

The true effect of CO₂ on silicate melt structure: Insights from NMR spectroscopy and molecular dynamic simulations

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Carbon dioxide (CO₂) is the second most abundant volatile implied in magmatic systems. The effect CO₂ induces on silicate melts structure is not clear.

We investigated the molecular structure of CO₂-bearing synthetic glasses with various compositions: basalt and nephelinite (~50 and 40 wt.% SiO₂, respectively). Glasses were synthesised at various pressure (50 MPa to 3.0 GPa) and temperature (1200-1500°C). We used Raman and Solid-State NMR spectroscopies to characterize the recovered glasses. First-Principle Molecular Dynamics (FPMD) simulations were conducted on the CO₂-bearing basaltic composition at high pressure (5.0 and 8.0 GPa).

The ²⁹Si NMR results on nephelinite glasses indicate an apparent increase in the degree of polymerization with increasing CO₂ content: NBO/Si decreases from 1.2 to 0.5 with the addition of 4.5 wt.% CO₂. This strong increase in glass polymerization is inconsistent with mass balance consideration. FPMD simulations point towards an absence of effect of CO₂ on melt polymerization: NBO/Si changes from 0.9 to 0.8 with the addition of 20 wt.% CO₂ at 8.0 GPa.

We propose that CO₂ dissolution scavenging available NBOs in the melt provokes a ²⁹Si NMR shift which mimics an apparent increase in glass polymerization.