USE OF ELECTRON LINAC FOR STUDY OF FISSION PRODUCT AND ACTYNIDE DIFFUSION THROUGH GLASS CERAMIC MATRICES

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The diffusion of cerium and uranium was investigated in glass ceramic matrices obtained in the gasostat. For activation of isotopes ^{140,142}Ce and ^{238}U the brake radiation from electrons of the linear accelerator was used. The threshold of detectability of elements was reached from 5 to 15 μg/g for a sample of the area 1 cm^2. The coefficients of diffusion in the grain material and at grain boundaries were measured, and their values for the pressing temperature of 910°C and pressure 100~MPa were $3.5\cdot10^{-9}$ cm^2/s in the grain and $1.2\cdot10^{-8}$ cm^2/s at grain boundaries.

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INTRODUCTION

The treatment with the spent nuclear fuel now assumes its storing at nuclear power plants (NPP) from 30 until 50 years with the subsequent disposal in deep geological media. At a disposal in deep geological rocks of a special importance are long-lived fission products: ^{99}Tc ($T_{1/2}$=2,1 10^5 years), ^{129}I ($T_{1/2}$=1,6 10^7 years), ^{135}Cs ($T_{1/2}$=2,3 10^6 years), ^{93}Zr ($T_{1/2}$=1,5 10^6 years), ^{126}Sn ($T_{1/2}$=10^5 years), ^{79}Se ($T_{1/2}$=6,5 10^4 years), ^{137}Cs ($T_{1/2}$=30,2 years), ^{90}Sr ($T_{1/2}$=28 years) and actinides ^{229}Pu ($T_{1/2}$=2,4 10^4 years), ^{240}Pu ($T_{1/2}$=6,5 10^3 years), ^{242}Pu($T_{1/2}$=3,8 10^5 years), ^{241}Am ($T_{1/2}$=432 years), ^{244}Cm ($T_{1/2}$=18 years). ^{99}Tc, ^{129}I, ^{135,137}Cs are the most toxic elements. They are dissolved in water, easily get into a biological cycle and can collect in various fabrics of organism. A burial volume of these elements reaches up 90% [1].

The failure of metal containers can result in release of fission products and actinides into environment. One of the promising approaches to preparation of NPP radioactive waste (RW) for a long disposal is their immobilization in glass ceramic matrices based on natural minerals (granite, kaolin etc.). The immobilization represents formation of protective matrice with the help gasostates. Therefore the study of migration of radionuclides in the process of gasostatic pressing is the important criterion of reliability of manufacturing the protective barriers. To study the carrying of the RW in the process of gasostatic pressing the high-effective nuclear-physical methods can be used [2-3]. The authors of [4] investigated uranium diffusion in granite. The penetration profiles of ^{233}U were obtained. The duration of the experiment because of a low specific activity of the given isotope was almost year and the problems of recycling radioactive waste were accompanied.

The use of nuclear-physical methods to study the diffusion of fission products and actinides in protective glass ceramic matrices is the purpose of the present work.

EXPERIMENTAL SET-UP AND METHODS

The interaction of oxides of uranium, cerium etc., in samples of ceramics obtained by hot pressing a material containing 70% of granite +30% kaolin is investigated. The given system was placed in gasostate for 3-5 hours at temperature 910°C and pressure 100 MPa. After mechanical removal of the tablet-tracer the glass ceramic was irradiated on a brake radiation of the powerful 23 MeV electron accelerator with a current of 700 μA. Further the sample layers were removed without loss of activity. The thickness of the removed layers varied from 2 up to 50 microns. Activity of the removed ceramic layer was measured with the help of Ge(Li)-detector of a volume 50 cm^3 and energy resolution 2.8 keV for a gamma radiation with energy 1333 keV. The reached threshold of detectability of elements was from 5 to 15 μg/g for a sample by the area 1 cm^2 [7-9].

RESULTS AND DISCUSSION

The typical spectrum of activity of the removed layer is given in Fig.1. The gamma radiation of elements Zr, Y, Rb, Ca, Na etc. is visible.

The diffusion of actinides was studied for various compositions of glass ceramic. In Fig.2 the distribution of cerium and uranium in the glass ceramic matrix from the sample consisting of 40% UO_2 and 60% CeO_2 is shown. Such a content of the matrix was chosen with the purpose of imitation of the behaviour of most toxic radionuclide – plutonium. For the given conditions of synthesis cerium on the is similar by behaviour to plutonium.

The calculation of diffusion of elements in a matrix was carried out by expression [5]:

$$\frac{\bar{C}(y)}{C_0} = erfc\left(\frac{\eta}{2}\right) + \frac{\eta\sqrt{D_{ma}t}}{b\pi}\chi$$

$$\chi \int_1^\infty \frac{d\sigma}{\sigma^{3/2}} \exp\left(-\frac{\eta^2}{4\sigma}\right)\left\{e^{-X^2} - \sqrt{\pi} * X * erfc(X)\right\}$$

$$X = \frac{1}{2}\frac{\sigma - 1}{\beta}, \qquad D_{ma}=D_m/(1+\rho_m\cdot Kd/\varepsilon_m), \qquad \eta = \frac{y}{\sqrt{D_{ma}t}},$$

$$\beta = \frac{\Delta}{\sqrt{D_{ma}t}}, \quad \Delta = \frac{aD_{fa}}{D_{ma}}, \quad D_{fa}=D_{fa}/(\varepsilon_m+\rho_m K_d),$$

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where a – fissure half-width, b – fissure half-interval, C_o– initial tracer concentration of solution, D_f – diffusion coefficient in the fissure, D_{fa} – apparent diffusion coefficient in a fissure, D_{ma} - apparent diffusion coefficient in the water pore of the matrix, K_d – adsorption coefficient, ε_m – porosity of matrix, ρ_m – density of matrix, y - distance from the surface of specimen in the direction of depth.

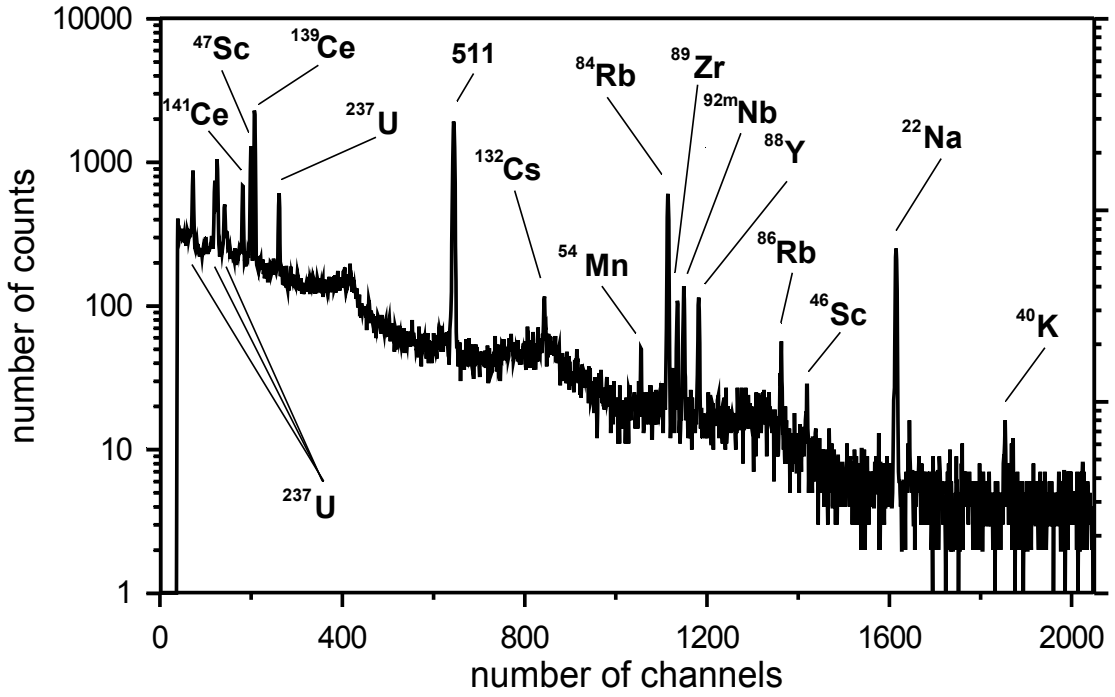

Fig. 1. Gamma - spectrum of activity of the removed layer

The first term of the right-hand part of the equation represents a direct diffusion into the grains and is a function of y only. The second term represents the contribution of diffusion through the fissure.

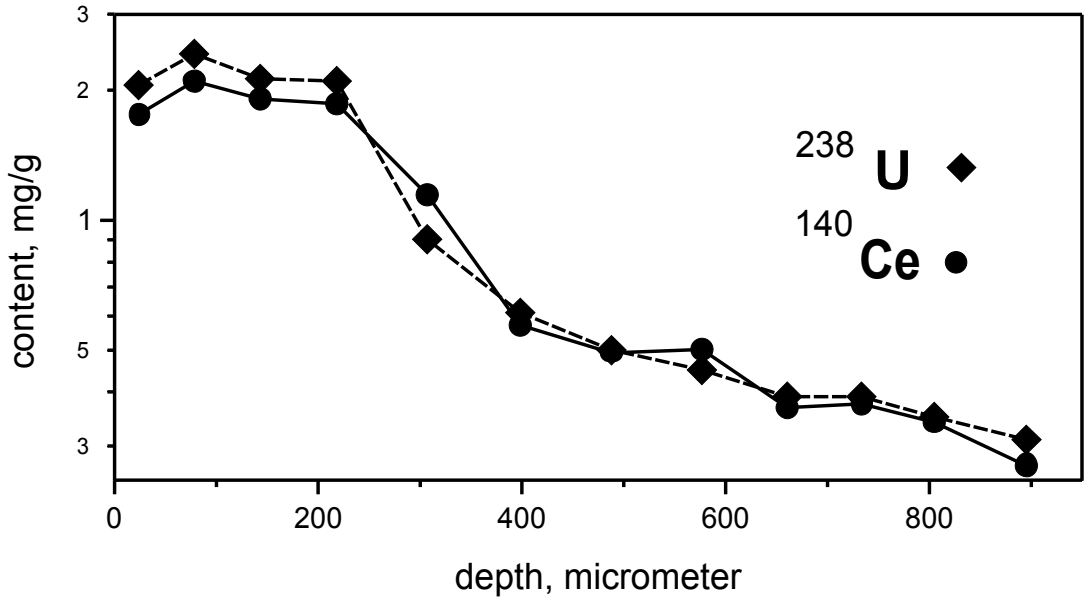

Fig. 2. The contents of uranium and cerium on the depth of glass ceramics

The coefficient of diffusion was determined by means of function minimization [5]:

$F=(\Sigma(y_{th}-y_{exp})^2/(k-s)$,

where y_{th}, y_{exp} - theoretical and experimental values of reaction yields, respectively, k - number of measurements, s - number of connections (number of degrees of distribution). The minimal function value was realized

for the value b=50 μm (radius of a grain). The values of coefficients for the given condition, given in Fig.2 was $3.5 \cdot 10^{-9}$ cm^2/s for diffusion in the grain and $1{,}2 \cdot 10^{-8}$ cm^2/s for diffusion through the fissure.

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ИСПОЛЬЗОВАНИЕ ЛУЭ ДЛЯ ИЗУЧЕНИЯ ДИФФУЗИИ ПРОДУКТОВ ДЕЛЕНИЯ И АКТИНОИДОВ В СТЕКЛОКЕРАМИЧЕСКИХ МАТРИЦАХ

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Методом снятия слоев изучена диффузия церия и урана в синтезированные посредством газостатического прессования стеклокерамические матрицы. Для активации изотопов $^{140,142}Ce$ и ^{238}U использовалось тормозное излучение от электронов линейного ускорителя. Предел обнаружения изотопов церия и урана составил 10^{-5} г/г. Были измерены коэффициенты диффузии в материале зерна и по границам, которые при температуре прессования 910 ^0C и давлении 100 МПа составили $3{,}5 \cdot 10^{-9}$ $см^2/с$ в зерне и $1{,}2 \cdot 10^{-8}$ $см^2/с$ по границам зерен.

ВИКОРИСТАННЯ ЛПЕ ДЛЯ ВИВЧЕННЯ ДИФУЗІЇ ПРОДУКТІВ ДІЛЕННЯ ТА АКТИНОЇДІВ У СКЛОКЕРАМІЧНИХ МАТРИЦЯХ

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Методом зняття шарів вивчена дифузія церію й урану в синтезовані за допомогою газостатического пресування склокерамічної матриці. Для активації ізотопів $^{140,142}Ce$ і ^{238}U використовувалося гальмове випромінювання від електронів лінійного прискорювача. Межа виявлення ізотопів церію й урану становила 10^{-5} г/г. Були вимірювані коефіцієнти дифузії в матеріалі зерна і по границях, що при температурі пресування 910^oC і тиску 100 МПа склали $3{,}5 \cdot 10^{-9}$ $см^2/с$ у зерні і $1{,}2 \cdot 10^{-8}$ $см^2/с$ по границях зерен.