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Influence of Rock Structure on Migration of Radioactive Colloids from an Underground Repository of High-Level Radioactive Waste

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Abstract: Studies of leaching of vitrified simulated high-level radioactive waste (HLW) evidence that most of actinides or their simulators enter leaching water in a colloidal form. In this paper, we consider a mechanism of colloid-facilitated migration of radionuclides from an underground repository of HLW located at a depth of a few hundreds of meters in fractured crystalline rocks. The comparison between data of field and laboratory measurements showed that the bulk permeability of the rock massif in field tests is much greater than the permeability of rock samples in laboratory experiments due to an influence of a network of fractures in the rock massif. Our theoretical analysis presents evidence that this difference can take place even in a case when the network is not continuous, and the fractures are isolated with each other through a porous low-permeable matrix of the rock. Results of modelling revealed a possibility of mechanical retention of radionuclide-bearing colloid particles in the frame of rock during their underground migration.

Keywords: radioactive waste; underground repository; radiocolloid; migration; ground water; fractures; mechanical retention

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1. Introduction

Observed climate changes and the necessity to reduce emission of greenhouse gases increases the significance of nuclear power engineering. Sustainable development of the atomic industry calls for a safe solution of high-level radioactive waste (HLW) management. The most effective and reliable approach to solving the problem at present is disposal of conditioned (solidified) HLW in underground repositories at a depth of several hundred meters [1–3]. Safety of such geological repositories is based on the multibarrier concept, which implies that both engineered and natural barriers provide for a reliable isolation of HLW in the repository from the biosphere. The engineered barriers include waste forms, canisters, and containers for solid HLW, backfill for holes in the repository where the canisters are disposed of, and repository construction units. The natural barrier is a rock massif between the loaded part of the repository and the biosphere [2,4]. The engineered barriers can degrade during the time period that is comparable with the half-life of many radionuclides from the HLW's composition [5]. As a result, after an initial period, the rocks can be considered as the main isolation barrier protecting the biosphere from HLW. The main hazard of radionuclides ingress from HLW to the biosphere is caused by their transport by groundwater, which flows in rocks through systems of connected pore and fracture voids [6–8]. These systems are called flow channels, the scales of which are considered in [9]. Reliability of HLW isolation depends on the time of radionuclides migration from the loaded part of the repository to the biosphere. Concentration of radionuclides in groundwater decreases during their migration to the biosphere due to radioactive decay. If the concentration in the groundwater at its leakage to the earth surface (or to a water reservoir or a river net) is less than a maximum allowable level, the natural barrier is reliable, and the repository is safe [1]. Therefore, duration of radionuclides migration through the rocks and then the migration velocity is of paramount importance for the repository safety.

This paper analyses the process of radionuclide migration from underground repositories to the biosphere via colloid-facilitated transport of radionuclides by flowing groundwater accounting for the mechanical retention of colloidal radionuclide-bearing particles.

Continuum-scale modeling of groundwater flow in fractured rock with an explicit treatment of rock fracturing is used in many safety assessments of hazardous underground radiation objects. One of the first such models was proposed by Barenblatt et al. [10]. Development of this approach as applied to contaminant transport in fractured rocks was presented by J. Bear [6], I. Neretnieks [7], and C.-F. Tsang [11]. Reviews of methods for continuum-scale modeling flow in fractured rocks are provided in [12,13]. Most attention is paid to the influence of fractures that form a connected network. However, there is an option that the fractures do not form a linked structure even in a case when elevated permeability of the rock indicates directly a significant influence of rock fracturing. The fracture network channelizes the groundwater flow in this case, but the fractures can be disconnected, and parts of the groundwater streamlines between the fractures run through the rock matrix.

A substantial ecological hazard can be caused by a release of radionuclides from the underground repository of HLW and their transport from the repository to the biosphere. The groundwater can carry radionuclides as ions and colloid particles (radiocolloids). Since the radiocolloids can be much more mobile in the underground medium than the radionuclide ions they can represent the most hazardous form of radionuclides migration [8]. The intervals of the rock matrix between the disconnected fractures along the same streamlines of the groundwater can serve as filters for the particles of radionuclide-bearing colloids. Analysis of this option is the objective of this study.

We show that (i) even a system of disconnected fractures can cause a difference by a few orders of magnitude between bulk permeability of rock massif and permeability of the nonfractured rock matrix; (ii) analysis of permeability measurement at the site of potential federal repository of HLW indicates that fractures in the rock are, at least, partially disconnected; (iii) intervals between the disconnected fractures along the same streamlines of the groundwater can be effective filters retaining radionuclide-bearing colloids. Analysis of the influence of disconnected fractures on the bulk permeability of rocks is carried out by methods of computer modelling.

2. Colloid-Facilitated Transport of Radionuclides by Groundwater

As a result of sorption on walls of the flow channels, cations of many radionuclides should move at a velocity that is much less than velocity of the groundwater [14,15]. Let us denote velocities of any contaminant and the groundwater as V_c and V_f, respectively. As a first approximation, one can assume that sorption is reversible and satisfies the linear equation

$$C_r = \rho K_d C, \quad (1)$$

where C_r and C are mass fractions of the contaminant in the rock and in the groundwater, ρ is groundwater density, K_d is a coefficient which characterizes sorption properties of the rock to the contaminant.

Then ratio between V_c and V_f satisfies the expression

$$V_c/V_f = 1/(1 + \rho_r K_d/\varphi), \quad (2)$$

where ρ_{r} is rock density and φ is rock porosity.

The higher sorption properties of the rocks to the contaminant (K_d), the less is the ratio V_c/V_f. If sorption of radionuclides on the rocks is absent, the ratio V_c/V_f tends to 1, which is the maximum value of this ratio except of particular cases that are considered in [16–20]. However, results of radiation monitoring at the sites of significant radioactive pollution showed that values of V_c/V_f were much higher than it was predicted on the basis of sorption properties of the rocks to the radionuclides [16–23]. Similar result was obtained in laboratory experiments [24]. Elevated values of the radionuclide's migration velocity as compared to its predicted values were explained by the assumption that groundwater carries radionuclides not only as a solute, but also in bound form as colloid particles (by definition, particles are called colloidal if their size ranges from 1 to 1000 nm), which are sorbed by the rocks to a lower extent than the radionuclide ions [25]. Colloid particles that carry radionuclides in groundwater are called radiocolloids. The colloids are subdivided into three main groups according to their origin: intrinsic colloids, primary colloids and pseudocolloids [26,27]. Intrinsic colloids consist of particles of colloid size that are composed to a significant extent of radioactive isotopes and their oxi- and hydroxides. Primary colloids represent colloid particles composed of leaching products of HLW vitreous form at its contact with groundwater. Pseudocolloids consist of colloid particles existing in the groundwater before contact with radioactive materials. At contact of the particles with polluted water radionuclides will be sorbed on these particles.

At present, immobilization of HLW on industrial scale is carried out using Al-P- or B-Si-glass [3,28,29]. Experiments on leaching of aged Na-Al-P-glass by water show that more than 95% of actinide simulators in the leaching products are attached to particles with diameter between 450 and 100 nm [30–32]. Thus, primary colloids represent more than 95% of actinide simulators' ingress into the groundwater. Former studies have demonstrated that formation of two kinds of radioactive colloids: (1) primary—at hydrothermal alteration of vitreous waste forms and (2) pseudocolloids—at expense of initial colloids of waters or derived from eroded bentonite buffer are typical processes in the environment of underground HLW repository [17–25,33–35]. Therefore, one can expect that practically all actinides resulting from nuclear waste forms corrosion can be carried by the groundwater in highly mobile colloidal form, which will potentially decrease the safety of the repository [32]. However, the probability exists that diameters of the primary colloid particles can be larger than apertures of the filtration channels, and the coarsest fraction of primary colloids will be mechanically retained. Analysis of this possibility is the main subject of this study.

3. Influence of Rock Fracturing

It is known that the bulk permeability of crystalline rocks is usually higher by 1–3 orders of magnitude than the permeability of rock matrix, which is obtained in laboratory measurements on small ($\cong 10^{-2}$ m in size) samples [36,37]. This is caused by the influence of fractures, the lengths of which are much larger than dimensions of interstitial voids and pores of the rock matrix. The latter focuses on the mainstream of the groundwater, which flows through the rock massif. Since they are much larger in size than rock samples used for laboratory measurement of permeability, laboratory measurements do not take into account the influence of fractures, and the permeability of small samples (i.e., permeability of the rock matrix) is much less than the bulk permeability of the massif. One can suppose in this case that mechanical retention of radiocolloid should be absent because the significant difference between bulk and sample permeabilities suggests that the fracture network and relatively large apertures of fractures are quite sufficient for free movement of radiocolloid particles along the fractures with the mainstream of the groundwater.

However, the elevated bulk permeability compared with the rock matrix permeability do only indicate on presence of fractures, although separate fractures can be disconnected and do not form a linked network through the rock massif.

Let us examine the influence of disconnected fractures on bulk permeability of rocks. We first consider a 2D cross-section of a fractured rock with porous matrix and chessboard ordering of the disconnected fractures directed along a mean flow of groundwater in the rock massif (Figure 1a) noting that the cross-section can be inclined in a general case.

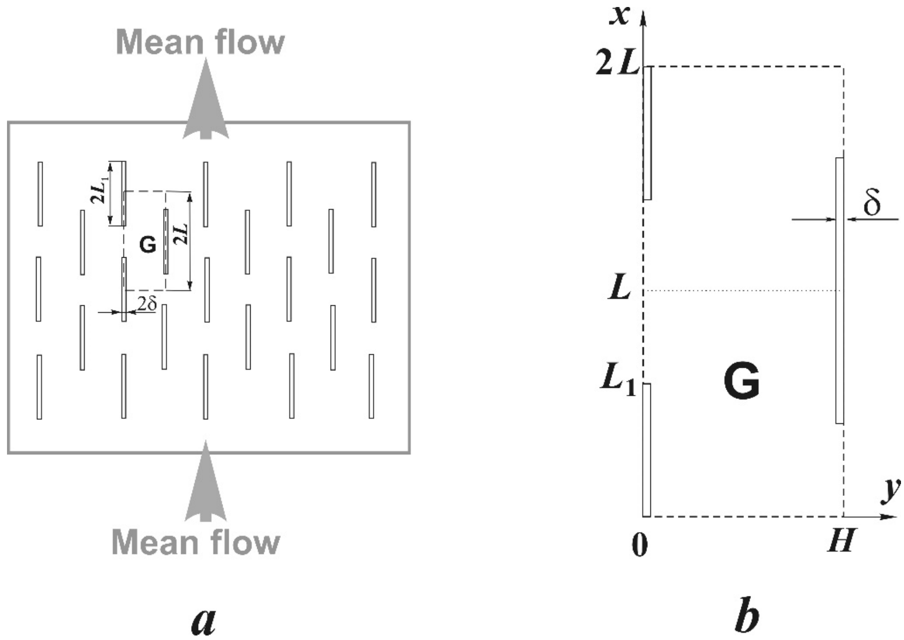

Figure 1. Diagram of porous–fractured rocks: (**a**) chessboard ordering of fractures in a porous permeable rock matrix; (**b**) recurrent cell of the porous–fractured medium at chessboard ordering of the fractures in the rock matrix.

Let us consider the groundwater flow in the recurrent cell G and introduce in it Cartesian coordinates as shown in Figure 1b.

The water flow in the porous matrix (outside the fractures) is governed by Darcy's law [15]

$$v_x = -\frac{k_m}{\mu}\frac{\partial p}{\partial x},\ v_y = -\frac{k_m}{\mu}\frac{\partial p}{\partial y}, \tag{3}$$

where $v_x,\ v_y$ are components of the Darcy's velocity, k_m is permeability of the porous matrix, μ is dynamic viscosity of the groundwater, $p = P + \rho g z(x, y)$, P is pressure, ρ is groundwater density, g is acceleration due to gravity, $z(x, y)$ is altitude of the point with coordinates x and y above any fixed horizontal plane. Since the cross-section is a plane, $z(x, y)$ is a linear function. If the cross-section is horizontal, then $p = P + const$.

Since the groundwater is practically incompressible, components of the velocity satisfy the continuity equation in the form

$$\frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y} = 0. \tag{4}$$

Hence, p satisfies the Laplace equation

$$\frac{\partial^2 p}{\partial x^2} + \frac{\partial^2 p}{\partial y^2} = 0. \tag{5}$$

We assume that the average velocity in cross-sections of fractures is governed by 1D Darcy's equation [6]. Then, the flow in the fractures is governed by mass balance equations in the form

$$\begin{array}{c} -\frac{\delta k_f}{k_m}\frac{\partial^2 p}{\partial x^2}\Big|_{y=0} = \frac{\partial p}{\partial y}\Big|_{y=\delta+0},\ 0 < x < L_1 \text{ or } 2L - L_1 < x < 2L; \\ \frac{\delta k_f}{k_m}\frac{\partial^2 p}{\partial x^2}\Big|_{y=H} = \frac{\partial p}{\partial y}\Big|_{y=H-\delta-0},\ L - L_1 < x < L + L_1 \end{array}, \tag{6}$$

where 2δ is fracture aperture, $2L_1$ is fracture length, $2L$ is length of the recurrent cell G.

As a result of mirror symmetry of the streamlines,

$$\begin{gathered}\frac{\partial p}{\partial y}=0,\ y=0,\ L_1<x<2L-L_1,\\ \frac{\partial p}{\partial y}=0,\ y=H,\ \{x<L-L_1 \text{ or } x>L+L_1\}.'\end{gathered} \tag{7}$$

Let us denote pressures at $x = 0,\ 2L$ as p_0 and p_1, respectively. It follows from the mirror symmetry of streamlines that p_0 and p_1 do not depend on y. Hence,

$$x=0,\ p=p_0;\ x=2L,\ p=p_1. \tag{8}$$

Equalities (6)–(8) are boundary conditions for the Equation (5). The boundary problem (5)–(8) was solved numerically by finite differences method of successive over relaxation. Since the considered porous–fractured medium consists of recurrent cells G, we can express the bulk permeability of the medium on the basis of the obtained numerical solution as

$$k_{bulk}=\frac{\mu}{H}\frac{2L}{p_0-p_1}\int_0^H v_x dy. \tag{9}$$

Results of k_{bulk} calculations can be approximated by the dimensionless expression

$$\frac{k_{bulk}}{k_m}=F(h,l,M)=\frac{h^2F_1(h,l,M)+0.328F_2(h,l,M)}{h^2+0.328}, \tag{10}$$

where

$$\begin{gathered}l=\frac{L}{L_1},\ h=\frac{H}{L_1},\ M=\frac{\delta k_f}{L_1 k_m},\\ F_1=1-\frac{4l}{\pi h}F_M\left\{\cos\left(\frac{\pi}{2l}\right)+\exp\left[-\frac{\pi(M+2.62)}{2l}\right]\right\},\\ F_2=1+\left[\frac{h}{2M}+\frac{0.439h^2}{h^{1.46}/(l-1)^{0.138}+0.439F_M F_l}\right],\\ F_M=M/(M+2.62),\ F_l=\exp\left\{-(l-1)^2\left[1+(l-1)^{3.57}\right]\right\}\end{gathered} \tag{11}$$

Comparison of k_{bulk}/k_m obtained numerically and calculated by approximating Formula (10) is shown in Figure 2.

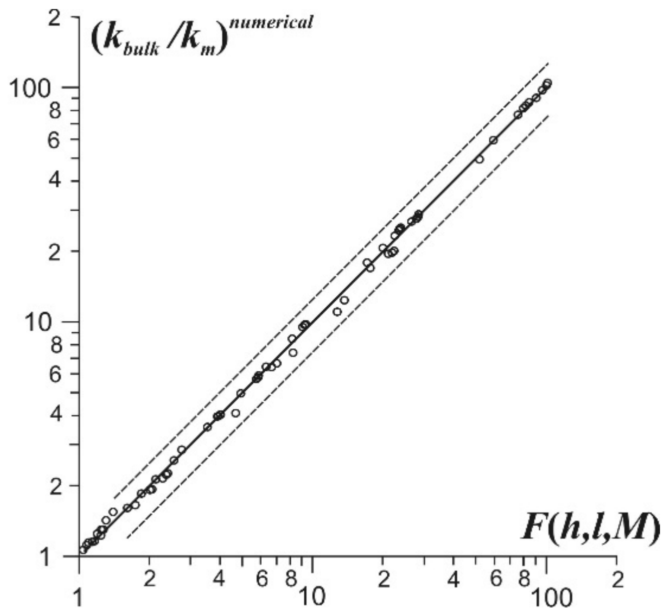

Figure 2. Comparison of bulk permeability values calculated numerically with approximating Formula (10). Dashed lines correspond to errors of ±25%.

The chessboard ordering of fractures is of particular importance for estimation of the influence of disconnected fracturing on bulk permeability of rock massifs. Let us consider disconnected fractures of equal length and aperture, which are disposed with equal spacing in rows, and distances between neighboring rows are equal. An example of this type of fracturing is the considered case of chessboard ordering of fractures. Another example is a rectangle-cluster ordering of the fractures (Figure 3).

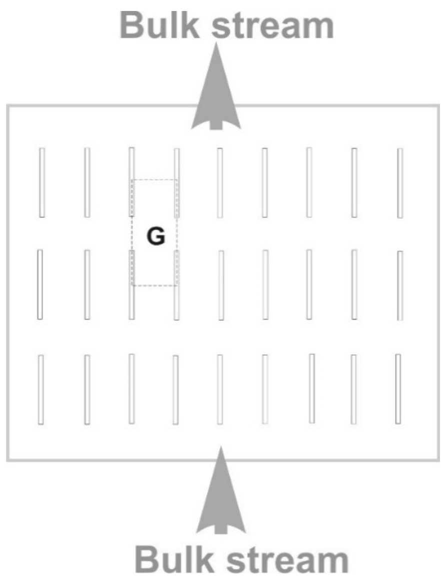

Figure 3. Rectangle-cluster ordering of fractures in porous rock matrix.

We show that chessboard ordering corresponds to an extremum in influence of such fracturing on bulk permeability of rocks. Let us enumerate fracture rows. In the case of chessboard ordering, middles of fractures in odd rows correspond to middles of intervals between fractures in even rows (Figure 4a).

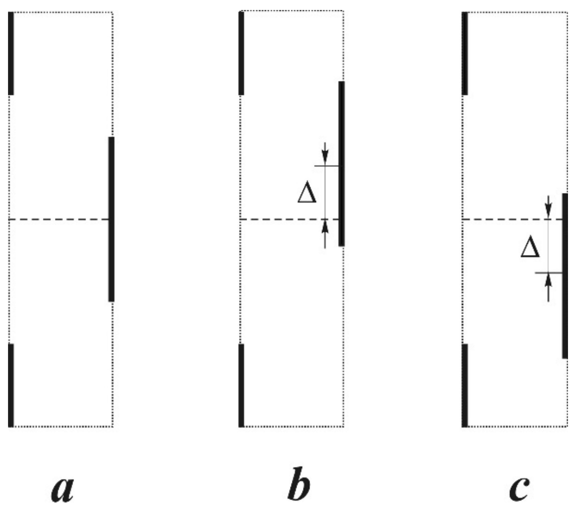

Figure 4. Modifications of chessboard ordering of the fractures. (**a**) chessboard ordering of fractures; (**b**) chessboard ordering shifted by Δ along the row direction; (**c**) chessboard ordering shifted by Δ in the opposite direction.

Let us shift odd rows by a distance Δ along the row directions (Figure 4b). We denote the bulk permeability of obtained medium as $k_{bulk}(\Delta)$. Then we consider shift of the odd rows from their initial position by a distance Δ in opposite direction, i.e., by $-\Delta$ (Figure 4c).

Permeability of the medium in this case is $k_{bulk}(-\Delta)$. Boundary problem (5)–(8) is linear. This implies in particular that the value of bulk permeability does not change if direction of the mean flow becomes opposite. Therefore, $k_{bulk}(-\Delta) = k_{bulk}(\Delta)$. Hence, $k_{bulk}(\Delta)$ is an even function, and

$$\frac{dk_{bulk}}{d\Delta}(0) = 0. \quad (12)$$

Therefore, $\Delta = 0$ (which corresponds to chessboard ordering of the fractures) is an extremum point of k_{bulk}.

In absolutely the same manner, we can show that rectangle-cluster ordering of the fractures is also an extremum point of k_{bulk}. Since k_{bulk} at chess-board ordering is much higher than in the case of rectangle-cluster ordering at the same values of h, l, M, it is reasonable to assume that chess-board and rectangle-cluster ordering correspond respectively to maximum and minimum influence of fracturing on k_{bulk} at the specified h, l, M.

One can see from Figure 3 that the bulk permeability can exceed the rock matrix permeability by more than two orders of magnitude even in the case of disconnected fractures. This implies that a significant difference between bulk permeability of the rocks and matrix permeability (which is measured in laboratory tests [30,31]) is not an argument in favor of an existence of a network of hydraulically connected fractures, which extends throughout the rock massif.

4. Characteristics of Fracturing of Nizhnekansky Massif Rocks (Eniseisky Site, Krasnoyarsk Region, Russia)

Granitoid massif Nizhnekansky is located in the Krasnoyarsk region (Russia). It is considered a potential territory for the development of a federal underground repository of high-level and intermediate-level nuclear waste with long-lived radionuclides [3,38]. Studies on the rock matrix were carried out on six samples from different sites and different depths of the granitoid massif. The core samples were 38–52 mm in diameter and about 150 mm in length. Petrographic and mineral–chemical studies have shown that the rocks are liable to metamorphic (amphibolite facies with quartz, feldspars, biotite, amphiboles) and low-temperature hydrothermal–metasomatic alterations (chloritization, sericitization and argillization), which are correspondingly accompanied by ductile (gneiss texture with characteristic foliation) and brittle (cataclastic, brecciated textures and microcracks filled by carbonate, chlorite, sericite and clay minerals) deformations [39,40]. A brief description of sample compositions and data on their permeability are presented in the Table 1, from which one can see that permeability of the samples (i.e., permeability of the rock matrix) does not exceed 3×10^{-18} m^2.

Table 1. Composition and permeability of rock samples [1].

Nos	Sample Index	Composition	Permeability, m^2
1	K 560.8	Granodiorite	1.488×10^{-18}
2	K 613.1	Porphyric adamellite	2.307×10^{-18}
3	I 142.6	Gneissic granite with metasomatic alterations	3.712×10^{-20}
4	I 491.7	Gneissic granite	8.201×10^{-19}
5	I 357.2	Quartz diorite	3.092×10^{-19}
6	I 504.6	Quartz diorite	9.595×10^{-19}

[1] Note: the sample number corresponds to depth of selection at the Kamenny (K) or Itatsky (I) sites.

Data of bulk permeability of rocks of the Yeniseisky site which is considered at present as the most promising place for development of the vitrified HLW repository are provided in [41]. Measurements were carried out in exploratory boreholes by pumping tests at depths up to 700 m. Results of the measurements are shown in Figure 5 (a and b where permeability k is expressed though hydraulic conductivity f as $k = f\mu/(\rho g)$, where ρ is groundwater density, g is acceleration due to gravity; $k \cong 1.16 \cdot 10^{-13} f$ in the considered case if permeability unit is m^2, and water conductivity unit is m/day, as in Figure 5b).

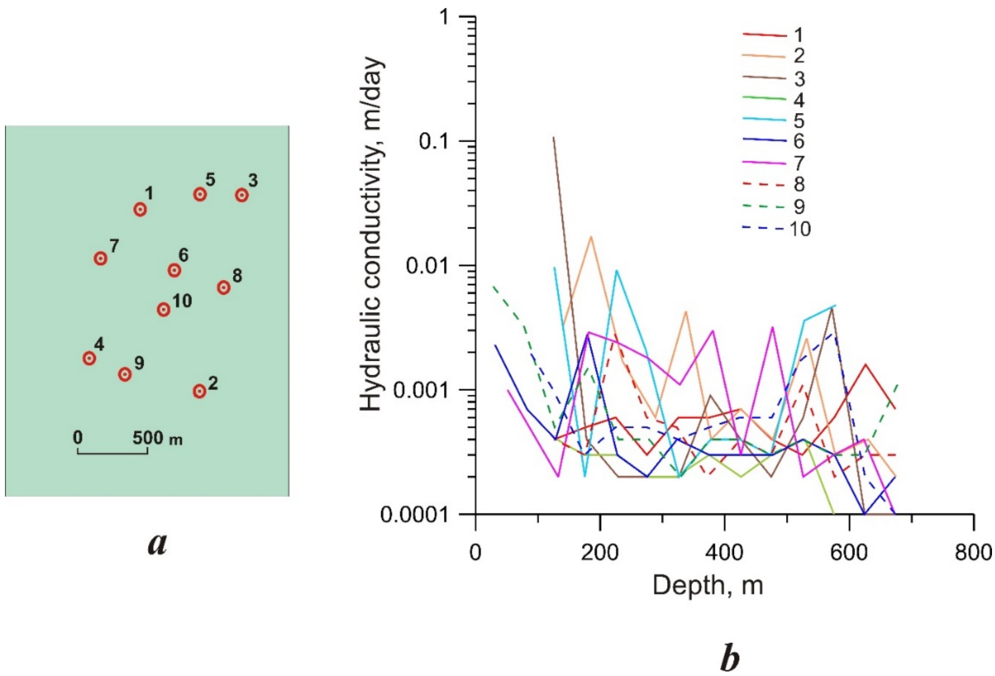

Figure 5. Data of pumping test at the site of the granitoid massif Nizhnekansky, which is selected for the development of the first federal underground repository of high-level radioactive waste in Russia. (**a**) Positions of the boreholes; (**b**) hydraulic conductivity of rocks.

5. Discussion

Recalculation of the data in Figure 5b shows that permeability of rocks does not exceed 10^{-15} m^2 beneath the depth of 300 m. This is higher by almost three orders of magnitude than the rock matrix permeability. Hence, one can expect that the bulk permeability is caused mostly by fractures. However, comparison of different curves in Figure 5b shows that peaks on one curve are absent at the same depth on curves obtained by tests in neighboring boreholes though the distance between them does not usually exceed 500 m. This evidences that fractures do not form hydraulically connected clusters (network) that extend throughout the whole massif [41]. Hence, a part of the groundwater flow path is within the rock matrix. Therefore, mechanical retention of radiocolloid is quite probable in this part of the Nizhnekansky granitoid massif, where research is being conducted to create the first federal underground repository of high-level nuclear waste in Russia.

6. Conclusions

The main mechanism of radioactive pollution propagation from an underground repository is caused by the transport of radionuclides by groundwaters to the biosphere. The groundwater flows in the earth's crust through connected systems of pore and fracture voids. These systems are called flow channels. Radionuclides can be carried by the groundwater as a dissolved component or in the form of colloid particles with attached radionuclides. Colloidal form can be more mobile in geological media than radionuclide ions. However, the rocks can mechanically retain radioactive colloid particles if dimensions of cross-sections of the filtration channels are less than dimensions of colloid particles. Hence, the possibility of mechanical retention of radioactive colloid remains even in the case of fracturing increasing bulk permeability of the rocks by a few orders of magnitude. This statement agrees with data from laboratory examination of rocks from the Nizhnekansky granitoid massif, which has been selected as a site for the construction of the first federal underground repository of vitreous high-level radioactive waste in Russia.

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