A Machine Learning-Derived Atomistic Potential for $Y_2Si_2O_7$

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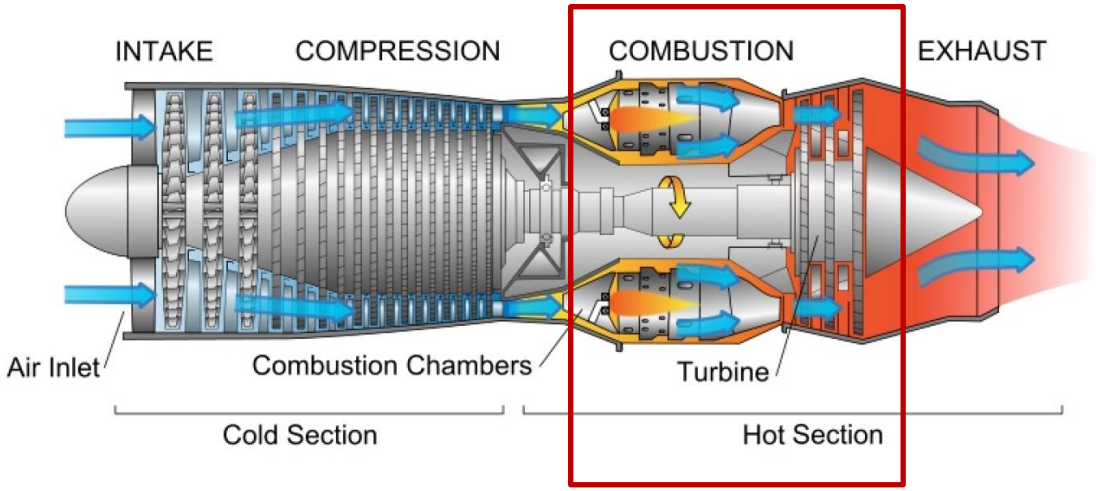
Outline

- Background
- Simulation Methods – Machine Learning-based Interatomic Potential for Molecular Dynamics (MLMD)
- Crystal Cell Optimization Calculations
- Thermochemical Property Calculations
- Thermomechanical Property Calculations – CTE
- Conclusions

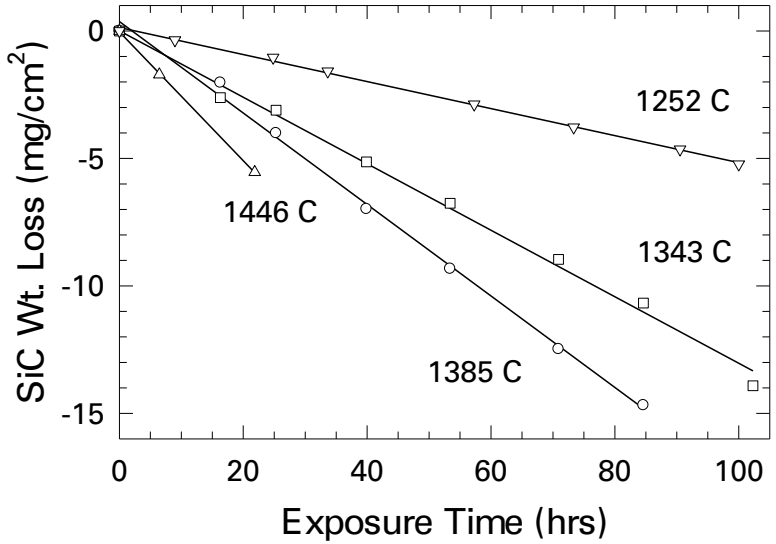
SiC/SiC Ceramic Matrix Composite (CMC) Components

- Replace metal alloy engine components with SiC/SiC ceramic matrix composites (CMCs)
- Increased efficiency and cost savings
  - Higher temperature stability
  - Lower density
- CMCs can degrade under O_2 and H_2O environments at high temperature (>800°C)



Target: 1482ºC

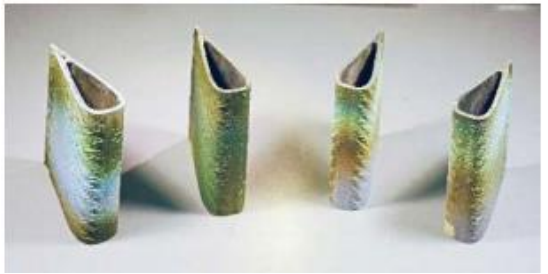
Weight loss of SiC in High-Pressure Jet Fuel Burner (6 atm, 20 m/s)



Robinson and Smialek, J Am Ceram Soc 82, 1817 (1999)
Zhu, et al. ICACC 2018
Jet engine diagram licensed under CC BY-SA 4.0, attributed to Jeff Dahl. https://commons.wikimedia.org/wiki/File:Jet_engine_numbered.svg

Environmental Barrier Coatings (EBCs) for SiC/SiC CMCs

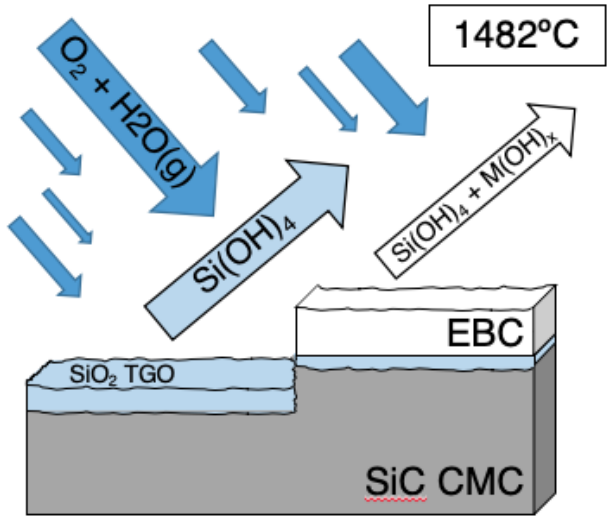
- CMCs can recess under O_2 and H_2O environments at high temperature (>800℃)
- Rare-Earth (RE) disilicates ($RE_2Si_2O_7$) are a promising class of EBCs
- EBCs can prevent H_2O diffusion
- Some formation of SiO_2 TGO still occurs with EBC



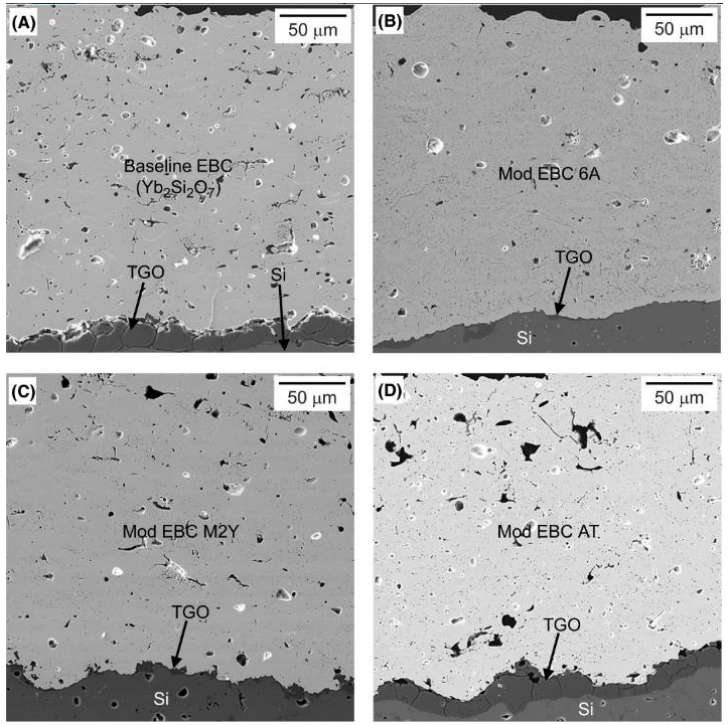
EBC-coated turbine vanes

$$SiC\ (s) + 3/2\ O_2\ (g) \longrightarrow SiO_2\ (s) + CO\ (g)$$

$$SiO_2\ (s) + 2\ H_2O\ (g) \longrightarrow Si(OH)_4\ (g)$$



EBC chemistry affects TGO growth and CMC recession

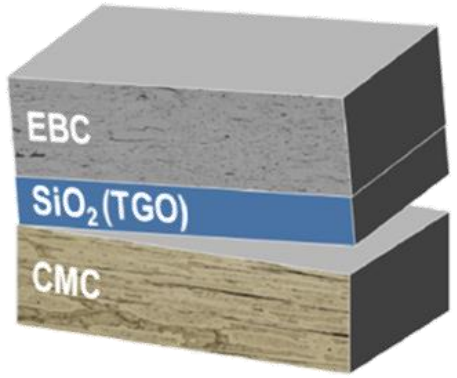


1000 h/1000 cycles at 1316℃ in 90% H_2O + 10% O_2

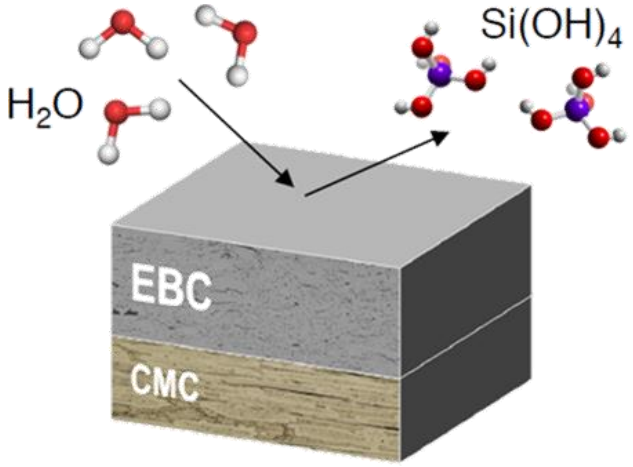
E. Opila et al. *J Am Ceram Soc* (**1999**)
K. Lee et al. *J. Am Ceram Soc*. (**2019**)

EBC Failure Modes

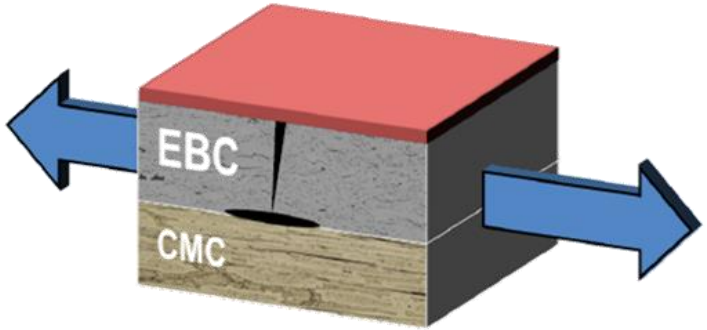
EBC lifetime and design requirements determined by combination of extrinsic failure modes



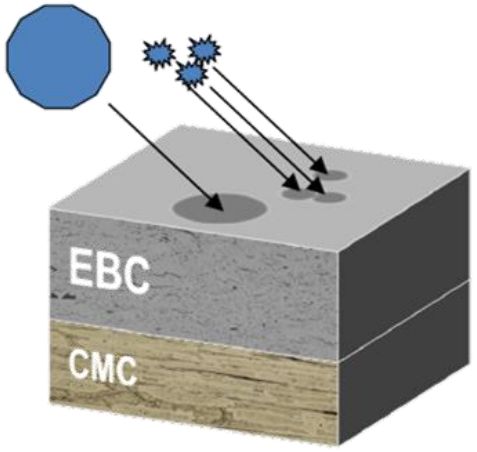
Steam Oxidation



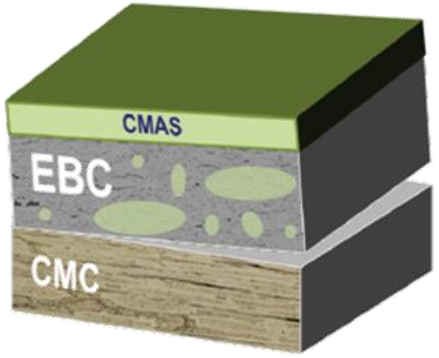
Hydroxide Formation/Recession



Thermomechanical Durability



Erosion and FOD

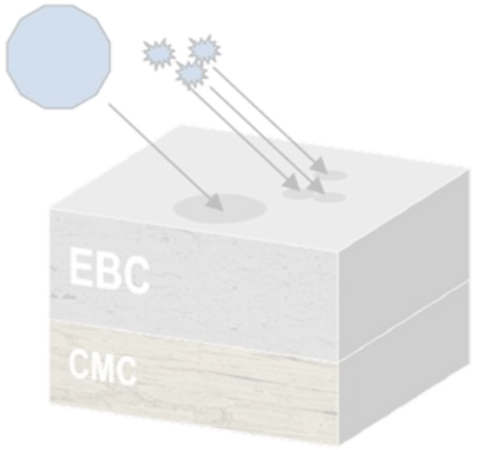


CMAS Attack & Infiltration

EBC Failure Modes

Steam Oxidation

Computational Simulations



Erosion and FOD

Thermochemical Properties

- Phase Stability
- Chemical Reactions

Thermomechanical Properties

- Mechanical Strength
- Thermal Expansion

Hydroxide Formation/Recession

CMAS Attack & Infiltration

Atomic-scale simulation methods for property calculations

DFT

- Electron-level theory
  - Cost scales with number of electrons
- Smaller simulation cells
  - Self-interaction errors?

Classical Molecular Dynamics

- Atom-level theory
  - Cost scales with number of atoms
- Bonding based on empirical trend fits
  - Generally, no bond breaking/formation

	DFT	Classical MD

Pros	• Higher accuracy
	• Generalizability
		• Time
		  • ~Hours
		• System Size
		  • Larger cells possible (~1,000s-10,000s of atoms)

Cons	• Time
	  • ~Days-Weeks
	• System Size
	  • Smaller unit cells (~100s of atoms)
		• Requires pre-parameterized potential
		  • Not as generalizable

Atomic-scale simulation methods

DFT

- Electro
  - Cost … electrons
- Smaller simulation cells
  - Self-interaction errors?

Classical Molecular Dynamics

- Atom-l
  - Cost … atoms
- Bonding based on empirical trend fits
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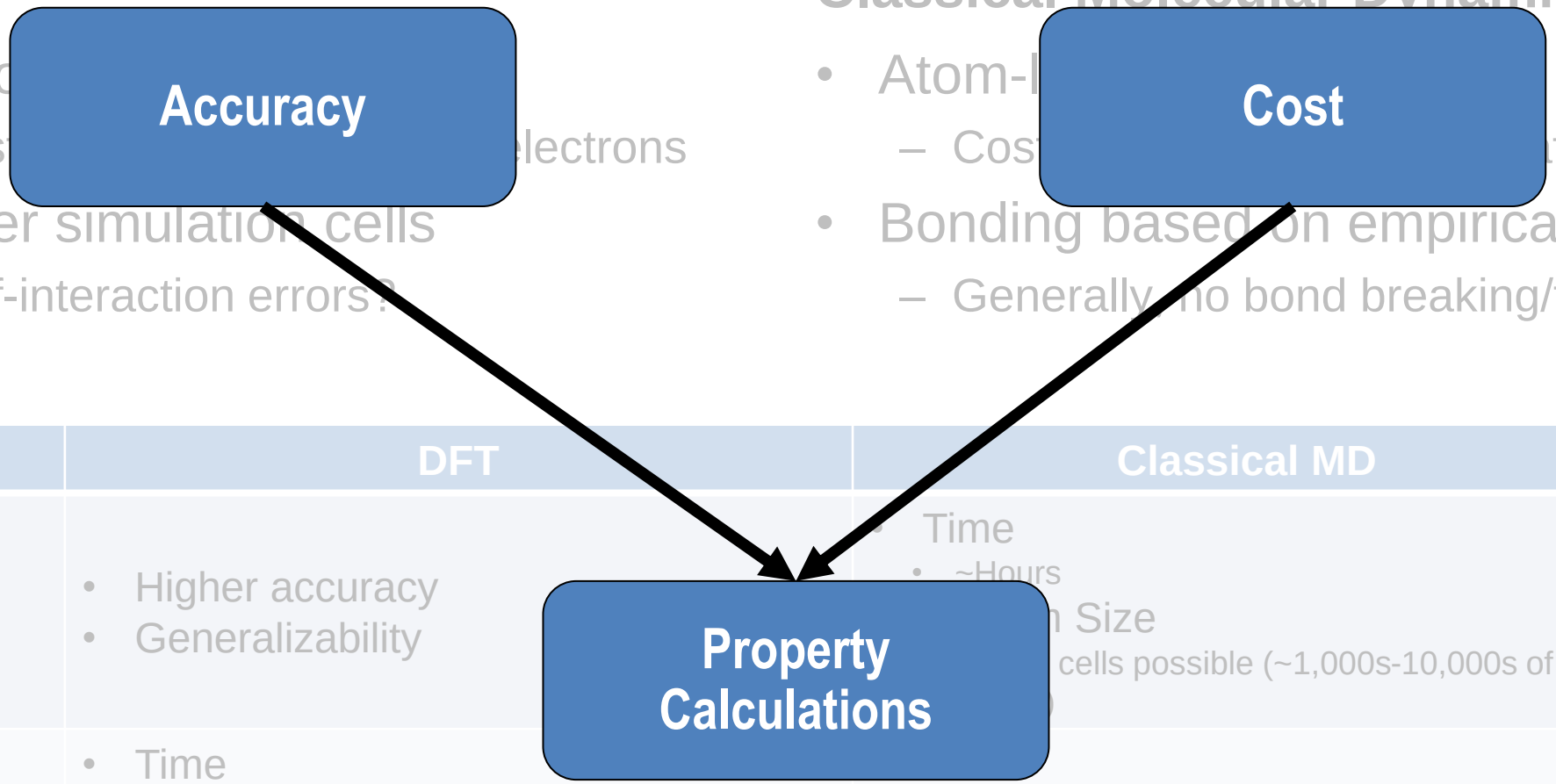


	DFT	Classical MD
Pros	• Higher accuracy • Generalizability	• Time • ~Hours • n Size cells possible (~1,000s-10,000s of
Cons	• Time • ~Days-Weeks • System Size • Smaller unit cells (~100s of atoms)	• Requires pre-parameterized potential • Not as generalizable

Machine learning approaches for crystalline materials

Direct Property Calculations

- Train ML model to directly predict property
- ML algorithm suitable for problem
  - Regression vs. Clustering
- Descriptors to capture system information
  - Crystal structure information
  - Atomic composition
  - Environmental variables
    - Temperature, Pressure, etc.
- Example: Neural network to predict CTE for rare-earth disilicates[1]

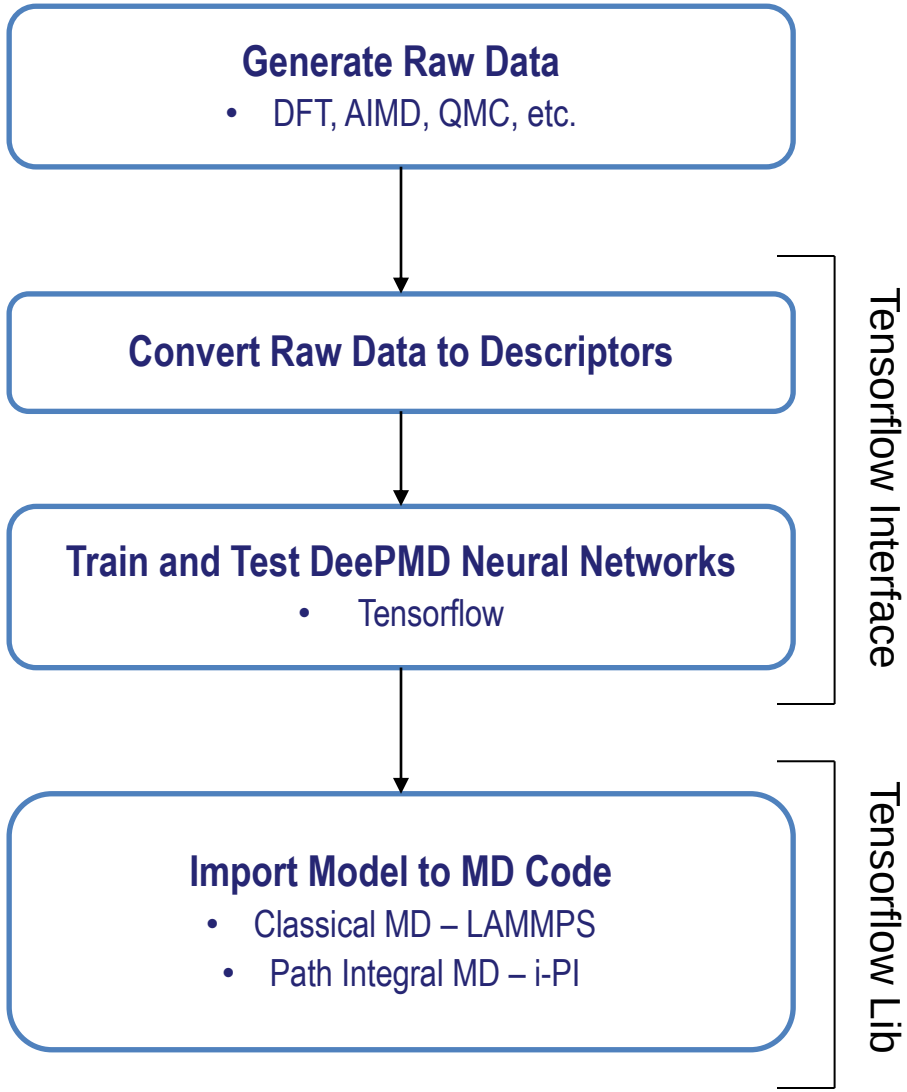
Indirect Property Calculations

- Train ML model to run simulations
  - Interatomic potentials
- Regression algorithms, typically
  - Almost always neural networks
- Descriptors often include atomic neighborhood information
  - Nearest-neighbor atoms within cutoff
  - Bonding information
- Example: NN-based interatomic potential for HfO_2

[1] Ayyasamy, et al. *J Am Ceram Soc.* **(2020)**
[2] Wu, et al. *Phys Rev B*. **(2021)**

DeePMD theory

- Deep Neural Network using descriptors to preserve translational, rotational, and permutational symmetries
- Descriptors dependent on atomic neighbor environment
  - Full information (radial and angular) included for first- and second-nearest neighbors
  - Radial information only for other atoms inside the user-defined cutoff radius
- Training data obtained from ab initio molecular dynamics (AIMD) simulations using DFT
- DNN used to calculate atomic energies; total energy is the sum of atomic energies.
- Forces and virial calculated using gradient of energy w.r.t. positions



Wang H, et al. *Comp Phys Comm*. (**2018**)

Molecular Dynamics: Minimization and Lattice Constants

Phase	Theory	a	b	c	α	β	γ
β C2/m	DFT (PBE)	6.91	9.06	4.78	90	101.97	90
	DFT (PBEsol)	6.85	8.97	4.74	90	101.79	90
	MLMD (PBE*)	6.90	9.05	4.77	90	101.95	90
	Expt[1]	6.88	8.97	4.72	90	101.70	90
γ $P2_1/c$	DFT (PBE)	4.75	10.90	5.63	90	96.18	90
	DFT (PBEsol)	4.71	10.81	5.57	90	95.98	90
	MLMD (PBE*)	4.74	10.88	5.62	90	96.14	90
	Expt[2]	4.69	10.84	5.58	90	96.03	90
	Expt[3]	4.69	10.86	5.59	90	96.01	90
	Expt[4]	4.66	10.78	5.54	90	96.06	90
δ $Pna2_1$	DFT (PBE)	13.80	5.09	8.20	90	90	90
	DFT (PBEsol)	13.62	5.03	8.12	90	90	90
	MLMD (PBE*)	13.77	5.07	8.19	90	90	90
	Expt[5]	13.81	5.02	8.30	90	90	90
	Expt[4]	13.69	5.02	8.17	90	90	90

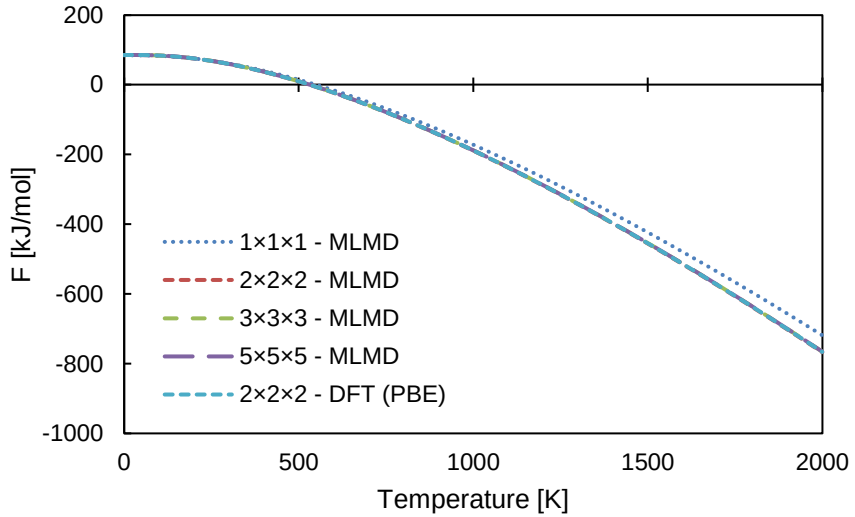
[1] Liddell, et al. *J Br Ceram Soc.* **(1986)**
[2] Dolan, et al. *Powder Diff.* **(2008)**
[3] Leonyuk, et al. *J Cryst Growth*. **(1999)**
[4] Smolin, et al. *Acta Crist B: Struct Cryst Cryst Chem.* **(1970)**
[5] Dias, et al. *Zeitschrift für Krist*. **(1990)**

Finite-Difference Phonon Calculations

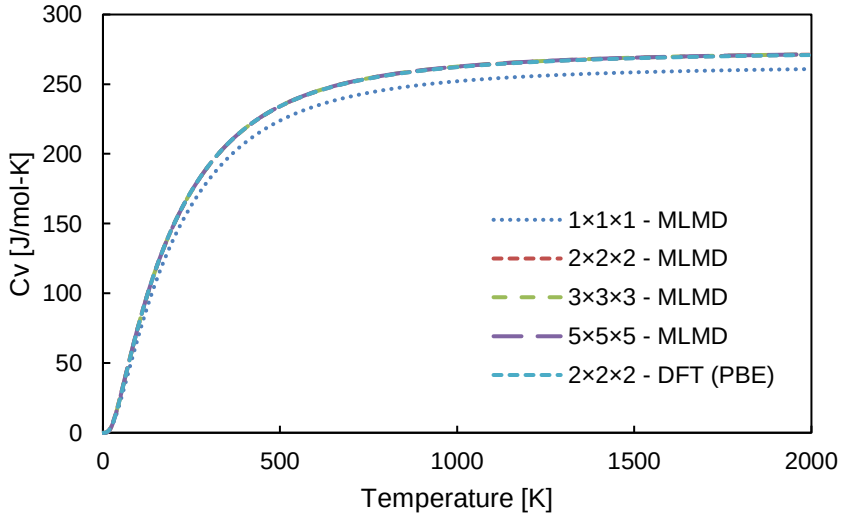
- Consistent results with DFT across supercell sizes
  - 1 × 1 × 1 (22 atoms)
  - 2 × 2 × 2 (176 atoms)
  - 3 × 3 × 3 (594 atoms)
  - 5 × 5 × 5 (2,750 atoms)

- Slight deviation between 1 × 1 × 1 cell in MLMD compared to DFT

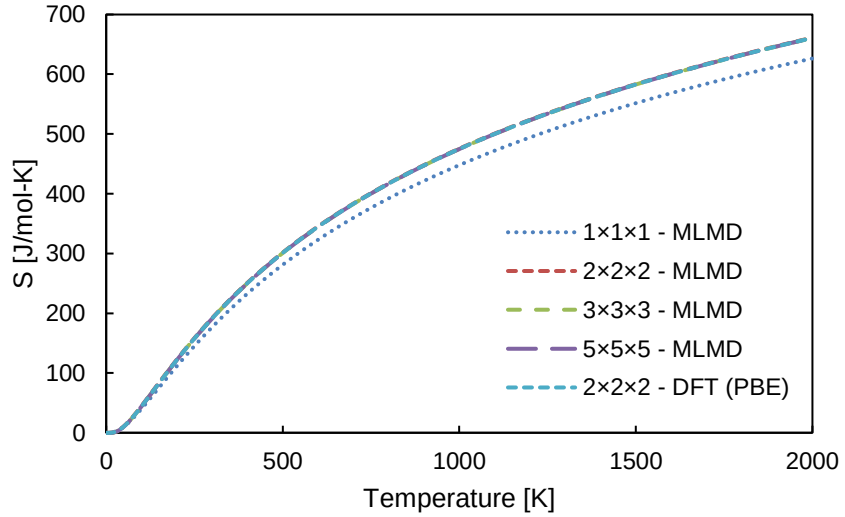
β-$Y_2Si_2O_7$ Free Energy



β-Y2Si2O7 Heat Capacity

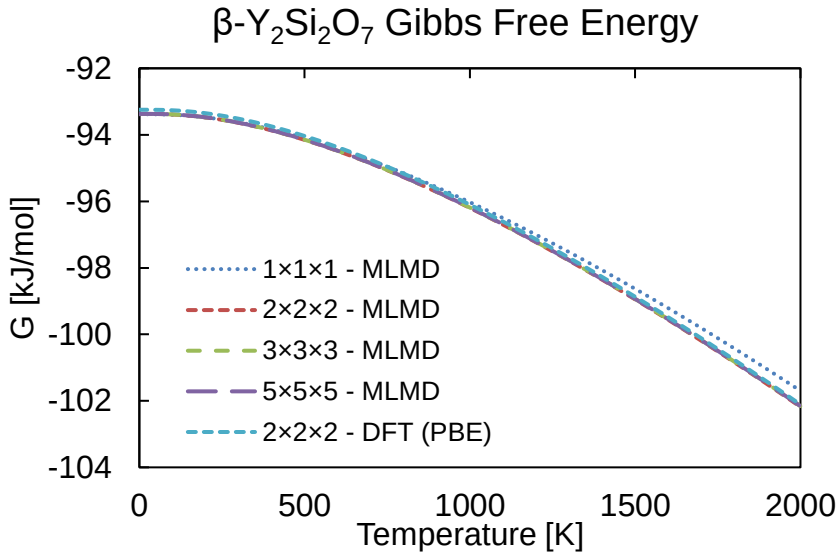


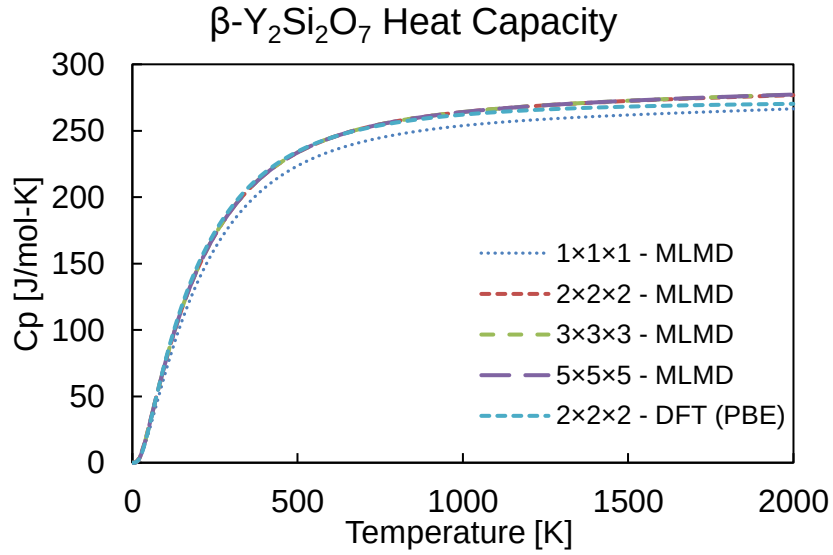
β-Y2Si2O7 Entropy



Quasi-Harmonic Approximation Phonon Calculations

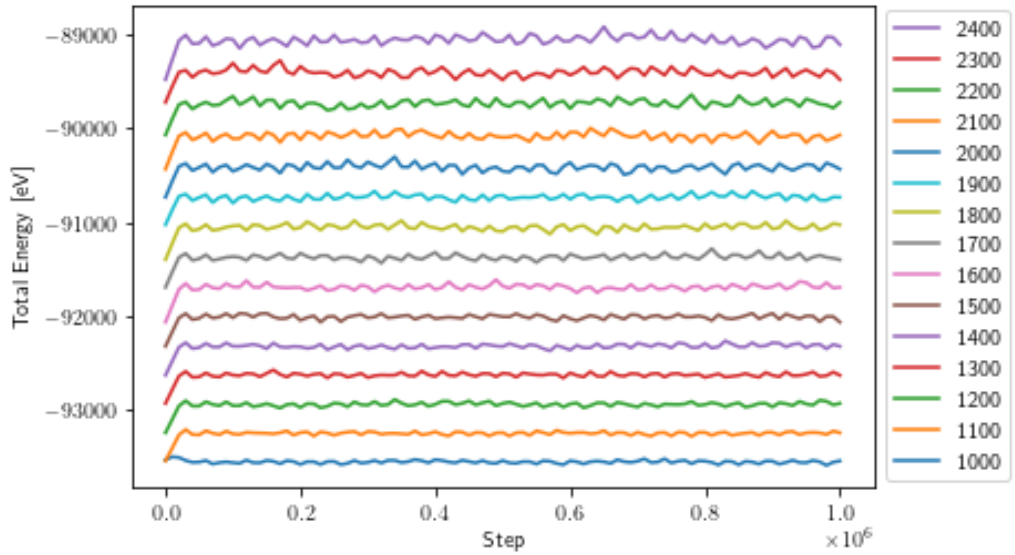
- Quasi-harmonic approximation considers non-equilibrium volume contributions
  - Span of volumes from 94% to 106% of equilibrium
- Slight deviation between $1 \times 1 \times 1$ cell in MLMD compared to DFT
- Can provide thermochemical properties required for phase stability and formation calculations
  - CALPHAD via Thermo-Calc software
- Larger supercell capabilities could enable studies of more complicated properties, simulation techniques, etc.

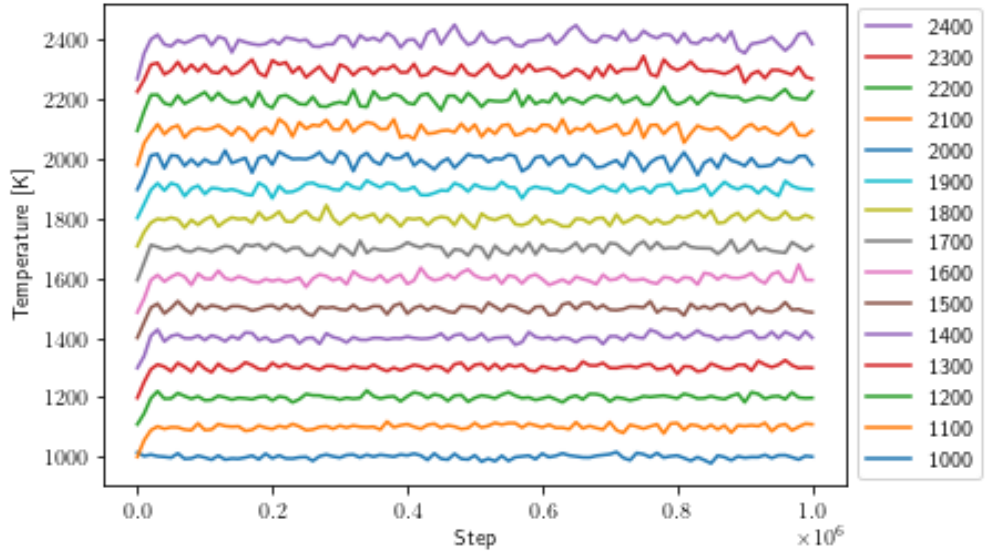




Long-duration and large-cell molecular dynamics

- Supercell size increased to 8 × 8 × 8 conventional unit cells
  - 11,264 atoms
- Total energy and temperature stable over a 2 μs simulation
- Cost-prohibitive cell size and timeframe for DFT





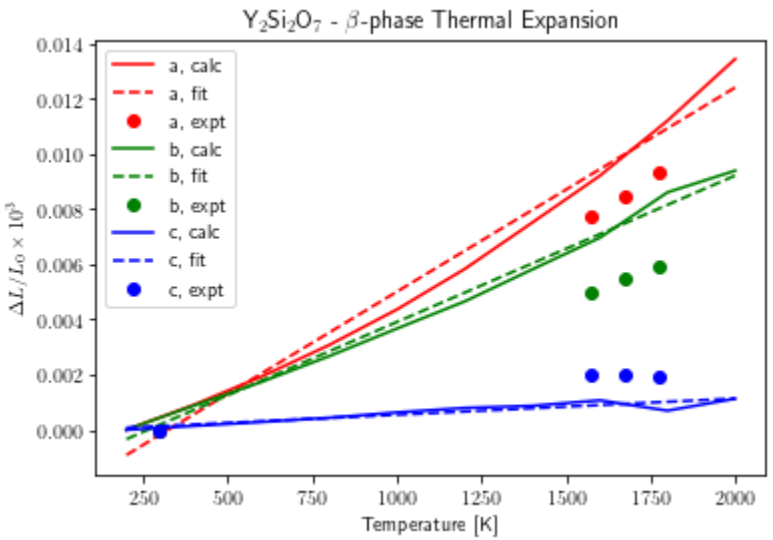
Timestep = 2 ps, Total Simulated Time = 2 μ

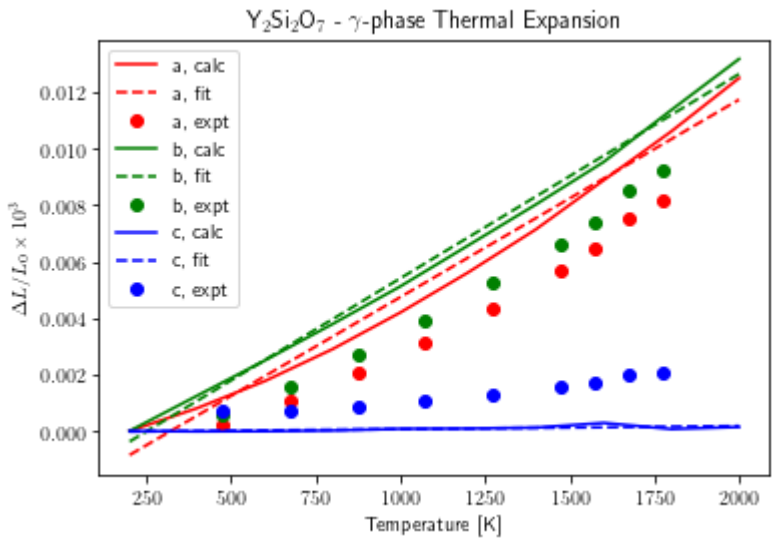
CTE calculations as function of temperature

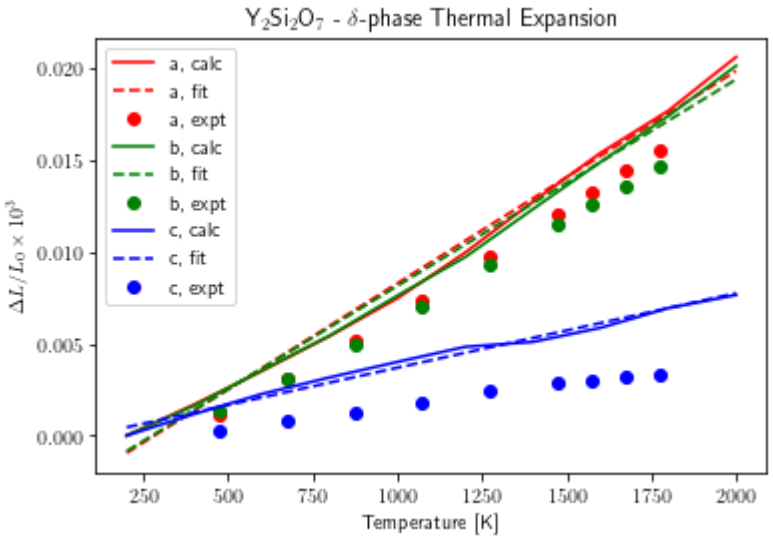
- Good agreement between MLMD simulations and experiment[1]
- Overprediction of *a* and *b* CTE, underprediction of *c* CTE

	CTE ($\times 10^6$ K^{-1})		
	β	γ	δ
a	7.4	0.1	4.1
b	5.3	7.2	11.5
c	0.6	7.0	11.2
Avg Bulk	**4.4**	**4.8**	**8.9**

Table 1: Linear lattice CTE and average bulk CTE of $Y_2Si_2O_7$ crystal phases







[1] Dolan, et al. *Powder Diff.* **(2008)**

Conclusions

- Machine learning was used to develop an interatomic potential for YDS
  - Potential was successfully used to calculate various properties of YDS including CTE
  - Results from MD match well with results from DFT

- MLMD enables calculation of phonon vibrational frequencies at similar accuracy but at substantial faster timeframes as compared to DFT (~hours vs. ~weeks)

- MLMD enables long-duration and/or large cell calculations that would be cost prohibitive with DFT
  - Simulations on the order of 10-100 ps were successfully completed
  - Simulations including $8 \times 8 \times 8$ conventional unit cells (11,264 atoms) of β-YDS were successfully completed

- Training of MLMD potentials is resource intensive, and training of potentials for additional materials is required. However, training is a one-time expense (per material), and potentials can be flexibly used in additional calculations.

Acknowledgements

- Environmental Effects & Coatings Branch at NASA Glenn Research Center
  - Dr. Bryan Harder – Wednesday, 8:00 AM in 335 David L. Lawrence Convention Center
    - Oxidation and Erosion Implications of CMAS on Environmental Barrier Coatings

- NASA Pleiades Supercomputer Cluster

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  - NASA Transformative Aeronautics Concepts Program (TACP)
  - NASA Aeronautics Research Mission Directorate (ARMD)