

Al and Fe speciation in natural chrysotile, lizardite and antigorite, from Fe *K*-edge XANES-EXAFS and ²⁷Al NMR spectroscopy

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Serpentine minerals are ubiquitous in altered oceanic lithosphere, and it is now well accepted that serpentinization processes are of major importance for understanding, e.g., prebiotic processes leading to the origins of life, abiotic hydrogen production, or mobility of metals in various geological contexts such as mid-ocean ridges, subduction zones or ophiolitic outcrops. Although the level of understanding has significantly increased over the past 10 years, thermodynamics remains today one of the main limitations to efficiently model these processes. In order to meet this challenge, a perfect understanding of the crystal chemistry of the main varieties of serpentine minerals (i.e., chrysotile, lizardite and antigorite) is necessary. Serpentine theoretical formula is $Mg_3Si_2O_5(OH)_4$ but several cationic substitutions are frequently observed in natural systems. Therefore a more complete formula may be written: $(Mg,Fe^{2+},Fe^{3+},Al)_3(Si,Al,Fe^{3+})_2O_5(OH)_4$; showing that Fe and Al potentially play an important role in the crystallization of these minerals.

This study proposes a fine characterization of the speciation of Fe and Al in a set of 23 magnetite-free serpentine samples, and to investigate the possible links between the micro-, and nano-structures. Iron speciation is derived from pre-edge, XANES and EXAFS spectra collected at the Fe *K*-edge, and Al speciation is derived from ²⁷Al NMR spectroscopy. Our results suggest a direct link between the mode of incorporation of trivalent cations and the microstructures observed. Chrysotile incorporates fewer trivalent cations than other varieties, which tends to preserve the so-called misfit between the TO layers, and therefore the tubular structure of the mineral. On the other hand, lizardite mainly involve trivalent Fe and Al Tschermak-type substitutions, which tend to flatten the mineral structure.