The nature of radiohaloes in biotite: Experimental studies and modeling

LUTZ NASDALA,1,* MARITA WENZEL,2† MICHAEL ANDRUT,3 RICHARD WIRTH,4 AND PETER BLAUM1

1Institute of Geosciences, Mineralogy, Johannes Gutenberg-University, D-55099 Mainz, Germany
2Institute of Solid State Physics, University of Technology, D-64289 Darmstadt, Germany
3Institute of Mineralogy and Crystallography, Geocenter, University of Vienna, A-1090 Vienna, Austria
4GeoForschungsZentrum Potsdam, D-14473 Potsdam, Germany

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/Fe2+ 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 (238U, 235U, and 232Th 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 Fe2+, Fe2+-Fe3+, and Fe2+-Ti4+ 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.

INTRODUCTION

Self-irradiation damage of natural, actinide-bearing minerals, finally leading to the metamict state, has been investigated and discussed in numerous papers. Metamict minerals (such as zircon, titanite, monazite, pyrochlore, and fergusonite) can be transformed from an initially crystalline to an amorphous state by long-term accumulation of damage resulting from α-decay events (e.g., Chakoumakos et al. 1987; Murakami et al. 1991; Meldrum et al. 1998). Most of this damage is caused by the recoil of the heavy daughter nuclei upon emission of an α-particle (4He core), whereas other related processes—such as α-radiation, spontaneous fission, etc.—make minor contributions to metamictization (e.g., Weber et al. 1994; Nasdala et al. 1996).

Radiation-damage phenomena, however, are also observed in minerals that do not contain major amounts of radionuclides in their structures. Such minerals may have undergone radiation-induced alteration due to long-term irradiation originating from their neighboring phases. In these cases, radiation damage is mainly due to α-particles, penetrating some ten μm into minerals (for comparison, recoil-nuclei ranges are on the order of hundreds of Å). Thus, strong α-emitters are typically surrounded by alteration haloes (usually called radiohaloes) with thicknesses corresponding to the travel distance of the α-particles.

The appearance of radiohaloes may vary appreciably and depends on the strength of the irradiation and particularly on the radiation-damaged mineral itself. Among the most widespread mineral species containing radiohaloes is biotite, where haloes are easily recognized by their significantly darker colors (“pleochroic haloes”; e.g., Gentry 1974). Haloes with similar coloration are also found in minerals such as cordierite, amphibole, fluorite, and chlorite. Radiohaloes in quartz appear...