

Iron oxidation and sulfur recycling in subduction zones: insights from multiphase fluid-rock reactions

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Chemical mass transfer in subduction zones is a fundamental aspect of plate tectonics on Earth, driving arc magmatism and the recycling of subducted materials to the surface. The reactive mechanisms between aqueous fluids released by slab dehydration and mantle minerals are not well understood, but are likely associated with redox transfer, which may explain the generally oxidized character of arc magmas. Here we use the Deep Earth Water (DEW) model to perform mass transfer calculations of sulfur-saturated slab fluids at variably oxidized conditions (FMQ+1 to FMQ+3) reacting with garnet and spinel lherzolite assemblages. Fe^{3+} -bearing multi-component phases are considered in DEW calculations for the first time, including Fe-Ti-Cr-Mg-Al oxides (ilmenite and spinel solid solutions), as well as clinopyroxene (esseneite, $\text{CaFe}^{3+}\text{AlSiO}_6$) and garnet (andradite, $\text{Ca}_3\text{Fe}^{3+}_2\text{Si}_3\text{O}_{12}$) components. Fluid-rock reaction heating paths are calculated at fixed pressure (2 and 3 GPa) and temperature increasing from 650 to 950°C.

At those subarc conditions, sulfate is very efficient at oxidizing iron and producing overall oxidized mineral products after reaction with the depleted mantle, whereas carbonate-rich fluids cause only marginal oxidation. Modelled Fe_2O_3 contents in spinel and clinopyroxene match those of natural data from variably metasomatized xenoliths. Modeled bulk rock Fe_2O_3 contents also compare well with, but do not reach the high concentrations observed in the most oxidized arc xenoliths. This is likely due to an incomplete characterization of Fe^{3+} -bearing mineral endmembers in our database (e.g. orthopyroxene, amphibole, mica).

Furthermore, despite oxidizing significant quantities of iron, sulfate-saturated fluids remain sulfate-rich owing to very high initial solubilities, implying that the sulfur isotope composition of oxidized slab fluids dominates the sulfur isotope signature of primitive arc magmas. We show that our calculated sulfur solubilities are generally consistent with the global fluxes of H_2O and S in and out of subduction zones.