Development of State of the Art Models and Software for the Prediction of Vapor-Liquid Equilibria.

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Abstract

The design of chemical processes requires detailed knowledge of the physical properties and phase behavior for the compounds of interest. Ideally, this information would be determined from experiments, however, experiments are costly and time consuming, and in some cases experiments may not even be possible at the temperatures and pressures of interest. Therefore, methods are desired that may be used to predict the phase behavior of multicomponent mixtures to a high degree of accuracy in the absence of experimental data. One such methodology is atom-based molecular simulations, such as Gibbs ensemble Monte Carlo or grand canonical Monte Carlo, combined with histogram reweighting techniques. Because the predictions of simulation are based on the fundamental interactions between atoms in molecules, the predictive capability is expected to be robust for conditions outside the training set used in the optimization of potential parameters.

In this talk, I discuss the use of computer simulation, and the development intermolecular potentials, for the prediction of vapor-liquid equilibria in complex mixtures. Force fields based on traditional 12-6 Lennard-Jones potentials are presented for n-alkanes, CO_2, H_2S, SO_2, and their mixtures. The limitations of the standard 12-6 Lennard-Jones potential are discussed, and new force fields based on a new functional form, the Mie potential, are presented for alkanes, fluorocarbons, alkenes, and noble gases. The predictions of the Mie potential for various physical properties and phase behavior are shown to be improved over the predictions over the ubiquitous 12-6 Lennard-Jones potential, especially for non-polar molecules.

In addition, I describe the development of a new open source Gibbs ensemble Monte Carlo (GEMC) simulation engine known as GPU Optimized Monte Carlo (GOMC) http://gomc.eng.wayne.edu, and its application to the simulation of vapor-liquid equilibria for systems containing tens of thousands of interaction sites. GOMC is parallelized with NVIDIA®'s CUDA™ API and employs an energetic decomposition scheme to distribute work to device threads. Performance is improved significantly compared to serial code for systems containing over 100,000 interaction sites. Fundamental issues regarding the parallelization of the Monte Carlo algorithm are discussed. GOMC is designed for interoperability with existing molecular dynamics simulations, such as NAMD, and data analysis tools, such as VMD.