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Calibration of a Raman technique for determining Mg numbers of serpentine solid-solutions

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Serpentinite-hosted hydrothermal systems have been proposed as a location for the genesis of life, and production of reduced gases (H_2, CH_4) during serpentinization has been identified as an energy source for active, and potentially ancient, microbial ecosystems. The source of these reduced gases is the oxidation of Fe and the coupled reduction of water to produce H_2, which can reduce inorganic carbon to produce CH_4. Tracing the fate of Fe during serpentinization is thus vital to understanding the fluxes of reduced gases from serpentinizing systems.

Raman spectroscopy is a rapid and non-destructive mineral identification technique that can also be used to determine relative proportions of end-members in solid-solutions. In the context of serpentinization reactions, this method has been used in literature to determine relative abundances of Fe and Mg end-members of both olivine and brucite from Raman peak shifts. A similar methodology has not been developed for serpentine group minerals. We posit that cation substitution of Mg^{2+} and $Fe^{2+/3+}$ in serpentine group minerals will contribute to a similar shift in spectral peaks and the identification of affected peaks could allow for rapid, quantitative analysis and mapping of serpentine composition.

To correlate Raman peak shifts with serpentine chemical composition, we performed co-located Raman and electron microprobe micro-analyses on iron-rich (XFe = ~0.2 – 0.6) serpentines from the Duluth Complex (USA). These analyses, combined with Raman analysis of nearly pure samples of the serpentine end-members antigorite, lizardite, chrysotile, greenalite, and cronstedtite, were used to assess the shifts in the characteristic serpentine peaks as a function of iron content and Fe^{3+} substitution. Initial results show minor peak shifts in the high and low spectral regions of the Duluth Complex samples in comparison to the standards, which suggests this technique has promise for mapping serpentine compositions.

Successful calibration of this Raman spectral technique will permit rapid identification of serpentine composition and potentially redox state. This technique will provide a cost-effective, non-destructive, and easily accessible alternative to current synchrotron-based techniques. The successful development of this technique may assist the interpretation of *in situ* measurements performed by the SHERLOC Raman instrument on the Mars Perseverance rover.