

超臨界岩石-水相互作用のための溶存種の熱力学データ：溶解実験と密度を用いた外挿

Thermodynamic data of aqueous species for supercritical water-rock interaction: dissolution experiments and density-based extrapolation

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Geothermal reservoirs under the supercritical conditions (supercritical geothermal reservoirs) have been taken greater attentions as the potential renewable energy in volcanic areas in the world. The chemical and physical processes of such supercritical regions are of special importance for various geological processes including understanding the formation of hydrothermal ore deposits, hydrological properties around brittle-ductile transitions of the crusts, and metamorphic and metasomatic processes. The thermodynamic data of the aqueous species at elevated temperatures and pressures has been constructed based on the Helgeson-Kirham-Flowers (HKF) model, where molar Gibbs energy is defined as a function of dielectric constant of water and species-specific Born parameter (e.g., Tanger and Helgeson, 1988), and the most common data is available through a software SUPCRT92. However, a large issue for understanding the supercritical water-rock interaction is a lack of thermodynamic data of charged aqueous species (ions and complexes) for the region near critical point of water and low water-density region (<0.35 g/cm³). In contrast to the electrostatic model like HKF model, the empirical density model has been proposed based on the fact that the log of molar Gibbs free energy of aqueous species shows a linear relationship with log of water density. Based on this model, Caciagli and Manning (2003) proposed the extrapolation of SUPCRT log K data to the high pressure regions; however, the effectiveness of this method to the low density region is not clear. In addition, the experiments for dissolution experiments are lacking for the low-density region.

In the present study, we tried the expansion of log K data set of the speciation and mineral dissolution into the vapor-like supercritical region (400, 420 degreeC, 20-35 MPa). We also conducted the new experiments for the mineral solubility measurements. The apparatus consist of 60 ml vessel with Ti inner wall, and pressure was controlled by syringe pump. We conducted the dissolution experiments with albite powder and mix powders of albite-K-feldspar-quartz. Then, for evaluating the validity of the extrapolated data, we compared the extrapolated logK of mineral dissolution equilibrium and the ion products (log Q) obtained from the solution chemistry of the experiments and extrapolated log K data set of aqueous species. The deviation of log K from log Q is systematic and less than 2 in log unit, indicating the effectiveness of this method as the first approximation.

Reference

Tanger, J.C., Helgeson, H.C., 1988. Am J Sci, 288, 19-98.

Caciagli N.C., Manning, C.E., 2003. Contrib. Mineral Petrol, 146, 275-285.

キーワード：熱力学データ、超臨界流体、岩石-水相互作用、水熱実験

Keywords: thermodynamic data, supercritical fluids, water-rock interaction, hydrothermal experiments