

Cristal de carbonate de 10 μm presque totalement dissout, à l'équilibre avec le fluide aqueux dans une cellule à enclume de diamant (300°C, 5 Gpa)

Solubilité des carbonates en zone de subduction

Isabelle DANIEL

Laboratoire de Géologie de Lyon

Carbonate minerals may be recycled into the mantle or dissolved in fluids in subduction zones. However, the evolution of carbonate minerals in equilibrium with aqueous fluids as well as the nature of the chemical species of dissolved carbon in the deep crust and mantle at high PT conditions are still unknown. We report an integrated experimental and theoretical study of the equilibration of carbonate minerals with aqueous solutions at the pressure and temperature characteristics of subduction zones. The fluid speciation was studied using in situ Raman spectroscopy in an externally heated diamond anvil cell. Solubilities were measured using X-ray fluorescence at the ESRF.

At equilibrium with the carbonate crystal, the Raman data show that bicarbonate is the most abundant species in low-pressure fluids whereas carbonate becomes progressively dominant at higher pressure. The relative amounts of dissolved carbonate and bicarbonate were estimated from the areas of the ν_1 and ν_5 symmetric stretching Raman modes of the carbonate and bicarbonate ions, respectively after correction from their Raman cross-sections.

The Raman data were used to constrain a theoretical thermodynamic model of the fluid speciation in equilibrium with carbonate minerals. Revised thermodynamic properties of aqueous CO_2 and HCO_3^- , data for the aqueous CaHCO_3^+ complex from 4 to 90°C, and estimated dielectric constants of water, enabled an equation of state characterization of the standard Gibbs free energy of CaHCO_3^+ at the conditions of the Raman study. At 300-400°C, the Raman speciation results were used to constrain equilibrium constants involving the carbonate ion. The results indicate that CO_2 is a minor species in fluids in equilibrium with aragonite at 300-500°C and $P > 1$ GPa. Instead, the CaHCO_3^+ species becomes important at high pressures until carbonate ion including the integration of the CaCO_3^0 complex become the dominant C-species. The model is validated against independent measurements of the solubility.

Jeudi 7 novembre 2013 à 11h

Salle de conférences d'ISTerre

OSUG-C, 1381 rue de la piscine, Campus Universitaire
Arrêt Tram B/C Bibliothèques universitaires