XAFS of Pu-doped zircon and monazite: Pu and U environment

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Ceramics are presently considered as a promising matrix for long-term immobilization of surplus actinides such as plutonium. Monazites and zircon are often regarded as the best candidates, since their natural analogues retain radioactive U and Th for billions years. However, behaviour of much more radioactive elements such as plutonium in these minerals is less well understood. Decay of a Pu nuclei produces highly energetic (> 4 MeV) alpha-particle and a recoil uranium, having an energy close to 100 keV. Whereas alpha-induced damage can be modeled by external ion implantation studies, influence of recoils is more difficult to study experimentally. In this work we report results of X-ray Absorption Spectroscopy investigation at U and Pu L_{III} edges of synthetic zircon and Eu-monazite, doped with 2.4 and 4.9 wt% of highly active isotope ^{238}Pu, characterized by relatively short lifetime of 87.7 years. The single crystals were grown in 2001 and 2003, correspondingly, and considerable damage of the crystalline lattice has already accumulated. Even recoil uranium is already detectable. Some results of investigation of these samples along with synthesis conditions are given in [Burakov et al., 1996, 2004].

The samples were packed into special hermetically sealed beryllium capsules and investigated in fluorescence mode at Structural Materials Science beamline at Kurchatov Center for Synchrotron Radiation. Zirconium foil was used for energy calibration.

The XAFS spectrum of Pu in the monazite does not show pronounced oscillations due to low symmetry of the mineral. According to the X-ray data, in both minerals plutonium enters the crystalline lattice as substitional atom. However, the lattice is somewhat distorted in comparison with the ideal structure. Results of detailed modeling of the local environment will be given in the presentation. The position of recoil uranium is not well constrained. Though local amorphisation around the end of the track is expected, partial lattice reconstruction is also possible.

In zircon only the spectrum of recoil uranium was recorded, since Pu and Zr edges are too close to each other to be reliably analysed.

Implications of our studies to problem of long-term actinide immobilisation in zircon- and monazite-based ceramics will be discussed.

References:
Burakov B., et al., (1996), MRS Symposiums Proceedings, 412, 33-38
Burakov B., et al., (2004), MRS Symposiums Proceedings, 824, 219-224