Veinlet-scale reactive transport during serpentinization: Implications for H_2 fluxes from oceanic serpentinites

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H_2 fluxes from mid-ocean ridges strongly controlled the redox evolution of Earth's ocean-atmosphere system through time. However, the impact of these emissions on global redox budgets is poorly understood because both the rates and extents of Fe oxidation during serpentinization of the oceanic lithosphere are currently unknown. In this study, we utilize a combination of thin section petrography, synchrotron micro-X-ray Fluorescence (μXRF) mapping, synchrotron micro-X-ray Absorption Near-Edge Structure (μXANES) spot analyses and mapping, synchrotron X-ray Computed Tomography (XRCT), Fourier Transform Infrared (FTIR) spectroscopic mapping, and combined Small and Ultra-Small Angle Neutron Scattering ((U)SANS) measurements in order to examine the veinlet-scale transport and oxidation of Fe during serpentinization of olivine in IODP drill-core samples from the mid-Atlantic Ridge. μXRF mapping reveals significant variation in the concentration of Fe from the olivine surface to the magnetite-filled veinlet center, which FTIR mapping suggests is associated with the transition from olivine at the veinlet edge, to Fe-brucite and Fe-serpentine, and finally to magnetite in the veinlet core. μXANES spectra show an accompanying shift in the centroid of the pre-edge peak, indicating changes in the oxidation state of Fe during this reactive transport process, which (U)SANS measurements show is facilitated by nanoscale porosity in the serpentine veinlet. The 2D μXRF, μXANES, and FTIR maps and spot analyses, combined with the optical petrographic visualizations and the 3D XRCT data sets, permit a quantitative description of the heterogeneity in the extent of Fe oxidation during serpentinization. Together, these data sets provide vital input data for the application of microcontinuum- and crustal-scale reactive transport models, which, in turn, facilitate the calculation of H_2 fluxes from serpentinizing environments throughout Earth history.