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The nature of radiohaloes in biotite: Experimental studies and modeling

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ABSTRACT

Several micro-techniques (confocal laser-Raman microprobe, optical absorption micro-spectroscopy, high-resolution transmission electron microscopy, electron microprobe analysis) were employed in the detailed characterization of radiohaloes in biotites from two Variscan rocks from Germany. The studied biotites are intermediate members of the phlogopite-annite series with Mg/Fe^{2+} ratios in the range 1.6–1.0. Radiohaloes in biotite resulted from the impact of ^{4}He cores (α-particles) emitted from actinide-bearing inclusions. Monte Carlo simulations yielded α (^{238}U, ^{235}U, and ^{232}Th series) penetration ranges in biotite between 12.5 and 37.3 μm, which are in reasonable agreement with the observed radii of radiohaloes in natural biotites. The coloration pattern of a radiohalo closely correlates with the calculated distribution pattern of point defects generated in displacive events. Calculated point defect densities in the range from $< 10^{-5}$ to at most 10^{-2} dpa (displacements per lattice atom) suggest that there are only scattered point defects in a mainly preserved biotite lattice. This is consistent with HRTEM studies that did not reveal any indication for initial volume amorphization in the haloes. However, general Raman band broadening and intensity loss suggest that the short-range order in radiohaloes is significantly disturbed. The darkened color of radiohaloes, when compared with the un-irradiated host biotite, is caused by increased light absorption over the complete visible range due to increased point defect density. No additional color centers were found, and the absorbances of the $^{VI}Fe^{2+}$, Fe^{2+}-Fe^{3+}, and Fe^{2+}-Ti^{4+}centers seem hardly to be changed. Both Raman and optical absorption spectra obtained from radiohaloes retain a clear orientational dependence. The results suggest that the formation of point defects rather than ionization is the main process causing the coloration of radiohaloes in natural biotites. The haloes represent an early stage of structural radiation damage, characterized by significantly disturbed short-range order but still widely preserved long-range order of the structure.