INVESTIGATION OF ACTINIDE SIMULATORS MIGRATION IN GRANITE AND TUFF IRRADIATED BY GAMMA-QUANTA

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Granite and tuff specimens have been γ- irradiated to the absorbed doses ranging from 10^7 to $3.0 \cdot 10^8$ Gy. Penetration profiles of cerium ^{139}Ce (actinide simulator) in granite and tuff specimens (before and after γ-irradiation) were measured by means of nuclear physics methods. The irradiation has led to an increase in the cerium diffusion coefficients by several orders of magnitude. The irradiation to $3.0 \cdot 10^8$ Gy has caused the mechanical fracture of granite whereas the tuff specimens have retained their integrity while showing the appearance of microcracks.

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1. INTRODUCTION

It is known that in the course of geological storage of high-level radioactive waste (HLW), such as spent nuclear fuel, etc., a failure of the metallic container (e.g. due to a mechanical damage or corrosion) may give rise to the HLW contact with subsoil waters [1]. Therefore, it appears rather important to clarify the mechanisms and the rate of radionuclide penetration into the surrounding matrix. It is known that in cases of abnormal destruction of the metal container after storage time ≤ 500 years, granite (or tuff) matrix, will be subjected mainly to γ-irradiation [2].The structure of irradiated rock will be different from that of intact natural rock resulting in the difference in actinide penetration rates depending on the γ-irradiation dose.

It was shown [3] ^{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, cracks. However, the influence of γ-irradiation on radionuclide migration in granite or tuff matrices has not been investigated to our knowledge.

The aim of the present work is to investigate the migration of ^{139}Ce (the actinide simulator) in the natural granite and tuff matrices γ-irradiated to different absorbed doses ranging from 10^7 to $3.0 \cdot 10^8$ Gy.

2. EXPERIMENTAL PROCEDURE

2.1. STARTING MATERIALS

Cores of granite species from the Yantsev deposit of the Ukrainian crystalline stratum and tuff species from the Yucca Mountain deposit (Nevada, USA) were used as materials to study. The cores were cut into experimental specimens with dimensions of 10×10×30 mm. The specimens were exposed to bremsstrahlung γ-irradiation produced at the electron linear accelerator.

2.2. IRRADIATION TECHNIQUE

As the 10 MeV electron beam passes through a tantalum converter, it generates γ-quanta and neutrons produced in the reaction ($\gamma + {}^{181}\mathrm{Ta} \rightarrow {}^{180}\mathrm{Ta} + n$). The filter system (aluminum and paraffin) provides a substantial decrease in the electron and neutron constituents of the converted beam so that practically a pure γ-quantum beam with average energy of 2.0…2.3 MeV is produced. The maximum absorbed dose rate in the present study was about 10^4 Gy/h. Temperature of granite/tuff specimens during irradiation was about 50°C.

2.3. ANALYTICAL METHODS

The migration ^{139}Ce deep into the rock matrix was investigated by several steps. First, the γ-tracer of ^{139}Ce was obtained using the following activation technique. Cerium oxide pellets (CeO_2) were irradiated with bremsstrahlung γ-quanta. In the course of irradiation, cerium isotopes (γ-tracers) were produced in the reaction $^{140}\mathrm{Ce}(\gamma,n) \rightarrow {}^{139}\mathrm{Ce}$ (the half-decay period, $T_{1/2}$=140 days). Then the pellets were dissolved in a concentrated acid H_2SO_4 to make eventually the solution with pH=1.8. The specimens under investigation (before and after irradiation) were placed in the solution and held for 180…550 hours at the room temperature. After that, the precise polishing was used to remove the material (layer by layer) from a free (uncoated with a protective compound) surface of the specimen. The thickness of the removed layers varied from 2 to 50 μ. The activity of the removed layer was measured with the use of a Ge(Li) γ-spectrometer. To estimate the migration of ^{139}Ce, the spectra obtained in the measurements were used to plot the distribution of γ-tracer concentration in the depth of the specimen. The coefficients of cerium diffusion into the rock matrix were calculated using the obtained concentration profiles.

Crystal-optical analysis was used to study the crystalline structure of granite and tuff matrices before and after irradiation.

3. RESULTS AND DISCUSSION

3.1. UN-IRRADIATED MATERIALS

It is known that the penetration of a diffusant (in our case, cerium) into the other material is determined either by the grain bulk or grain boundary diffusion mechanisms [4]. These two mechanisms of diffusion (bulk and grain boundary) are described by equations (1) and (2), respectively [5]:

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

where C is the tracer concentration; C_0 is the initial tracer concentration, D is the diffusion coefficient, x is the distance from the specimen surface, t is the time.

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

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

Two characteristic regions, namely, 0 to 60 μ and 60 to 900 μ, have been established during the analysis of the curves showing the concentration distribution of cerium in the depth of the matrix (Fig.1).

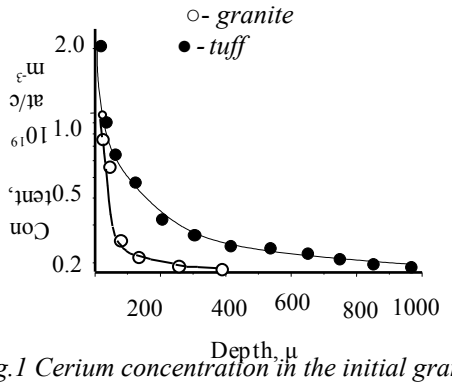

Fig.1 Cerium concentration in the initial granite and tuff samples

The first region is adequately described by Eq.(1). This means that the main mechanism of cerium diffusion both in granite and tuff up to a depth of 60 μ is the bulk diffusion, possibly modified by the presence of structural microdefects (pores and cracks). The average effective diffusion coefficients of cerium in the initial granite and tuff specimens at a depth of 0…60 μ were found to be $1.2 \cdot 10^{-15}$ m^2/s and $2.9 \cdot 10^{-15}$ m^2/s, respectively.

In the 60…900-μ range, the cerium concentration distribution for all granite and tuff specimens is better described by Eq.(2), and so the penetration of cerium in this region is controlled by the grain boundary diffusion mechanism. The average effective diffusion coefficients of cerium diffusion were found to be ~$2.5 \cdot 10^{-13}$ and $1.6 \cdot 10^{-10}$ m^2/s for granite and tuff, respectively. Note that the grain boundary diffusion is much faster in tuff than in granite due to the higher porosity of tuff, which has been confirmed by the crystal-optical analysis.

3.2. IRRADIATED MATERIALS

The variations in the state of boundaries of natural materials under irradiation may substantially facilitate the penetration of cerium by the grain boundary diffusion mechanism. Besides, the structural defects formed under irradiation in the bulk (pores and cracks) may also facilitate the bulk diffusion. These expectations have been confirmed in our experiments.

Comparison of Figs.1 and 2 shows that after irradiation dose of 10^7 Gy (1 Grad), both granite and tuff specimens show substantial increase in the cerium penetration.

For irradiated granite specimens, the average effective diffusion coefficient was found to be $7.5 \cdot 10^{-14}$ m^2/s (initial $1.2 \cdot 10^{-15}$ m^2/s) and $4.2 \cdot 10^{-8}$m^2/s (initial $2.5 \cdot 10^{-13}$ m^2/s) at penetration depths up to 60 μ and 1600 μ, respectively.

For irradiated tuff specimens, the average effective diffusion coefficient was found to be $6.2 \cdot 10^{-14}$ m^2/s (initial $2.9 \cdot 10^{-15}$ m^2/s) and $2.5 \cdot 10^{-8}$ m^2/s (initial $1.6 \cdot 10^{-10}$ m^2/s) at penetration depths up to 60 μ and 1800 μ, respectively.

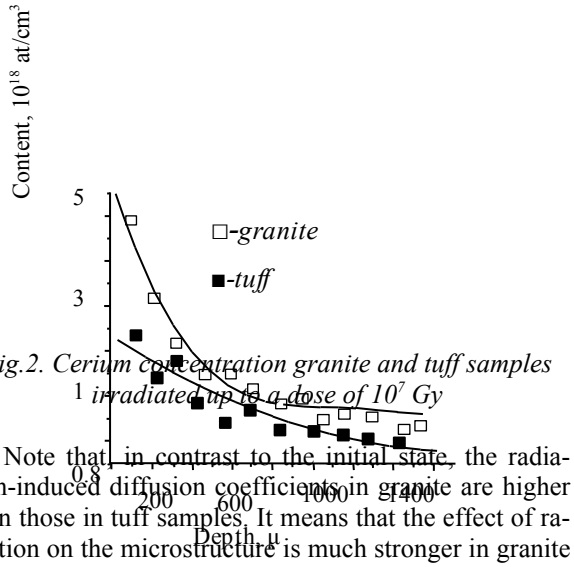

Fig.2. Cerium concentration granite and tuff samples irradiated up to a dose of 10^7 Gy

Note that, in contrast to the initial state, the radiation-induced diffusion coefficients in granite are higher than those in tuff samples. It means that the effect of radiation on the microstructure is much stronger in granite than in tuff. This conclusion has been confirmed by further irradiation up the maximum absorbed dose of $3.0 \cdot 10^8$ Gy.

At this dose, the coefficients of bulk and grain boundary diffusion in tuff were found to be $3.16 \cdot 10^{-14}$ m^2/s, and $1.26 \cdot 10^{-8}$ m^2/s, respectively, which were comparable with those obtained after the irradiation dose of 10^7 Gy. So, there is a tendency to saturation of diffusion coefficient growth with irradiation dose increasing beyond 10^7 Gy. The tuff specimens have retained their integrity, although the appearance of microcracks has been revealed by the crystal-optical analysis.

In contrast to that, under γ-irradiation to the maximum dose of $3.0 \cdot 10^8$ Gy, structural changes in the granite specimens have caused their mechanical fracture into smaller pieces. The penetration of cerium through the fractured granite specimens has not been measured but evidently it should be much higher than the penetration through the whole tuff matrix.

3.3. ESTIMATION OF THE ACTINIDE PENETRATION IN TUFF SURROUNDING HLW

In our experiments, cerium is the analog of actinides, namely, plutonium. We have used the radiation-induced Ce diffusion coefficients obtained in our experiments, $D = 1.26 \cdot 10^{-8}$ m^2/s, in order to estimate the penetration depth of plutonium isotopes in natural tuff massifs. The time dependence of the radionuclide penetration depth is given by $\lambda_{diff}(t) = 2 \cdot (Dt)^{1/2}$ [5]. The characteristic time, during which the actinides preserve the radioactivity, is usually taken as $10 \cdot T_{1/2}$, where $T_{1/2}$ is the half-decay period. The estimated values of $\lambda_{diff}(10 \cdot T_{1/2})$ for different Pu isotopes are different due to the differ-

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ence in their $T_{1/2}$, which are as follows (in years): 14.4 (^{241}Pu), 87.7 (^{238}Pu), 6537 (^{240}Pu) and 24000 (^{239}Pu). Accordingly, $\lambda_{diff}(10 \cdot T_{1/2})$ is given by (in meters) 15.13 (^{241}Pu), 37.34 (^{238}Pu), 322.34 (^{240}Pu) and 617.62 (^{239}Pu).

Therefore, the recommended distance of the HLW storage place from the ground surface in tuff is about or larger than 600 m.

4. CONCLUSION

Penetration profiles of cerium ^{139}Ce (actinide simulator) in the granite and tuff specimens before and after γ-irradiation have been measured by means of nuclear physics methods. On the basis of the measured cerium concentration distribution in the depth of specimens, cerium diffusion parameters have been estimated.

It is shown that the bulk diffusion modified by the presence of structural microdefects (cracks and pores) is the dominant mechanism of cerium diffusion to a depth of 60 μ in both granite and tuff. In the 60…900 μ range, the penetration of cerium is controlled by the grain boundary diffusion mechanism.

The grain boundary diffusion of cerium is shown to be much faster in tuff than in granite due to the higher porosity of tuff.

The irradiation of natural material specimens to doses of 10^7 Gy has led to an increase in the diffusion coefficients, which became larger in the granite than in tuff samples. It means that the effect of irradiation on the microstructure is much stronger in granite than in tuff.

A tendency towards saturation of diffusion coefficient growth in tuff with irradiation dose increasing beyond 10^7 Gy has been noted. The tuff specimens have retained their integrity under γ-irradiation to the maximum dose of $3.0 \cdot 10^8$ Gy, although the appearance of microcracks has been revealed.

In contrast to that, under γ-irradiation to the maximum dose of $3.0 \cdot 10^8$ Gy, structural changes in the granite specimens have caused their mechanical fracture.

The recommended distance of the HLW storage place from the ground surface in tuff is about or larger than 600 m.

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ИССЛЕДОВАНИЕ МИГРАЦИИ ИМИТАТОРОВ АКТИНИДОВ В ГРАНИТЕ И ТУФЕ, ОБЛУЧЕННЫХ ГАММА-КВАНТАМИ

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Образцы гранита и туфа были облучены гамма-квантами до доз 10^7…$3.0 \cdot 10^8$ Гр. С помощью ядерно-физических методов получены профили проникновения церия ^{139}Ce (имитатора актинидов) до и после облучения. Облучение привело к значительному увеличению коэффициентов диффузии церия. Облучение до дозы $3.0 \cdot 10^8$ Гр привело к механическому разрушению образцов гранита. Образцы туфа сохранили свою целостность, но в них выявленны нарушения структуры за счет появления микротрещин.

ДОСЛІДЖЕННЯ МІГРАЦІЇ ІМІТАТОРІВ АКТИНІДІВ У ГРАНІТІ ТА ТУФІ, ОПРОМІНЕНИХ ГАМА-КВАНТАМИ

В.І. Дубінко, А.М. Довбня, С.Ю. Саєнко, М.П. Дікий, Е.П. Шевякова, В.М. Грицина, А.Є. Сурков, О.П. Березняк, Ю.В. Ляшко, О.П. Медведєва

Зразки граніту і туфу було опромінено гамма-квантами до доз 10^7…$3.0 \cdot 10^8$ Гр. За допомогою ядерно-фізичних методів отримані профілі проникнення церію ^{139}Ce (імітатора актинідів) до і після опромінення. Опромінювання зразків природних матеріалів привело до значного збільшення коефіцієнтів дифузії церію. Опромінення до дози $3.0 \cdot 10^8$ Гр привело до механічного руйнування зразків граніту. Зразки туфу зберегли свою цілісність, але у них виявлені порушення структури за рахунок появи мікротріщин.