Developing Machine-Learning Force Fields

For Amorphous SiO_2 in Lithium-ion Batteries

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Introduction

Lithium-ion batteries (LIBs) power everything from smartphones to electric vehicles, but limited battery life remains a major challenge. **Amorphous Silicon Oxide** (SiO_2) is a highly promising anode for LIBs because its capacity (~1965 mAh/g) far exceeds the traditional graphite (~372 mAh/g). It is also abundant, cost-effective, and eco-friendly [1]. However, amorphous SiO_2 suffers a massive ~200% volume expansion during charge up (Fig. 1 & 2), causing breakdown of the structure. To fix this, we must first understand how the structure changes at the atomistic scale. Because physical experiments cannot capture such atomistic structural changes, we must rely on computer simulations (Fig. 3).

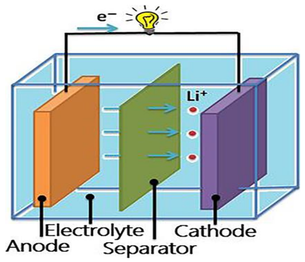


Figure 1. Representation of charging and discharging of lithium in a battery.



Figure 2. Atomic model representation of the effects of volume expansion [3].

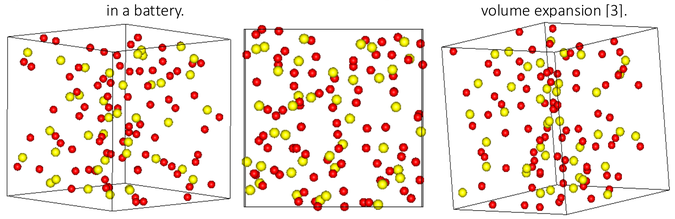


Figure 3. Accurate computational models of amorphous SiO_2

To study the structural changes of amorphous SiO_2, we use **Ab Initio Molecular Dynamics (AIMD)**. AIMD combines quantum mechanical electronic structure calculations with molecular dynamics simulations to calculate energies and forces on the fly to predict the structure, dynamics, and properties of the material. While highly accurate, such calculations takes a long time.

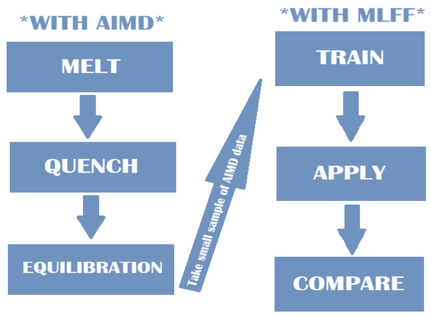
Machine Learning Force Fields (MLFF) can resolve this limitation. Instead of calculating heavy physics equations at every step, a MLFF is trained using data from a small sample of the AIMD simulations. Once trained, the MLFF can accurately predict energies and forces, enabling the rapid determination of material structures and properties at a fraction of the time of AIMD calculations.

Methodology

Using **MedeA** and **Vienna Ab initio Simulation Package (VASP)**, we performed AIMD simulations to execute a melt-quench-equilibration procedure to generate amorphous SiO_2, capturing both **high-energy states (HES)** and **low-energy state (LES).**

We ran **MLFF training simulations** on these structures, varying in size to provide a structurally diverse training dataset. Once we verified the energy and force errors were sufficiently small, we **applied** the MLFF to study the structure of amorphous SiO_2.

To evaluate the applied MLFF model, we compared its energy, force, and stress predictions against the AIMD reference data. We also analyzed **pair distribution functions (PDF)** and **angle distributions functions (ADF)** to verify that the simulated atomic structures are consistent with experimental data.



Scheme 1. Simplified representation of the procedures

Results and Discussions

MLFF Training errors

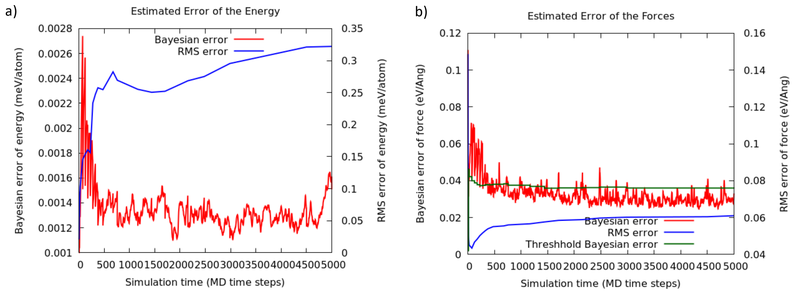


Figure 4. Errors between MLFF model and AIMD in a) energy and b) force

MLFF vs. AIMD Applied to HES SiO_2

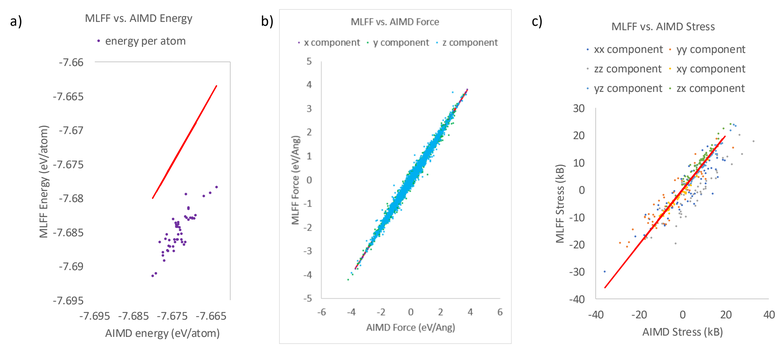


Figure 5. Difference between MLFF vs. AIMD of HES SiO_2 structure in a) energy, b) force, c) stress

MLFF vs. AIMD Applied to LES SiO_2

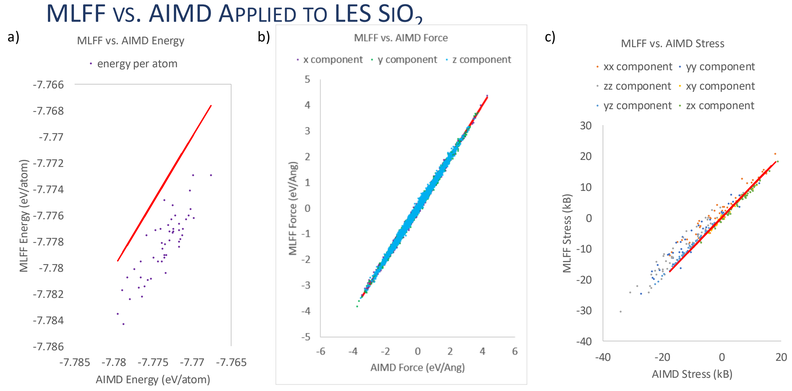


Figure 6. Difference between MLFF vs. AIMD of LES SiO_2 structure in a) energy, b) force, c) stress

PDF and ADF Predicted by MLFF

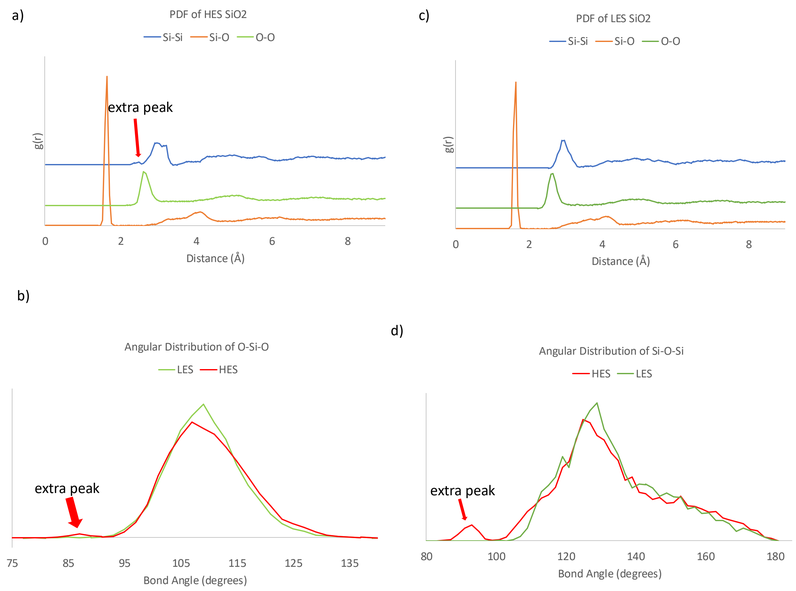


Figure 7. PDF for Si-Si, Si-O, and O-O bonds of a) HES and (c) LES, and ADF for O-Si-O and Si-O-Si in b) HES and d) LES.

Structural validation: MLFF vs. Experimental data

Structure Data	MLFF	Experimental	Percent Error (%)
Si-O Bond Length (Å)	1.65	1.61	2.48%
Si-Si Bond Length (Å)	2.97	3.08	3.57%
O-O Bond Length (Å)	2.67	2.63	1.52%
avg Si-O-Si Bond Angle (degrees)	146.8	140 - 150	1.24%
avg O-Si-O Bond Angle (degrees)	109.3	109.4 - 109.7	0.23%

Table 1. Comparison of MLFF and Experimental Data [2]

Conclusions

- MLFF completed the simulations in 15 minutes, while AIMD took 57 hours
- MLFF maintained minimum error across energy, force, and stress when applied to HES or LES
- MLFF structural results closely matched experimental data
- MLFF has proved its reliability to predict the structures of amorphous SiO_2
- Moving forward, we will incorporate lithium into the model to address the ~200% volume expansion problem

References and Acknowledgement

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