Transferable top-down CGMD parameterization for linear polymers

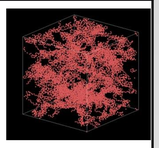
Graduate Student Fellows: Chanwook Park[1], Heather White[1], Ruijie Zhu[2]
Faculty Advisors: Zhengtao Gan[1], Sinan Keten[1]
(1) Northwestern University Department of Mechanical Engineering, 2145 Sheridan Rd, Evanston, IL 60208
(2) Northwestern University Department of Materials Science and Engineering, 2220 Campus Drive, Evanston, IL 60208

Introduction

All-atomistic molecular dynamics (MD) simulations based on the ab-initio force field have been regarded as accurate but computationally expensive simulation methods. To reduce this cost, reduced-order modeling techniques have been developed. One such method is coarse-graining (CG) where several atom are combined into a single pseudo-atom. One of the largest challenges in creating CGMD models is that most force fields are non-transferable [1] and parameterizing a new CGMD forcefield usually takes months. Thus, there exists a compromise between and time cost for developing a such a force field [1].

The CGMD research community needs a computationally cheap and labor-saving method for force field parameterization that accurately predicts user-defined target properties among a wide range of chemical species.

Materials and Methods



Linear polymer chain initial coordinate generation

In order to run an MD simulation, and initial structure is needed. To accomplish this, a semi-self-avoiding random walk was written in Python where, given the density and bond length, a corresponding melt of linear polymer chains is created.

Automated equilibration

Every MD system takes a different amount of time to equilibrate based on the geometry and force fields of each individual instance. To factor this into the automated process, a LAMMPS [2] protocol inspired by Auhl et. al. [3] was created that evaluated the mean square internal distances (MSID) of the polymer chains repeatedly during equilibration. If the root mean squared deviation of the average MSID value between subsequent evaluations is less than a user-specified tolerance, the equilibration terminates.

Database

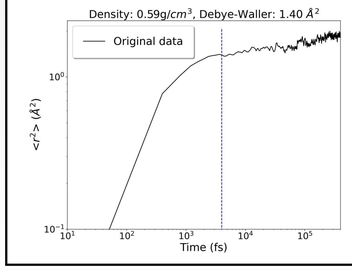
The polymer database consists entries with three sets of descriptions: polymer specific parameters, force field parameters and target properties. The first two parameters are updated iteratively based on the recommendations of gaussian process model, and the target properties are calculated from the simulation results of CGMD runs.

Neural Network

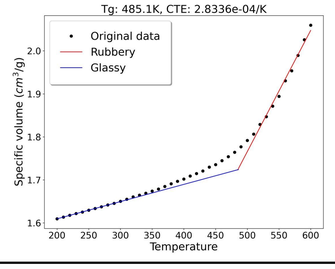
The neural net, which is trained on the user-specified entries in the database, functions as a surrogate model that replaces CGMD runs. It enables accurate prediction of target properties of interest.

Results

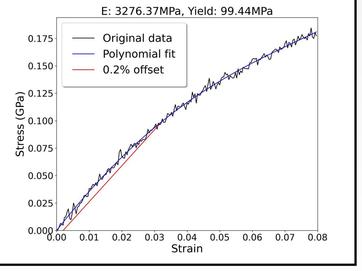
- **Dyamics test data**



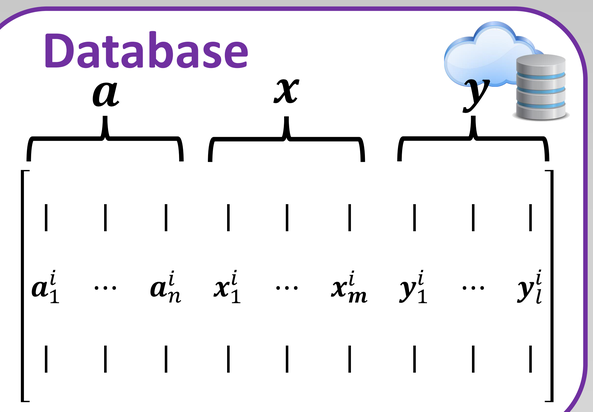
- **Thermal test data**



- **Mechanical test data**



Database



$\boldsymbol{a}$: polymer specific parameters (monomer molecular weight, chain length, polydispersity, etc.)
$\boldsymbol{x}$: force field parameters
$\boldsymbol{x}^*$: optimized force field parameters
$\boldsymbol{y}$: general target properties
$\bar{\boldsymbol{y}}$: target properties ($\bar{\boldsymbol{y}} \subset \boldsymbol{y}$)
$\bar{\boldsymbol{y}}_0$: all atomistic (or exp.) target properties
$\bar{\boldsymbol{a}}_0$: polymer specific parameters of target material

Target material ($\bar{\boldsymbol{a}}_0$) : Polyethylene (PE), MW: 20,000 g/mol

Target properties ($\bar{\boldsymbol{y}}$): density, diffusivity, elastic modulus

Training

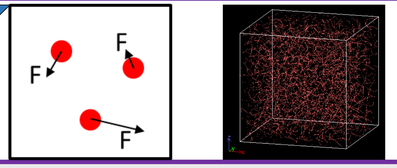
Training data $\begin{bmatrix} | & | & | \\ \boldsymbol{a}^i & \boldsymbol{x}^i & \bar{\boldsymbol{y}}^i \\ | & | & | \end{bmatrix}$

$\bar{\boldsymbol{y}} = CGMD(\boldsymbol{a}, \boldsymbol{x}) \rightarrow \bar{\boldsymbol{y}} = NN(\boldsymbol{a}, \boldsymbol{x})$

Optimization (parameterization)

$$\boldsymbol{x}^* = \arg\min_{\boldsymbol{x} \in S} \|NN(\bar{\boldsymbol{a}}_0, \boldsymbol{x}) - \bar{\boldsymbol{y}}_0\|^2$$

Simulation



Conclusions

We proposed a general-purpose data generation and modeling framework for linear polymers in CGMD simulations. The iteratively updated database serves as input to the neural network model, which takes polymer specific parameters and force field parameters as input and makes direct prediction of target properties, thus allowing for efficient property prediction. We foresee that this framework will speed up the search for linear polymers with desired properties.

Acknowledgements

We thank the Predictive Science and Engineering Design Graduate Program for providing funding for this project. Additionally, we express sincere gratitude to Profs. Zhengtao Gan and Sinan Keten for their guidance, as well as to Profs. Wing Liu and Wei Chen for their support. Finally, we thank Adetoye Adekoya for organizing our regular meetings.

References

[1] Yang et al., The Journal of Physical Chemistry B, 2014.
[2] Plimpton et a., Journal of computational physics, 1995.
[3] Auhl et a., The Journal of chemical physics, 2003

Northwestern | ENGINEERING