

Solubility of fluorine and chlorine in nominally anhydrous mantle minerals: Implications for mantle metasomatism and arc magmas

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Subduction fluxes and recycling of halogens are important for interpreting the fate of halogen budget of altered subducting slab, availability of ligands for complexing in aqueous fluids, origin of high-pressure brines, and volatile signatures of arc magmas. We investigated solubilities of fluorine and chlorine in forsterite, enstatite and pyrope, and halogen partitioning between aqueous fluids and these minerals by piston-cylinder experiments at 1100 °C and 2.6 GPa. The chlorine solubility in forsterite, enstatite and pyrope is very low, 0.2-0.9 ppm, and it is independent of the fluid salinity (0.3-40 wt. % Cl). The fluorine solubility is 16-31 ppm in enstatite and 24-52 ppm in pyrope, also independent of fluid salinity. Forsterite dissolves 246-267 ppm up to a fluid salinity of 1.6 wt. % F. At higher fluorine contents in the system, forsterite is replaced by the minerals of the humite group. The fluid-mineral partition coefficients are 10^1 - 10^3 for fluorine and 10^3 - 10^6 for chlorine. The latter values are approximately three orders of magnitude higher than those for hydroxyl partitioning suggesting a gradual increase in the fluid salinity during fluid percolation through the mantle wedge. Energetics of fluorine incorporation in forsterite and the forsterite-humite chemical equilibria in the system Mg_2SiO_4 - MgF_2 were further explored by first principles computations. The fluorine solubility in forsterite strongly increases with temperature, from 0.01 ppm F at 500 K up to 0.33 wt. % F at 1900 K and 0 GPa. By contrast, the effect of pressure on the fluorine solubility is very small, producing a decrease by a factor of two to three at 12 GPa. Consequently, partition coefficients of fluorine between forsterite and aqueous fluid (or silicate melt) are expected to increase with increasing temperature and decreasing pressure. When fluids or melts pass through the mantle wedge, fluorine will most efficiently be stored in the high-temperature portions of the wedge, promoting mantle metasomatism beneath the arc, and it will be released when the metasomatized mantle is advected to colder regions or to higher pressures.

Diffusion and microbial consumption of oxygen in an acidic geothermal iron-oxide mat

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The role of dissolved oxygen as a primary electron acceptor for microbially mediated iron oxidation was investigated within the primary flow path of an acidic geothermal spring in Norris Geyser Basin, Yellowstone National Park. Previous data has suggested that Fe(II)-oxidizing microbial populations (e.g., *Metallosphaera yellowstonensis* and potentially other novel members of the domain Archaea) represent the primary-producers within these microbial communities. Consequently, the availability of oxygen is hypothesized to limit microbial Fe(II)-oxidation and primary-productivity in this system. In situ measurements of oxygen profiles were obtained perpendicular to the direction of convective flow across the aqueous phase-Fe(III)-oxide interface using oxygen microsensors. Dissolved oxygen concentrations drop below detection by ~ 600 μ m into the Fe(III)-oxide mat, indicating reactive oxygen consumption and defined spatial gradients. Evaluation of the oxygen flux across the liquid-mat boundary showed that convection was negligible compared to diffusive transport in the mat. Reaction-diffusion models were evaluated assuming both zero and first-order reaction kinetics. The in situ measurements and models suggest that the rate of oxygen consumption exceeds the rate of diffusion. Thus, microbially mediated Fe(II)-oxidation in this system is likely limited by oxygen diffusion, resulting in an active surface layer of Fe(III)-oxide biomineralization.