

The speciation of carbon dioxide in silicate melts from *in situ* infrared measurements

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Carbon dioxide may dissolve in silicate melts either as molecular CO₂ or as carbonate. The ratio of the two species in quenched glasses strongly depends on composition. While molecular CO₂ predominates in silica-rich, polymerized glasses, only carbonate can be detected in depolymerized basaltic glasses. However, the diffusion coefficient of CO₂ in rhyolitic and basaltic melts is nearly the same at identical temperatures, despite the fact that CO₂ speciation in the quenched glasses is very different. This suggests that the speciation observed in quenched glasses may not be representative for CO₂ speciation in silicate melts.

In order to determine CO₂ speciation directly in the silicate melt, we measured the infrared absorption spectra of silicate melts inside an externally heated diamond cell to more than 1000 °C. Samples include an alkali silicate melt with a degree of polymerization similar to a basalt and a dacite melt. Type II a diamonds of reduced thickness were used to minimize absorption due to the anvils. Measurements to about 700 °C were carried out using a globar source, experiments at higher temperature were carried out at the ANKA synchrotron source in Karlsruhe.

Only carbonate was observed in the alkali silicate melt to the highest temperatures studied. In the dacite melt, molecular CO₂ and carbonate were found to coexist in the melt. The quantification of speciation was complicated by the fact that the extinction coefficients of both molecular CO₂ and of carbonate were found to significantly decrease with temperature. This effect can be attributed to the changes in the population level of the vibrational states with temperature.

The high-PT stability of apatite: Constraints on transport and storage of phosphorus and halogens in the Earth's upper mantle

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High-PT experiments were performed in a range 3-15 GPa and 850-1450°C in order to constrain the stability field of OH- and Cl-apatite and to study the partitioning of phosphorus between apatite or its high-P breakdown-product and the coexisting phases. The following starting materials were used: (1) an average MORB, (2) an Fe-free simplified MORB equivalent in the system SiO₂-Al₂O₃-MgO-CaO-Na₂O-TiO₂ and (3) a fertile lherzolite. The starting materials were doped with 3% synthetic OH-apatite or Cl-apatite and with a trace element mix. In the average MORB, OH-apatite is stable to 7 GPa/950°C coexisting with grt + cpx + SiO₂ + TiO₂. From 7.5-11 GPa and 950-1200°C, OH-apatite is replaced by tuite (γ-Ca₃(PO₄)₂ [1]) coexisting with the same assemblage. At higher P and T, no phosphate-phase was found due to increasing partitioning of phosphorus into coexisting grt or grt + melt. The P₂O₅-concentration in grt buffered by apatite, tuite or quenched melt is between 0.2-0.3 wt% but rises to 0.6-0.8 wt% in the absence of a phosphate-phase. Compared to MORB, the lherzolite system shows notable differences in the stability of phosphate-phases and in the P₂O₅-concentration of coexisting silicates. Apatite is stable to slightly higher pressures of ≥7.5 GPa/≥1000°C and was found to coexist with tuite at 8.5 GPa/1000°C in an assemblage ol + opx + cpx + grt. In addition, apatite in the lherzolite incorporates significant amounts of Mg and Fe, reaching a total of 4.0 wt% at 8.5 GPa. This may be a reason for its increased P-stability in bulk (3). By comparison, (MgO+FeO) in eclogitic apatite does not exceed 1.0 wt% at 7.0 GPa. In coexisting apatite and tuite, the latter strongly partitions Sr and REE. The P₂O₅-concentration in lherzolitic grt is always higher than in MORB-grt, ranging from 0.7 wt% at 6 GPa/1100°C coexisting with apatite to 1.9 wt% at 13 GPa/1200°C in the absence of a phosphate-phase. The results of this study show that apatite in both MORB and lherzolitic bulks has a much smaller PT-stability field than previously assumed [1] and, consequently, that halogens cannot be transported and stored in apatite to depths > approx. 250 km.

References

- [1] Murayama, J.K., Makai, S., Kato, M., Kumazawa, M. (1986) *Phys. Earth. Planet. Inter.* **44**, 293-303.