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Polytypism in semi-disordered lizardite and amesite by low-dose HAADF-STEM

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Abstract

Serpentine minerals exert important controls on the physical properties of ultramafic rocks and have the potential to influence deformation phenomena in fault zones and to control the release of water in subducted slabs. Sheet serpentine generally, and lizardite and amesite specifically, can adopt alternative crystallographic stacking arrangements called polytypes. Polytypism has been extensively studied in fully ordered crystals, but it remains largely enigmatic in the more common semi-disordered crystals that in long-range analyses such as X-ray diffraction only exhibit random combinations of 0***b*** and ±1/3***b*** interlayer shifts. To date, atomic-resolution imaging to identify locally ordered polytypes has been precluded by the beam-sensitive nature of this hydrous magnesium silicate mineral. Here, we employed low-dose high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) to study the polytypic structure of semi-disordered lizardite and amesite. Because the electron dose was as low as ~6000 $e^-/Å^2$, it was possible to directly resolve oxygen atomic columns and all the cations with a resolution of ~1 Å and reveal the short-range order. For lizardite, we identified long-period non-standard polytypes, including examples with 3, 4, 8, and 9 layers stemming from the ordering of the octahedral tilt along the ***a***-axis. For amesite, we found short-range ordered polytypes with periodicities of up to 42 Å stemming from the ordering of interlayer shifts along the ***b***-axis. The resolution was sufficient to determine the relative abundance of $6R_2$, $6R_1$, $2H_1$, and $2H_2$ polytypes in amesite to be 46.1, 29.6, 7.7, and 1.9%, respectively. This is contrary to the expectation that the most common form of amesite is the $2H_2$ polytype, which may be more likely to form macroscopic crystals suitable for conventional X-ray diffraction-based studies. We conclude that HAADF-STEM methods open the way for the characterization of beam-sensitive minerals and to resolve the structural details of less well-ordered (but possibly more abundant) minerals at a unit-cell scale.

Keywords: Polytype, HAADF-STEM, amesite, lizardite, serpentine