APPLICATION OF NUCLEAR-PHYSICS METHODS FOR STUDYING THE RADIONUCLIDE TRANSPORT IN GRANITE ROCKS

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The results of studying the migration of ^{169}Yb as actinide-simulator in granite specimens in pristine state and preliminary irradiated with various γ-irradiation dose up to $3{,}0*10^9$ rad are represented and briefly discussed. Method of γ-spectrometry was used in order to determine the distribution of γ-emitting tracer isotope ^{169}Yb in granite. Estimations based on analysis of ytterbium bulk distribution are given to determine the diffusion coefficients of radionuclides in granite rock.

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1. INTRODUCTION

It is known that geological disposal of radioactive high-level wastes (HLW) or nuclear spent fuel will eventually result in contact between groundwater and the radioactive wastes upon failure of disposal metal container (e.g. mechanical breach or metal corrosion). Therefore, studying the mechanisms of radionuclides release from spent fuel and glass waste forms for HLW upon contact with aqueous solutions is being urgent now. These investigations usually include two directions: studying of oxidation and dissolution of specimens of really used UO_2-fuel or unirradiated chemical analogue of spent fuel - SIMFUEL (simulator of fuel) [1-5] and studying of radionuclide migration in natural rocks intended as repository material for radioactive waste disposal [6]. The main attention is paid to studying the behaviour of long-lived radionuclides and, in the first turn, actinides, e.g. U, Pu, Np, Am, Cm. It was shown in particular, that the amount of leached actinides in leach solution reduced when crushed granite was present under dynamic leaching tests in deionized water (Soxhlet method) and that concentration of radionuclides in solution is greatly influenced by sorption phenomenon in present of granite [3,4].

The closest to direction of used methods is the work of Japanese authors, describing the migration of radionuclides through granite of the local deposit [6]. Researches included the study of radionuclide migration by means of building penetration profiles of ^{233}U in granite during 58 and 252 days after contact with nitrate solution of uranium, containing ^{233}U as tracer isotope and simulator of actinides. Alpha-autoradiography was carried out to obtain the distribution of uranium concentration in granite matrix. It was shown that ^{233}U migration depends both on the mineral composition of natural granite (quartz, plagioclase, feldspar, biotite), and its internal structure, in particular, micro and macro porosity, micro and macro cracks. The influence of external radiation streams as well as absorbed doses on radionuclide migration in granite is not investigated there.

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It is known, that in case the spent fuel container fails, i.e. after a storage time less than 500 years, the radiation field surrounding the fuel will be constituted essentially by γ-radiation, but after storage time ≥ 500 years - by α-particles [7]. So, granite, in cases of abnormal destruction of the metal container after storage time ≤ 500 years in the first turn will be influenced mainly by γ-irradiation. Actinides dissolved from spent fuel or HLW will interact with granite rock, differing from intact natural granite rock by changing the structure and composition on account of magnitude of absorbed γ-dose.

The study of granites of Korostenian deposit from the Ukrainian crystalline shield is of most interest now because it is located near the "Shelter" unit and is being considered as a perspective place for disposal of long-lived radioactive wastes in Ukraine [8].

Here the results of the first experiments on investigation of the transport of ^{169}Yb in pristine and γ-irradiated (up to doses about 10^9 rad) granite are presented.

2. CONDITIONS OF EXPERIMENT

A piece of Korostenian granite was cut into a test specimens in the form of blocks with the sizes of 10x10 mm in cross section and 30 mm in length. Each block was covered with shellac adhesive besides one surface.

Following steps realized studying the migration of radionuclides as actinide simulators into granite matrix. Pellets of Yb_2O_3 were irradiated by bremsstrahlung obtained in Electron Linear Accelerator (ELA). In the course of irradiation the tracer ^{169}Yb-isotope was produced in accordance with reaction: ^{168}Yb (n,γ) ^{169}Yb (half-decay period $T_{1/2}$ = 30,7 days). Mass of ytterbium oxide pellet after irradiation was 0,1 g. Then the pellet was dissolved in concentrated HCl acid with the volume of 0,2 ml and finally the solution with pH = 1,8 was prepared. Obtained solution (with the volume of about 40 ml) was transferred into thermoresistance flask supplied with reverse motion refrigerator. Experimental granite blocks were placed inside flask. The flask was being heated by water steam within 32 hours. Then each of granite blocks was being washed in distillated water

within 24 hours and dried out at 60°C in the drying box. Then layers were removed by means of precision grinding of the uncovered surface of block (to escape the loss of tracer). The thickness of removed layers varied from 2 μm to 50 μm. Material of the removed layer was used for γ-spectrometry with the Ge(Li)-detector. ^{169}Yb gamma-intensity was measured within 20 min.

Studying of the radionuclide migration was being made on granite specimens in both natural pristine and γ-irradiated state. Irradiation has been carried out up to magnitudes of absorbed dose $0{,}3*10^9$, $1{,}0*10^9$ and $3{,}0*10^9$ rad, exposure times were respectively 7, 21 and 60 days. Realization of these severe conditions may be of greater interest to estimate chemical and radiation stability of granite matrix under conditions of radioactive waste disposal. The selected range of absorbed doses corresponds to some international standards on tests of radiation strength of protective materials of geological repositories (in particular, materials to be exposed to absorbed doses ranging from 10^5 to 10^{10} rad).

3. IRRADIATION TECHNIQUE

To realize the conditions of external γ-irradiation effect of the granite specimens, the bremsstrahlung of the ELA was used. Scheme of irradiation is shown on Fig. 1.

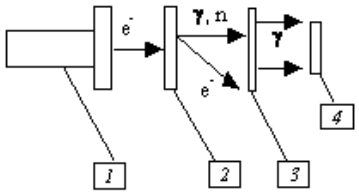

Fig. 1. *Scheme of γ-irradiation in electron accelerator: e^- -electrons, n-neutrons, 1 -electron accelerator, 2 - Ta-converter, 3 - filter for electrons and neutrons, 4 - granite specimen.*

The beam of electrons with energy of 20 MeV while going through converter (tantalum) generates the stream of γ-quantums, i.e. bremsstrahlung. Neutrons, which appear in converter ($\gamma + {}^{181}Ta \rightarrow {}^{180}Ta + {}^{1}n_0$) have isotropic distribution. The further placed system of filters (aluminium and paraffin) allows to reduce the electron and neutron components of the radiation beam, obtaining practically pure stream of γ-qauntums with average (by spectrum) magnitude of energy 2,0-2,3 MeV. The maximum magnitude of absorbed dose rate corresponds to ~ 10^6 rad/h. The similar scheme of irradiation is realized to get γ-emitting tracers on the base of ^{169}Yb used for γ-spectroscopy. The process of irradiation for this purpose was carried out up to the absorbed dose of 10^8 rad.

4. ANALYTICAL METHODS

Crystal-optical analysis with using of polarized microscopes and immersion liquids was done to study the crystalline structure of granite matrices before and after tests. Morphology of grains, grain sizes, grain boundary states and the extent of modification of internal structure were investigated.

5. RESULTS AND DISCUSSION

Characteristic γ-spectrums of tracer isotope ^{169}Yb in removed layers are shown in Fig. 2 and Fig. 3. It should be noted, that both spectrums are different in most extent due to appearance of gamma-activity lines of ^{84}Rb and ^{47}Ca, which is connected with induced activity of specimens during irradiation by bremsstrahlung. Thickness of removed layer regarded to Fig. 2 was about 2 μm. However, that was insufficient for measuring of induced activity in irradiated specimens. Therefore, further the layers with thickness more than 2 μm (i.e. 50 μm) were used with simultaneous measuring of their γ-spectrums.

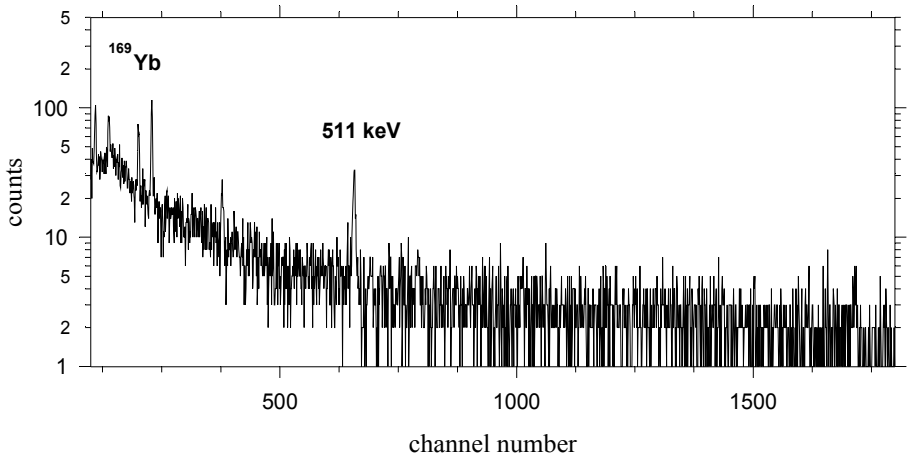

Fig. 2. *Typical γ-spectrum of natural granite.*

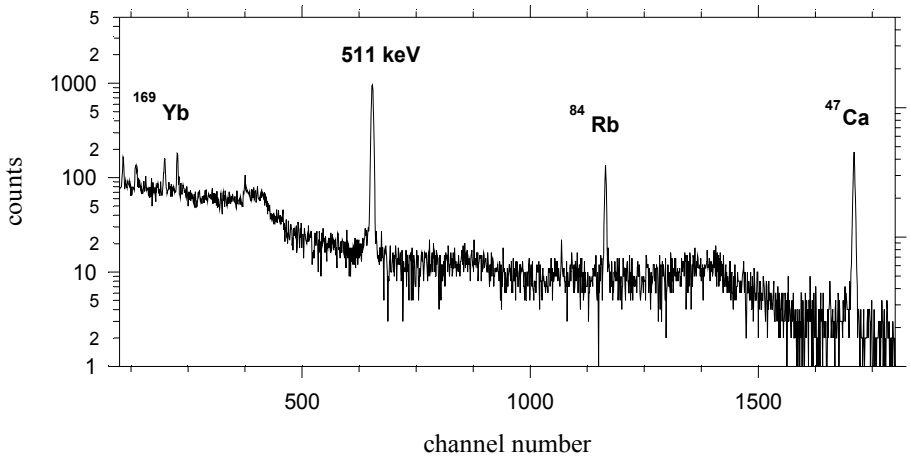

Fig. 3. *Typical γ-spectrum of irradiated granite (dose 3,0*10⁹ rad).*

The absolute magnitudes of ytterbium concentrations on each removed layer of pristine and irradiated specimens were determined by calibrated measurements of γ-spectrums with the use of ^{137}Cs as standard γ-emitted source. The maximum magnitude of ytterbium concentration was determined as $3{,}0*10^{20}$ at/cm^3 that corresponded to the first layer (i.e. thickness 50 μm) of block irradiated up to external dose of $3{,}0*10^9$ rad (Fig. 4.).

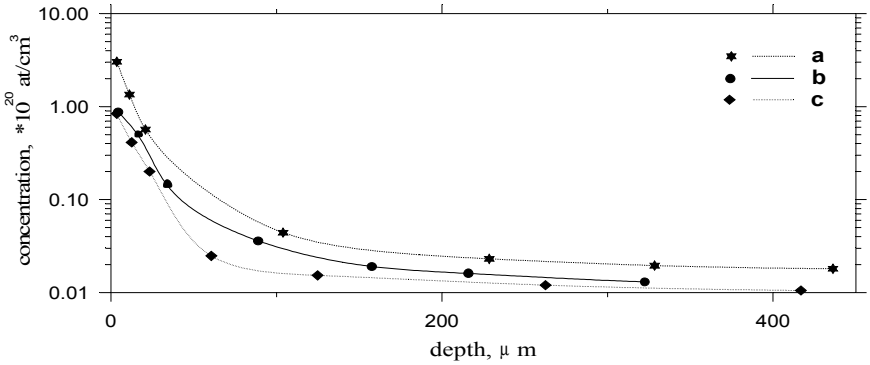

Fig. 4. *^{169}Yb concentration in specimens of granite;* ***a*** *- irradiated by dose of 3,0*10⁹ rad,* ***b*** *- pristine state with pegmatite structure,* ***c*** *- pristine state with uniform grain structure.*

Curves of ytterbium concentration on various specimens of granite blocks are shown in Fig. 4 as function of matrix depth. To explain character of the curves, one can give the following arguments. The total diffusant mass in a body matrix is known to be determined by both grain volume diffusion and grain boundary diffusion. The grain boundary diffusion is usually of importance for natural minerals [9]. Besides this, structure defects (pores, cracks) may also enhance the penetration of diffusant inside the matrix. This means that the diffusant enters the rock matrix far from the surface mainly along grain boundaries (under absence of obvious micro or macro defects). The both processes of volume grain and grain boundary diffusion may be described by the following two equations, respectively (1) and (2) [9]:

$$C(x,t) = \frac{C_0}{\sqrt{2\pi Dt}} \cdot \exp\left(-\frac{x^2}{4Dt}\right), \qquad (1)$$

C - tracer concentration;
C_0- initial tracer concentration;
D - diffusion coefficient;
x - distance from specimen surface.

$$C(x,t) = C_0 \cdot erfc\left(\frac{x}{2\sqrt{Dt}}\right), \qquad (2)$$

where

$$erfc(x) = 1 - \frac{2}{\sqrt{\pi}} \cdot \int_0^x \exp\left(-u^2\right) du,$$

$$u = \frac{x}{2\sqrt{Dt}}.$$

Based on the above-mentioned explanation, analysis of concentration profiles presented on Fig. 3 was carried out. Two characteristic regions of 0-60 μm and 60-400 μm were revealed. The first one is in a good agreement with classic grain volume diffusion (penetration inside the bulk of grains). It was determined by crystal-optical analysis that the grains of minerals, forming the granite rock, i.e. plagioclase and feldspars were exposed to interaction with tracer solution to a greater extent than grains of quartz. This may be explained by taking into consideration that in the natural granite rock

the quantity of microcracks in plagioclase and feldspars is higher than that in quartz.

Curves of profiles for natural unirradiated granite blocks (b, c, Fig. 4) are satisfactorily described by Eq. (1). Magnitudes of diffusion coefficients were $1,4*10^{-15}$ and $1,09*10^{-15}$ m^2/s respectively. On the other hand, curve of ytterbium concentration profile regarded to granite block irradiated to dose of $3,0*10^9$ rad is different from profiles regarded to natural pristine blocks. In this case, the characteristic profile view of irradiated block (a, Fig. 4) cannot be described by grain volume diffusion because Eq. (1) does not correspond to obtained experimental results. This can be explained by microstructure changes and accumulation of micro defects (pores, cracks) occurred under action of irradiation. Therefore, diffusion of ytterbium into deep granite block with depth up to 60 μm occurs mainly on these microdeffects. To achieve mathematical accordance, the correct coefficients in Eq. (1) for estimating diffusion coefficients were introduced. Average magnitude of ytterbium diffusion coefficient on depth from surface 0-60 μm for specimen of irradiated block was $3,2*10^{-15}$ m^2/s.

In the range 60-400 μm the concentration profiles for all specimens (both pristine and irradiated) have similar nature, connected with grain boundary diffusion, which is described quite precisely by Eq. (2). It is noteworthy that in irradiated granite specimens up to doses more than $1,0*10^9$ rad, the diffusion coefficient is 10^{-12} m^2/s that is higher than for classic grain boundary diffusion. Based on the results of crystal-optical analysis, one may suppose that it is connected with the modification of grains and change of condition of grain boundaries during irradiation.

6. CONCLUSION

The methods to study radionuclide (actinide simulators) migration under conditions simulating the situation of destruction of metal disposal container in the geological repository for spent fuel, further interaction of spent fuel with groundwater and transport of leached actinides into environmental geological medium are proposed. To create the simulated γ-irradiation on the side of nuclear spent fuel, the bremsstrahlung of electron accelerator is used.

Penetration profiles of ^{169}Yb (as actinide simulator) in specimens of natural granite rocks were obtained with the use of nuclear-physics methods. Diffusion characteristics for Yb-isotopes were estimated from the concentration profiles, taking into consideration the microstructure of granite rock. The observed diffusion coefficients for ytterbium were: for the depth 0-60 μm - about $1,09\text{-}1,4*10^{-15}$ m^2/s concerning unirradiated specimens and $3,2*10^{-12}$ m^2/s as to irradiated specimens; for the depth 60-400 μm - about $1,0\text{-}1,2*10^{-8}$ m^2/s that is practically same for both kinds of the specimens.

The radiation streams are shown to influence the process of radionuclide migration in granite on account of both changing of internal structure of rock matrix during irradiation and enhancing of grain volume diffusion. The magnitude of characteristic dose of γ-irradiation ($3,0*10^9$ rad), which is a reason for evident, increasing of radionuclide transport was determined. As such magnitudes of irradiation dose by spent fuel in real situation are hardly possible, the release of actinides on account of their transport in granite rocks will be determined first of all by the structure of natural granite.

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