

Molecular simulations enable studies of the impact of microscopic processes on macroscopic phenomena.

In order to be able to apply simulations successfully within natural science and process engineering, appropriate molecular models have to be present. The foundation of a simulation to predict physical properties quantitatively is the so-called force field. The latter describes both intramolecular and intermolecular interactions.

The main difficulty lies in the parameterization of a force field, especially of the intermolecular part. To do this manually is extremely time consuming, as for each set of force field parameters, a numerically complex molecular simulation has to be performed. Already two to four hours are required for the simulation of a very small system, e.g. consisting of 1000 small molecules, during one nanosecond on a modern parallel high-performance computer. In this thesis, a new automated parameterization scheme based on the formulation and solution of a mathematical optimization problem is proposed. While intramolecular degrees of freedom and partial atomic charges can be calculated by quantum mechanics, the calibration of parameters describing intermolecular interactions, is not trivial at all. Within the optimization procedure, properties resulting from a molecular simulation like density, enthalpy of vaporization, self-diffusion coefficient, and vapor pressure, are fitted to their respective experimental reference data. As molecular simulations exhibit a high complexity and the resulting properties are affected by statistical noise, the optimization problem is solved by both already existing and novel, efficient, and robust numerical algorithms. Another goal of this thesis is to evaluate the optimized force fields with regard to their physical and chemical applicability.