

Microstructural and compositional evolutions during transformation from biotite to berthierine: Implications for phyllosilicate alteration processes

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ABSTRACT

The composition and microstructure of phyllosilicates are prone to transformation due to their great sensitivity to surrounding physicochemical changes. Berthierine $[(R^{2+}, R^{3+}, \square)_6(\text{Si}, \text{Al})_4\text{O}_{10}(\text{OH})_8]$ (\square represents octahedral vacancy) is a typical ferromagnesian phyllosilicate that commonly occurs in feruginous rocks of shallow-marine habitats and has been used as an indicator of local depositional and/or hydrothermal activity in marine environments. However, little is known about the formation and mineralogy of non-marine berthierine, particularly in volcanic systems. Using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM), we have identified a berthierine twin structure within weakly altered biotite in a rhyolite from Long Valley, California, U.S.A. The presence of nanoscale Fe-rich layers in the host biotite is revealed by energy-dispersive spectroscopy and electron energy loss spectroscopy (EELS). The HAADF-STEM pictures with atomic resolution demonstrate that the Fe-rich layers are composed of twinning berthierine layers rather than a single chlorite layer. The transformation of biotite to berthierine requires the dissolution of a tetrahedral (T) layer and the introduction of a new TO (O represents octahedral sheet) structure into the biotite stacking sequence, resulting in substituting one biotite layer (i.e., TOT) with two twinning berthierine layers (i.e., TO-OT). Observations based on morphology indicate that the transformation began at biotite defect locations (such as screw dislocation, edge dislocation, and microcleavage fracture), concurrent with the rearrangement of metal cations. During the fluid alteration of biotite, berthierine was produced via an interface-coupled dissolution-precipitation process. The EELS analyses further demonstrate that the Fe-rich biotite promotes the production of berthierine as the principal alteration product in low-temperature environments. Additionally, this study suggests that the combination of HAADF-STEM and EELS is effective for identifying nanominerals and elucidating their formation and alteration mechanisms.

Keywords: HAADF-STEM, EELS, biotite, berthierine twin, alteration mechanism; Isotopes, Minerals, and Petrology: Honoring John Valley