

Phase Behavior of Methane Hydrates in Confined Media

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Hydrate phase equilibria for the binary methane and water mixture were measured by high pressure differential scanning calorimetry in mesoporous and macroporous silica that had controlled pore size ranging from 4 nm to 200 nm. Oscillating dynamic method was used to form hydrates and determine the decomposition conditions of methane hydrates. Significant shifts of dissociation temperature were observed and fitted by Gibbs-Thomson equation. The experimental data up to 50 MPa indicated the interfacial energy in the confined media showed a pressure dependency. The enthalpy of hydrate fusion in confined media were experimentally measured and calculated by Clapeyron equation. Both methods indicated that the enthalpy of fusion was independent of pore size. The effects of interfacial energy on phase equilibria were investigated quantitatively by grafting of silica surface. The dissociation temperatures of methane hydrates in hydrophobic and hydrophilic silica were significantly different. Methane hydrates in strong hydrophobic porous silica tended to behave as bulk methane hydrates in terms of the equilibrium temperature. The measured phase equilibria data, interfacial energy and enthalpy were considered as important parameters for different gas hydrate simulation models.