

Phase Equilibrium and Interfacial Properties Between Liquid and Hydrate or Ice Phases Obtained by Molecular Simulations

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Molecular simulation is a powerful tool to obtain the physical properties at a wide range of thermodynamic conditions for multi-phase or multi-component system, such as phase equilibrium condition or interfacial tension. The direct coexistence technique has been recently developed to compute the phase equilibrium from molecular simulations, then we applied it for the interfacial system containing liquid water and methane hydrate or hexagonal ice phases. All simulations were performed on the Infiniband PC cluster or Graphic Processing Unit cluster because the direct coexistence technique requires a larger system size than other techniques. For one-component, ice/water system, we established a new procedure to accurately predict the phase equilibrium conditions using an iterative scheme. The uncertainty of our procedure was examined by comparing with other technique to compute the phase equilibrium condition, such as the thermodynamic integration (TI) method, and we found that our procedure gives the more precise equilibrium temperature at fixed pressure than the TI method. The interfacial tensions at different contact planes were also examined, and compared with the experimental data. The potential model dependencies on these properties were studied to find the best water model to reproduce the experimental available data. For two-component, hydrate/water system, we compute the solubility of the solute molecule as well.