

Influence of radiation damage on the crystal structure of monazite

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The safe disposal of nuclear waste is an intergenerational issue which needs to be addressed. The incorporation of radionuclides into crystalline solid phases is a potentially efficient mechanism for immobilizing radionuclides in future radioactive waste repositories. In particular, the immobilization of specific waste streams containing the minor actinides (Np⁴⁺, Am³⁺, Cm³⁺) or plutonium (Pu^{3+,4+}), by incorporation of the radioactive nuclei into crystalline solid phases may be advantageous compared to glass matrices, which may be less resistant to leaching and disintegration [1-3]. Monazite-type compounds are considered suitable due to their chemical and structural flexibility and radiation stability [4].

In order to better understand structural changes due to radiation damage, monazite single crystals with different chemical compositions (Nd, Pm, Sm)PO₄ were irradiated at the UNILAC beamline of GSI Helmholtzzentrum Darmstadt using 1.7 GeV Au ions and a fluence of 1e13 ions per cm². Subsequently, the single crystals were characterized by Raman spectroscopy, secondary electron microscopy and single crystal X-ray diffraction. The obtained results, e.g. the penetration depth of the ions, are compared with results of numerical SRIM (Stopping and Range of Ions in Matter) calculations [5].

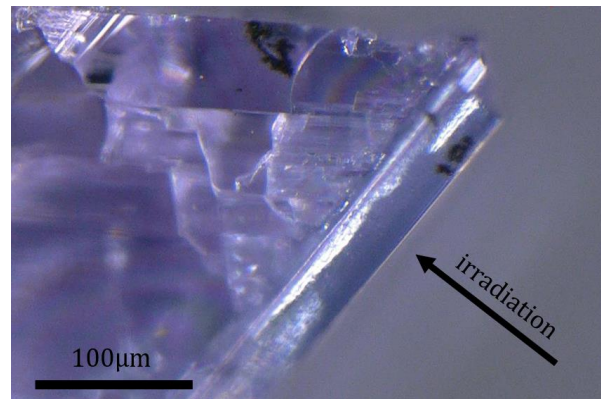


Figure 1 Cross section of irradiated NdPO₄ irradiated with 1.7 GeV Au ions showing the amorphization of a ~55 μm surface layer

The irradiation of monazite with 1.7 GeV Au ions results in an embrittlement of the crystals and the formation of a glassy X-ray amorphous surface layer of about ~55 μm thickness (Figure 1), which correlates well with the projected range from the SRIM-2010 calculations [5]. The irradiation results in a significant broadening of the Raman modes, and further changes in the lattice dynamics. The analysis of diffraction data obtained from the irradiated samples is currently in progress.

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