RADIATION RESISTANCE OF GARNET-TYPE ACTINIDE HOST PHASES

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Key words: *Actinides, lanthanides, ferrite garnets, radiation resistance, amorphization*

Actinides, lanthanides, Fe and Zr, extracted from liquid high-level wastes (HLW) are proposed to solidify in durable crystalline phases (matrices) and then dispose in underground repositories [1-4]. Ferrites with garnet-type structure (*Ia3d*, Z=8) are one of the prominent host phases for An-Ln fraction [5]. Garnets with general formulae $A_3Al_5O_{12}$ (A=An, Ln) are considered as matrices for Al,Ga,Pu-containing wastes from conversion of weapon Pu and inert matrix for transmutation of long-lived actinides as well [6]. One of the most important properties of actinide host phases is radiation stability of their structure. Radiation resistance is characterized by critical dose of irradiation (D_c), which accumulation leads to full structure amorphization, and critical temperature (T_c), above which amorphization does not occur with increase of irradiation dose. Radiation stability of crystalline phases can be estimated by incorporation in the phase structure short-lived actinides (Cm-244, Pu-238, Am-241), by heavy ion irradiation of synthetic compounds and studying of natural U,Th-containing mineral analogues as well. There are not garnets with high amounts of U and Th in nature. Only recently results of investigation of natural garnets with 20 wt.% of uranium oxide have been published [7]. However radiation stability of these minerals cannot be determine because of minerals age, 2.5 Ma. There are not sufficient amount of accumulated radiation destructions in their structure. Thus actinide incorporation in garnet structure allows to model processes which will take place in matrices with real wastes during many thousands years of disposition.

Ferrite $(Ca_{1.5}Gd_{0.91}Cm_{0.09}Th_{0.5})ZrFe_4O_{12}$ and aluminate $Y_{2.88}Cm_{0.12}Al_5O_{12}$, doped with 2-3 wt.% 244-Cm ($T_{1/2}$=18 years), were studying. Curium distribution in the samples was studied by its surrogate – Sm. Data obtained show that all Cm will be included in garnet-type phases. Samples were produced by pressing (25°C, 200 MPa) and sintering of initial components for 4-5 hours at 1350°C (ferrite garnet) and 1450°C (aluminate garnet). Two intermediate stages of pressing and sintering were carried out to synthesis of aluminate garnet. Samples were examined with X-ray diffraction (XRD). According to XRD data samples are composed for the most part of garnets. There are small amounts of thorianite and corundum in Fe- and Al-system respectively. Due to small amount of additional phases compositions of garnets are considered to be close to ideal formulae.

XRD patterns getting just after synthesis of samples are revealed all garnets reflection (fig. 1). Dimension of garnet unit cell is 12.01 Å and 12.65 Å for Al- and Fe-garnet respectively. There are several changes in XRD patterns of ceramics with increase of observation time: intensity of garnet reflections decreases, their wide enlarges and less intensive reflections disappear. Garnet reflections are displaced into area of lower 2θ° values due to expansion of unit cell dimensions of these phases. Volume expansion of garnet cell is 0.9 % and 2.1 % for ferrite and aluminate respectively.

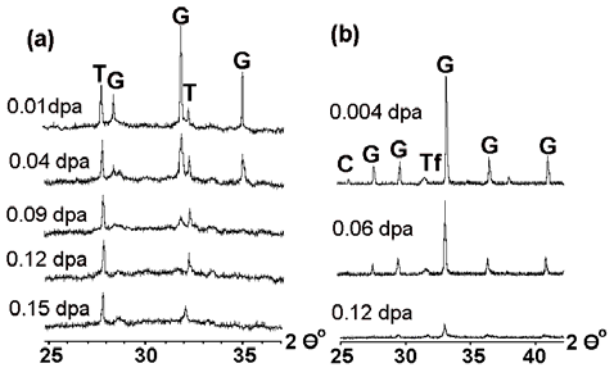

Fig.1. Radiation induced structural damage of (a) Fe-garnet and (b) Al-garnet with increase of accumulated dose observed on XRD patterns of ceramics. G – garnet, T – thorianite, C – corundum, Tf – teflon

Observed changes of XRD patterns are due to accumulation in the garnet structure of radiation induced damages which are results of interactions between atoms of garnet lattice and products of α-decay (α-particles and mainly α-recoil nuclei). Value of D_c for ferrite with garnet-type structure is about 0.15–0.17 displacements per atom (dpa). Aluminate with 0.21 dpa does not become amorphous.

Radiation stability of garnets with different compositions (natural silicate and synthetic ferrite) previously was estimated by Kr^+ irradiation [8, 9]. Values of critical dose lie within interval 0.15–0.2 dpa.

Dependence of critical dose on temperature was earlier studied for garnet with the composition close to $(Ca_{1.5}GdTh_{0.5})ZrFe_4O_{12}$ [10]. Value of T_c for this garnet is 547°C. Radiation stability of Al-garnet with formulae $Y_3Al_5O_{12}$ was studied by neutron irradiation in reactor at 540°C [6]. There are not any changes in garnet structure at 4 dpa. When accumulated dose increase up to 17 dpa only isolated point defects are observed in garnet structure. This fact is explained by achievement of critical temperature. Therefore garnet structure doesn't destroy with increase of accumulated dose. Based on data of Al-garnet behaviour at 25°C (present study) and 540°C [6] one can conclude that curves of dependence of amorphization dose on temperature for Fe- and Al-garnets are coincide (fig. 2).

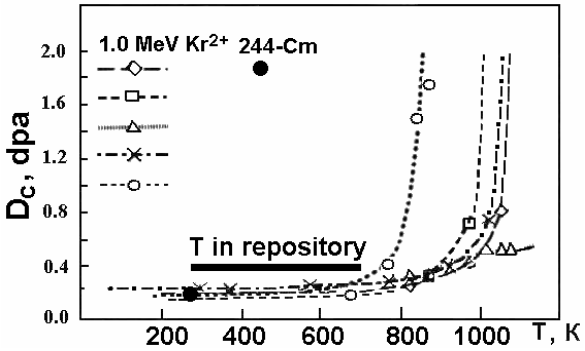

Fig.2. Amorphization dose of synthetic garnets under ion-irradiation [9] and incorporation of Cm-244 as function of temperature

Aluminate garnet is more resistant to radiation from Cm-244 than ferrite. Radiation stability of ferrites and aluminates with garnet-type structure are close to well known actinide matrices based on Ti-pyrochlore and monazite [11–12].

The research was supported by the Russian Foundation for Basic Research (project 08-05-00024-a)

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Electronic Scientific Information Journal "Vestnik Otdelenia nauk o Zemle RAN" № 1(27)'2009
ISSN 1819 – 6586
Informational Bulletin of the Annual Seminar of Experimental Mineralogy, Petrology and Geochemistry – 2009
URL: http://www.scgis.ru/russian/cp1251/h_dgggms/1-2009/informbul-1_2009/geoecol-8e.pdf

Published on July, 1, 2009