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Nanoscale characterization of chrysocolla, black chrysocolla, and pseudomalachite from supergene copper deposits of Atacama Desert in northern Chile

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Abstract

We present the first textural and chemical characterization at nanometer scale of chrysocolla [$(Cu_{2-x}Al_x)H_{2-x}Si_2O_5(OH)_4{\cdot}nH_2O$], black chrysocolla (a Mn-rich variety of chrysocolla), and pseudomalachite [$Cu_5(PO_4)_2(OH)_4$] from two distinct supergene copper deposits from Atacama Desert in northern Chile. These minerals are the most common copper minerals found in the supergene deposits associated with copper porphyries from Atacama Desert. However, the lack of nanoscale morphological information prevents a deeper understanding of their formation process. Nanoscale characterization using transmission electron microscope (TEM) imaging allows further characterization of the structural states of chrysocolla, black chrysocolla, and pseudomalachite, offering valuable insights into their genesis. Chrysocolla and black chrysocolla are not single crystals but assemblages of Cu nanoparticles embedded in an Si-rich amorphous matrix. Scanning TEM (STEM) images reveal that chrysocolla consists of rounded Cu-rich nanoparticles embedded in an amorphous matrix, while black chrysocolla consists of rounded Cu-rich nanoparticles with few needle-shaped Mn-rich particles, all embedded in an amorphous matrix. The richness in nanoparticles defines a layering that mimics the colloform texture observed in optical microscopy. In contrast, pseudomalachite is a massive polycrystalline mineral consisting of a juxtaposition of large nanocrystal grains of ~500 nm. The STEM-electron energy loss spectrometry (EELS) spectra show that copper in chrysocolla and black chrysocolla is in a reduced state. This suggests that chrysocolla and black chrysocolla form under reducing conditions, probably just below the water table. Alternatively, it could be that water table oscillation allows for the cyclical precipitation of Cu^0-rich nanoparticles and oxidized copper-rich silicates. Conversely, pseudomalachite crystallization requires oxidative conditions. The oxidation state variations, from chrysocolla (Cu^0) to pseudomalachite (Cu^{2+}), certainly occur during the episodic switch of the water table linked to tectonic events or climatic changes. The findings also have implications for the U-Pb dating of supergene copper deposits, since black chrysocolla and pseudomalachite can incorporate significant U contents. The different structural states of the three minerals may explain their different behaviors regarding U and Pb mobility and, therefore, the preservation of the U-Pb chronometric signal.

Keywords: Chrysocolla, black chrysocolla, pseudomalachite, transmission electron microscope, nanoscale characterization