

The effect of composition and pressure on the structure of carbonate-silicate melts using *in situ* X-ray diffuse scattering

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Carbonatites are carbon-rich magmas that are crucial participants in the deep-Earth carbon cycle. The structure of carbonate-silicate liquids at high P-T conditions helps determine their physical and chemical properties at depth, but little is known about these liquids. To examine the liquid structure of carbonate-silicate binary systems as a function of pressure and composition, we performed *in situ* X-ray scattering experiments at HPCAT (Advanced Photon Source, Argonne National Laboratory) using a Paris-Edinburgh press. Mixtures from the CaCO₃-CaSiO₃ (calcite-wollastonite) and CaCO₃-Mg₂SiO₄ (calcite-forsterite) binary systems were used to simulate mantle silicate carbonatites. Samples were loaded using the experimental setup of Yamada et al [1], and held at 1800 °C and a pressure of either 3 or 6 GPa while energy dispersive X-ray scattering spectra were recorded.

Pair distribution functions for pure calcite liquid calculated from the scattered X-ray intensities confirm the presence of an ionic liquid, as found by previous investigators. [2,3] The silicate-rich liquids show evidence of significant polymerization, even in melts rich in Mg₂SiO₄ starting material. For silicate-rich mixtures in both binary systems, the extent of silica polymerization, as measured by scattering at the nearest neighbor Si-Si distance of 3.3 Å, increases, goes through a maximum at ~40 wt% carbonate, and then decreases as the carbonate content is increased. Experiments at 3 and 6 GPa for fixed bulk compositions reveal that rising pressure increases the extent of silica polymerization in carbonatite melts, implying that decompression during buoyant ascent decreases the viscosity of the liquid. Thus, both pressure and carbonate-silicate ratio are important controls on the polymerization state of silica in carbonate-silicate melts, while the Si:O ratio appears to have little influence.

[1] Yamada, A. *et al* (2011) *Rev. Sci. Instr.* **82**, 015103. [2] Waseda & Toguri (1977), *Metall. Trans. B* **8B**, 563-568 [3] Benmore *et al* (2010), *Phys. Rev. B* **82**, 224202.