APPLICATION OF A PROTON MICROPROBE TO STUDY DIFFUSION PROFILES OF Ce IN SYNTHETIC ALUMINOSILICATES

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The procedure of measuring the diffusion profiles of Cerium by means of a proton microprobe was developed. These profiles in the synthetic aluminosilicate matrix synthesized on the base of kaolin and red granite were measured.

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1. INTRODUCTION

The methods of nuclear physics are widely applied for investigation of materials on accelerated particle beams. In the present work these methods were used to study glass ceramics, which can be applied for disposal of radioactive waste (RAW).

Development of nuclear power in Ukraine is accompanied by the RAW accumulation. According to the forecasts for 2025 a total volume of RAW to be stored in Ukraine will be approximately 70000 m^3 [1]. In this connection in Ukraine a conception for construction of a geological RAW depository has been adopted. Before disposal of high-level radioactive wastes into the underground depository it is necessary to reprocess wastes with subsequent their immobilization in the matrix from a material based on natural rocks.

For immobilization of high-level RAW it is intended to apply synthetic aluminosilicates (glass ceramics) as a promising material. Long-term forecasting of the rate of radioactive element release from the geological depository with the use of such materials requires carring out studies on the processes of radionuclide diffusion and migration in synthetic aluminosilicates. Generally, for such investigations Cerium is used as an actinide imitator. In paper [2] the diffusion profiles of Cerium in glass ceramics were measured by sequential removal of layers from the material sample with measuring the gamma-spectra from the material of each layer after gamma-activation.

There are available also other methods of profile measuring deeply in the depth (tens and hundreds of microns). In particular, the investigations can be carried out by means of a microprobe (see, for example, [3-8]).

The goal of the present work is to develop the procedure of measuring the diffusion profiles of Cerium by means of a proton microprobe and to measure these profiles in the synthetic aluminosilicate matrix synthesized on the base of kaolin and red granite.

2. MATERIALS AND METHODS

The procedure of sample preparation for investigations consists in the following. The samples of granite and kaolin were milled in the mechanical vibratory mill to obtain a powder with a particle size less than 40 μm. The weighted powder samples were filled into the press-mould. The press-mould and the movable dies were made of graphite. To prevent the interaction between the material of the filler and the material of the crucible the latter was lined with a molybdenum foil. After that, the powder filler was subjected to vibrational compaction on the vibration-testing machine and to pressing at room temperature under pressure of 40 MPa. Sintering of samples under pressure was realized by direct passing of the electric current through the press-mould. Hot pressing was carried out at a temperature of 1050°C under pressure of 40 MPa. The treatment duration was 15 min.

The aluminosilicate samples were obtained in the form of a pellet. The material density was 2.5… 2.7 g/cm^3. By the same method using the CeO_2 powder we sintered a pellet of the imitator material containing Cerium, i.e. the element imitating Uranium and transuranium elements when studying their mass transfer.

For simulation of the processes of mass transfer two pellets (of aluminosilicate and material-imitator) were placed in interior of the metallic capsule where they were brought into close contact with polished surfaces. The capsules were sealed by the electric welding and after subjected to the gasostatic pressing at a temperature of 920°C during 30 min.

After treatment in the gasostatic press these capsules with the pellets of aluminosilicate and cerium oxide were annealed in the temperature range from 600 to 750°C in vacuum during 10 hours. As a result of annealing some amount of Ce from Cerium oxide pellet were migrating throught the interface into the depth of

the aluminosilicate pellet. The pellets were drawn out from the pressed capsule with the help of a diamond tool. Then the pellets were separated and cleaved in the direction transversal to the former interface of the pellets.

To measure the diffusion distribution of Cerium a proton microprobe based on the duplet of magnetic quadrupole lenses [9,10] and an X-ray spectrometer were used. A 10-micron proton beam with energy of 1.8 MeV and a current of 2 nA was applied.

Beam monitoring was performed by counting of protons backscattered from the target periodically overlapping the beam of protons (interruption frequency was of about 1 s^{-1}). The monitoring unit was placed between the diaphragms of the collimator forming the beam before the port into the duplet of magnetic qudrupole lenses. The X-ray spectra were measured using the detector made of high-pure germanium of BDRG-05165 type (product of the firm "Baltic Scientific Instruments").

The detector was arranged at an angle of 135° relatively to the beam direction. The distance from the sample to the detector was 20 mm.

The spectrometric path was chosen basing on standard modules "VECTOR". The resolution of the spectrometer was 155 eV along the Mn K_α line. Spectra were recorded by the computer-added spectrometric amplitude-digital converter ADC-8K-B2 produced by the firm "Aspect".

The profile of Cerium distribution in the aluminosilcate matrix was measured by moving discretely the microprobe beam in the direction of the transverse cleavage of the surface from the former interface into the depth of the pellet material. After beam extraction into the given point of cleavage the X-ray spectrum was measured. A similar technique of profile measurement was applied earlier in works [3,4]. In this case the Cerium concentration at the corresponding depth, counted from the separation surface, was determined along the Ce L_β line. A pellet of compacted Cerium oxide was used as a standard.

Fig. 1 demonstrates the cleavage surface of the aluminosilicate sample and the area of measurement of local Cerium concentrations.

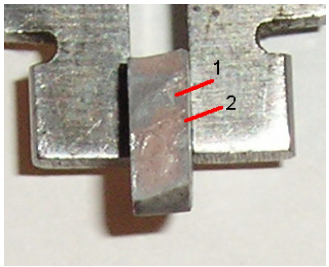

Fig. 1. *1 - the cleavage surface of the alumino-silicate sample (×5). 2 - the area of measurement of local Cerium concentrations*

3. DISCUSSION OF RESULTS

Fig. 2 shows the characteristic X-ray spectrum obtained from the cleavage surface of the aluminosilicate pellets after separating it from the pellet-imitator of CeO_2. In the spectrum observed are the characteristic lines of such elements as Cl, K, Ca, Ti and Fe contained in the composition of the initial matrix, and the lines of Cerium diffused into aluminosilicate from the imitator material.

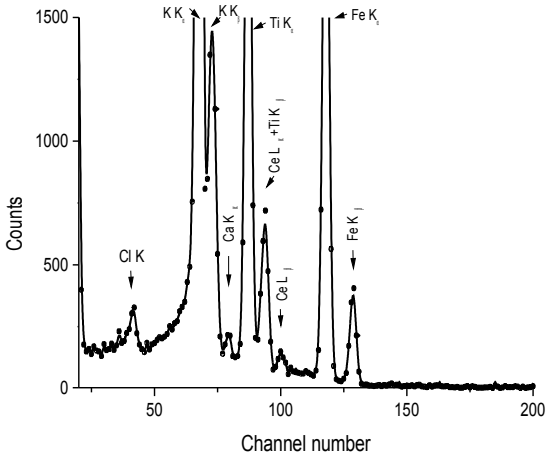

Fig. 2. *The characteristic X-ray spectrum obtained from the aluminosilicate pellet*

The diffusion profile of Cerium in the aluminosilicate matrix measured with help of the proton microprobe is presented in Fig. 3.

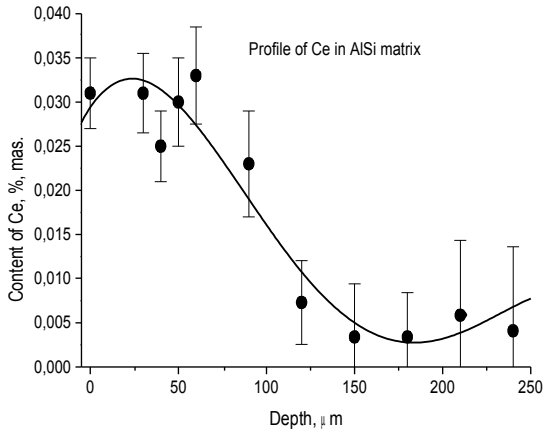

Fig. 3. *Distribution of Ce in the aluminosilicate matrix (annealing temperature 700°C, annealing time 300 hs). Points -experimental data, solid line - result of smoothing*

From the distribution obtained it can be concluded that the maximum content of Cerium in the aluminosilicate matrix subjected to diffusion annealing is approximately 0.03%, mas. The depth of Cerium penetration into the matrix is of about 130 μm. At a depth of 150 μm the content of Cerium decreases up to the level of 0.003%, mas. Evidently, such a content of

Cerium is characteristic for the initial material of aluminosilicate, as this level is remained at a depth more than 200 μm.

4. CONCLUSIONS

The investigations have shown that a proton microprobe can be a useful tool for measuring the concentration profiles of imitators of actinides in materials being applied for immobilization of actinides. One of its advantages is that the profiles can be measured practically at an unlimited depth. It is reached by the microbeam scanning over the transverse cleavage surfaces of samples under study.

The developed technique also can be easily applied for investigation of migration processes of such elements as Strontium, Iodine, Cesium and Uranium into the depth of synthetic aluminosilicate saples.

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ПРИМЕНЕНИЕ ПРОТОННОГО МИКРОЗОНДА ДЛЯ ИССЛЕДОВАНИЯ ПРОФИЛЕЙ ДИФФУЗИИ Ce В СИНТЕТИЧЕСКИХ АЛЮМОСИЛИКАТАХ

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Отработана методика измерения диффузионных профилей Ce с помощью протонного микрозонда и проведено измерение таких профилей в синтетической алюмосиликатной матрице, синтезированной на основе каолина и красного гранита.

ЗАСТОСУВАННЯ ПРОТОННОГО МІКРОЗОНДА ДО ДОСЛІДЖЕННЯ ПРОФІЛІВ ДИФУЗІЇ Ce У СИНТЕТИЧНИХ АЛЮМОСИЛІКАТАХ

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Відпрацьовано методику виміру дифузійних профілів Ce за допомогою протонного мікрозонда і проведено вимірювання таких профілів у синтетичній алюмосилікатній матриці, синтезованої на основі каоліну і червоного граніту.