

"Computational molecular engineering by reliable molecular modelling and massively-parallel molecular simulation"

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Adjusting force field parameters, while aiming at an accurate representation of various thermophysical properties, constitutes a multicriteria optimization problem. Since different objectives are generally conflicting, a single optimal solution cannot be determined.

Instead, a multicriteria optimization algorithm is employed to identify the Pareto set, i.e. the set of parameter combinations that represent rational compromises between conflicting objectives. Multi-criteria optimization is applied here to two-center Lennard-Jones plus point quadrupole models, focussing on carbon dioxide as a test case.

Furthermore, the physical basis of the CO₂-N₂O analogy is examined by molecular simulation. The CO₂-N₂O analogy is an approximation which, in its most common form, states that the ratio of the physical solubilities of N₂O and CO₂ (in some solvent of interest) is the same as that in pure water at the same conditions. In many technically relevant cases, the physical solubility of carbon dioxide is difficult or impossible to determine experimentally, since physical and chemical effects, which occur simultaneously, cannot be separated. The CO₂-N₂O analogy is then employed as a workaround. However, molecular simulation only partly confirms the validity of this approximation.

Homogeneous nucleation of carbon dioxide is considered by molecular simulation of large systems, regarding both the pure fluid as well as mixtures with carrier fluids, such as air, which do not participate in the phase transition significantly. Canonical ensemble molecular dynamics simulation using the Yasuoka-Matsumoto method is applied to nucleation in supersaturated vapors that contain more carbon dioxide than at saturation. Furthermore, bubble formation in metastable liquid carbon dioxide at negative pressure is simulated. Scale bridging results are obtained by using the `ls1 mardyn` program, which is shown to scale efficiently on up to 146000 cores, reaching a speedup factor of 133000 by massive parallelization.