Quantification of CO_2 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 Mean Ocean Ridges via exsolution of CO2 vesicles in ascending tholeiite 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 CO_2 into carbonate ion, CO_3^{2-}. 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 CO_2 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 CO_2 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 CO_2 and CO_3^{2-} 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

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

Microbial community structures in shallow-sea, deep-sea, and terrestrial hydrothermal systems

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The shallow-sea vent fluids and beach sediments at Vulcano Island (Italy) have yielded more culturable hyperthermophiles than any other hydrothermal system—marine or terrestrial. A seemingly insignificant hot spring in Yellowstone National Park (USA) was touted as harboring "remarkable" archaeal and bacterial diversity [1,2]. But how similar are the microbial community structures between sites? And can we quantitatively link the variability between sites to geographical isolation or geochemical parameters such as pH; temperature; pressure; salinity; or oxygen, sulfide, hydrogen, and organic carbon concentrations. In addition, do shallow-sea, deep-sea, and terrestrial hydrothermal systems share a significant number of taxa? And how do the microbial community compositions in these three fundamentally different types of high temperature ecosystems correlate with a wide range of geographical and geochemical parameters?

Our preliminary data analysis of ~1200 archaeal 16S rRNA gene sequences (assigned to ~300 operational taxonomic units (OTUs) with >90% similarity) from 5 terrestrial, 6 shallow-sea, and 17 deep-sea hydrothermal systems shows that ~85% of the OTUs are unique to one of the three types of hydrothermal systems. Only 2% are shared between 'terrestrial' and 'deep-sea' and between 'terrestrial' and 'shallow-sea'; 11% are shared between 'deep-sea' and shallow-sea'. Only 1 OTU (or 0.3%) of those investigated was found in all three types of systems—this was a sequence related to the Thermoproteales, an order of obligately or facultatively anaerobic hyperthermophiles known for carrying out sulfur respiration both chemolithotrophically (with H_2) or heterotrophically. A dendogram of sequences binned by sample showed a clear clustering of the terrestrial, the shallow-sea, and the deep-sea systems. Within the deep-sea systems, the samples from sediments, chimneys, vent fluids, and in situ colonization systems clustered separately. We hypothesize that multivariate analyses of geochemical and microbiological datasets can be used to identify major geobiological patterns and putative environmental causes.

[1] Barns *et al.*, (1994). PNAS 91:1609. [2] Hugenholtz *et al.*, (1998). J. Bact. **180**:366.