Solubility of Carbon and Hydrogen in Melts at High Pressures and Reduced Conditions

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Important characteristics of the Earth's mantle, such as the temperature of its solidus at a given depth and the compositions of mantle-derived magmas (e.g. silica-saturated vs. silica-undersaturated), are dependent on the prevailing oxidation state and the solubilities of volatile species. In particular, C-H-O species are known to have large effects on mantle solidus temperatures and magma compositions. Magmas on the Earth's surface are generally oxidized ($fO_2 \sim$ FMQ) and CO₂-bearing. However, the source region of these magmas - the convecting upper mantle - is likely to be significantly reduced at depth, probably ~ FMQ-2 to FMQ-3 log units for basalt source regions and < FMQ-3 log units for kimberlite source regions. The stable C-H-O volatile phase under these fO2 conditions will be a CH4-H2±H2O fluid, rather than CO₂-H₂O. Calculations suggest that upper mantle fO2 may decrease by as much as 0.6 log fO2 units per GPa (relative to FMQ). The question arises as to how oxidized magmas can be extracted from a reduced source.

In order to tackle this question, we have undertaken an investigation into the role of C-H-O volatile species and their interaction with silicate melts. Initial experiments have been conducted at 2.8 GPa with silicate melts in the system forsterite-nepheline-quartz (Fo-Ne-Qz), which represents a simple analogue to natural basalts and peridoitie melting. Experiments were first conducted in order to locate the olivine-orthopyroxene two-phase boundary in the system Fo-Ne-Qz+CH₄-H₂O fluid. Increasing pressure shifts this boundary to increasingly silica-poor compositions, but pure H₂O and CH₄ fluids have the opposite effect and produce increasingly silica-rich compositions. CH₄-H₂O fluids significantly lower the 2.8 GPa olivine-orthopyroxene cotectic (~ 1260°C compared to ~ 1490°C under volatile-free conditions) but have little effect on the cotectic composition. Jadeite glass was then saturated at 2.8 GPa and 1240°C with CH₄-H₂O fluid. The glass was analysed by FTIR spectroscopy to determine the solubility of C-H-O volatiles. The volatile solubility of CH₄-H₂O fluid in equilibrium with Jadeite-Ferrosilite (Jd₅₀Fs₅₀) melt was compared at 0.5 and 2.8 GPa and 1240°C. Measured and calculated fluid species ratios generally agree at 2.8 GPa. CO₂/CO₂+H₂O ratios show a marked increase at 0.5 GPa compared to 2.8 GPa, with Fe³⁺/Fe²⁺ ratios mirroring this increase. The experiments demonstrate an important redox exchange equilibrium by which degassing of carbon-rich fluids containing small CH₄ contents increase Fe+++/Fe++ in the decompressing melt.