Hydrate Model for CCS Relevant Gases Compatible with Highly Accurate Equations of State – I. Parameter Study and Model Fitting

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In this study, a gas hydrate model for pure CO_2 hydrates [1] was extended to eight hydrate formers, which are especially relevant in CCS applications. The hydrate model was adopted from the model of Ballard and Sloan [2], which is based on the van der Waals and Platteeuw [3] theory (vdWP). The new model is compatible with highly accurate equations of state (EoS) for other phases than hydrate given in the form of the Helmholtz energy or the Gibbs energy. A detailed analysis of the main parameters of the vdWP-based model including the specific hydrate volume, the Langmuir constant, and the EoSs employed for other phases was carried out. A new correlation based on the Murnaghan equation of state for solids [4] was developed for the volume of gas hydrates. The correlation provides reasonable prediction for the lattice parameter over wide pressure and temperature ranges for a large variety of sI and sII hydrates. This represents an important improvement compared to other correlations considered in some previous vdWP-based hydrate models. A comprehensive procedure for fitting the hydrate-model parameters (the reference properties, and the Kihara potential parameters) with the Levenberg-Marquardt algorithm was developed. The chemical potential of water for three-phase equilibria with hydrates and various hydrate composition data (hydration number and cage occupancies) can be used in a multi-property fit to determine the model parameters. The uncertainties of the experimental data were thoroughly studied in order to define a dimensionless quality criterion for the multi-property fit.

References

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