Predicting the High-Temperature Oxidation Response of Nickel Superalloys Using CALPHAD-Enhanced Machine Learning

Aditya Sundar

NETL Support Contractor

Disclaimer

This project was funded by the United States Department of Energy, National Energy Technology Laboratory, in part, through a site support contract. Neither the United States Government nor any agency thereof, nor any of their employees, nor the support contractor, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Authors

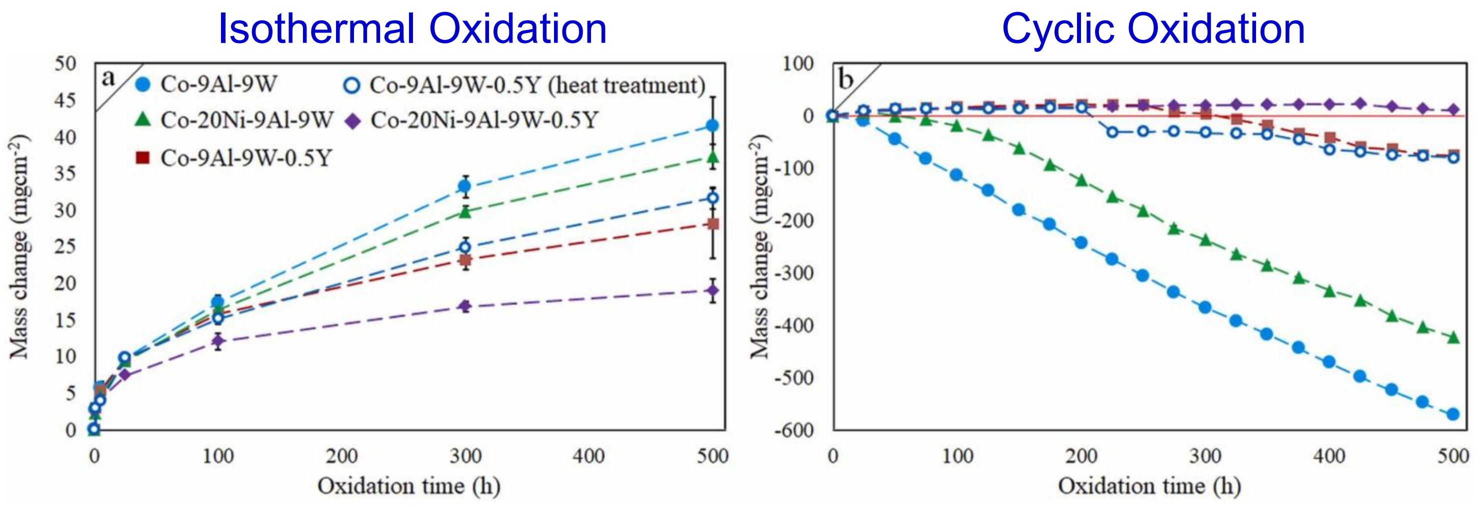
Aditya Sundar[1,2]; William Trehern[1]; Casey Carney[1,2]; Richard Oleksak[1]; Michael Gao[1]

[1]National Energy Technology Laboratory, 1450 Queen Avenue SW, Albany, OR 97321, USA

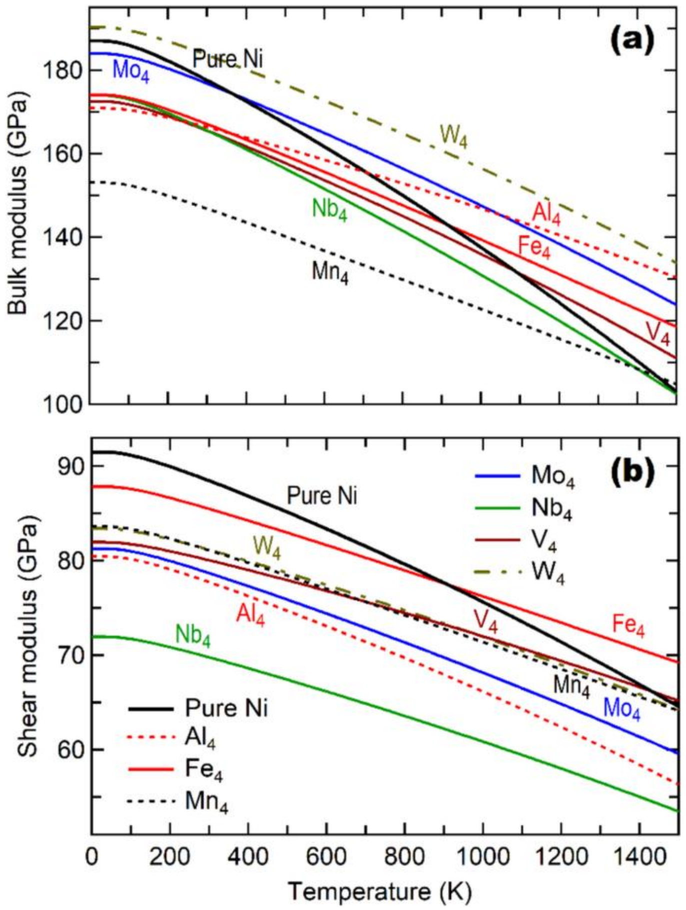
[2]NETL Support Contractor, 1450 Queen Avenue SW, Albany, OR 9732, USA

The High-Temperature Oxidation Challenge

- Strength depletion at higher temperatures
- Coefficient of Thermal Expansion (CTE) mismatch between oxide and alloy
- Cyclic oxidation promotes stress buildup, mass loss, and spallation
- Corrosive environments like sCO_2 and molten salts accelerate failure



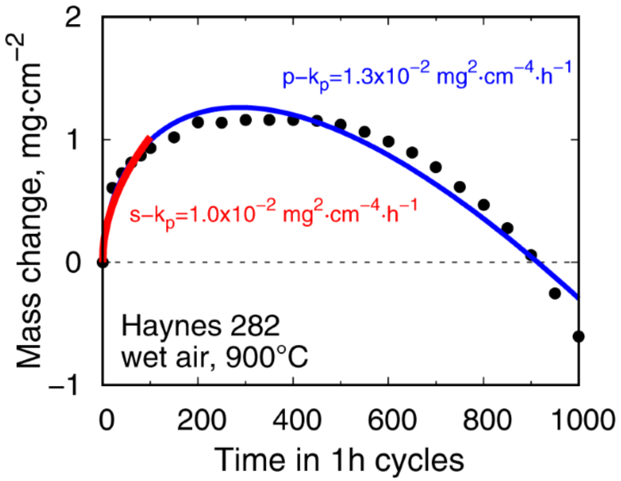
Migas et al., Corrosion Science Volume 208, November 2022, 110674.

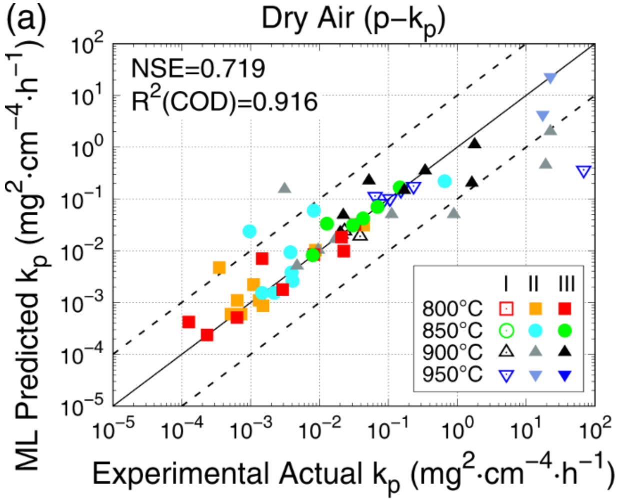


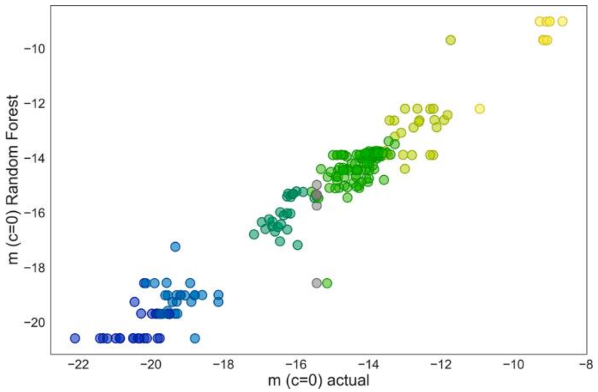
Shang et al, High Entropy Alloys Mater. **3**, 307–321 (2025).

Modeling and Machine Learning Approaches

Modeling the Parabolic Oxidation Rate

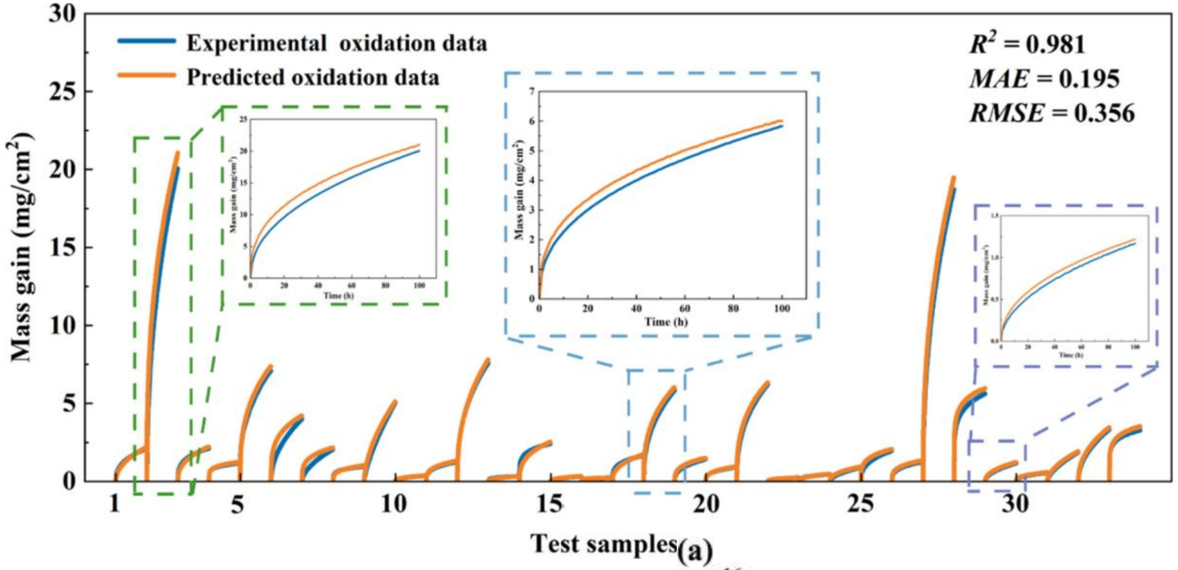




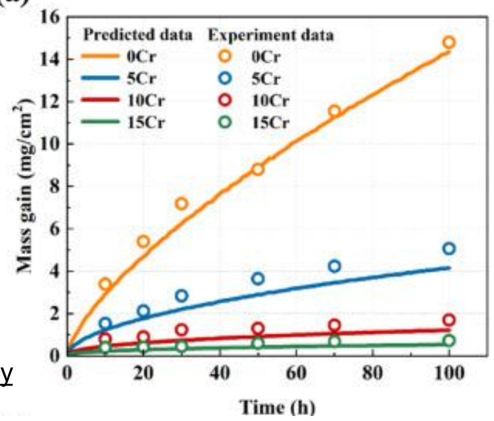


Peng et al., npj Materials Degradation **volume 5**, Article number: 41 (2021).
Taylor et al., npj Materials Degradation volume 5, Article number: 38 (2021).

Modeling the Mass Change Directly as a Time Series



- Can capture non-parabolic trends
- Co-alloys with controlled 100 h experiments
- Only mass-gain trends



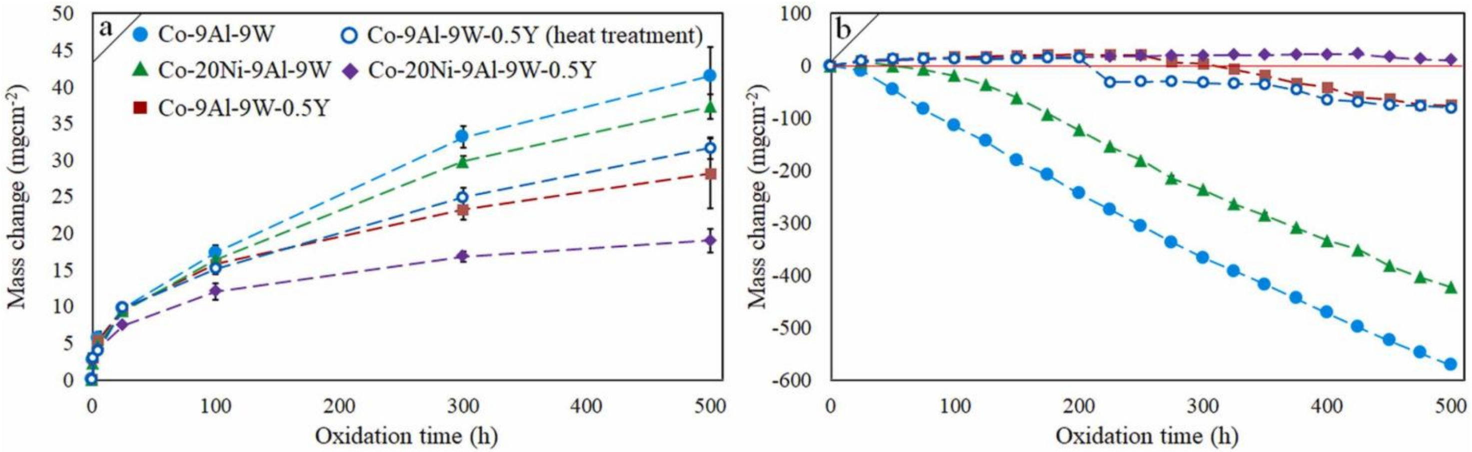
Pei et al., Journal of Materials Science & Technology Volume 235, 10 November 2025, Pages 232-243.

Open Challenges

- Prior studies on highly controlled datasets with simple features → x, T
- Data is heterogeneous → variable exposure time, number of time-points, isothermal vs. cyclic, etc.
- Quantitatively modeling the impact of thermal cycling → dominant at high temperatures
- Lack of clear model benchmarks

5 measurements in 500 h

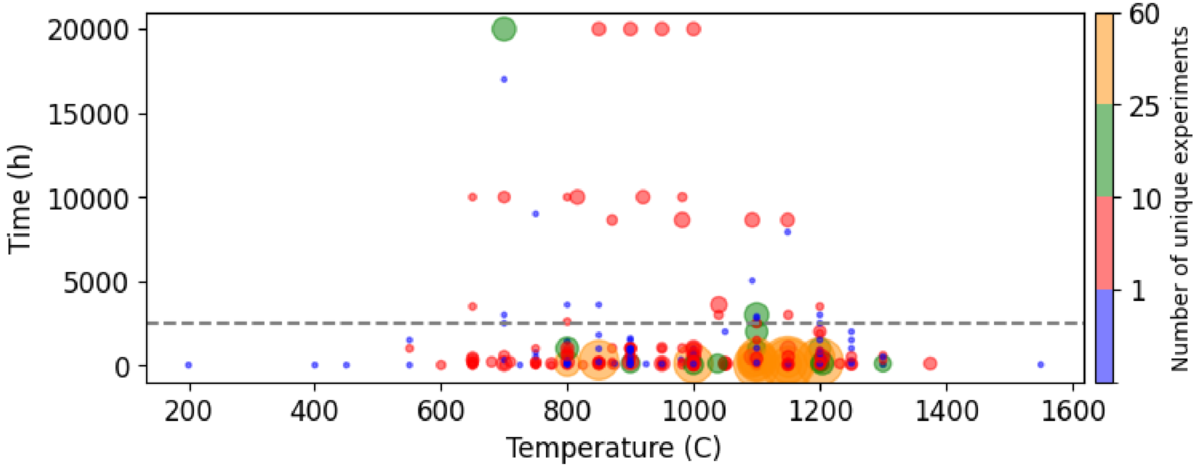
20 measurements in 500 h

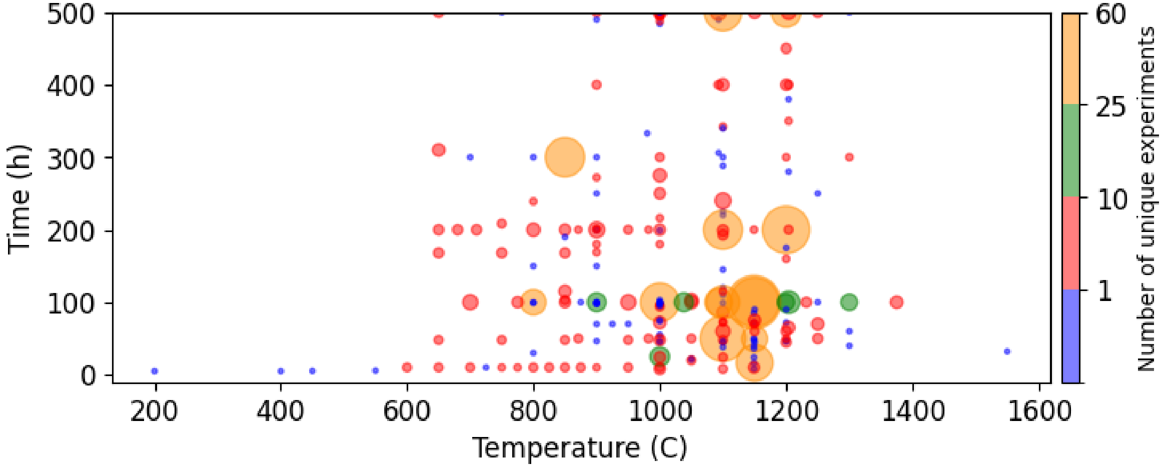


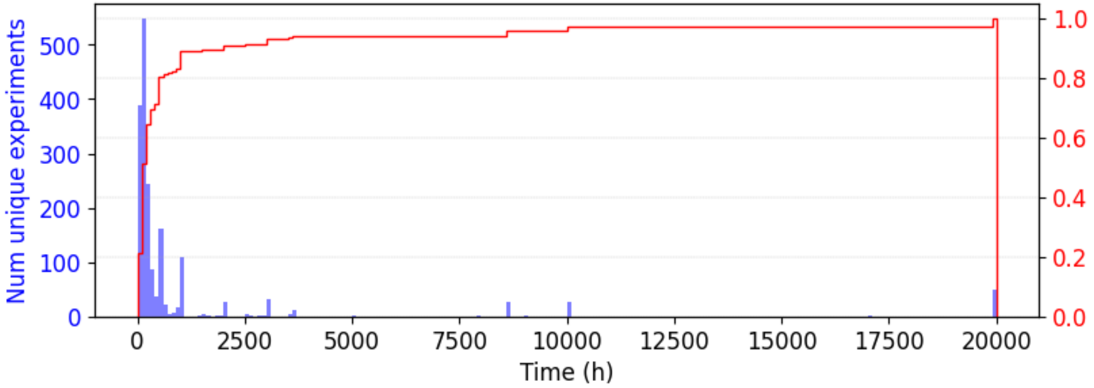
Migas et al., Corrosion Science Volume 208, November 2022, 110674.

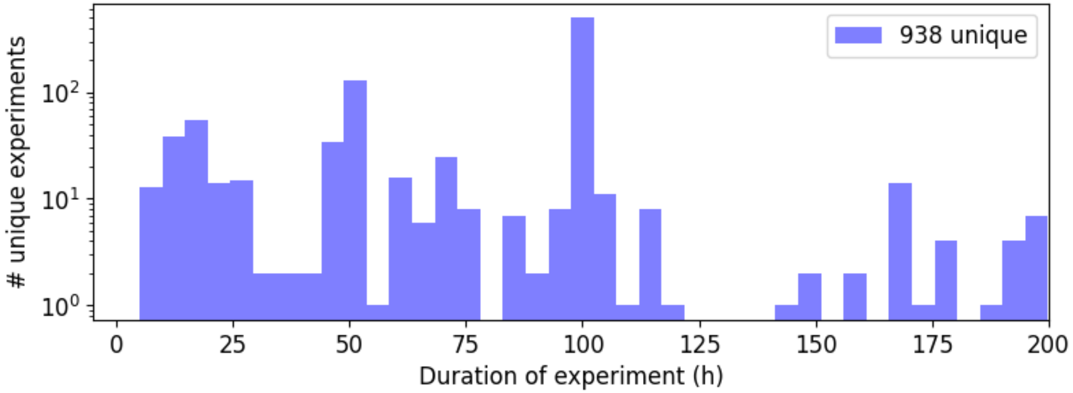
Dataset Curation

Air and O_2 Atmospheres – 1,827 Unique Experiments – Ni Alloys with 33 Elements



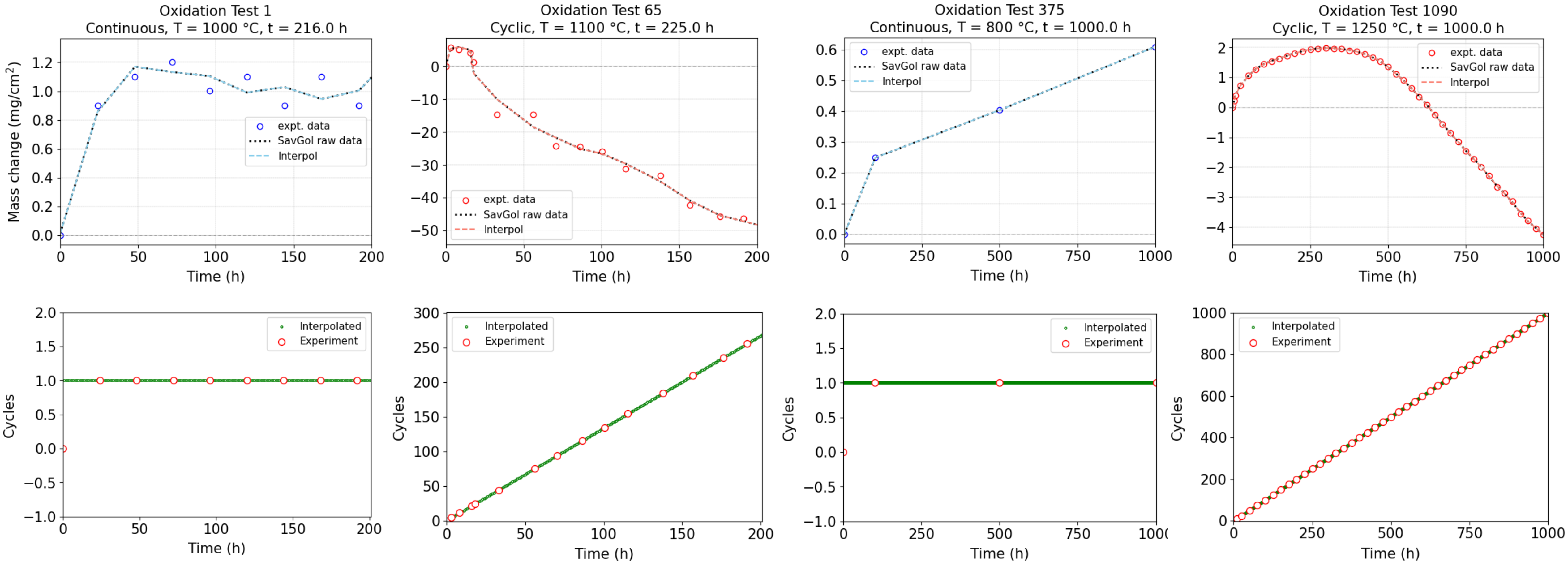






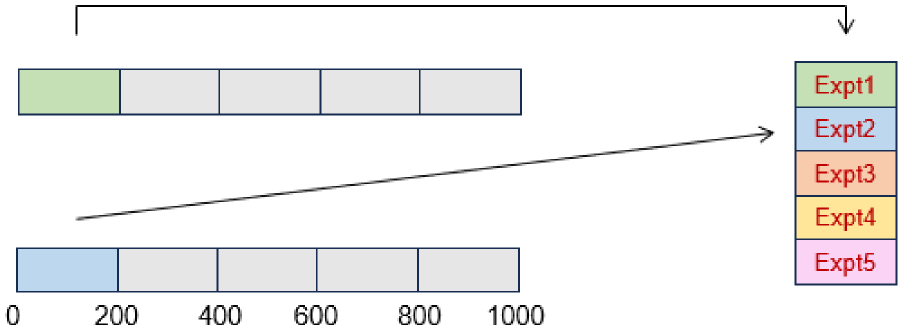
Experiments have variable number of mass-change measurements → Highly heterogeneous data

Synthetic Data to Improve Homogeneity

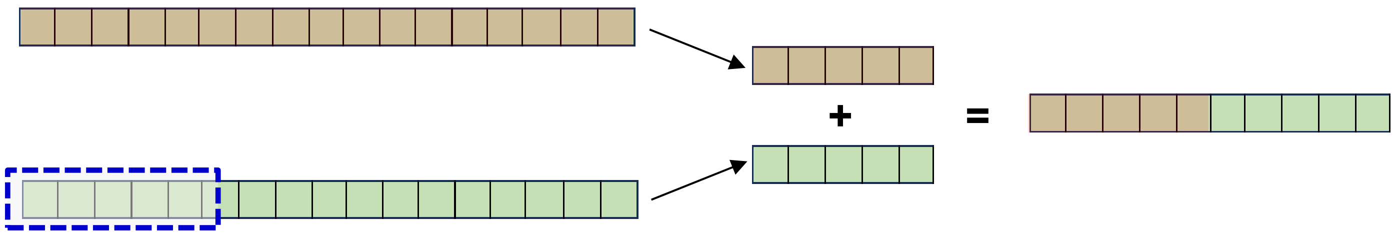


Sampling data at 1 h intervals

Selecting the Best ML Model



Neural network (NN) for time-independent features: $x, T, \Delta\alpha$



Convolutional neural network (CNN) for time-dependent features: $t, n(t), p(t)$

Cycle number (t)

CTE difference

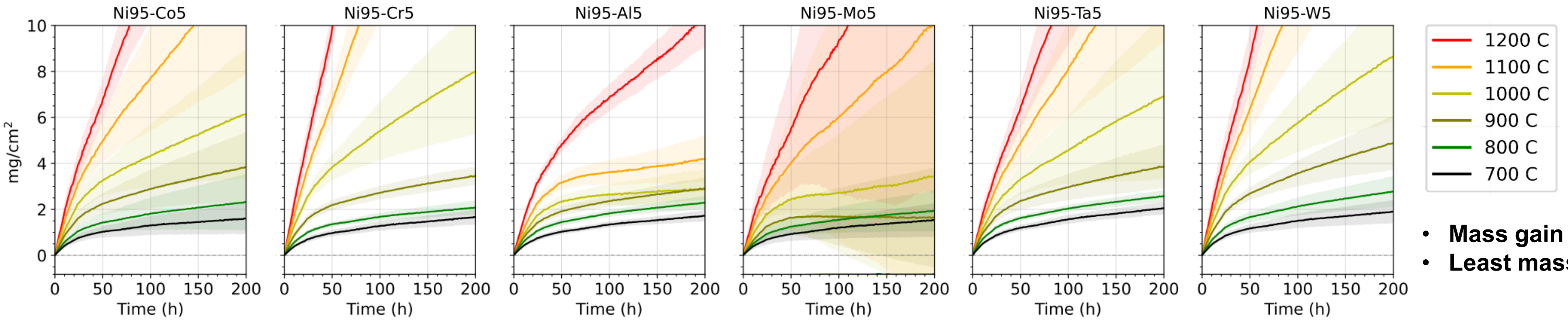
$$p = n^{1/2} \frac{(\Delta T)^2 . f . E_{ox} . (\Delta\alpha)^2 . (1 - \nu)}{\gamma_F}$$

E_{ox}, ν, γ_F: Oxide properties assumed constant for all samples

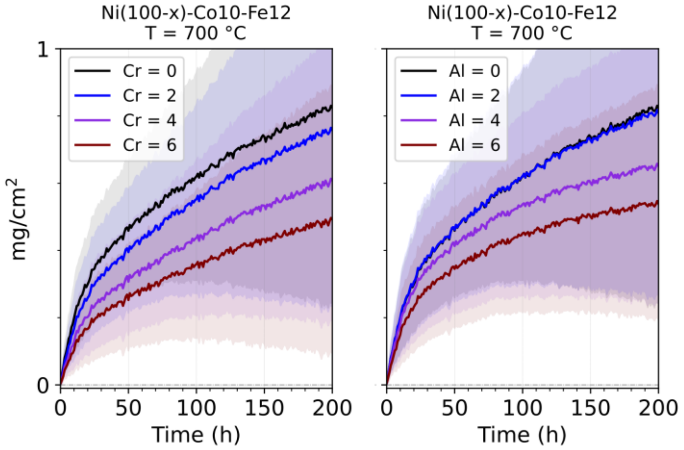
<u>Best model based on</u>:

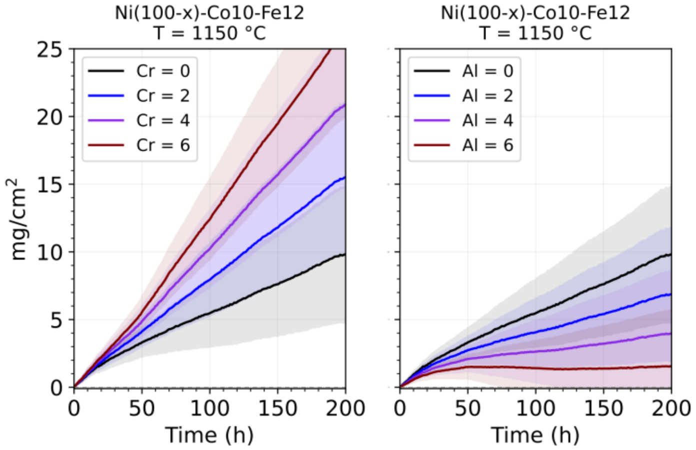
- ~Zero $\Delta\mathrm{m}$ at t=0
- Continuous mass change curves
- No spikes/jumps near end-points

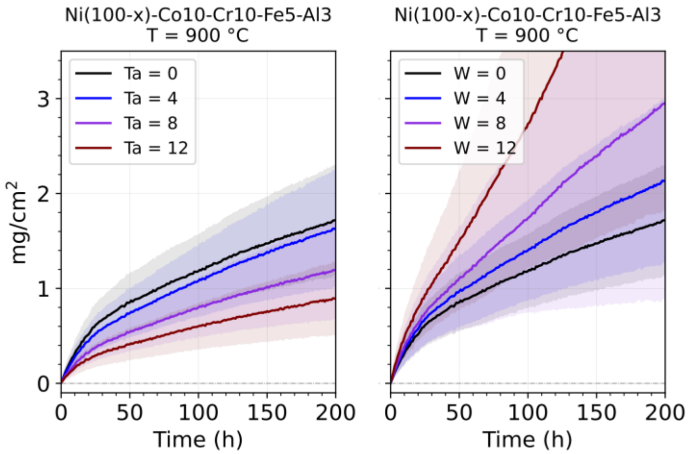
Validating the CNN+NN Model on Simple Alloys



- **Mass gain increases with T**
- **Least mass gain at 1200 °C for Ni-Al**





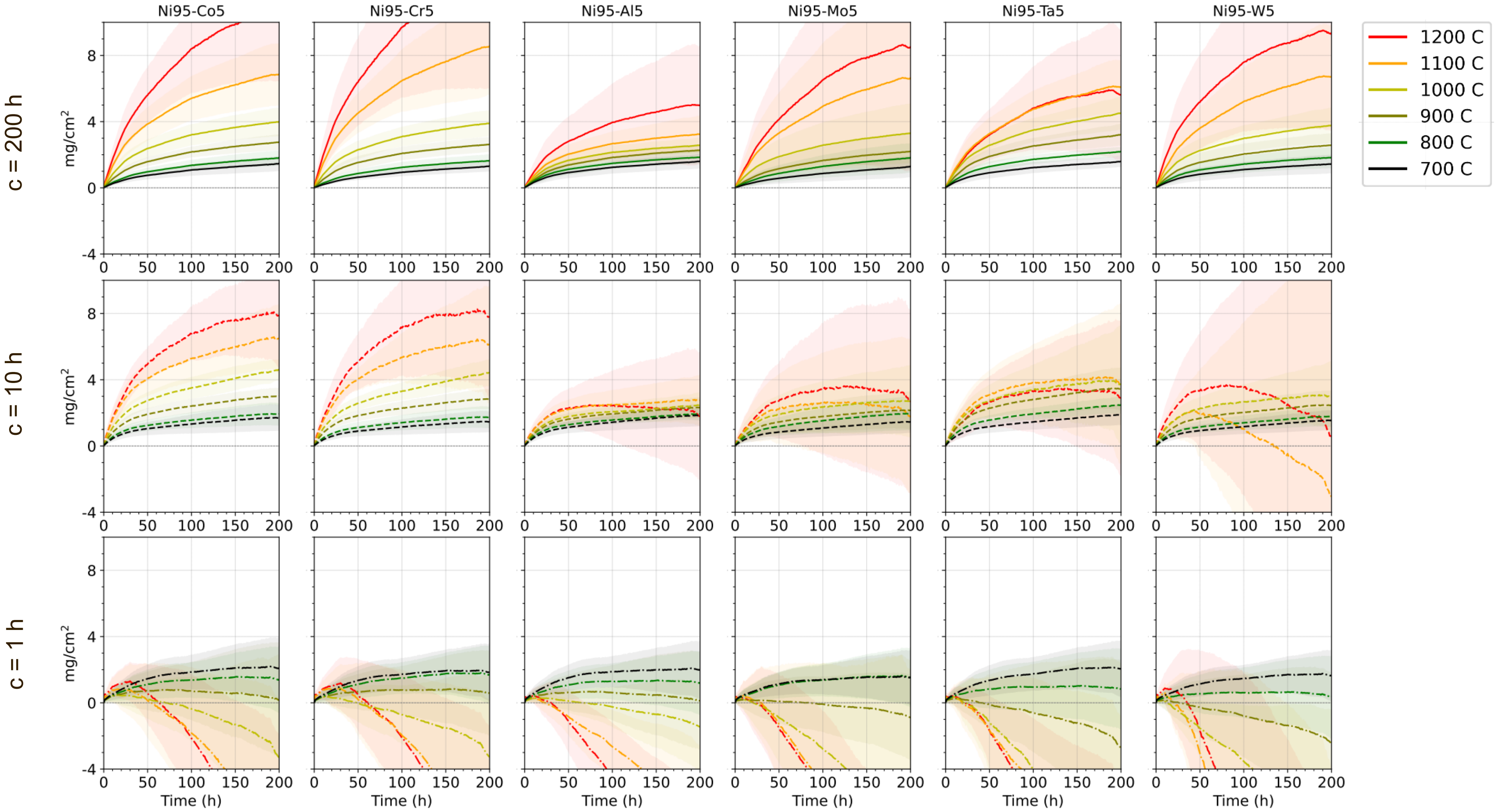


- **Compact Cr and Al oxides at 700 °C**

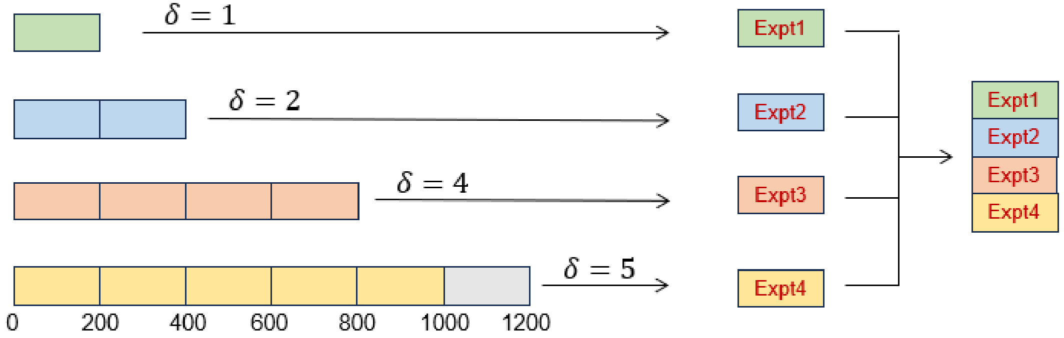
- **Compact Al oxide at 1150 °C**
- **Rapid oxidation for Cr**

- **Ta addition promotes compact oxides**
- **W addition promotes rapid growth**

Modeling Variable Cycling Rates



Handling Experiments with Variable Time-Exposure



- Each experiment has equal number of time-points
- No bias toward long-time experiments

CNN+NN feature set	Features (NN)				Features (CNN)				Number of features
	x_{1-33}	T	α	f_{1-17}	t	n	p	M	
f1	✓	✓	✕	✕	✓	✓	✕	✕	36
f2	✓	✓	✓	✕	✓	✓	✓	✕	38
f3	✓	✓	✓	✓	✓	✓	✓	✕	55
f4	✓	✓	✓	✓	✓	✓	✕	✓	55
f5	✓	✓	✓	✓	✓	✓	✓	✓	56
f6	✓	✓	✓	✓	✓	✓	✕	✕	54

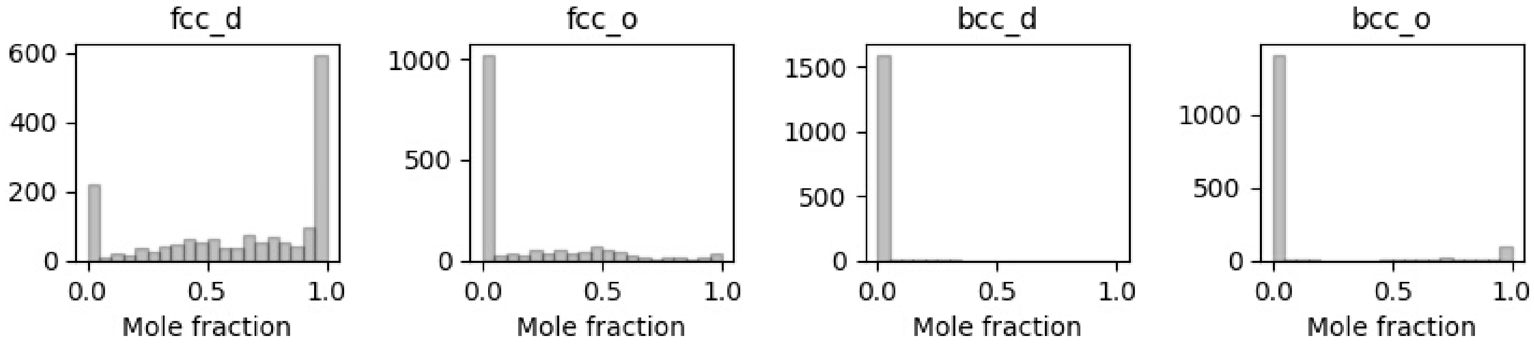
CALPHAD-Based Thermophysical Features

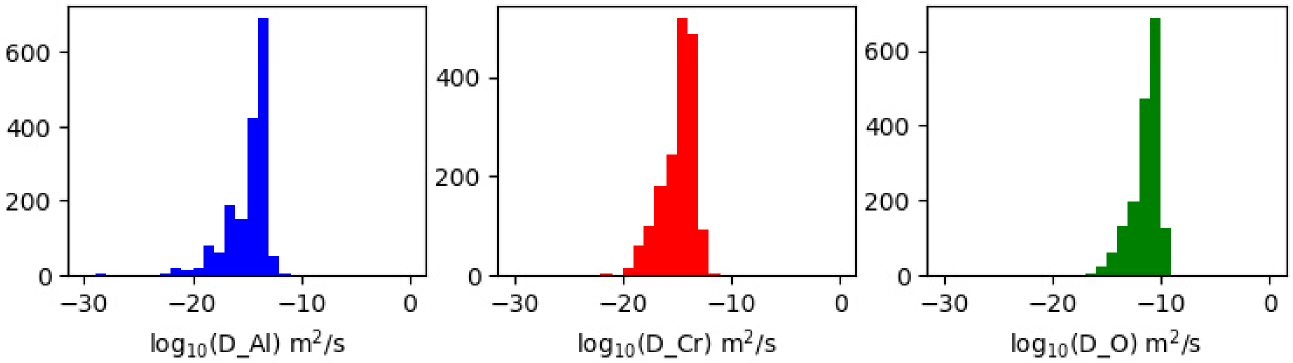
16 features are calculated at the test temperature

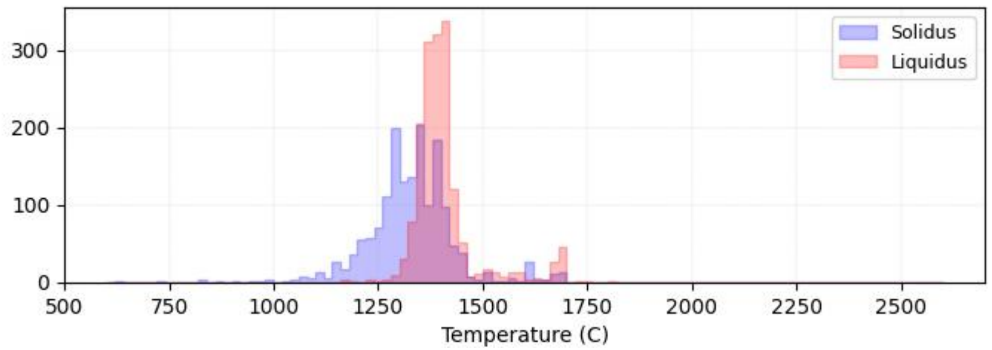
- Phase fractions of six phases (Body Centered Cubic (BCC) ordered, BCC disordered, Face Centered Cubic (FCC) ordered, FCC disordered, Hexagonal Close Packed (HCP), and precipitates)
- Thermal conductivity, thermal diffusivity
- Solidus, liquidus, volume
- Diffusivity of Al, Cr, and O
- Activity of Al and Cr

1 feature at room temperature

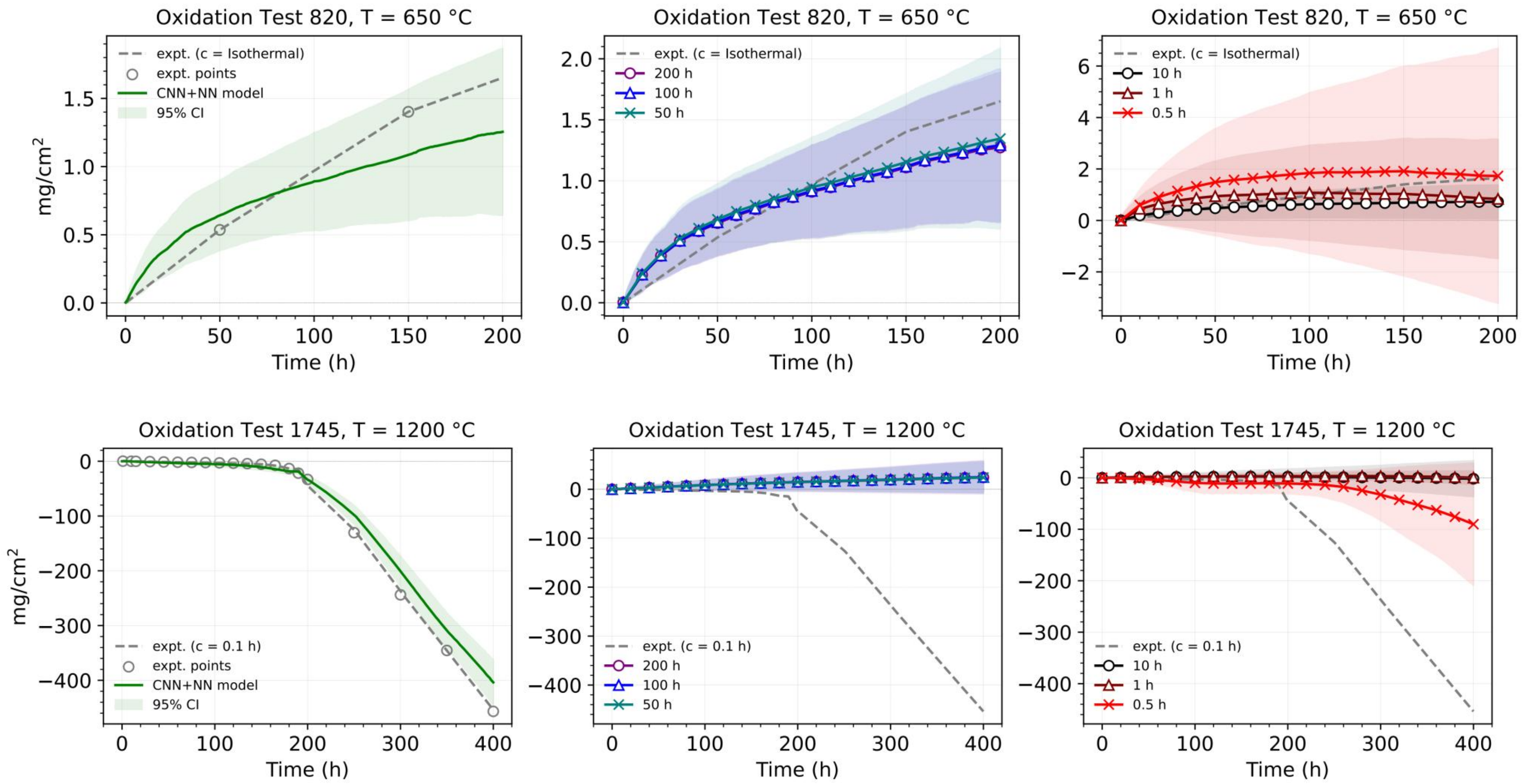
- Volume



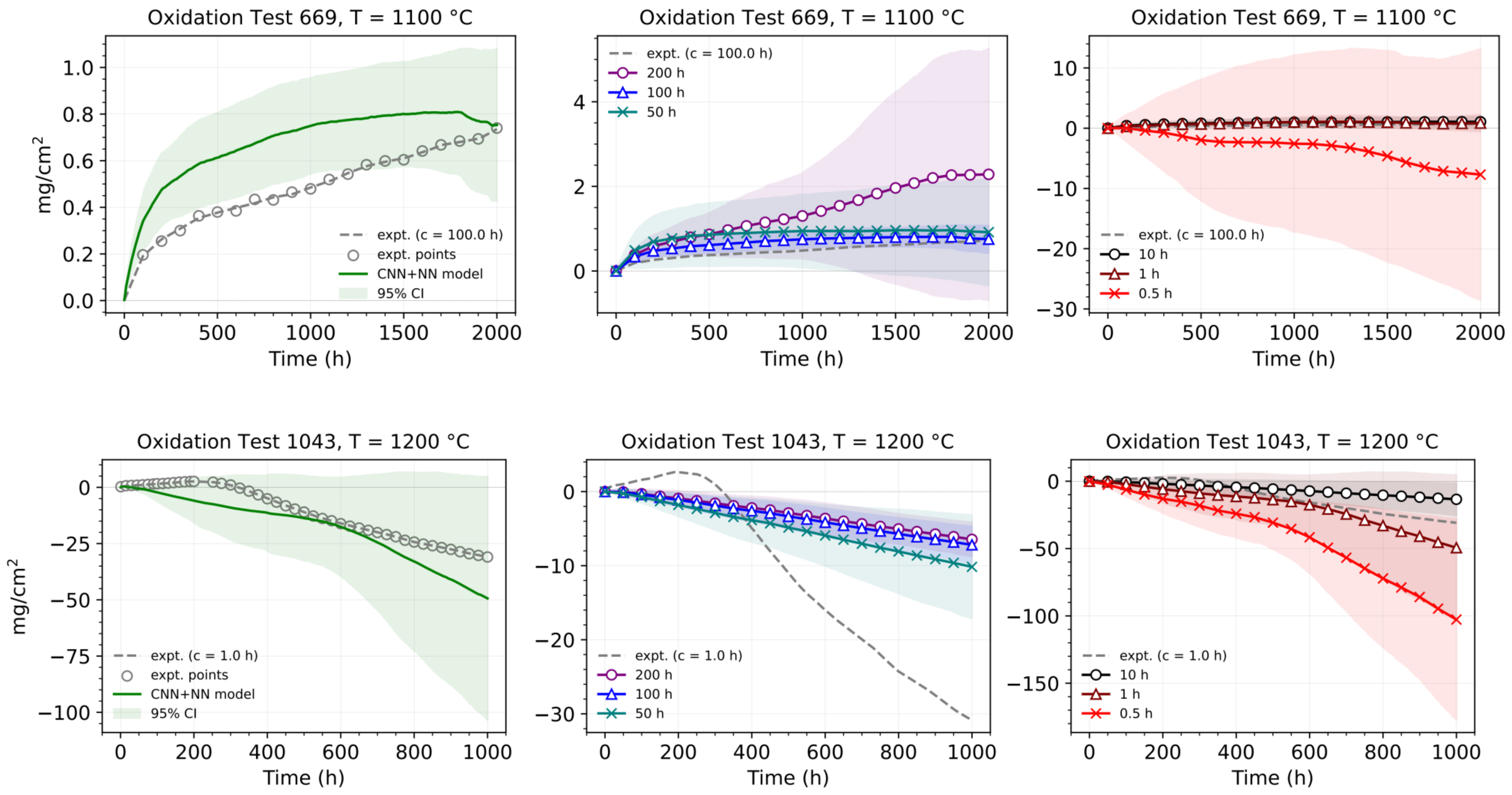




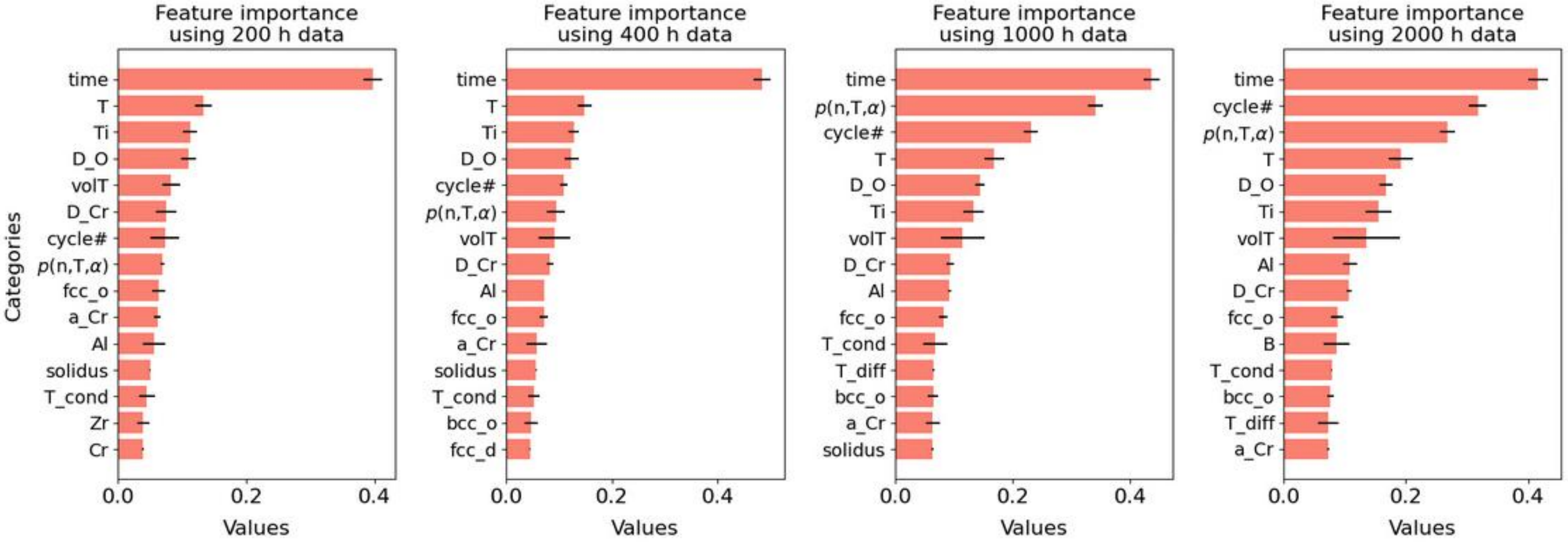
Predictions with Final Model



Predictions for Long-Time Experiments

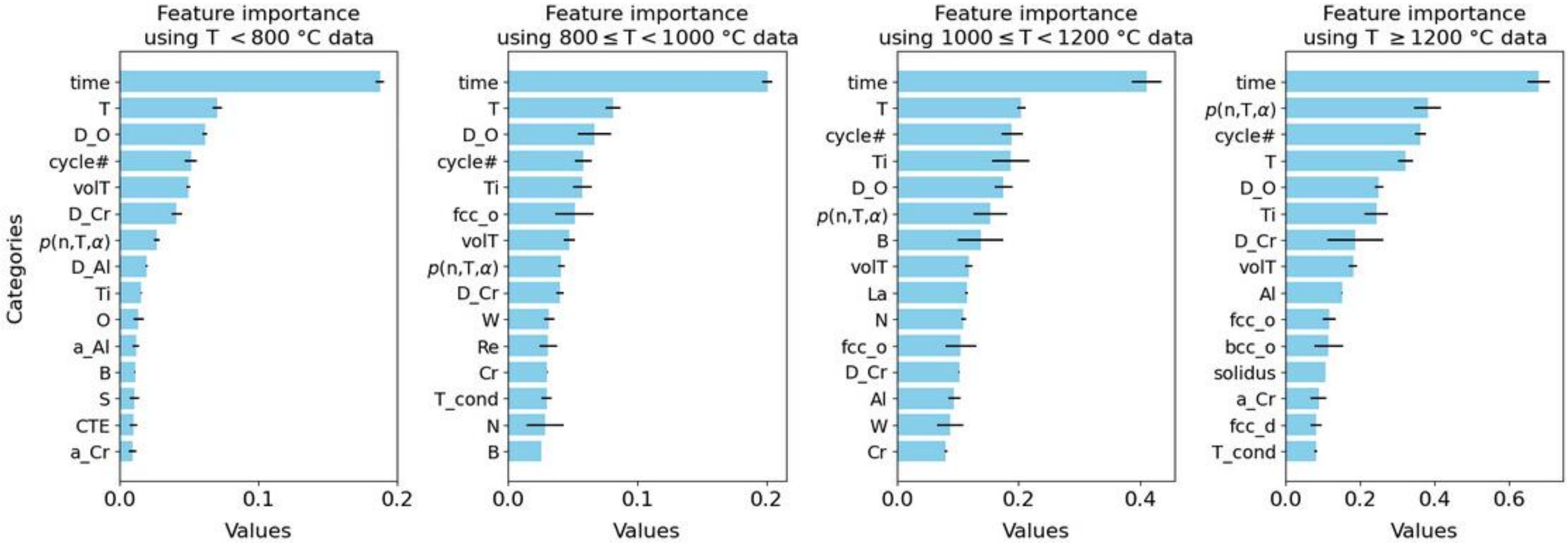


Feature Importance from Shapley Additive exPlanations (SHAP): Time Intervals



- Several CALPHAD features are prominent (diffusivity, activity, solidus, etc.)
- $p(n, T, \alpha)$ and n gain importance at longer times

Feature Importance from SHAP: Temperature Intervals



- $p(n, T, \alpha)$ and n gain importance at higher temperatures

Conclusion and Ongoing Work

- CNN+NN model accurately models mass change experiments
- Extensive benchmarking on model 2-6 element Ni-alloys
- Model capable of simulating variable cycling rates
- CALPHAD-features provide interpretable ML results

In Progress:

- For most samples, experimental data is only available at a particular cycling condition
- Validating predictions at different cycling rates is not so straightforward