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Quantification of CO₂ dissolved in silicate glasses and melts using Raman spectroscopy: implications for geodynamics

Quantification of CO₂ dissolved in silicate glasses and melts using Raman spectroscopy: implications for geodynamics

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Understanding Earth degassing is fundamental in global studies of our planet history, as well as in studies of its recent climate. Degassing occurs mainly at Mid Ocean Ridges via exsolution of CO₂ vesicles in ascending tholeiitic magma, and probably begins at some 30 km under the ridge. Therefore, a precise knowledge of how carbon solubility varies during ascent from the source region is mandatory, a process for which the effect of pressure remains poorly known. A pressure increase induces melt compression, known to diminish argon dissolution with respect to Henry's law at pressures above ~10 kbar, but this effect is poorly documented for carbon where things are complicated by the transformation of CO₂ into carbonate ion, CO₃²⁻. Early experimental investigations on carbon solubility in various silicate melts up to ~20-30kbar have shown that Henry's law is not followed at high pressures.

We have performed an experimental study of C dissolution in basaltic melts using high-pressure facility in Clermont-Ferrand (France). Analysis of dissolved C was performed using a micro-Raman spectroscopy. Dissolved carbon appears as clear bands due to carbonate ions (an intense peak at ~1100 cm⁻¹ and a doublet in the 1350-1600 cm⁻¹ region), molecular CO₂ being not detectable. Calibration of Raman spectroscopy for quantitative analysis was done by preparing standards at atmospheric pressure and analyzing them using a stable isotope mass spectrometer.

The results show that carbon concentration increases steadily with increasing pressure, a behavior consistent with (rare) previous studies on basaltic melts. We also have performed molecular dynamics simulations to investigate the dissolution of CO₂ in a silicate melt. The calculated solubility is consistent with the data, which help understanding how pressure acts on fluid and melt, and yield insight into the details of how CO₂ and CO₃²⁻ interact with the melt network. However, the fact that the carbon solubility in a MORB is continuously increasing with pressure is somewhat surprising, and will be discussed.

This work has shown that

- (i) Raman spectroscopy can be used to quantify C content in natural samples
- (ii) The C solubility measured in basaltic melt exhibits a behavior with pressure different from that exhibited by rare gases.
- (iii) Our results have important implications concerning the history of the atmosphere degassing and structure of the mantle.

キーワード: CO₂, Raman spectroscopy, silicate glasses and melts

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