

A multi-scale approach to partially crystalline gem materials: unravelling the complexity of chrysocolla

Monico S.*¹, Marinoni N.¹, Gatta G.D.¹, Adamo I.², Prosperi L.², Conconi R.³⁻⁴, Capitani G.³ & Bernasconi A.⁵

¹ Dipartimento di Scienze della Terra “A. Desio”, Università di Milano.

² Istituto Gemmologico Italiano, Milano.

³ Dipartimento di Scienze dell’Ambiente e della Terra, Università di Milano-Bicocca.

⁴ UMET Unité Matériaux et Transformations, CNRS UMR 8207, Université de Lille, France.

⁵ Dipartimento di Scienze della Terra, Università di Torino

Corresponding author email: sara.monico1@unimi.it

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Partially crystalline materials used in gemmology include a wide range of mineral species with distinctive compositional and structural properties, influencing their appearance, durability, and aesthetic value. A notable example is chrysocolla, classified as a hydrated copper silicate ($\text{Cu}_{2-x}\text{Al}_x(\text{H}_{2-x}\text{Si}_2\text{O}_5)(\text{OH})_4 \cdot n\text{H}_2\text{O}$; $x < 1$) (Dold et al., 2023) and compositionally described as a colloidal system of hydrated silica and copper oxides. Chrysocolla occurs in various colours influenced by impurities and, in its blue-green hues, has been valued since antiquity as a pigment and, in jewellery, as a gem material for amulets and ornaments. Due to its intense colouring and unique aesthetic aspects, it remains widely appreciated in gemmology today. Chrysocolla was officially recognised as a mineral species by the International Mineralogical Association (IMA) in 1969. However, its nature remains debated, as it is still unclear whether it constitutes a distinct mineral phase, an aggregate of several phases, or a colloidal gel. Research has often produced discordant results due to its inherent complexity and pseudo-amorphous character, which make it analytically challenging (Frost et al., 2012) and unstable under certain conditions. These aspects complicate its identification and classification in mineralogy and gemmology. This study aimed to characterise chrysocolla in terms of composition, mineralogy and microstructure, through a multi-methodological approach. To this end, an ad hoc analytical protocol was developed to highlight the peculiarities of the material without compromising its stability. Distinctive aspects of chrysocolla not previously observed were reported for samples from two mining localities — Capo Calamita, Capoliveri (Italy), and Cornwall (Lebanon County), Pennsylvania (USA) — which were investigated using both conventional and high-resolution techniques. Particular focus was placed on chemical composition, through point and spatial surveys employing electron microprobe analysis with wavelength-dispersive spectroscopy (EMPWDS). Multivariate analysis of the data allowed for the identification of relationships between compositional variability and formation conditions. The local atomic

arrangement was studied using the Pair Distribution Function (PDF) analysis based on synchrotron radiation data, giving new insights into bond distances and structural coherence. Given the partially crystalline nature of chrysocolla, in which distinctive properties are manifested at the level of local domains, the investigation was extended to the nanoscale using transmission electron microscopy (TEM). The results provided new insights into the compositional, microstructural, and textural properties of chrysocolla, helping to better define its distinctive characteristics. Moreover, the study introduced new methodological approaches for its recognition and classification, with significant implications in both mineralogical and gemmological fields.

Dold B. et al. (2023) - Genesis of the exotic chrysocolla - “copper pitch/wad” - atacamite/brochantite ore at the Exótica (Mina Sur) deposit, Chuquicamata, Chile. *Mineralium Dep.*, 58(3), 569-591, <https://doi.org/10.1007/s00126-02201147-7>.

Frost R.L. et al. (2012) - Thermogravimetric analysis, PXRD, EDX and XPS study of chrysocolla $(\text{Cu,Al})_2\text{H}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot n\text{H}_2\text{O}$ structural implications. *Thermochim. Acta*, 545, 157-162, <https://doi.org/10.1016/j.tca.2012.07.011>.