

The effect of pressure on the structural environment of trace elements in silicate melts

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Trace element partitioning between minerals and silicate melts depends on temperature, pressure and composition (including $a\text{H}_2\text{O}$, $f\text{O}_2$ etc.). The sites of trace elements in minerals are easier to model than the sites in melts because the former depend on readily-determined properties such as bulk and Young's moduli and can be observed using X-ray or neutron diffraction. The highly disordered nature of silicate melts makes them more challenging to study, with X-ray absorption spectroscopy (XAS) one of the principal techniques to study local order [1]. Models such as lattice strain theory ignore the characteristics of the melt and focus exclusively on the properties of the crystal and the ionic radii of the trace elements relative to the lattice site in the mineral of interest.

Although melts and glasses are often referred to as 'amorphous', this is something of a misnomer. Spectroscopic studies have demonstrated that, for major elements, there are systematic variations in the degree of polymerisation and coordination environment of the constituent elements as a function of composition, pressure and temperature. These structural characteristics are much better characterised for major elements than for trace elements.

We have used XAS to examine the local structural environments of seven trace elements (Rb, Sr, Y, Zr, Nb, Cd, In) in silicate glasses annealed at the glass transition temperature at a range of pressures (0.001 – 6 GPa) for a single melt composition in the system CMAS. Data were collected in fluorescence mode at the Australian Synchrotron and processed using Athena and Artemis.

The elements studied vary significantly in their response to changes in pressure: e.g. Nb shows negligible differences in coordination number and radius, whereas Zr and Y change both as a systematic function of pressure. Furthermore, the coordination changes in Zr appear to be non-linear, with a greater rate of change above 4 GPa. These results demonstrate the potential for inter-element fractionations due to changes in the availability and geometries of different sites within a silicate melt across the range of pressures studied.

[1] Calas et al., 1987. Phys. Chem. Minerals 15, 19-29.