EXPERIMENTAL TESTS ON THE MIGRATION OF LONG-LIFE RADIONUCLIDES IN TECTONICALLY DISTURBED HARD ROCK MASS

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ABSTRACT

Underground mine workings of the Krasnoyarsk Mining & Chemical Combine (MCC) offer a unique opportunity to study radionuclide migration in the zone of development of rocks left intact of physical weathering.

Experimental filtration and migration testing studies are being designed and carried out in various tectonic zones, with different secondary mineralization in the fractures.

Laboratory studies on the processes of interaction between solutions containing plutonium, americium and neptunium and gneiss of fracturing and schist formation zones have been started. The results indicate that neptunium and plutonium are less sorptive ability than americium under elevated temperature and pressure.

Background and Site Geology

Underground mine workings of the Krasnoyarsk Mining & Chemical Combine confined to the hard rock massif composed of a metamorphic complex of Pre-Paleozoic age, offer a unique opportunity to study radionuclide migration in the zone of development of rocks left intact of physical weathering.

The areas under investigation are located at the depth of 230-260 m from the earth surface, which is considerably lower than the exogenetic fracturing zone of about 80 m thickness. Rock mass tectonic parameters and geologic structure for one of the experimental sites are given in Fig. 1.

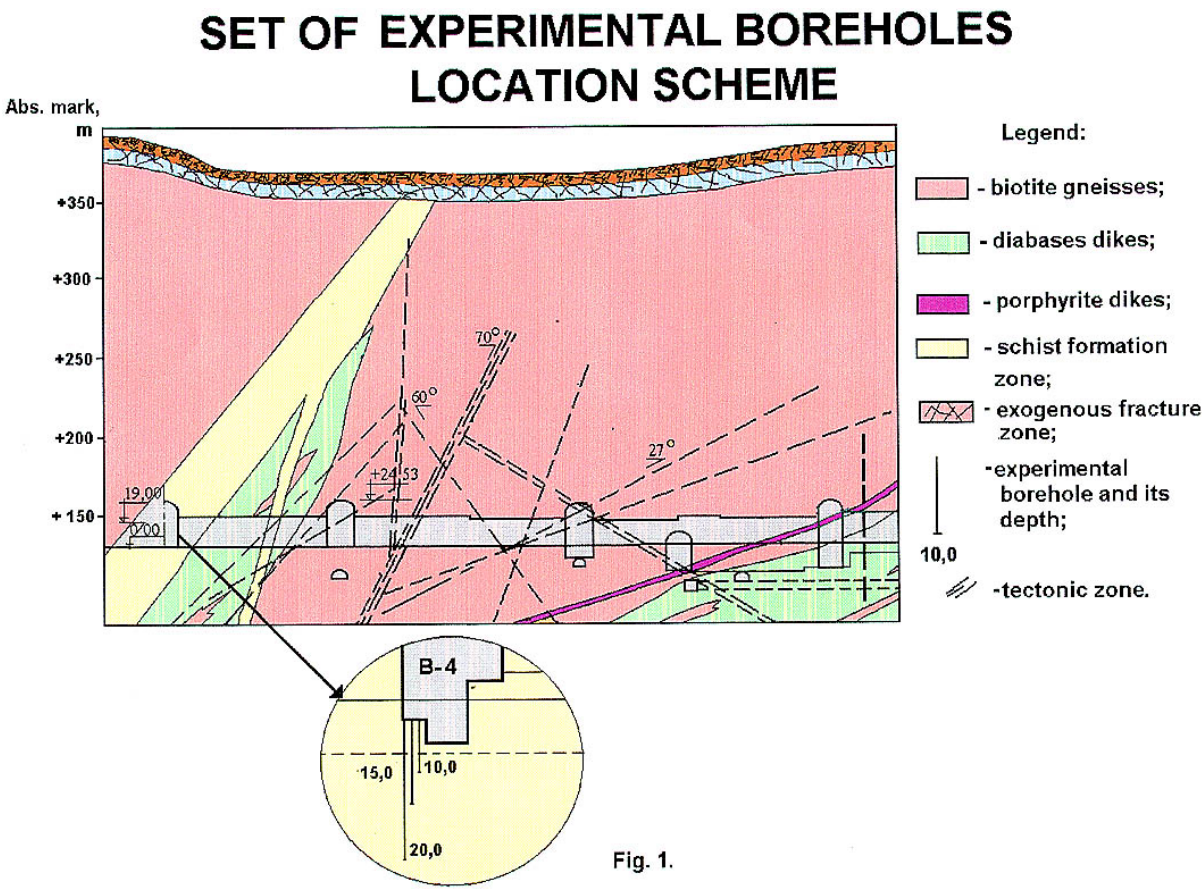


Fig. 1.

Grey, dark-grey, and green-grey biotite plagioclase gneisses of homogenous structure are most widely spread in metamorphic complex (up to 90 %), with more rare banded and spotty structures. Metamorphic rocks including amphibolites, micaceous gneisses, quartzito-gneisses, biotite and biotite chlorite slates are less common.

The rock mass also includes intrusive dikes of diabase, amphibolites and porphyrites. Dyke length varies to tens of meters and dike thickness of about 0.8-1.5 m with a maximum thickness of 15-20 m.

The major elements of rock mass tectonic structure are fractures and rupture dislocations. Fractures are shear or pressure-origated, and grouped mainly in two steep (60-90^0) systems extending to north-north-east and west-north-west.

Rupture dislocations often have zonal structure: a central crush zone with outer schist formation zone. Crush zones are from tens centimeters up to 2-10 meters thick and are composed of intensively crushed and ground host rocks, with planes of substitutes filled with secondary clay.

From a hydrogeological point of view, it is especially important to note that the rocks in the crush zones are cemented by clay or carbonate materials in ratio ranging from 1:0.01 to 1:0.5 and are practically watertight.

In contrast, the zones of schist formation and dike contacts with the host rock are flooded, though their permeability is of the order of 10^{-2} to 10^{-3} m/day. Schist formation zones are composed of cataclasite gneisses with platy partings (plate thickness varies from 1-2 to 10-25

cm), with some gneisses being converted into talc-chlorite slates and talc-chlorite mass with zoning structure. In this case, the presence of secondary minerals created an additional anti-migration barrier on the radionuclide' pathway.

Fracture displacement can be either West-East or North-South oriented, the latter being younger shifting West-East oriented fractures and dikes with amplitude not exceeding 0.5-2.0m.

It should be noted, that fractures outside the gneiss mass rupture dislocations are normally filled with chlorites, carbonates, iron hydroxides, zeolite and clayish minerals. Along with their low openness (or full closeness) this defines good rock mass isolating properties as a whole.

Ground Water at the MCC

These tectonic structures as well as mining and geological conditions define the character and degree of the rock mass flooding at the MCC. As major part of the rock mass is permeable and permeability of tectonically disturbed zones amounts to 0.002 – 0.0007 m/day, the rock mass can be considered relatively dry at the facility location site.

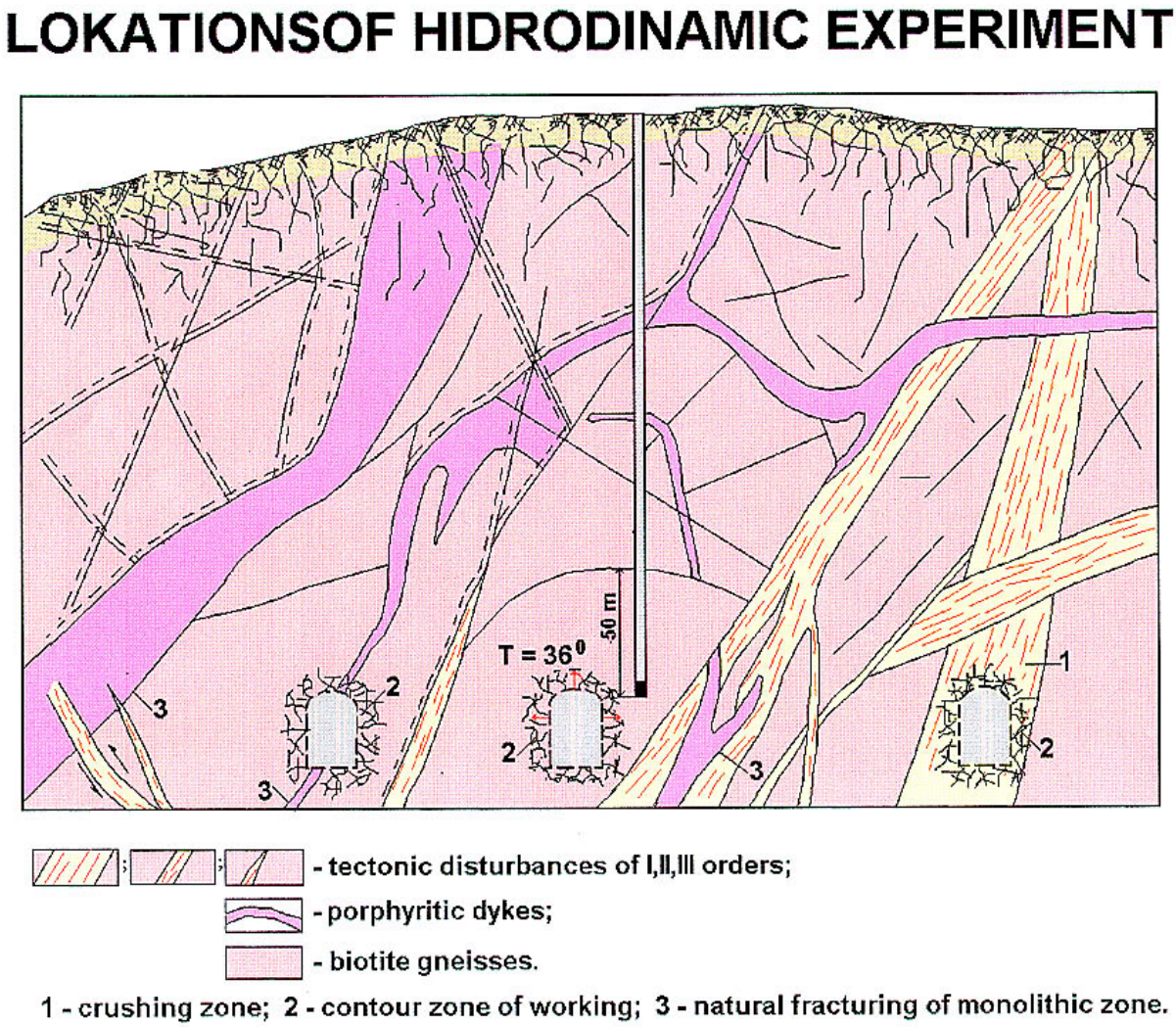
There are two general types of ground waters in the rock mass: fracture–ground waters confined to the zone of exogenetic fracturing and fracture-vein waters. The fracture–ground zone is about 80 meters thick, and fracture-vein waters are confined to linear elongated zones of tectonic disturbances and spread at least down to 260-300 m depth. Flooding of tectonically disturbed zone at the location marks of workings is very low (water inflows do not exceed 0.5 l/min and commonly range from 0.05-0.2 l/min).

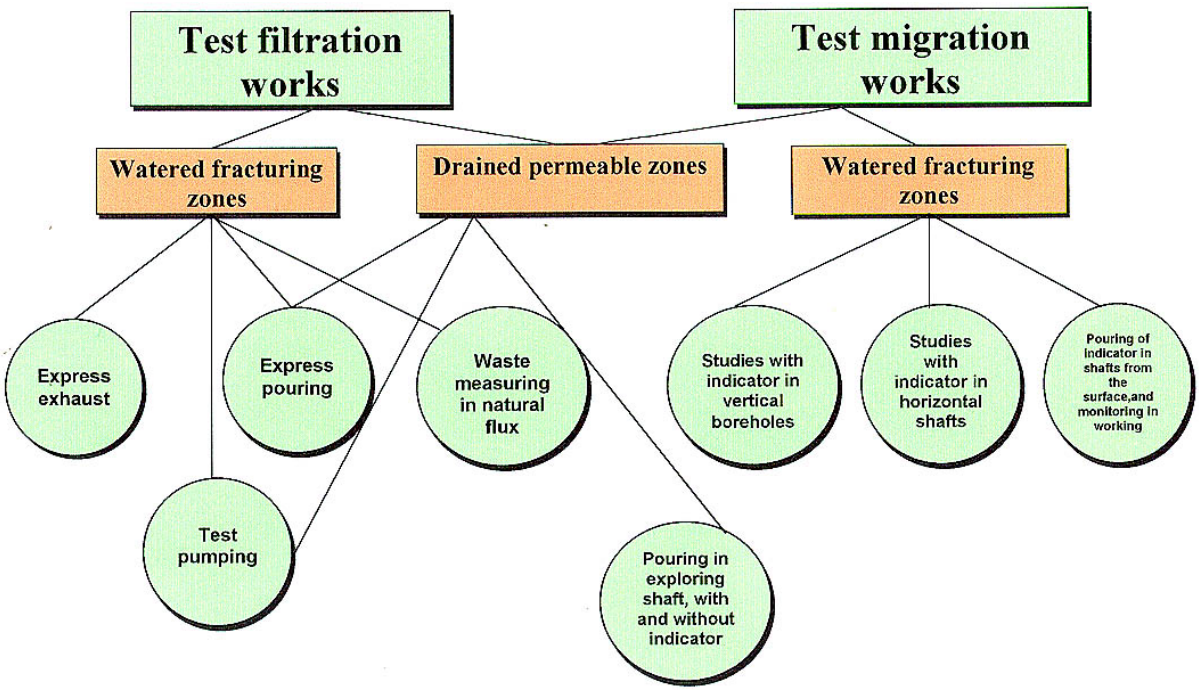
Planned Field Studies

However, with the use of traditional blast techniques in the construction of workings, a zone of artificial fracturing was formed around them which was not completely plugged during hydroisolating works. Coefficient of filtration here amounted to the order of 10^{-1} m/day.

The analysis of disturbed regime of ground waters in the MCC rock mass at the locations marks of workings shows three ways of possible radioactive contaminant migration to the local erosion base – the Yenisey river. They are a zone of mining-induced fractures, zone of tectonic disturbances and dike contacts with the host rock.
Each pathway of possible radionuclide migration is studied separately, according to a specially selected methodology. Considering a large amount of workings drifted at different levels, all existing varieties of the rock mass can be studied, as well as different natural and mining-induced fractures and sites with different secondary mineral fracture fillings. Certainly, such experiments will take several years, and for to-day we can speak only about the preliminary stage of works. The general research scheme is given in Fig. 2. Analysis of processes affecting migration of long-life radionuclides, such as Pu^{239}, Np^{237}, $U^{235,238}$, Am^{241} , shows that the processes under consideration are aimed at decreasing the isotope concentration in an aqueous phase. For rock mass with linear low permeability zones, the main processes which lead to the reduction in fracture-vein waters activity will be two-dimensional hydraulic dispersion (dilution) and radionuclide absorption by a solid phase (sorption, ion exchange) in pores and fractures. Of less importance are radioactive decay (not including Am^{241}) and dif-

fusion (both inside rock fragments in the crush zones and into monolithic rocks bounding linear tectonic disturbances).





The experimental studies conducted at the MCC are focused on obtaining hydrodynamic and geomigration parameters of the first two main processes.

Geomigration is studied by stages, interpretation of the results obtained at the first stage allowing the subsequent studies to be planned and implemented. The studies were initiated in linear tectonic zone (zones of fracturing and schist formation) oriented towards the Yenisey river and thus having a known direction of fracture-vein water flow. Additional experiments are planned for water-saturated and dewatered (but water permeable) fracturing zones. A complex of investigations on hazardous-filtration and migration is planned to be carried out in vertical and horizontal (or inclined) wells in water saturated zones.

A set of experimental hydrogeological wells have been drilled in Working B-4 within the zone of schist formation, the upper part of well log consisting of superimposed zones of mining-induced fracture and schist formation (Fig. 4). The set of experimental wells will be used for express-pumping, single-well tests with colored neutral trace indicator; indicator–electrolyte tests according to the method of charged body (to determine rate and direction of natural water flow) and resistivity metering method. Starting the neutral indicator in fracture-vein water flow and tracing it through observation wells will allow assessment of longtitudinal and lateral hydrodispersion. Followed by the determination of the values of coefficient of filtration, the parameters of longtitudinal and lateral hydrodispersion, and active fracture disturbance and natural flow rate, the solutions containing radioactive Pu^{239} and U^{238} will be introduced to obtain values of kinetic coefficients characterizing heterogeneous reaction of absorption.

SCHEME OF TEST BOREHOLE LOCATION IN WORKING B - 4

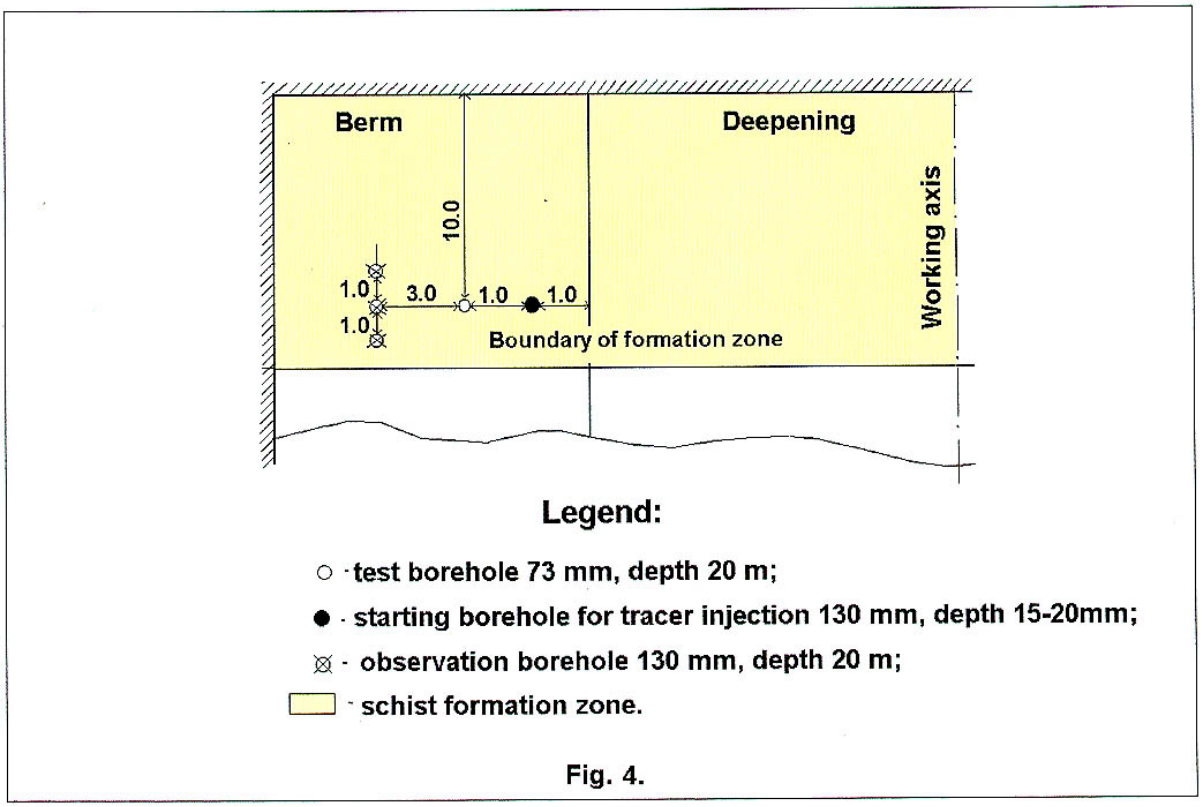
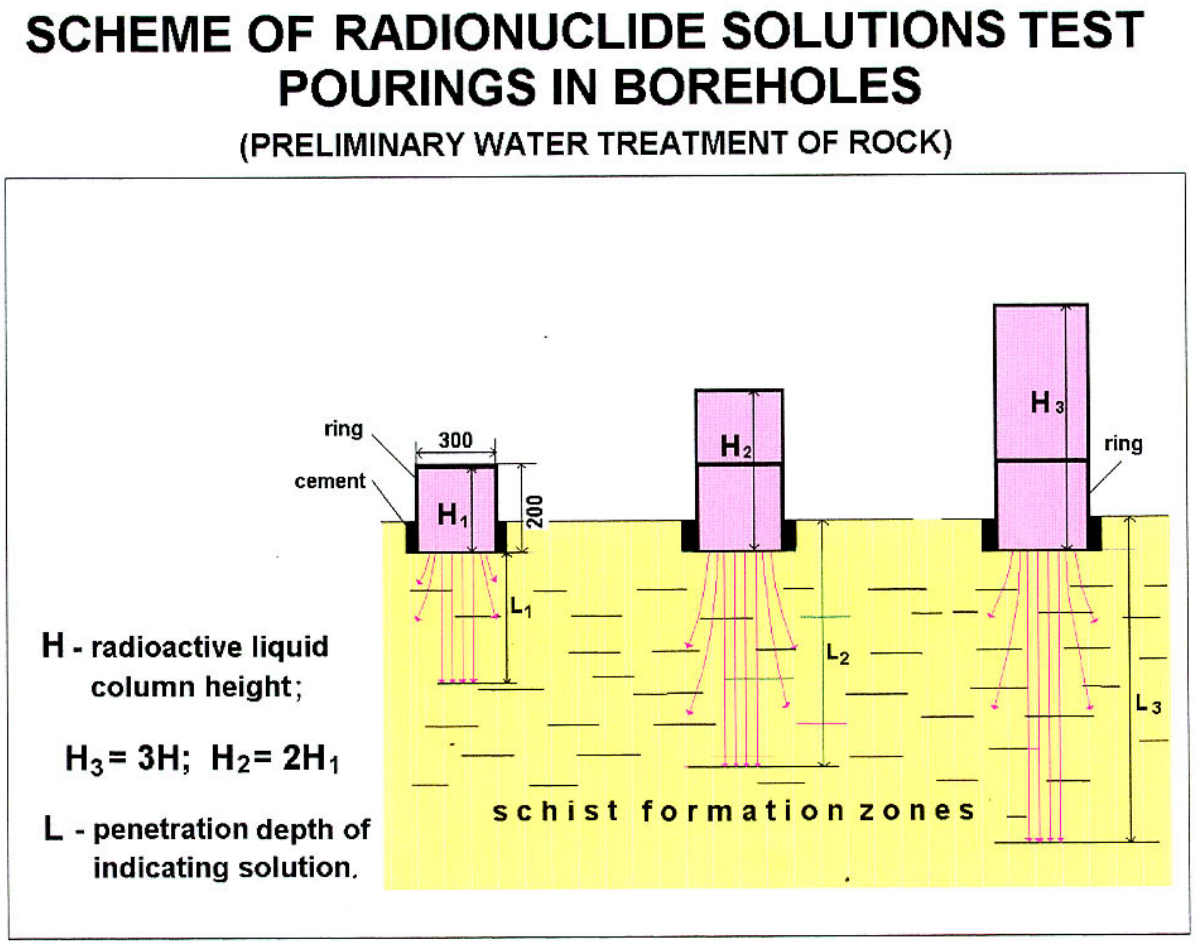


Fig. 4.

A similar series of experiments will be performed in horizontal wells drilled in the P-2 working wall at different elevations. In this case indicator will be introduced in vertical fracture-vein water flow.

A special set of tests in bore pits will be carried out in drained permeable fractured zones. The scheme of tests is given in Fig. 5. At the first stage the rock is watered up to stabilization of absorption discharge, then a neutral colored tracer with a real or analog radioactive solution containing complex contaminants is introduced. Then the bore pit bottom is drilled and the values of coefficient of hydrodispersion and sorption rate constants for each long-life radionuclide (Pu^{239}, Np^{237}, U^{238}) are calculated by the radioactive isotopes contents in a solid phase and core color. The test is repeated on the adjoining bore pits with the radioactive solution concentrations exceeding the first one by a factor of two and four. The experimental results are applied for empirical determination and static models creation. In case the solid phase proves to be saturated with radionuclide, then sorption coefficient for isotope can be determined. Otherwise, it can be determined by the additional laboratory tests.



Laboratory Studies

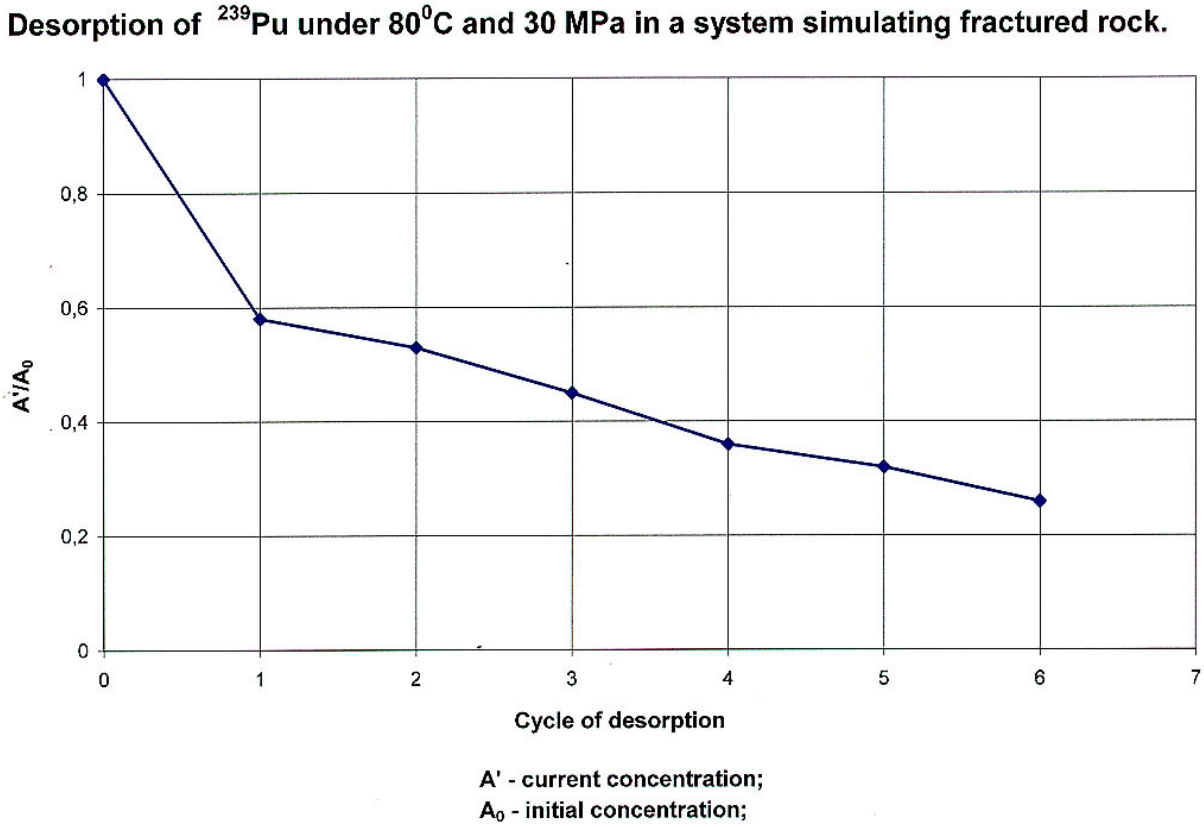
Laboratory scale tests of physical and chemical interaction of long-life migrating radionuclides with rocks of tectonic disturbance zones are carried out on cores of observation and trial wells in parallel to the field tests.

A series of experiments were performed to study Pu, Np and Am fixation strength on unaltered gneisses. The tests were conducted at laboratory installation which enables the conditions in crystalline rock mass at the depths down to 1000 m at elevated temperature (80^0 C) and pressure (30MPa) to be simulated to a certain extent. The water flow rate was assumed not to exceed 0.1 m/year.

In the course of the experiments, three 5 mm disks of the rock were placed into a volumetric compression chamber; both surfaces of the central disk sorbed plutonium in one series of the test, and Am and Np in a second series. In slits between the disks there was a film of the model water. Chamber heating up to 80^0C caused gradual increase of pressure up to 30 MPa. Portion of water between the disks was pressed out. Plutonium sorption on the central disk surfaces from water with the initial plutonium specific activity of $6 \cdot 10^6$Bq/l took place at 80^0C and atmospheric pressure.

Surface sorption capacity amounts to 850 Bq/cm^2. Desorption was effected at a ratio of water volume to disk surface area as 1.7:1 (cm) in several cycles. While studying radionuclide behavior under atmospheric and increased pressure, their distribution between the original disk,

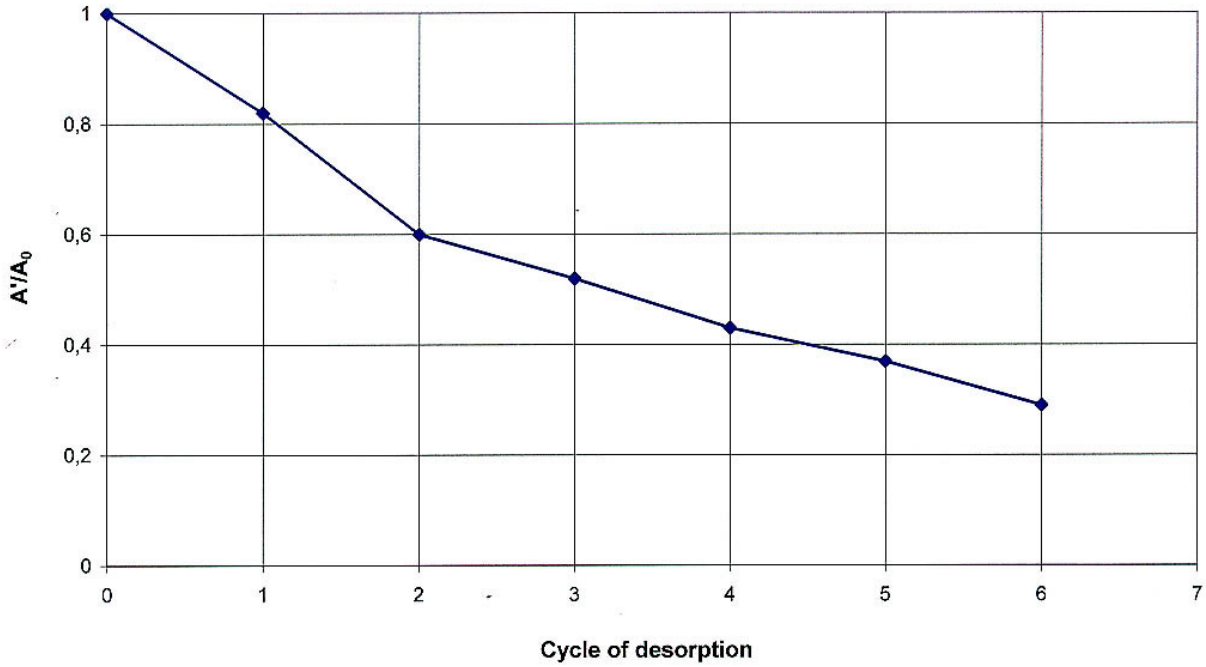
the clean disks and a liquid phase pressed out from fractures was determined. The test results are given in Fig. 6.



Comparison of the plots in this figure with the similar plots obtained under atmospheric pressure shows that plutonium desorption was intensified under high pressure, with general laws of the process remaining constant.

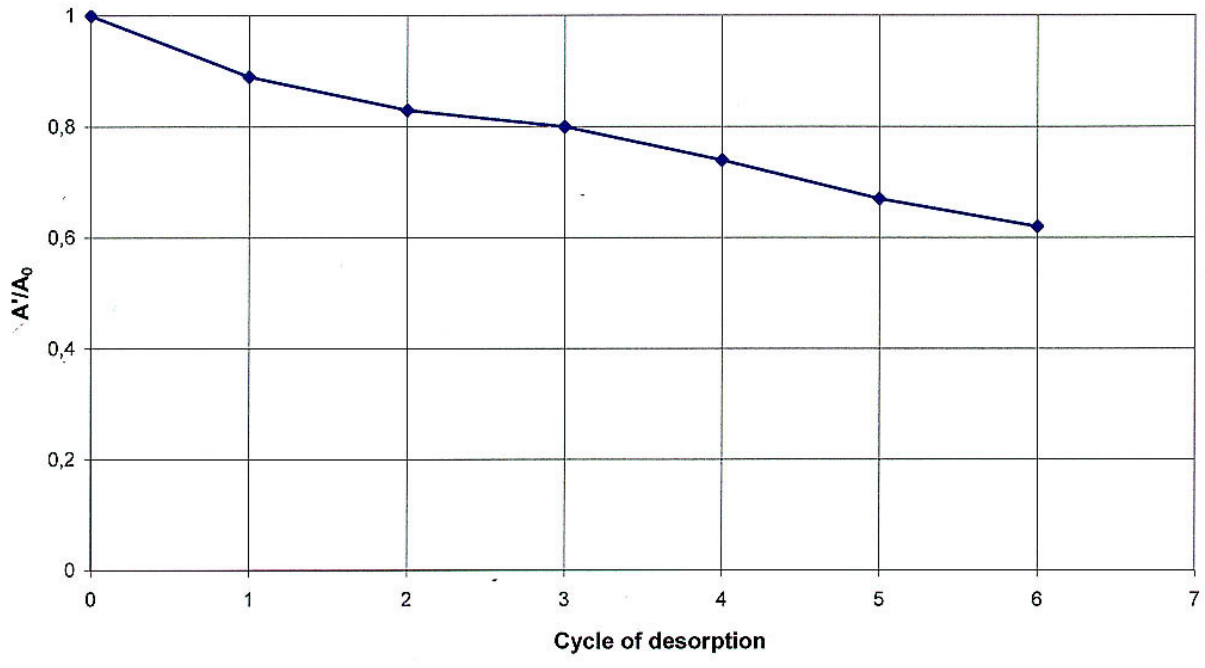
Am^{241} and Np^{237} fixation strength from water was studied simultaneously, with the initial specific activity of Np - $3.5 \cdot 10^5$Bq/l, and Am – $8.25 \cdot 10^5$ Bq/l. The surface sorption capacity amounted to 4 Bq/cm^2 and 144Bq/cm^2 respectively. The test results for desorption are shown in Fig. 7 and 8.

Desorption of ^{237}Np under 80^0C and 30 MPa in a system simulating fractured rock.



A' - current concentration;
A_0 - initial concentration;

Desorption of ^{241}Am under 80^0C and 30 MPa in a system simulating fractured rock.



A' - current concentration;
A_0 - initial concentration;

The results indicate that americium is more strongly sorbed on the gneiss surface; thus 35.8 % of neptunium and only 5.8 % of americium were recovered after the first desorption cycle. Under high temperature and pressure the behavior of neptunium is similar to that of plutonium.

Upon completion of all tests the gneiss disks were studied with the radiographic method and mineralogical analysis of specimens was made. The zones enriched in chlorite and biotite show the highest sorption capacity. It should be noted that the laboratory tests are preliminary. They represented radionuclide sorption on artificially-made surfaces.
Further laboratory and field-scale investigations on natural fracture surfaces healed with secondary minerals, will make it possible to run quantitative assessments of kinetic parameters for heterogeneous reactions in the radionuclide containing solutions with the rocks of tectonically disturbed zones. And finally, in situ tests will be performed on the rocks with naturally occurred crush zones, schist formation zones and artificial fracture disturbance zones which enable empirical model of complex radioactive contaminants migration to be obtained.

CONCLUSIONS

1. The Krasnoyarsk MCC underground facilities offers a unique opportunity to determine in situ hydrogeological parameters of tectonically disturbed zones of various gneisses and morphology in unweathered rocks.
2. Geomigration tests, both planned and in progress will enable the development of field-scale distribution models of long-life radionuclides (both determined and stochastic).
3. Laboratory scale tests allowed fixation strength of plutonium, neptunium and americium on gneisses of crush zones to be estimated. Americium was found to be more strongly fixed by rock, and neptunium and plutonium were less sorptive.
4. Surface sorption capacity of the intact gneisses at a high temperature (80^0C) and initial activity of $6 \cdot 10^6$Bq/l –Pu^{239}, $3.5 \cdot 10^5$Bg/l – Np^{237}, $8.25 \cdot 10^5$ Bq/l –Am^{241} was calculated at 850 Bq/cm^2, 4 Bq/cm^2, 144 Bq/cm^2, respectively.
5. In the future it is necessary to continue the tests on sorption-desorption of long-life radionuclides on natural rock surfaces of tectonically disturbed zones, considering that only the in-situ experiments will provide an opportunity for quantitative assessment of the field-scale migration.

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