Accelerating crystal structure prediction and liquid electrolytes simulations using actively trained moment tensor potentials

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1. Introduction

Methods that rely on density functional theory (DFT) have established themselves as work tools in modern computational material science. Nevertheless, these methods often prove to be too costly for performing calculations related to the crystal structure search or calculations of the temperature dependencies of the physical properties (for example, thermal conductivity or viscosity) of liquid electrolytes. Machine-learned interatomic potentials (MLIPs) offer a more cost-effective alternative that can achieve accuracy comparable to that obtained with DFT simulations.

2. Accelerating simulations using moment tensor potential

In this work, we show how machine-learned Moment Tensor Potentials (MTPs) [1, 2] can be used as accurate interatomic potential models for applications such as crystal structure prediction and liquid electrolyte simulation. A key feature of our approach [3, 4] is the integration of an active learning strategy (based on the maxvol algorithm [5]) during the construction of the training datasets, which allows us for the development of a reliable and precise MLIPs, while keeping the training datasets small.

2.1 Crystal structure prediction

A typical approach to predict the most energetically favorable crystal structures is based on the iterative procedure of generating candidate structures followed by their local optimization and ranking [6]. We will present our approach, which accelerates this procedure, by generating a reliable MTPs, for efficient materials search. We applied it to both inorganic solids [7] and molecular crystals [3]. As an example, Fig. 1 shows a convex hull of the Zn-Cu-Pd system, where each read circle represents a novel structure hitherto unknown to the Materials Project database. Fig. 2 shows good agreement between DFT-based search of the polymorphs of glycine and MTP-based search. All the aforementioned developments are implemented in our code Sputnik (structure prediction using theoretical krystallography), which is already publicly available at [8] and will be soon released [7].

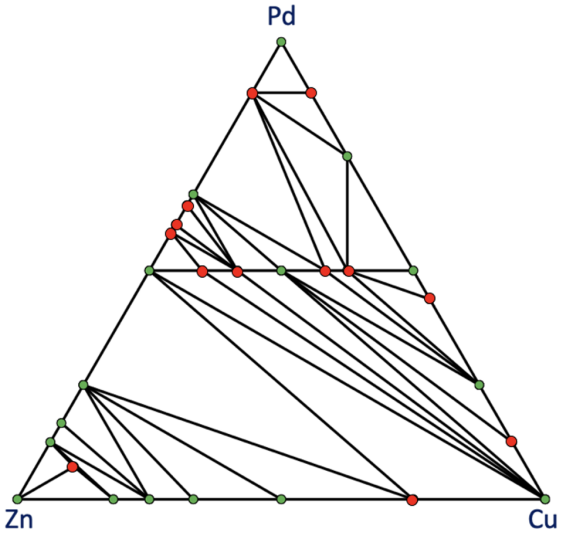

Fig. 1: Convex hull of the Zn-Cu-Pd system. Red circles represent novel compounds, not presented in the Materials Project Database.

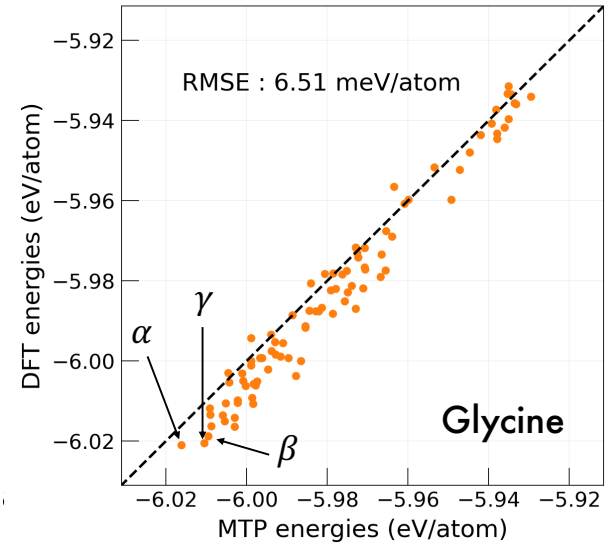

Fig. 2: Comparison between DFT-calculated and MTP-calculated energies of the most energetically favorable (α, β, γ) glycine polymorphs.

2.2 *Liquid electrolytes simulations*

Another application of the actively-trained MTPs is related to simulations of temperature dependence of physical characteristics such as density, viscosity, thermal and electrical conductivities of liquid electrolytes. MLIPs allow one to perform large-scale molecular dynamic simulations and compute the aforementioned properties with substantial accuracy, as we have recently demonstrated by calculating properties of liquid LiF-NaF-KF melt [4]. However, one of the limitations of many modern MLIPs is associated with the lack of long-range interactions description, which might become a problem during the simulations of molecular electrolytes, which are widely applied in industry. We will show several methodological advancements of the explicit inclusion of long-range interactions in MTP construction [9], which allowed us to accurately compute the viscosities of several molecular electrolytes as demonstrated in Fig. 3.

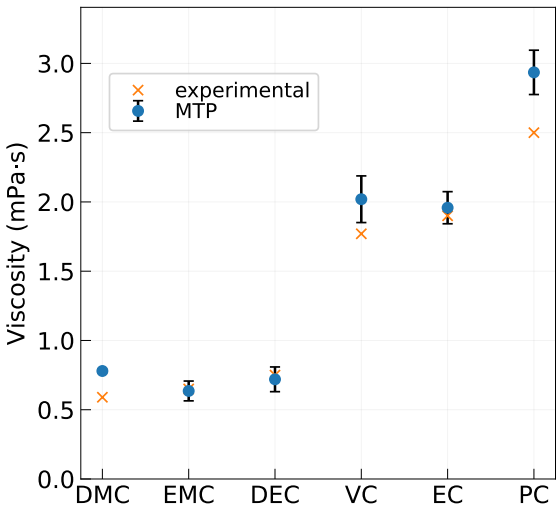

Fig. 3: Viscosity calculated using Green-Kubo method during molecular dynamics simulations with machine-learned MTP. Carbonates are labeled as: DMC – dimethyl , EMC – diethyl methyl, DEC – diethyl, VC – vinyline, EC – ethylene, PC – propylene.

2.3 *Related works*

Several works recently showed the application of machine learning methods to the inorganic crystal structure prediction problem [10, 11] – our work might be considered as complementary to the aforementioned works. The use of MLIPs for liquid electrolyte simulations is a recent trend; see, for example, work [12].

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