## Biotite dissolution processes and mechanisms in the laboratory and in nature: Early stage weathering environment and vermiculitization

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## ABSTRACT

Biotite dissolution in the laboratory and in nature was examined and compared to elucidate certain aspects of the weathering processes. Batch dissolution experiments of fresh biotite  $[(K_{0.91}Na_{0.01})(Mg_{0.40}Fe_{2.07}Mn_{0.05}Al_{0.14}Ti_{0.19})(Si_{2.82}Al_{1.18})O_{10}(OH)_2]$  in granite were carried out at 150 °C for 1 to 56 days. Examination by scanning electron microscope (SEM) equipped with an energy dispersive X-ray spectrometer (EDS) revealed that, in the early stage, dissolution proceeds from the edges of crystals inward and secondary minerals such as Fe oxide are precipitated mostly at the edges, with only a few secondary minerals found on the basal surfaces. A dissolution experiment using a mixture of biotite and muscovite, done at 150 °C for 7 days, indicated that hematite crystals formed mostly at the edges of biotite but not on muscovite. This observation suggests that released Fe is precipitated before it diffuses into the bulk solution. Because a dissolution rate at the edge is larger by two orders of magnitude than at the basal surface (Turpault and Trotignon 1994), precipitation before diffusion of dissolved elements to bulk solution well explains the preferential secondary mineralization at the edges in the laboratory experiments. SEM-EDS of fresh to slightly weathered biotites revealed that early stage weathering proceeds in the same way as in the laboratory with the edges being preferentially weathered and secondary minerals being precipitated mostly at the edges. Because of the similarity between the occurrence of secondary minerals in the laboratory experiments and in nature, the laboratory results elucidate the early stage weathering conditions, namely that (1) supersaturation with respect to secondary minerals in a solution occurs around biotite, i.e., the solution is poorly connected to a main flow pathway of water, and (2) once supersaturation is achieved, secondary minerals are precipitated mainly at the edge before some released elements diffuse into the solution. The immobility of water immediately adjacent to primary minerals partly explains the large difference in dissolution rate between the laboratory and natural samples.

A phlogopite dissolution experiment was done at 150 °C for 5.5 days to examine the effect of biotite composition on vermiculitization. High-resolution transmission electron microscopy revealed that vermiculite layers are formed by layer-by-layer transformation within phlogopite after 5.5 day dissolution but not within biotite after 56 day dissolution or within slightly weathered biotite. A comparison of our data with results of other studies indicates that the formation of vermiculite layers between biotite layers occurs even in the early stage when biotite is not Fe-rich [more than 0.8 Mg per  $O_{10}(OH)_2$ ]. When biotite is Fe-rich [less than 0.4 Mg per  $O_{10}(OH)_2$ ]—like the present one—released Fe is precipitated as, for example, Fe hydroxide, and vermiculite is rarely formed because of limited availability of Mg. Thus, a higher Mg content in biotite facilitates the formation of vermiculite, at least in the early stage. Because vermiculite dissolves at a much slower rate than biotite, Mg-rich biotite dissolves at a slower rate than Fe-rich biotite. In the late stage, biotite dissolution still continues from the edge and within biotite, which results in a fine comb-like texture for weathered biotite; vermiculite occurs as a domain within chemically and structurally altered biotite as well, as at the edge even during weathering of Mg-poor biotite.