A Thermodynamic Consideration of the Equilibrium of CO_2-Hydrate in Seawater

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Thermodynamic equations of stability and solubility of carbon dioxide hydrate were developed using the Pitzer method [1] with regard to the proposed disposal of CO_2 in the ocean. The effect of high CO_2 concentration on phase equilibrium was taken into account using the Pitzer method. Dissociation pressures were calculated for temperature and salinity ranges of 273 to 283 K and 0 to 40 for temperature and salinity respectively. The calculation of dissociation pressures has been approximated as a function of temperature and salinity by empirical equation. The solubility of carbon dioxide hydrate was calculated for the same temperature and salinity ranges as well as for hydrostatic pressures (Pdis) up to 50 MPa. It was found that the solubility of the hydrate increases with increasing temperature and decreasing hydrostatic pressure. An empirical equation describing hydrate solubility as a function of salinity, temperature, and pressure was obtained. In comparison with liquid CO_2, the hydrate reduces the major environmental impact of ocean CO_2 disposal, as seawater equilibrated with hydrate was found to have a higher pH than that of liquid CO_2 by more than a 0.15 pH unit.

[1] K.S. Pitzer, *Ionic interaction approach: Theory and data correlation,* In: Activity Coefficients in Electrolyte Solutions. K. S. Pitzer Ed.; 2nd Edition; CRC Press: Roca Raton Ann Arbor Boston London, 1991, p. 75-153.