Limitations of cluster-trained MLIPs for liquid density and diffusivity

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Machine-learned interatomic potentials (MLIPs) based on quantum-mechanical (QM) data are often used as a means to combining the performance of classical force-fields with the accuracy of electronic structure methods. Training on clusters instead of periodic snapshots of liquid structures could potentially permit the use of more advanced QM methods, such as hybrid-DFT or even coupled cluster calculations. Given the emerging interest in fine-tuning foundation models [1, 2], this could prove to be a way to raise the level of theory on such models with a limited effort.

The main challenges with cluster data-based MLIPs are that (i) cluster-solvent interactions are truncated in the training data (**Fig. 1**), which can translate to *out-of-domain* extrapolation if subsequent MLIPs are employed in MD simulation, and (ii) *the local atomic neighbour density in the gas-phase clusters can be quite different compared to a liquid* (not a full, or representative, atomic coordination). However, since MLIPs employ a cut-off radius, it is unclear how large these differences will be, e.g., compared to MLIPs trained on periodic data.

The goals of this work are to:

1. For selected organic liquids (EC and EMC), compare the densities and self-diffusivities resulting from MD simulations with MLIPs that have been trained on periodic data using different DFT functionals (i.e., DFT benchmarking with the help of "periodic MLIPs").
2. Compare the densities and self-diffusivities resulting from periodic- and cluster-based MLIPs.

There have been reports of success in the literature [3, 4] regarding MLIP training on cluster-data and applying the MLIPs to MD simulation of electrolytes. In the two cited references, the MLIPs treated Coulomb interactions explicitly. Our study considers solvents (no ions!) and use MACE-MLIPs which are not range-separated.

This work highlights important aspects that need to be taken into account when cluster-trained MLIPs are used for MD simulations of bulk liquids. We find that:

- All MACE-MLIPs (periodic- and cluster- based ones) produced stable 1 ns NPT trajectories.
- Even so, MACE-MLIPs trained on cluster data were found to be very sensitive to both the random training seed and to data selection, making the DFT functional benchmarking task a futile effort. (With the periodic-data-trained MLIPs, this worked well.)

Method details. In this work, MACE-MLIPs intended for molecular dynamics (MD) simulation of ethylene carbonate (EC) and ethyl methyl carbonate (EMC), were trained on two publicly available datasets—one based on periodic structures [5], the other based on gas phase cluster data [3]. The periodic training sets were labelled with PBE-D2, PBE-D3 and B97-D3 whereas the cluster-based training data was labelled with the B97-D3 and ωB97X-D3 functionals.

Figure

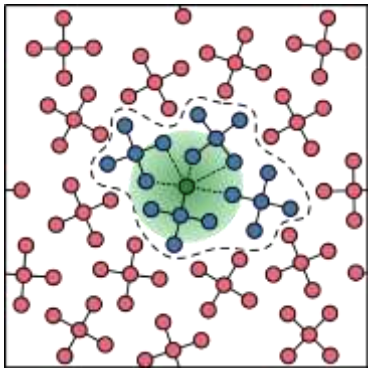

Figure 1. Important contributions to forces and energies are missing in clusters (within dashed line) if they are extracted from their liquid environment (red molecules). The green disc highlights the local atomic environment around a central atom, defined by the MLIP cut-off.

References

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