Structural changes in Ln-monazites under swift heavy ion irradiation

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The safe disposal of nuclear waste is one of the intergenerational issues which needs to be solved. A potential route to effectively immobilize radionuclides could be realized by their incorporation into crystalline solid phases in future radioactive waste repositories. In particular, the immobilization of specific waste streams containing minor actinides (Np, Am, Cm) or plutonium in crystalline solid phases may be advantageous compared to glass matrices, which may be less resistant to leaching and disintegration (Donald et al. 1997; Ewing 1999; Lumpkin et al. 2006). Due to their radiation stability and chemical and structural flexibility, monazite-type compounds are considered suitable matrix materials (Schlenz et al., 2013).

To better understand structural changes due to radiation damage, synthetic monazite single crystals with different chemical compositions (La, Nd, Pm, Sm)PO₄ were irradiated at the UNILAC beamline of GSI Helmholtz Centre Darmstadt using 1.7 GeV Au ions and fluences

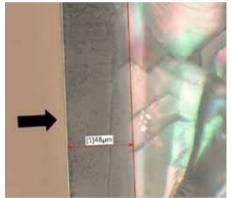


Figure 1. Cross section of a $(Pr,Nd)PO_4$ monazite crystal, prepared parallel to the direction of the irradiation indicated by the black arrow

of up to 1e13 ions/cm². The irradiated single crystals were characterized by Raman spectroscopy, secondary electron microscopy and single crystal X-ray diffraction.

The irradiation of monazite with 1.7 GeV Au ions results in an embrittlement of the crystals and the formation of a glassy surface layer of about ~48 μ m thickness (Fig. 1), which correlates well with the projected range of ~44 μ m according to SRIM-2013 calculations (Ziegler et al. 2010). The irradiation results in a significant broadening of all Raman modes up to the complete disappearance of the symmetric stretching mode v₁ and further changes in the lattice dynamics. X-ray diffraction experiments revealed the amorphization of the surface layer.

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