Machine Learning Interatomic Potentials for Pyrolysis of Polysiloxanes and Properties of SiCO Ceramics

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Abstract

We present Moment Tensor-based (MTP) Machine Learning Interatomic Potentials (MLIPs) fit to DFT energies, forces, and stresses. The purpose of this potential is to simulate the reactive conversion from polysiloxane precursors to SiCO ceramics. In this study, we give examples of low temperature vibrational calculations and both small and large-scale high temperature reactions using the MLIP.

Introduction

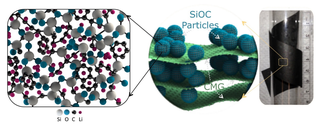
SiCO Polymer Derived Ceramics

- Polysiloxane precursors
- Pyrolyzed between 400-1000 °C
- Continued reactions > 1200 °C

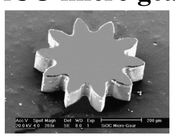
SiCO Ceramics

- Excellent thermal and chemical stability
- Interesting electrical properties
- Creep resistance
- Anode material for Li^+ and Na^+

Li^+ anode material



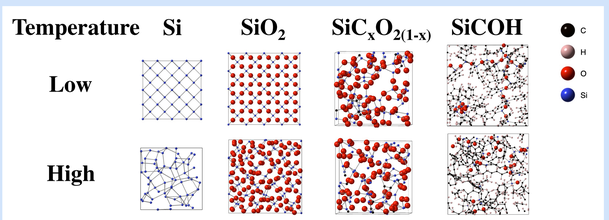
SiCO micro gear



Goal

Simulating systems Si/C/O/H

- Low temperature behavior
- High temperature behavior
- Reactive conversion

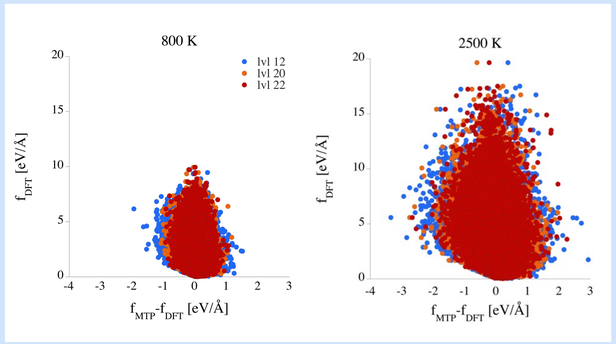


Method

- Machine learning with mlip-2
- DFT calculations with VASP
- Classical MD with LAMMPS
- vDOS calculations with VASPKIT

Validation

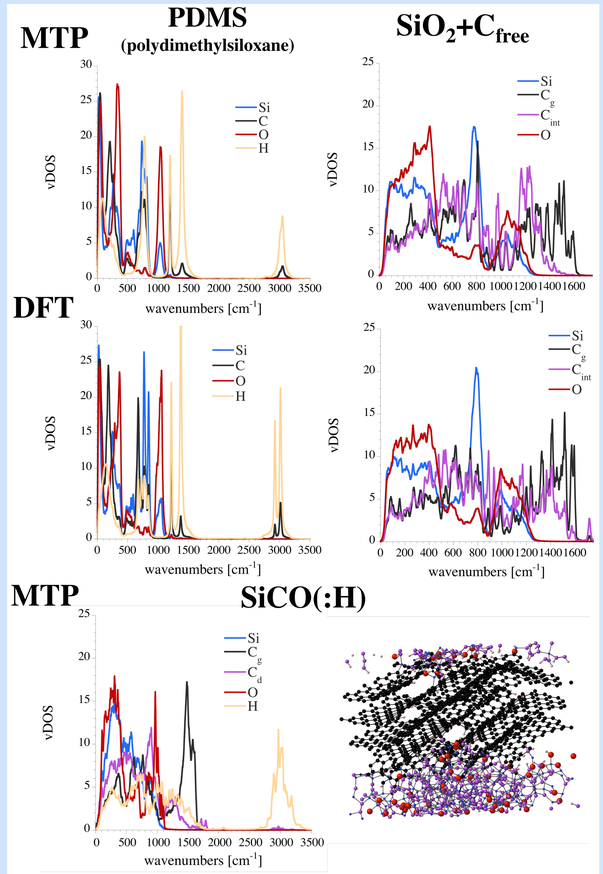
- Compare errors in forces (MTP-DFT)
- Impact of complexity and temperature



Vibrational Spectra

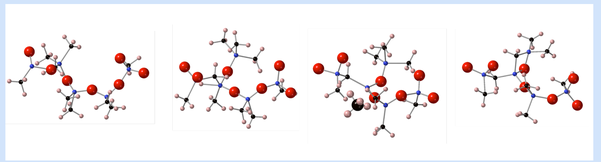
Vibrational Density of States (vDOS)

- Compare MTP to DFT
- Low temperature, 300 K
- Experimental analog – Spectroscopy

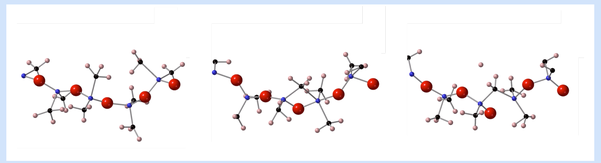


Localized Reactions

Methane Production



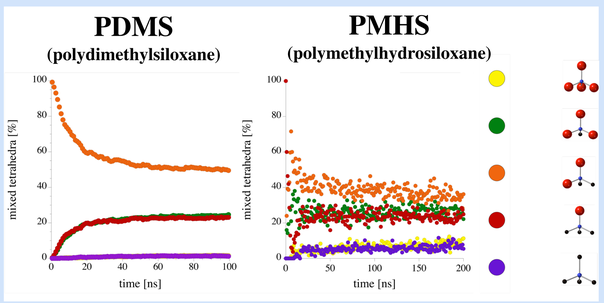
Si-CH_2-Si Formation



Large-Scale Simulations

Mixed Tetrahedra

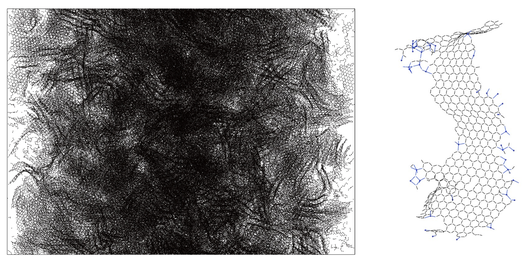
- Reactive simulation with gas removal
- Formation of mixed Si tetrahedra
- Approaches the ideal distribution
- Comparable to Si^{29}-NMR Experiments



Free carbon phase

- Simulation of ~ 1 million atoms
- High carbon content (PDPhS)
- Formation of free carbon phase
- 10 ns in ~ 1.5 weeks of wall-clock-time

PDPhS – simulated @ 2500 K
(polydiphenylsiloxane)



Great

Accurate

DFT-like behavior at low and high temperature

Fast

Capable of long simulation times with large systems

Useful

Vibrational calculations and reaction simulations

Room for Improvement

Imperfections

- Low reactivity of H_2O and CH_4
- Si and C defects
- Uneven force error distribution

Solutions

- More configurations
- Reactive intermediates
- SiCO glass and SiCO + C_{free}

Summary

Developed MLIP for Si/C/O/H

- Validated MLIPs of differing complexity
- Compared vDOS results of MLIP vs. DFT
- Simulated reactive conversion of polysiloxanes to SiCO ceramics

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