Fe-oxidation, H_2 generation and microbial activity in peridotites undergoing serpentinization

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The energy availability in rock-hosted subsurface ecosystems is often strongly dependent on Fe(II)-bearing minerals as the source of reducing power. However, it is often difficult to predict the rates and mechanisms of Fe-oxidation, and the production of soluble energy sources such as H_2 or formate, during low temperature water/rock interaction. Yet the fluxes of geologically-derived electron donors are a key control on *in-situ* microbial activity, such as methanogenesis, acetogenesis and sulphate reduction. Therefore, one of the key questions to address is the solubility and reactivity of Fe(II), and the mechanisms of electron-transfer from Fe-bearing minerals in reactions with H_2O, CO_2 or stronger oxidants.

We have developed an integrated approach to quantitatively image Fe distribution and oxidation state within peridotites undergoing multiple stages of water/rock interaction. Synchrotron-based micro X-ray fluorescence spectroscopy multiple energy mapping has been optimized for the pre-edge region of the Fe K-edge, in order to determine microscale variability in Fe(III)/Fe_T across reactive interfaces. Fe redox mapping is combined with hyperspectral Raman imaging and EMPA mapping to determine the Fe oxidation state distributions across all the mineral phases. We use this approach to characterize partially-serpentinized rocks recently obtained through drilling in sites of active alteration in the Samail Ophiolite in Oman. In this system, we infer that metastable Fe(II)-bearing brucite may regulate the H_2 activity across regimes with highly variable water/rock ratio and fluid chemistry. Ferroan brucite is abundant at depth, and is lost through reaction with near surface fluids. Brucite is converted to a diversity of Fe(II/III)-bearing phases, such as Fe(III)-bearing layered double hydroxide minerals, as well as magnetite and Fe(III)-rich serpentine. Coupling the quantitative imaging to depth variations in bulk mineralogy and aqueous geochemistry has demonstrated where brucite is an important reservoir for Fe(II) to react with water to produce H_2. We will also discuss how these mineralogical observations are related to the distribution and activity of hydrogenotrophic organisms in the serpentinite-hosted subsurface biosphere.