

Does fluid transport in the mantle wedge determine radiogenic isotope ratios in arc magmas?

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Radiogenic isotope ratios are commonly used to distinguish the relative inputs of subducted oceanic crust, subducted sediments, and mantle wedge material to the generation of arc magmas. Most mixing models assume that fluids (whether they be aqueous, silicate melts, or supercritical) faithfully retain the isotopic ratios of their slab sources during transit to the melt source region in the mantle wedge. This assumption drives much of the physical modeling of fluid transport in the mantle wedge. Relaxation of this assumption, however, allows us to explain some otherwise puzzling features of arc magmas, and to test the robustness of isotopic tracers under a variety of circumstances. For example, modeled slab-derived fluid compositions based on measured arc magma compositions indicate that some Sr isotope compositions in fluids are considerably less radiogenic than would be expected for altered oceanic crust, or a mixture of crust and sediment. In such cases we must conclude that a) the Sr contribution from the slab has been over-estimated, b) the composition of the subducted crust is not what we believe it to be, or c) the composition of the slab-derived fluid has been altered prior to arrival at the melting source region. Simple numerical models are used to show that fluids released from the slab can exchange Sr with the mantle wedge if reactive transport is allowed, causing the Sr isotope ratios in the fluids to be subtly decreased toward mantle values prior to reaching the magma source region. The extent of exchange depends on the cumulative rock/fluid ratio experienced during transport. At the same time, the Pb isotope ratios of the initial fluids will be retained. Thus Pb is a robust tracer of slab (both crust and sediment) inputs to arc magmas, while Sr isotope ratios in fluids may not necessarily reflect those in the subducted slab.

The role of intermediates during metal carbonation of forsterite in wet supercritical CO₂

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The capture and storage of carbon dioxide and other greenhouse gases in deep geologic formations represents one of the most promising options for mitigating the impacts of greenhouse gases on global warming. As a result there has been a focus on evaluating mineral-fluid interaction for subsurface CO₂ storage in aqueous solutions in contact with supercritical CO₂ (scCO₂). However, at the interface with the caprock over the long term, interactions with neat to water-saturated non-aqueous fluids are of equal if not more importance given recent observations that wet scCO₂ is immediately reactive with steel, cement, and certain mineral assemblages. In such a scenario, it is of vital importance to understand the mechanistic role of small quantities of water dissolved into the scCO₂ fluid in mineral carbonation reactions.

Here we present results on the metal carbonation of forsterite (Mg₂SiO₄) as a function of water content in scCO₂ solutions conducted at 80°C and 80 atm pressure. NMR and TEM analysis of the reaction products showed that at high water contents where an aqueous solution was present forsterite was converted into magnesite and an amorphous SiO₂-like reaction product. As the amount of added water became limiting, intermediate reaction products could be identified until at zero added water no reaction products were observed. Intermediate reaction products identified both by NMR, XRD, and molecular simulation is dypingite [Mg₅(CO₃)₄(OH)₂•5H₂O] and a complex mixture of partially hydrated/hydroxylated Q1 and Q2 silica, the latter of which implies consumption of water via hydrolysis of Mg-O-Si linkages. If insufficient water is available, the reaction does not proceed far enough to form magnesite and SiO₂ reaction products. Water in excess of this limit yields the crystalline reaction products, a process that in turn is expected to liberate water. Hence, for a given fluid/forsterite ratio there is a water concentration threshold above which water serves in a catalytic role for the carbonation process.