Machine learning approaches for rare-earth silicate environmental barrier coating thermodynamic property predictions

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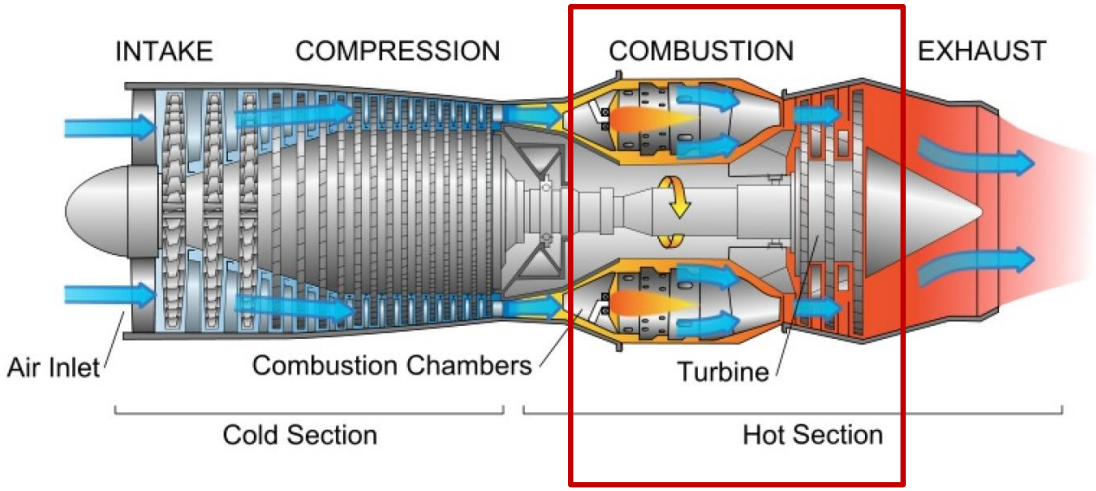
SiC/SiC Ceramic Matrix Composite (CMC) Components

- 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)

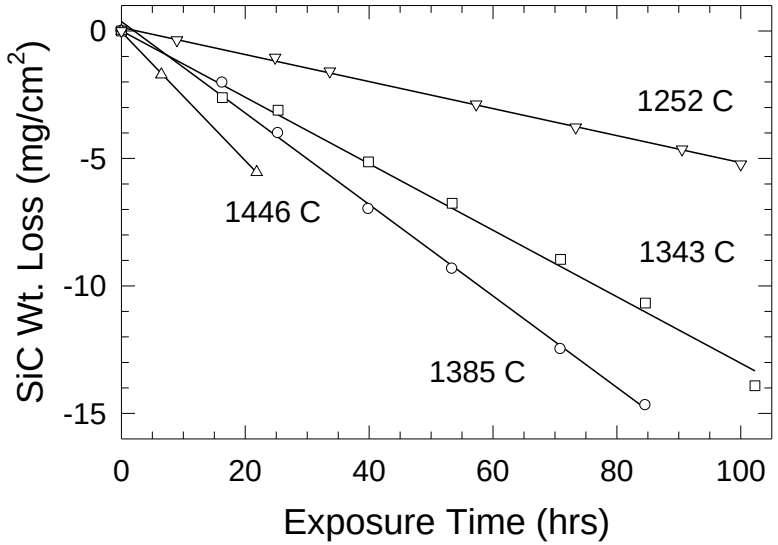


Target: 1482ºC

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

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

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

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 reduce H_2O diffusion
- Some formation of SiO_2 TGO still occurs with EBC

EBC-coated turbine vanes

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

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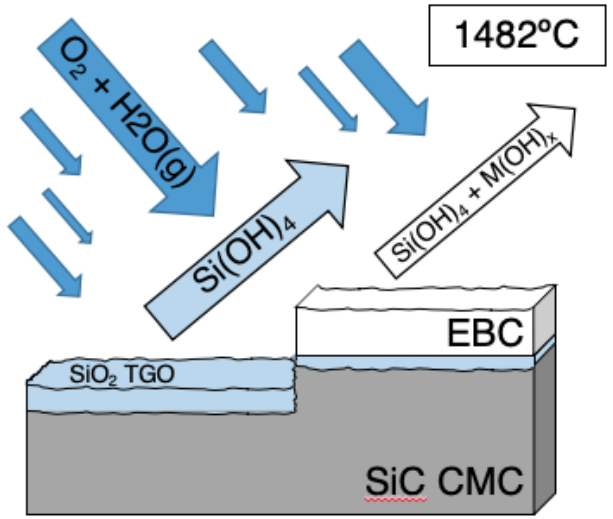
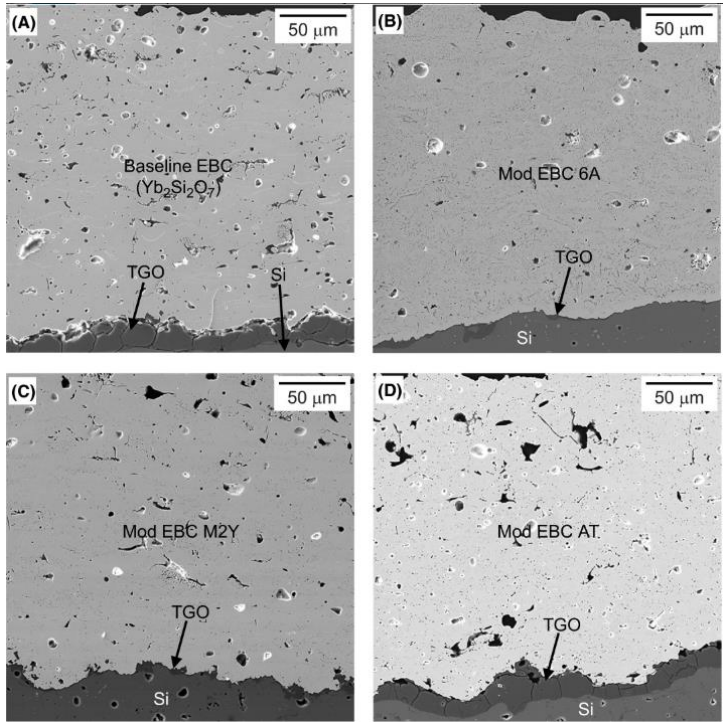


EBC chemistry affects TGO growth and CMC recession



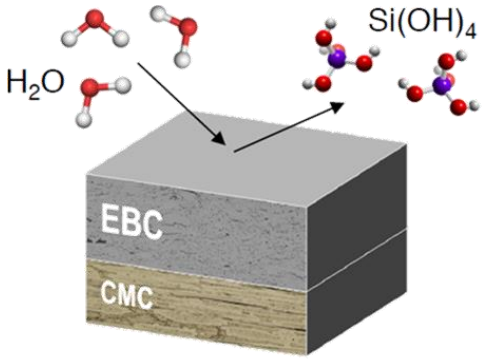
1000 h/1000 cycles at 1316℃ in 90% H_2O + 10% O_2

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

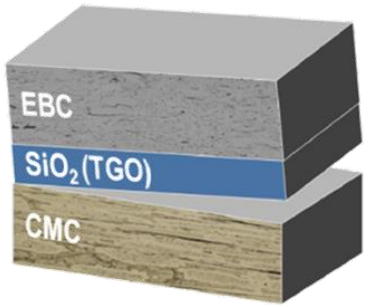
EBC Failure Modes and Design Criteria

Intrinsic Material Selection Criteria

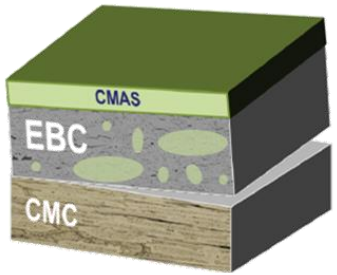
- Coefficient of thermal expansion (CTE)
- Sintering resistance
- Low H_2O and O_2 diffusivity/solubility
- Phase Stability
- Low Modulus
- Limited coating interaction



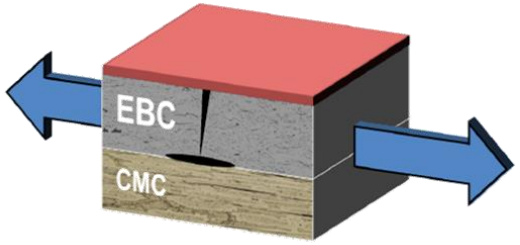
Hydroxide Formation/Recession



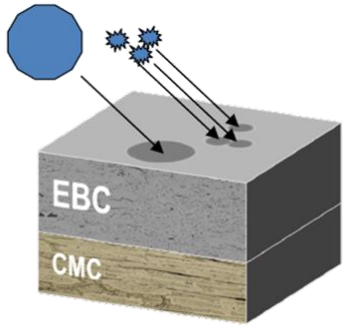
Steam Oxidation



Calcia-Magnesia-Alumina-Silicate Attack & Infiltration



Thermomechanical Durability



Erosion and FOD

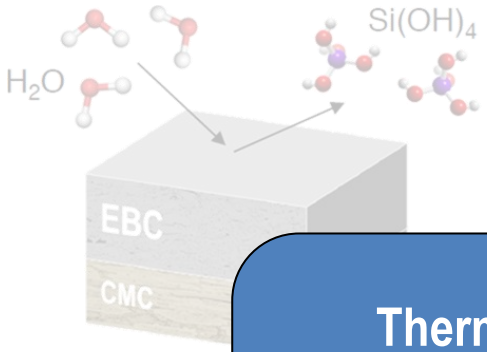
EBC lifetime and design requirements determined by combination of extrinsic failure modes

EBC Failure Modes and Design Criteria

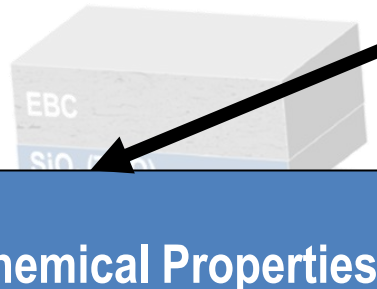
- Coefficient of therm
- Sintering resistanc
- Low H_2O and O_2 dif

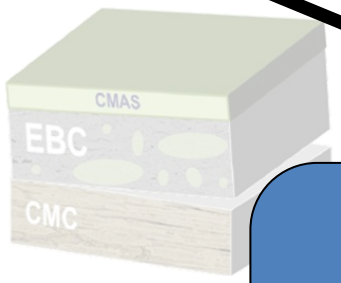
Stability
odulus
d coating interaction

Computational Simulations

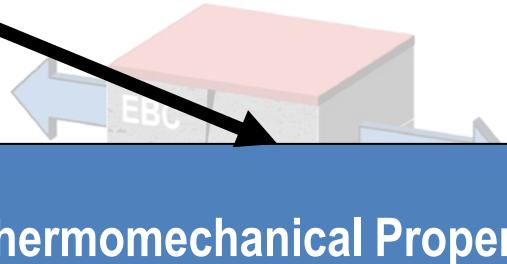


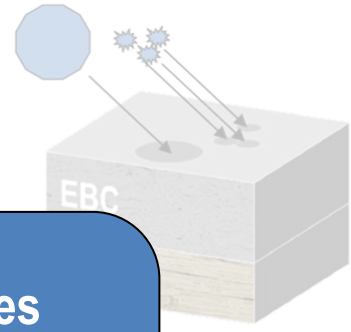
Hydr
Formation





-Magnesia-Alu
Attack & Infiltr





on and FOD

Thermochemical Properties

- **Phase Stability**
- **Chemical Reactions**

Thermomechanical Properties

- **Mechanical Strength**
- **Thermal Expansion**

etime and
nents determined by
combination of extrinsic
failure modes

Atomic-scale simulation methods for property calculations

Density Functional Theory

- 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
		  • Just don't exist

Atomic-scale simulation methods

Density Functional Theory

- Electro
  - Cos lectrons
- Smaller simulation cells
  - Self-interaction errors?

Classical Molecular Dynamics

- Atom-l
  - Cos toms
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Accuracy

Cost

Property Calculations

	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
- 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
- Descriptors often include atomic neighborhood information
  - Nearest-neighbor atoms within cutoff
  - Bonding information
- Example: NN-based interatomic potential for HfO_2

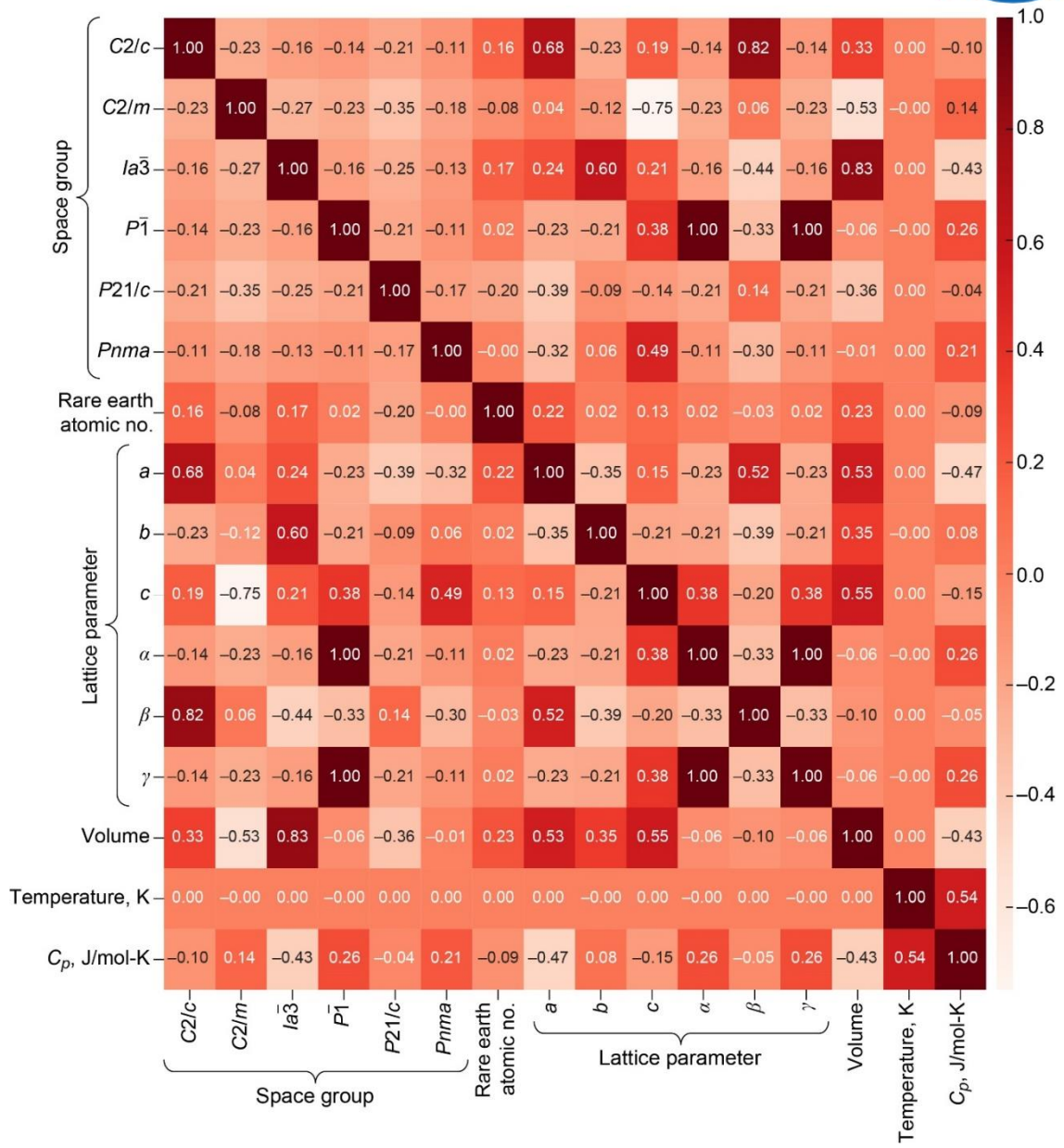
	Direct	Indirect
Pros	• Can describe more materials • Can be easier to train	• Can enable prediction of multiple properties
Cons	• Single output property	• More intensive to train • Often limited to a single material

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

DIRECT MACHINE LEARNING PREDICTIONS

Feature Selection for Neural Network Model

- NN model to predict heat capacity (Cp) for RE oxides, monosilicates, and disilicates
  - Candidate EBC materials
- Features selected to simply include geometric and chemical information
  - Space group, lattice parameters, RE cation, and temperature were used as input features
- Correlation Matrix analysis performed to determine most impactful features
- Features with a correlation coefficient of 1.0 were neglected from model training
  - Reduces overfitting
  - $P\bar{1}$ space group and γ angle were removed

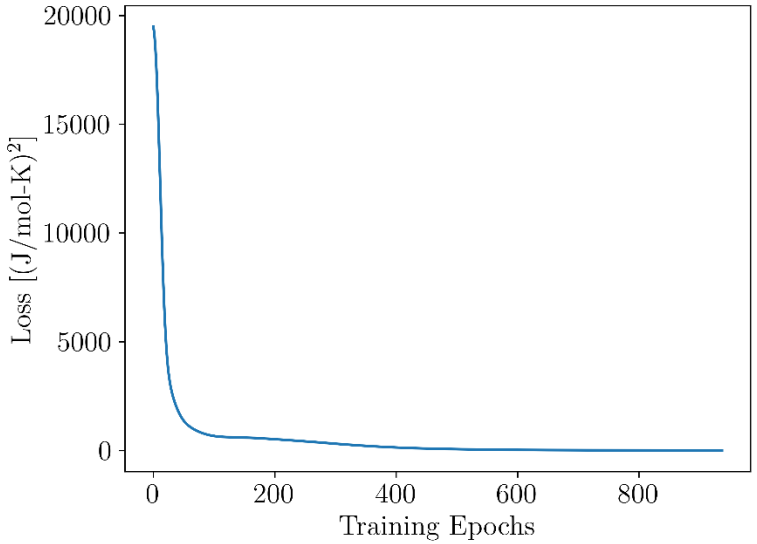


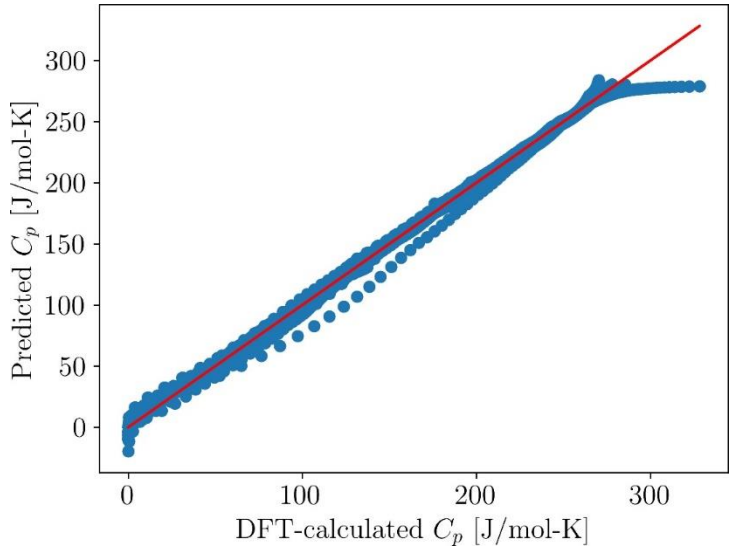
Hyperparameter Optimization and Model Training

- Trained on DFT data
- Model was trained using “leave-one-group-out” cross validation where each material C_p-T dataset was a group
- Optimized model structure:

Parameter	Tested	Optimal
Hidden Layer Architectures	(1), (10), (100), (1,1), (10,10), (100,100), (1000,1000), (1000,100), (10,10,10), (100,100,100), (1000,1000,1000), (1000,100,10)	(100)
Activation function	Logistic, RELU	RELU
Solver	LBFGS, SGD, Adam	Adam
Learning rate update method	Constant, adaptive	Adaptive
L2 regularization constant, α	0.0001, 0.001, 0.01	0.01

- Optimized model RMSE:
  - 5.12±3.37 J/mol-K

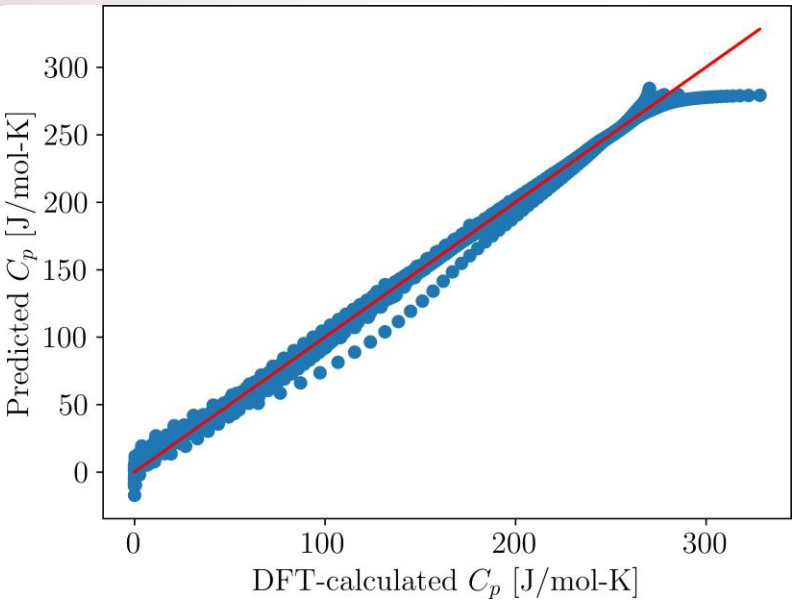


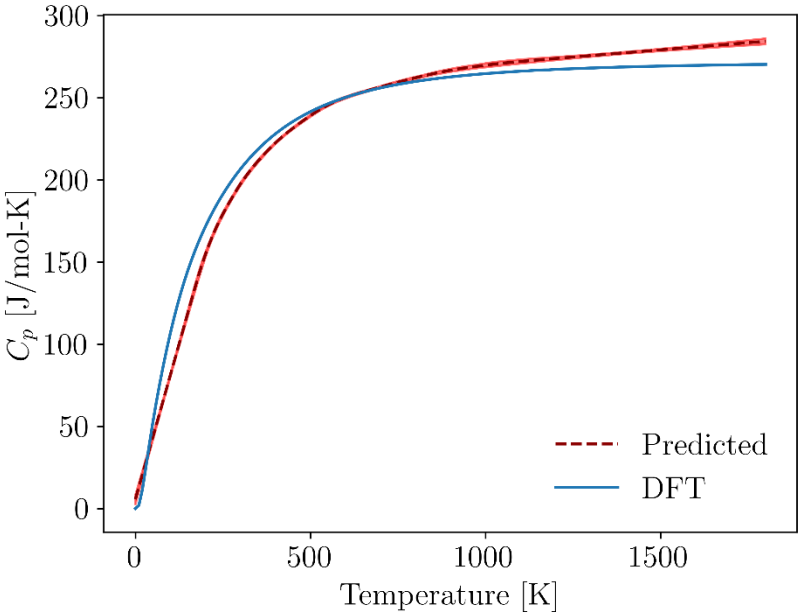


Bodenschatz. *NASA/TM-20230004256*. **(2023)**

Bagged Ensemble Model

- After determining the optimal model structure, the model was re-trained using the bagging ensemble regressor model
  - 50 models were trained using randomized subsets of C_p data
- The ensemble model can be used to calculate the average prediction of all the individual models
- The standard deviation of the ensemble of models can be used to show an error estimate of the model prediction capabilities
- The Out-of-Bag score for the overall accuracy of the ensemble model is 0.997, and the mean standard deviation over the temperature range is 1.1 J/mol-K

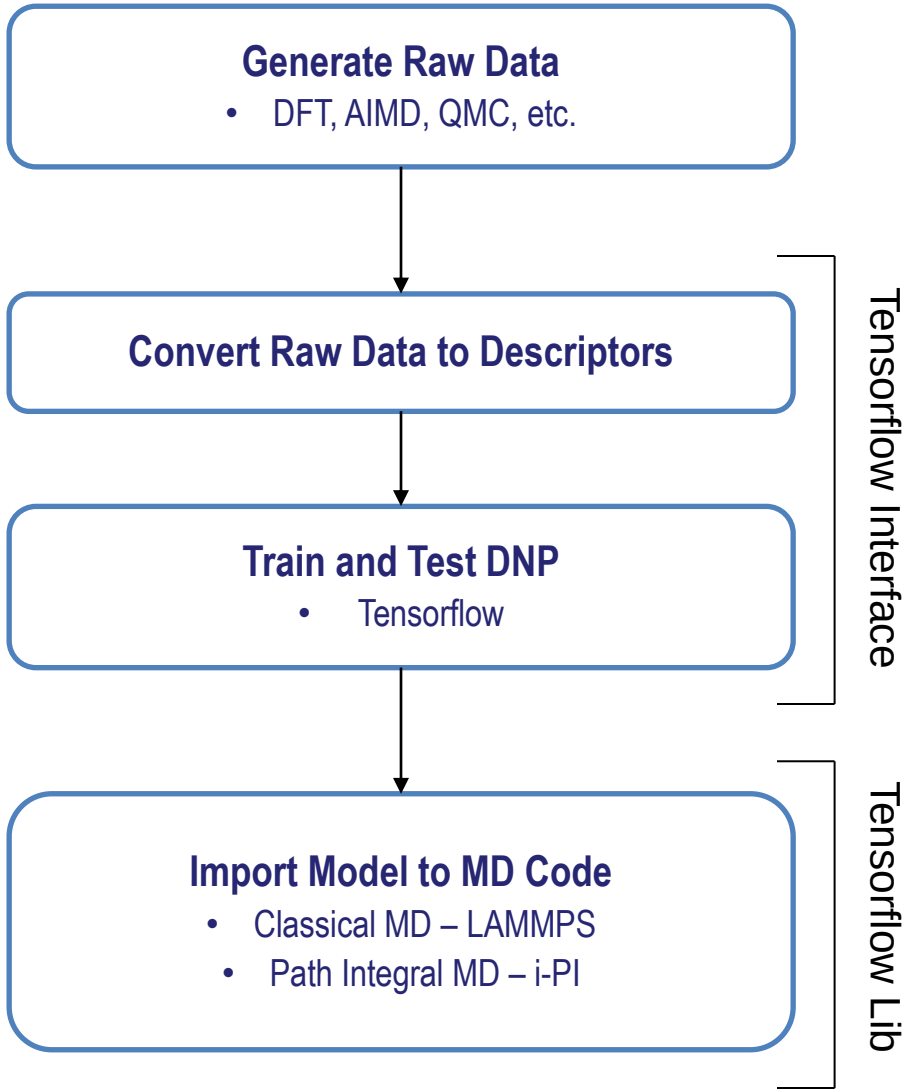




INDIRECT MACHINE LEARNING PREDICTIONS

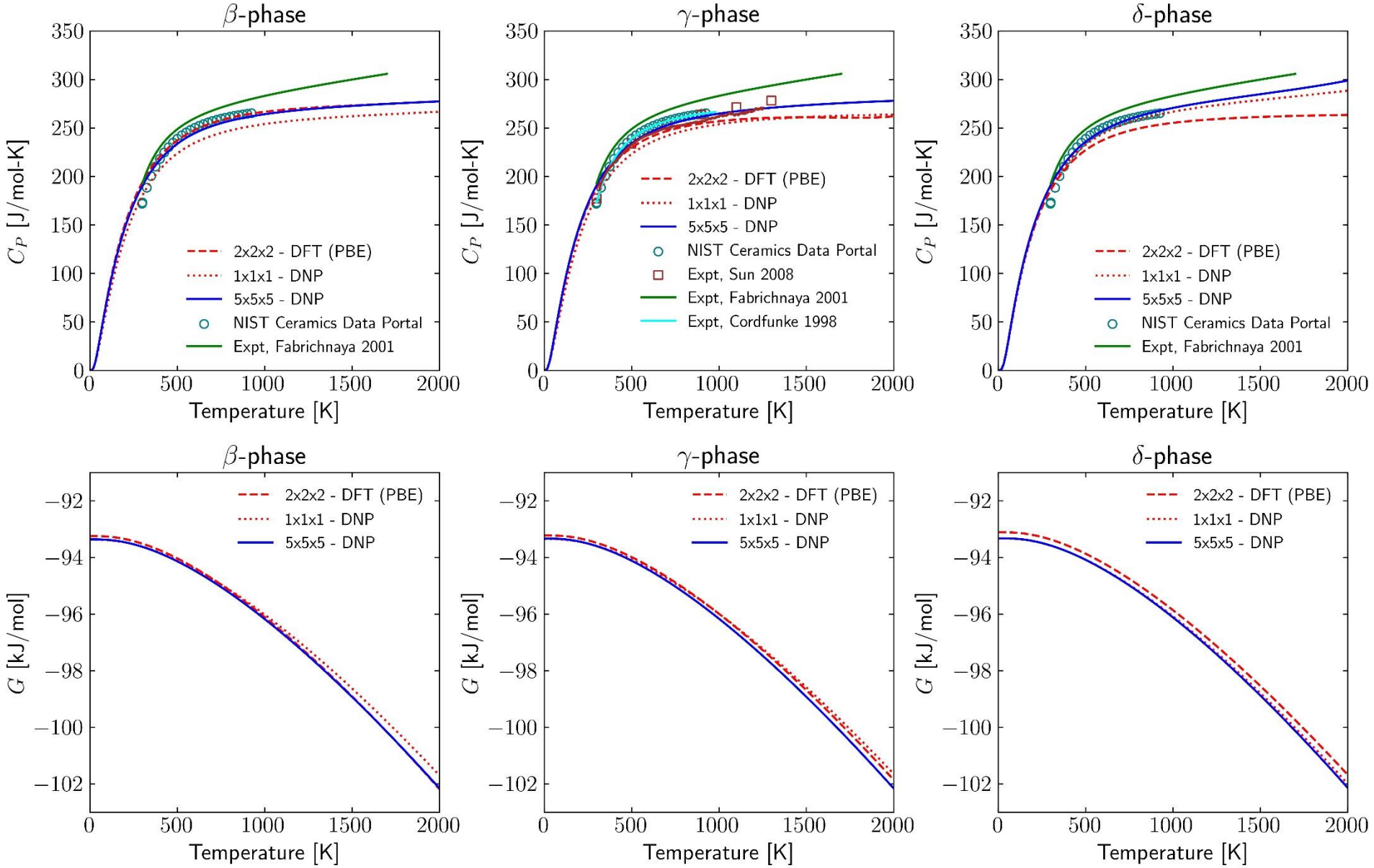
Training an Interatomic Potential from DFT Data

- Deep Neural Network Potential (DNP) 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
  - VASP, Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional
- DNP 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**)

$Y_2Si_2O_7$ Predicted Properties From MLMD Potential

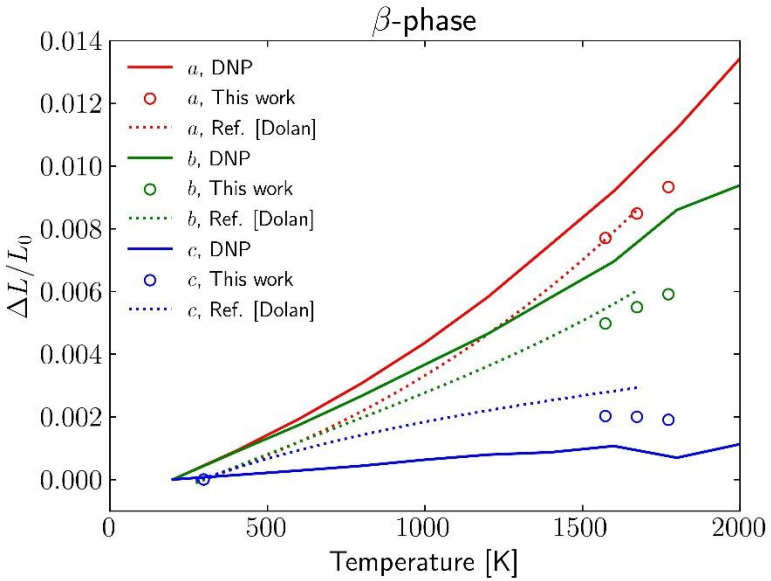


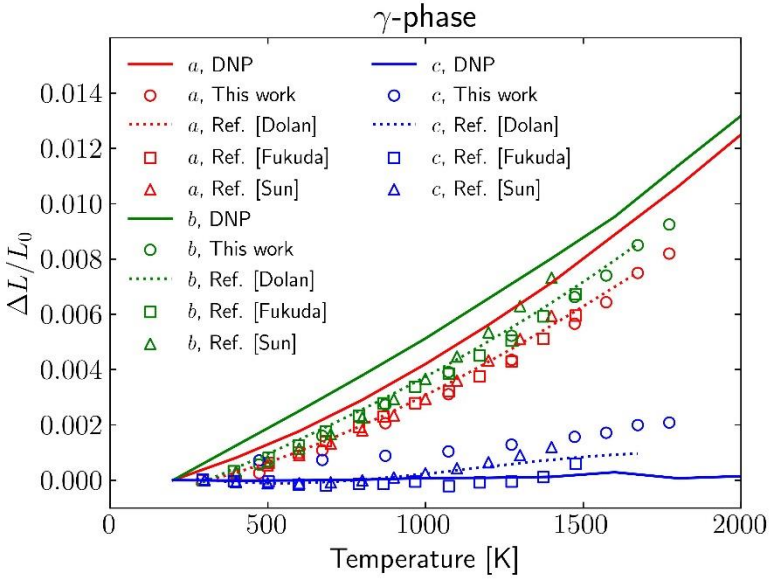
CTE calculations as function of temperature

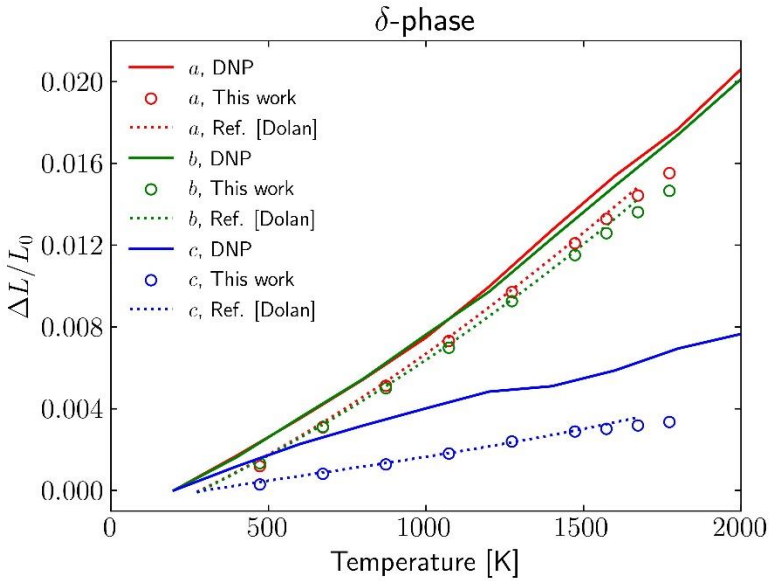
- Good agreement between DNP simulations and experiment[1]
- Overprediction of *a* and *b* CTE, underprediction of *c* CTE

	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 (4.1[1])**	**4.8 (3.9[1])**	**8.9 (8.1[1])**

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







Dolan, et al. *Powder Diff.* **(2008)**
Fukuda, et al.
Sun, et al.

Conclusions

- Two machine learning approaches were used to predict thermodynamic properties for environmental barrier coating candidate materials
  - Direct prediction of properties via training a neural network model
  - Indirect prediction of properties via training an interatomic potential

- Training a neural network model enabled prediction of Cp across material classes
  - RE oxides, monosilicates, and disilicates
  - Training did not require a very large dataset (only 25 unique crystal phases)
  - Bagging ensemble method provides error estimate for model

- 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
  - Calculation of phonon vibrational frequencies accelerated compared to DFT (~hours vs. ~weeks)
  - Training DNPs 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

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- NASA Pleiades Supercomputer Cluster

- Funding
  - NASA Transformational Tools and Technologies (TTT) project
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