Machine Learning Interatomic Potentials for the Conversion of Polysiloxanes to SiCO Ceramics

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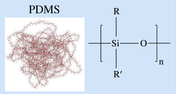
Introduction

Polysiloxanes

- Si-O backbone with organic substituents
- Silicones for sealants, lubricants, etc.
- Hydrophobic/oleophilic porous materials
- Material coatings

PDMS



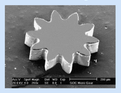
Polysiloxane aerogel

Silicon Oxycarbides

- Glass with Si-O/Si-C bonds and free carbon
- Thermal, mechanical, and chemical stability
- Creep resistant
- Biocompatible
- Interesting electrical properties

SiCO glass

SiCO micro gear



Motivation

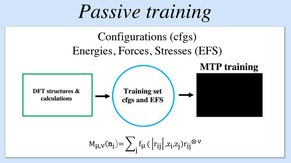
Motivation: Accurate quantum chemical methods can only be used for small models (<2000 atoms). However, amorphous models and reaction simulations require much larger representations (>>10,000 atoms).

Hypothesis: Novel Machine Learning Interatomic Potentials (MLIPs) with DFT-level accuracy can handle simulations with model sizes comprising millions of atoms. Thanks to previously shown success with Si-containing systems (Si, SiC, SiO_2), we suppose the same approach will work for more complex systems (SiCO/SiCOH).

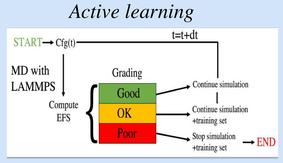
Method

Machine learning with moment tensor potentials (MTPs)

Passive training



Active learning



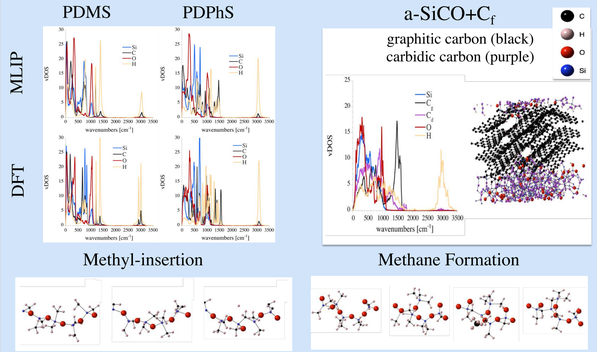
Software and code

- Machine learning with mlip-2
- aiMD simulations and DFT calculations with VASP
- MLIP simulations with LAMMPS and mlip-2
- Structure visualization with CrystalMaker
- Vibrational spectra computed with VASPKIT
- Weight-based gas removal script:

$$p_{gas}=p_{ref}\sqrt{\frac{M_{H_2}}{M_{gas}}}$$

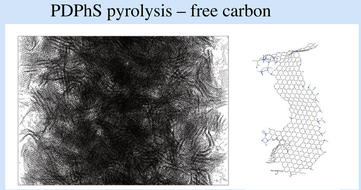
MLIP Preliminary Results

- Low temperature vibrations (vDOS)
- Localized reactions: methane and Si-CH_2-Si formation
- High temperature reactions and pyrolysis simulations



PDPhS pyrolysis – free carbon

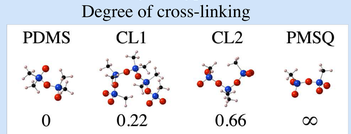
~ 1 million atoms
10 ns pyrolysis
2500K, 2kbar



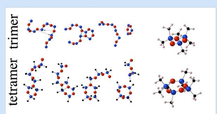
Cross-Linking Study

- Degree of cross-linking: T/D
- Degradation into cyclic oligomers depending on T/D
- Higher T/D -> better thermal stability/greater mass retention
- Using PDMS as a sacrificial template

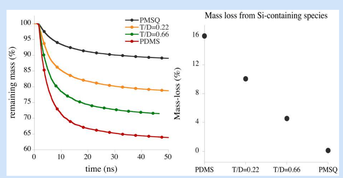
Polymer types
Degree of cross-linking



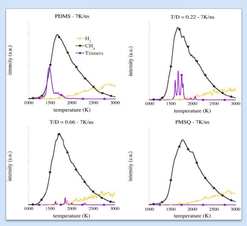
Cyclic oligomer formation



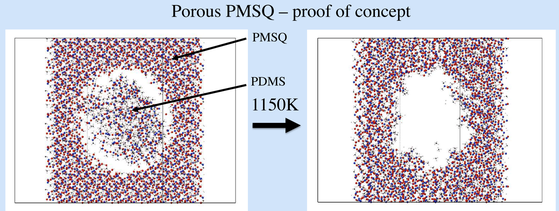
Mass loss/retention



Gas detection



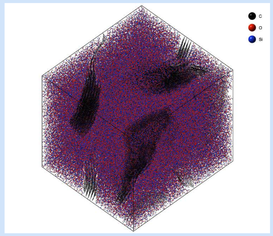
Porous PMSQ – proof of concept



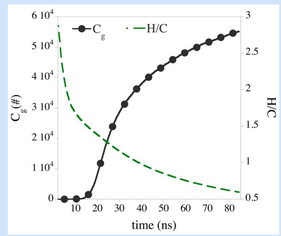
Genesis of Free Carbon

- Free carbon formation in PDMS
- Multi-process growth mechanism
- Impact of model size and temperature

PDMS after 85ns @ 2500K
(Initially 1 Million atoms)



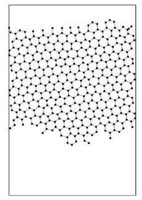
C_g = graphitic carbon
Starting at H/C ~ 1.5



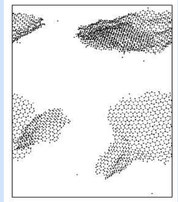
50ns @ 2500K

10^4 atoms | 10^5 atoms | 10^6 atoms

ONE



SOME



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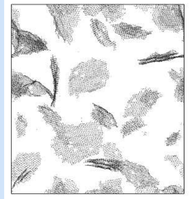
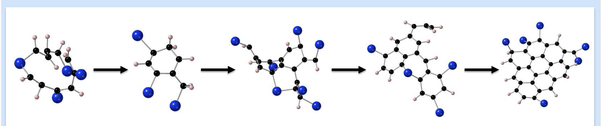
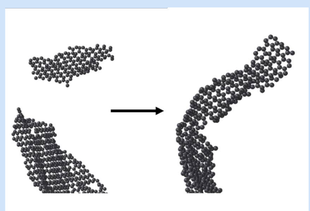
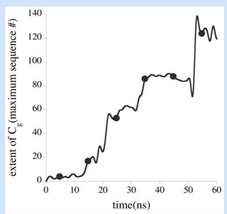


Step-by-step growth

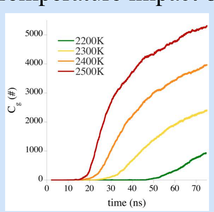


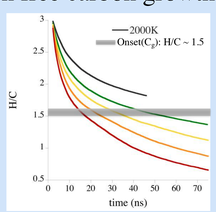
Percolation – quick extension of sheets





Temperature impact on free carbon growth

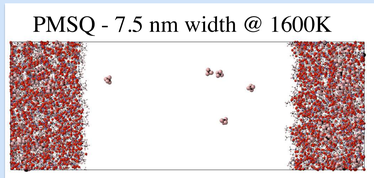




Future Work

- Room for improvement of the potential. Adding more structures for training
- Scale-up to larger models. Simulate systems with more than 10 million atoms
- Large-scale thermal conductivity studies with different amounts of free carbon.
- More in-depth study of gas formation. Analyze gasses diffusing through a surface

PMSQ - 7.5 nm width @ 1600K



Summary

- Developed a new MLIP using MTPs for simulating the pyrolysis of polysiloxanes
- The MLIP demonstrates DFT-level accuracy at a fraction of the computational cost
- Probed the impact of cross-linking on polysiloxane thermal stability
- Explored mechanisms of free carbon formation during polysiloxane pyrolysis

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