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SPECIAL COLLECTION: NANOMINERALS AND MINERAL NANOPARTICLES

Dissolution-reprecipitation and self-assembly of serpentine nanoparticles preceding chrysotile formation: Insights into the structure of proto-serpentine

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Abstract

Any poorly crystalline serpentine-type mineral with a lack of recognizable textural or diffraction features for typical serpentine varieties (i.e., chryotile, lizardite, and antigorite) is usually referred to as proto-serpentine. The formation of the so-called proto-serpentine seems ubiquitous in serpentinization reactions. It is related to dissolution-precipitation of strongly reactive particles prior to true serpentine formation (e.g., in veins where both chrysotile and proto-serpentine are described). However, the structural characteristics of proto-serpentine and its relation with serpentine crystalline varieties remain unclear. In this study a model describing the transformation from proto-serpentine to chrysotile is presented based on experimental chrysotile synthesis using thermogravimetric analyses, transmission electron microscopy, and high-energy X-ray diffraction with pair distribution function analyses. The combination of the high-resolution TEM and high-energy X-ray diffraction enables to resolve the local order of neo-formed particles and their structuration processes occurring during pure chrysotile formation (i.e., during the first three hours of reaction). The formation of individual nanotubes is preceded by the formation of small nanocrystals that already show a chrysotile short-range order, forming porous anastomosing features of hydrophilic crystallites mixed with brucite. This is followed by a hierarchical aggregation of particles into a fiber-like structure. These flake-like particles subsequently stack forming concentric layers with the chrysotile structure. Finally, the individualization of chrysotile nanotubes with a homogeneous distribution of diameter and lengths (several hundreds of nanometer in length) is observed. The competitive precipitation of brucite and transient serpentine during incipient serpentinization reaction indicates that both dissolution-precipitation and serpentine-particle aggregation processes operate to form individual chrysotile. This study sheds light into mineralization processes and sets a first milestone toward the identification of the factors controlling polymorph selection mechanisms in this fascinating system.

Keywords: Proto-serpentine, pair distribution function, chrysotile, stacking, simulation