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Combined Raman and photoluminescence spectroscopic investigation of He-irradiation effects in monazite

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We present first results of a study addressing effects of the corpuscular self-irradiation on stress/strain and optical properties (in particular the photoluminescence behaviour) in monazite–(Ce). For this, natural samples, and synthetic $CePO_4$ crystals mildly doped with Nd^{3+}, were irradiated with 7.7 MeV He ions, which are analogues of alpha particles generated in the $^{214}Po \rightarrow {}^{210}Pb$ decay event (^{238}U decay series). Light-ion irradiation was preferred over heavy-ion irradiation (i.e., $\sim$100 keV heavy ions as analogues of alpha recoils) for two reasons. First, MeV He ions penetrate much deeper into mineral targets (tens of micrometres vs. a few hundred angstroms), resulting in irradiated volumes that are "measurable" using micro-spectroscopy techniques. Second, the depth distribution profiles of nuclear and electronic energy losses vary appreciably in the case of MeV He ions. The latter is most helpful in assigning observed effects to either point defects created or target ionisation. The irradiations were done at the 3 MV Tandem accelerator of the Helmholtz-Zentrum Dresden-Rossendorf (Germany). The fluences applied varied in the range 10^{13}–10^{17} He/cm^2.

The degree of disturbance of the short-range order in the samples irradiated was evaluated from the broadening of the $\nu_1(PO_4)$ Raman band (symmetric PO_4 stretching; A_{1g} mode) near 970 cm^{-1} [1,2]. The majority of the damage created was observed near 28 μm into the monazite–(Ce) targets, i.e., near the ends of the helium trajectories. Here, the $\nu_1(PO_4)$ Raman band may broaden appreciably, from 2.2 cm^{-1} (in the case of synthetic $CePO_4$) to well above 20 cm^{-1}. The generation of additional damage was also observed upon He irradiation of naturally radiation-damaged monazite–(Ce). By contrast, there was no indication of any annealing effect of the MeV He ions irradiated in these samples (compare [3]; a similar effect of alpha-assisted annealing was suspected by [4] in the discussion of fission tracks). Sharp luminescence emissions of REE centres with $4f$ electronic structure, such as the $^2H_{3/2} \rightarrow {}^4I_{9/2}$ emission of Nd^{3+} near 11300 cm^{-1} (885 nm wavelength), were found to show similar broadening behaviour. This may potentially be used to develop a luminescence-based calibration for the non-destructive quantification of the structural irradiation damage on a micrometre-range. In addition, a brownish-orange, broad-band defect emission is created at low and depleted at moderate damage (similar to observations on irradiated zircon by [5]). The presence of this emission may be considered as a simple indicator of low degrees of damage, for instance in the evaluation of heavy-mineral concentrates.

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