In situ obvervation of critical phenomena of geofluids

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Geofluid was chiefly composed from mixture of H2O-CO2 solution, and additional electrolytes were contained in the geofluid. Interaction between geofluid and geomaterial has great role for chemical and mechanical processes in the Earth interior. Water-rock interaction was inferred by fluid condition such as under sub-critical or supercritical, and pressure and temperature even in the supercritical region.

Experimental observation of critical behavior of critical temperature and pressure of multicomponent geofluid was carried out for discussing geofluid- rock interaction, and comparison of the critical values (temperature and pressure) to the calculated thermodynamic value by equation of state (EOS) and empirical equation was performed.

Transparent window (sapphire: 25.5 mm diameter, 13 mm in thickness) was installed with pressure vessel. Maximum temperature is 500C and pressure is 40 MPa. Filling ratio of selected solution was 50%. After overheating, we conformed monophase state through the sapphire window, and then we can observe meniscus during extremely slow cooling process. Some differences between the values (critical temperature and pressure) in heating and in cooling process were observed. The critical values of pure water and pure CO2 in the cooling process is coincide to the value in literatures. Therefore, the value in the cooling process is suitable data for critical temperature and pressure for the solution.

With increasing concentration of electrolyte, critical point shifts to higher temperature and pressure, and with increasing CO2 mole fraction in the solution, critical points moved to lower temperature but higher pressure. Degree of shift is depended on ionic strength and chemical species.

Several equation of state (EOS) for solution containing some electrolytes have been proposed. EOS of Duan et al. (1995) was used here, and additional compute program by Dr K. Hoshino (Hiroshima University) was used to obtain calculated value of critical temperature and pressure based on EOS. Recently Shibue (2003) proposed new empirical equation to estimate of thermodynamic properties of multicomponent solution. We also compared calculated critical values to experimental ones. We describe temperature and pressure of the critical point for geofluid and model ocean water in the session.

Duan et al. (1995), Geochim. Cosmochim. Acta, 59, 2869-2882
Shibue Y., (2003), Fluid Phase Equilibria, 213, 39-51.