From Oligomers to entangled Polymers: How transferable are Machine Learning Interatomic Potentials?

Mirko Fischer[1], Andreas Heuer[1]
[1]Institute for Physical Chemistry, University of Münster, Corrensstraße 28/30, 48149 Münster, Germany

mirko.fischer@uni-muenster.de

Machine Learning Interatomic Potentials (MLIPs) have emerged as a powerful tool in computational chemistry and materials science, connecting highly accurate quantum chemistry (QM) calculations with fast and efficient molecular dynamics (MD) simulations [1].

Recent studies have demonstrated high accuracy and reliability of MLIPs for crystal and amorphous materials as well as for small organic molecules, including the description of reactions [2]. However, so far only few studies have focused on macro-molecules such as polymers [3].

A key challenge arises from their large molecular size, which makes reference calculations for training the MLIP on the full polymer system computationally demanding. Alternatively, MLIPs can be trained on much smaller oligomer clusters and only afterwards are applied to the polymer system.

It has already been demonstrated that potentials, which are trained on oligomers of a specific chain length, are transferable to oligomers of different chain lengths with respect to the prediction of correct potential energies and forces that act on individual atoms. [4]

However, due to their short chain length, oligomers are typically not entangled, which is an important property of polymer melts. The entanglements lead to altered and reduced dynamics of individual segments within a polymer chain due to heavily restricted possible movement. Moreover, the dynamics are governed by slow relaxation processes and require generally long simulation times to be fully resolved.

A key property to characterize dynamics is the mean squared displacement (MSD), for which a more complex behavior in the subdiffusive regime is observed in entangled polymers compared to non-entangled oligomers and polymers. In particular, there is an additional regime where the MSD is proportional to $t^{0.25}$. [5]

In this study we investigate whether MLIPs that are trained on small non-entangled oligomers can reproduce the dynamics of entangled polymer systems.

Therefore, we apply an active learning-based workflow that utilizes the Atomic Cluster Expansion (ACE) framework [6,7,8,9], which provides a complete local descriptor of the atomic environment in conjunction with linear regression, as well as the LAMMPS MD software [10] to fit ACE potentials for small oligomers from scratch.

Instead of computationally costly QM reference calculations, we employ cheaper MD reference calculations with classical force fields, enabling nanosecond-long and cost-efficient trajectories for small oligomer and larger polymer systems.

The fitted ACE potentials are then applied first to longer non-entangled oligomers and subsequently to entangled polymers. We characterize the performance of the potentials by checking first if they reproduce correct densities and structural properties, which is a premise for correct dynamics. In a second step we investigate the dynamics in more detail by evaluating the MSD.

As an outlook and prove-of-principle the insights gained from this analysis can be used to identify an optimal procedure for collecting training data from small alkane clusters, which balances accuracy, transferability and training costs, to fit an accurate ACE potential for polyethylene based on DFT-reference data.

References

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Figures

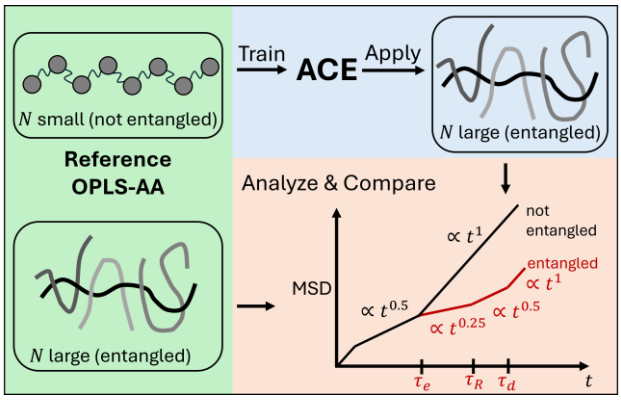

Figure 1. Schematic sketch of the training and simulation procedure and effect of entanglements on the MSD.