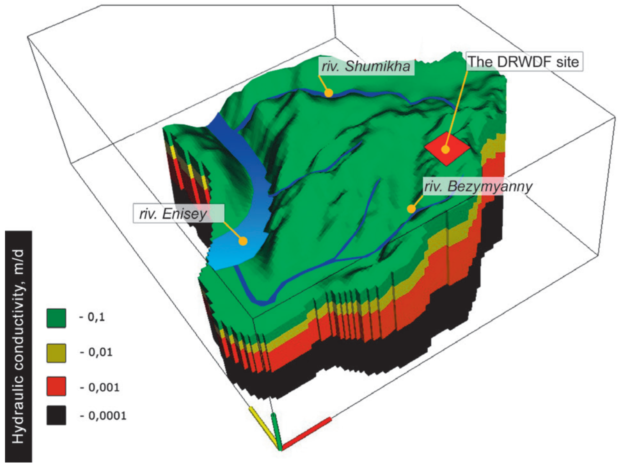


Fig. 5. Conceptual 3D hydrogeological model

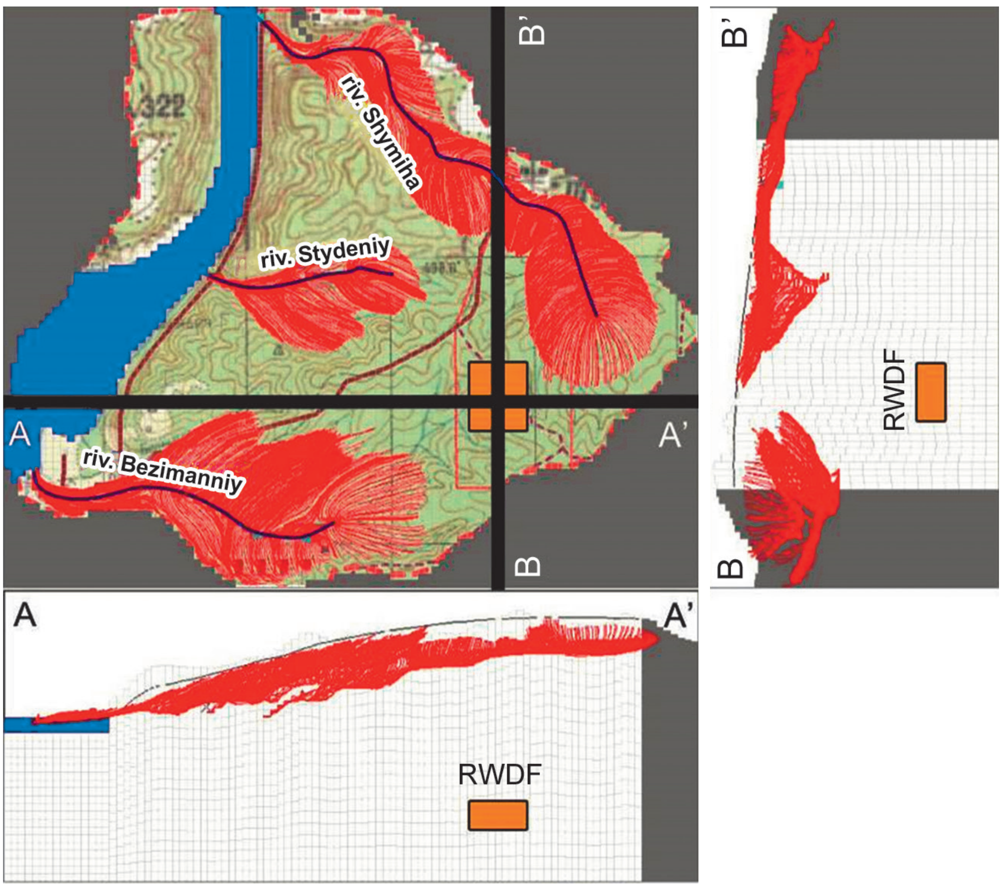


Fig. 6. Zones of infiltration waters capture by surface water flows (the rectangle shows the potential site location)

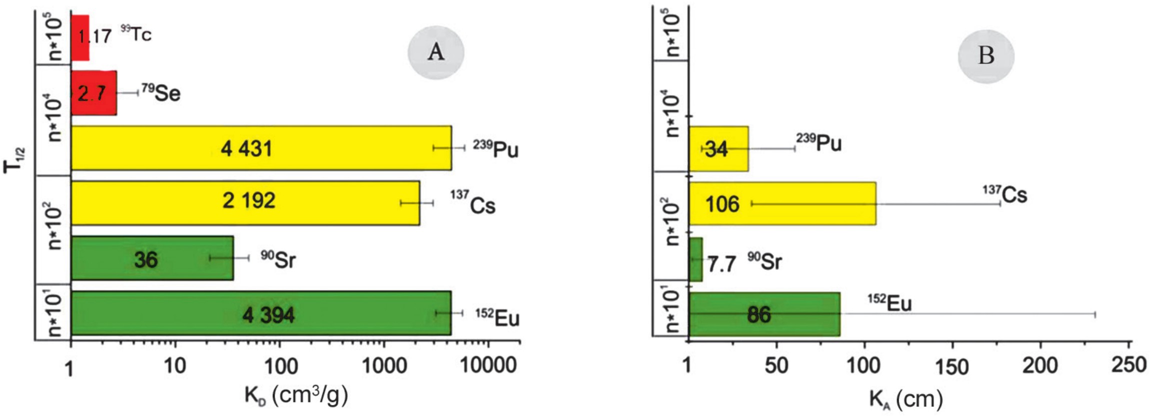


Fig. 7. Experimentally defined sorption distribution coefficients of ^{137}Cs, ^{90}Sr, ^{79}Se, ^{99}Tc, ^{152}Eu, ^{239}Pu: A — for disintegrated material of fracture filling in host rock; B — for surfaces of fractures of monolithic samples

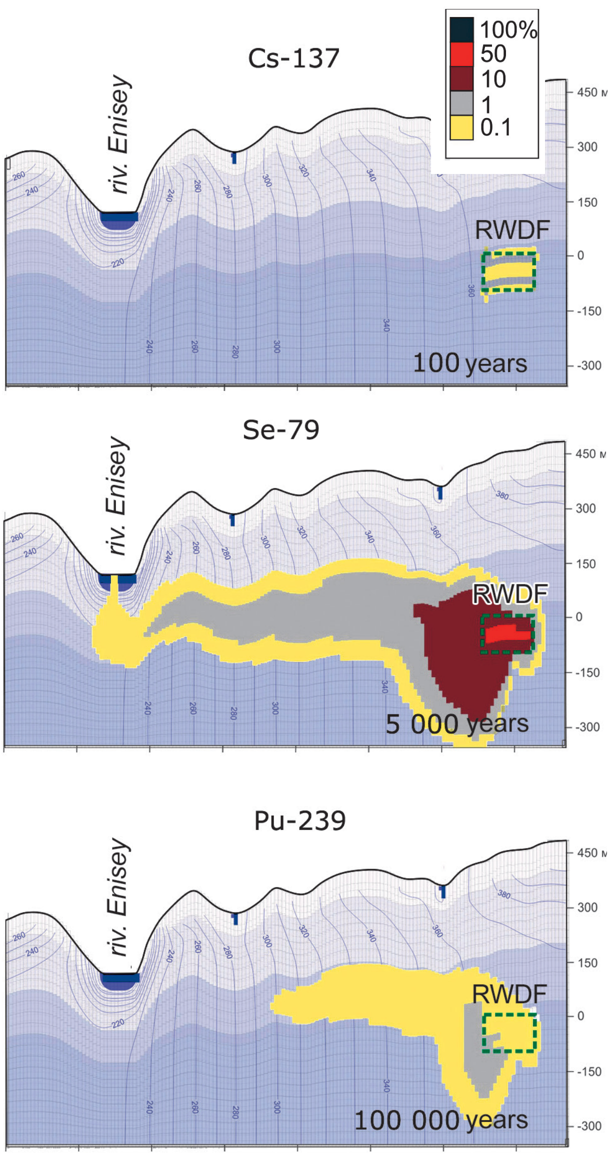


Fig. 8. Configuration of underground waters contamination envelopes in relative concentrations; time shown for the time since release beyond the engineered barriers

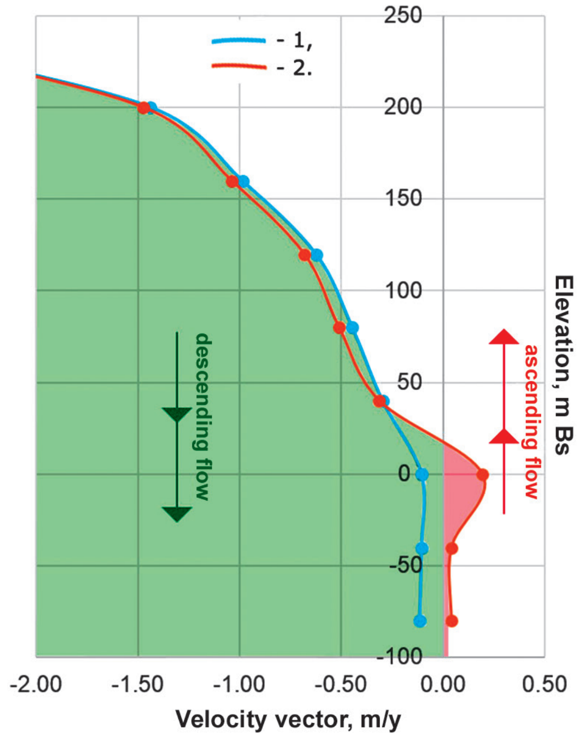


Fig. 9. Flow rate profile assuming the absence of thermal convection (curve 1) and the presence of thermal convection (curve 2)

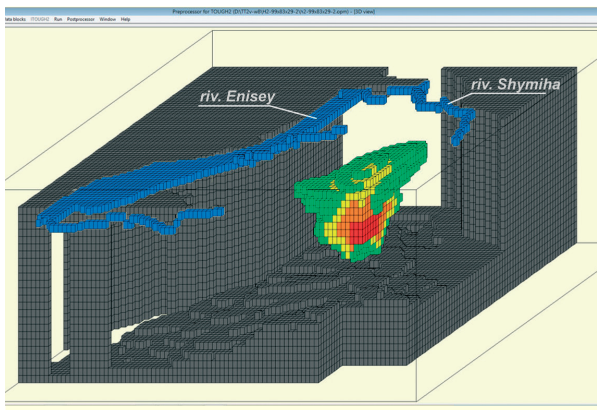


Fig. 10. Plume of underground hydrosphere contamination by hydrogen (dissolved form)

in RWDF would reach 80—90 °C in 55—65 years after RW disclosure. The heat distribution has a stable spherical form, indicating that conductive (diffusion) heat transfer mechanism dominates in the crystalline host-rocks. The heat plume dimensions are less than 100 m. The circulating cell formed by the heat field does not have substantial effect on the flow rate field (Fig. 9). Therefore, the migration calculations performed earlier for isothermal conditions do not need to be corrected.

The modeling shows that the gas phase spreads in the DRWDF in the vertical direction and remains within the repository contour. For approximately 100 years, the plume of non-dissolved hydrogen stabilizes, and reaches the level of 220 m — about 200 m below the surface. The hydrogen saturation is insignificant.

The gas does not reach the daylight surface; it is transformed to a dissolved form.

The dissolved gas phase (Fig. 10) is of highest interest, as the scale of spreading of the dissolved part is much larger than the gas phase spreading plume.

The simulation shows that in the upper (most permeable) parts of the cross-section, the dissolved hydrogen is engaged in the movement of groundwater in predominantly horizontal direction.

Conclusion

The preliminary analysis indicates that from the safety case point of view, there are no factors that may have unfavorable impact on the two considered facilities. The reliability of this conclusion may be additionally confirmed by further studies of geological and hydrogeological conditions, mechanisms and parameters controlling the transport of radionuclides in the rock massif, as well as by further advances of mathematical tools.

The safety case for RWDF at fairly small depth in clay masses (Sosnovoborsky district conditions) would benefit from geomechanical simulation of aquifers tapping in the clay roof to forecast the evolution of man-caused fracturing over the mining areas.

It is reasonable to develop a network of offsite monitoring wells for DRWDF in the Nizhne-Kansky granite-gneiss massif to study the general structure of subsurface flows and the water balance of the territory. It is critical to use and develop the models of new generation taking into account the continuum principle (“discrete/channel” character of liquid flow in fractured medium).

Integration of local process models at the facility (near field) and large low-scale regional model (far field) is required for justification of all design solutions, as well as calibration of models using additional information (isotopes, hydrology, satellite data) with sensitivity analysis. Particularly, special attention should be paid to physical-chemical processes, multicomponent aqueous solutions and the formation of colloids.

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