American Mineralogist, Volume 91, pages 1346–1354, 2006

A model to predict phase equilibrium of CH_4 and CO_2 clathrate hydrate in aqueous electrolyte solutions

ZHENHAO DUAN* AND RUI SUN

State Key Laboratory of Lithospheric Evolution, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing, 100029, China

ABSTRACT

A thermodynamic model to predict phase equilibrium of methane and carbon dioxide hydrate in aqueous electrolyte solutions is presented. Using the Pitzer model to account for the variation of water activity due to electrolytes and dissolved gas in aqueous solutions, we extended the model based on ab initio molecular potential developed recently by us for the CH_4-H_2O and CO_2-H_2O binary systems to the CH_4 (or CO_2)-H_2O-salts system. Comparison of the model with extensive experimental data indicates that this model can accurately predict the phase equilibrium of CH_4 hydrate and CO_2 hydrate in various electrolyte solutions (such as aqueous NaCl, KCl, $CaCl_2$, NaCl + KCl, NaCl + $CaCl_2$ solutions, and seawater) from zero to high ionic strength (about 6 *m*) and from low to high pressures.

Keywords: Methane hydrate, carbon dioxide hydrate, thermodynamic stability, electrolyte solution, phase equilibrium, formation conditions