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Atomic-resolution transmission electron microscope evidence for the mechanism by which chlorite weathers to 1:1 semi-regular chlorite-vermiculite

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ABSTRACT

Atomic-resolution transmission electron microscope (TEM) images reveal that II*bb* (β = 97°) Mg,Al,Fe-chlorite from Koongarra, Australia, transforms to vermiculite via a range of intermediate chemical and structural states. Semi-quantitative analysis of contrast in atomic-resolution images of layer silicates with ~1.4 nm basal spacings indicate that the interlayers range from brucite-like to having ~0.3–0.6 interlayer cations per formula unit. Octahedral cations (predominantly Mg and Fe) tend to be removed from every second interlayer, leading to semi-regular 1:1 interstratifications of chlorite-vermiculite. Further loss of interlayer cations is accompanied by partial to complete interlayer collapse in the vacuum of the TEM. Resulting intergrowths of chlorite and semi-regular 1:1 chlorite-vermiculite retain the primary chlorite orientation, morphology, and sense of octahedral tilt in 2:1 layers. Although vermiculitization is a continuous process that occurs by a solid-state mechanism, the reaction involves important structural modifications. Atomic-resolution [010] images indicate initial loss of interlayer cations is accompanied by ~**a**/3 shifts of 2:1 layers and cations in brucite-like interlayers. Displacements of interlayer cations change the interlayer stacking from II*bb* to I*ab* and shift of the following 2:1 layer converts it from I*ab* to the I*aa*. Displacements are driven by the lower energy of *a*-type interactions when vacancies occur in sites above tetrahedral cations. Shift of a 2:1 layer alters the subsequent interlayer from II*bb* to II*ab*. Stabilization of every slightly altered second interlayer by introduction of *a*-type stacking explains development of semi-regular 1:1 chlorite-vermiculite interstratifications. Displacements occur before significant modification of interlayer electron density can be detected in high-resolution images. This observation is consistent with previously reported inhibition of layer shifts by low interlayer charge. Layer displacement may occur by an elastic process (no rupture of bonds within the 2:1 layer) at the tip of the growing vermiculite portion of the intergrowth. Removal of cations from the chlorite-vermiculite junction may be facilitated by rapid diffusion along the vacancy-rich interlayer. Mg is removed in solution, Fe is precipitated locally in aggregates of nanocrystalline Al-, Si-, and P-bearing goethite.