Efficient Force Field Tuning for Predicting Thermophysical Properties

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Molecular simulation is a powerful tool for studying the thermophysical properties of materials and shows great promise for screening vast molecular design spaces. However, utilizing molecular simulation in this capacity requires accurate force fields. Developing force fields has historically been a laborious, time-consuming endeavor. Though off-the-shelf force fields offer accurate predictions for some systems, they inevitably lack quantitative accuracy across the extraordinary range of chemistries found in the natural and synthetic world. Further manual parameter tuning is often necessary to ensure the model has the required accuracy for the molecule(s) and properties of interest.

In this work we propose a machine learning -enabled automated force field optimization framework. We show integrating Gaussian Process surrogate models and support vector machine classifiers facilitates rapid tuning of force fields and provides a quick and efficient route to accurate, physics-based molecular models. In our case studies, we tune van der Waals repulsion-dispersion interaction parameters to reproduce experimental property measurements. In our first demonstration case, we optimize force fields for two hydrofluorocarbons, HFC-32 and HFC-125, for liquid and vapor densities, vapor pressures, and enthalpies of vaporization. We can find at least 26 HFC-32 and 45 HFC-125 force field parameter sets in a timeframe of weeks which give less than 5% mean absolute percent error in all of the properties of interest. Additionally, we find that these parameter sets are able to predict transport and critical properties accurately for HFC-32 and HFC-125 without further tuning. In our second demonstration case, we apply our framework to develop a force field to predict solid properties of ammonium perchlorate, including lattice parameters, unit cell structure, hydrogen bond distances, angles, and symmetry. Multiple parameter sets have been found that outperform existing force fields in reproducing experimental observations of the listed quantities.