Uncertainty Quantification of Non-bonded Potentials for Prediction of Thermophysical Properties with Molecular Simulation

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Meaningful estimates of uncertainty are essential for molecular simulation to be a quantitative tool in thermophysical property prediction. As vapor-liquid equilibria is highly sensitive to the non-bonded potential functional form (Lennard-Jones 12-6, Mie n-m, Buckingham exponential-6, etc.) and the corresponding non-bonded parameters, the present study focuses on the parameterization and uncertainty quantification of the non-bonded potential. Unfortunately, uncertainty quantification methods (such as Bayesian inference) are computationally infeasible when molecular simulations must be performed with tens of thousands of non-bonded parameter sets. The novel methodology employed in this work reduces the computational cost of force field parameterization and uncertainty quantification by up to three orders of magnitude. The computational acceleration is achieved by reweighing configurations that are sampled with a small set of reference force fields to predict physical property values for parameter sets that are not simulated directly.

The proposed methodology enables the simultaneous optimization and uncertainty quantification of several non-bonded parameters to the large amount of experimental data available at the National Institute of Standards and Technology (NIST) Thermodynamics Research Center (TRC). Furthermore, advanced Bayesian methodologies are employed to ensure that the parameters are not overfit to the training data set by simultaneously investigating several non-bonded potential functional forms. This improves the transferability of the force field when compared to data at different state points.