Predicting Thermal Conductivity and Oxide Scale Thickness in ZrB_2 Composites: A Machine Learning Approach

M.TECH THESIS DEFENCE

by

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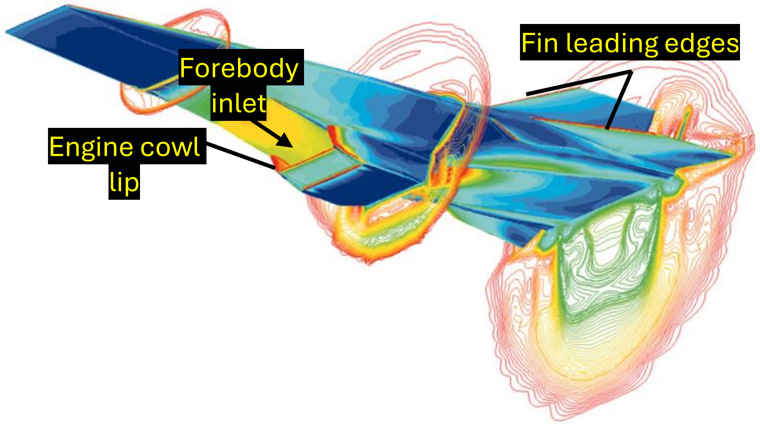
Presentation outline

- Need for *UHTCs', classification* and *properties of UHTCs'*
- Review of studies on thermal conductivity and improving oxidation resistance of ZrB_2 and its composites
- *Objectives* of this study and *work plan* incorporating *synthetic minority oversampling technique (SMOTE)*
- *Machine Learning methodology* for predicting thermal conductivity and oxidation resistance
- Results and Discussion : *Machine Learning Findings and Analysis*
- Conclusive Remarks : Model performance and statistically affecting factors
- Future scope of work : Connecting *experimental and Modelling* framework with *Bayesian optimization*

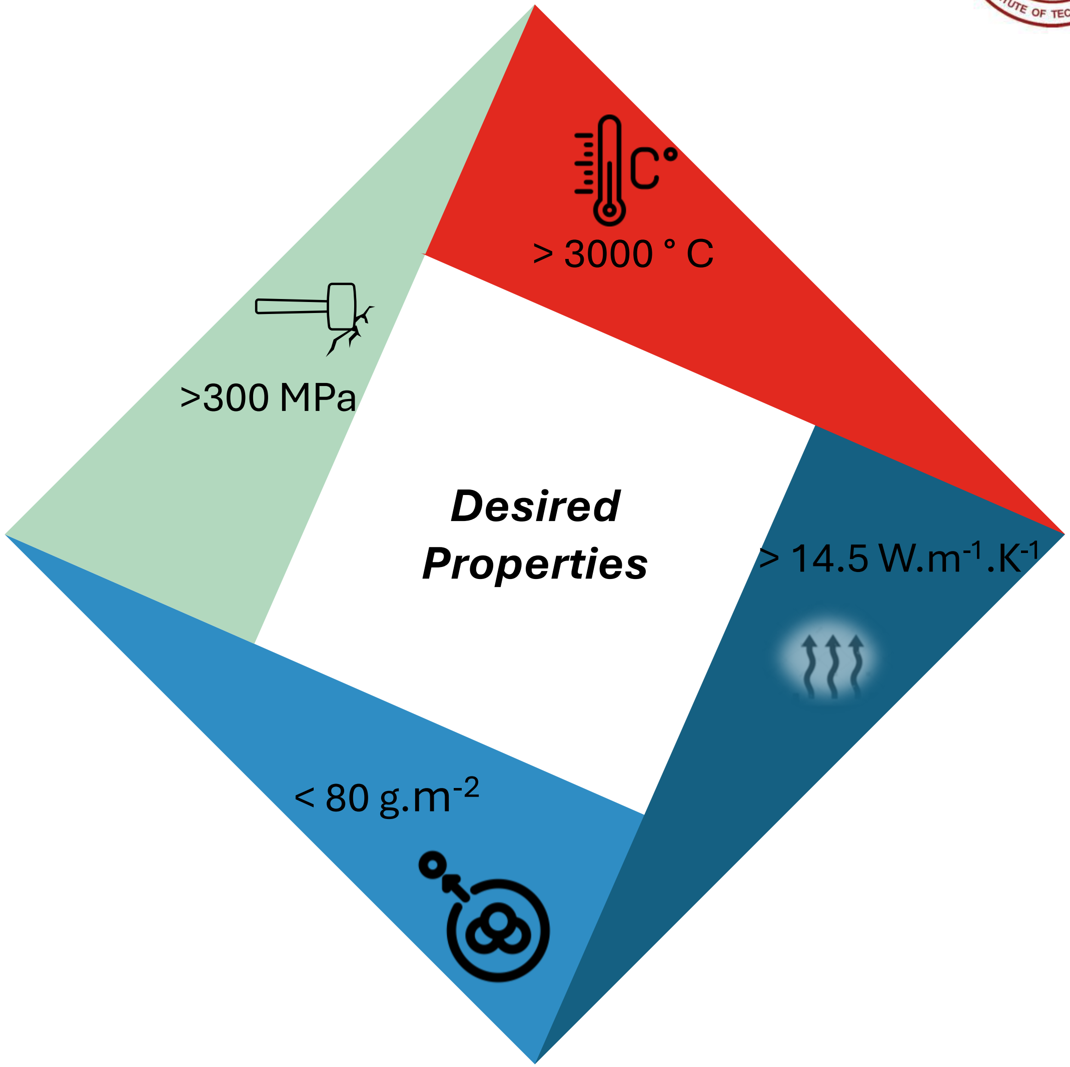
Need for Ultra High Temperature Ceramics (UHTCs)

Characteristics of hypersonic flow



a. Mach 7 hypersonic CFD simulation: heat transfer

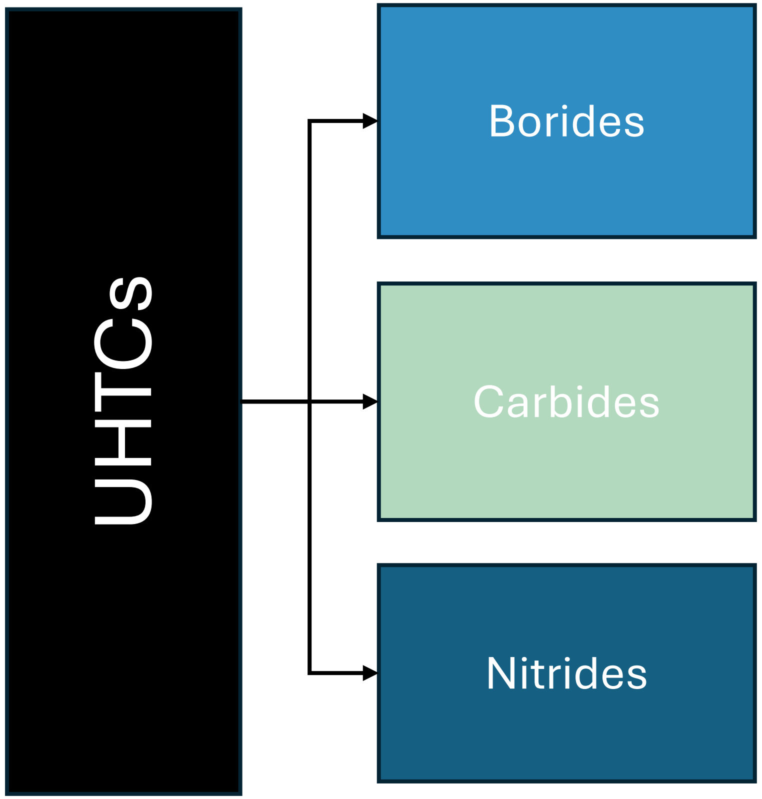
b. X-51A : record-breaking Mach 5+ scramjet flight (210 sec)



Aerospace advances need advanced materials for hypersonic flight and re-entry

Nature Mater 15, 804–809 (2016), Int. J. Appl. Ceram. Technol. 8 (4) (2011) 832–847, Aerospace Lab (3) (2011) 1–11, J. Eur. Ceram. Soc. 19 (13) (1999)

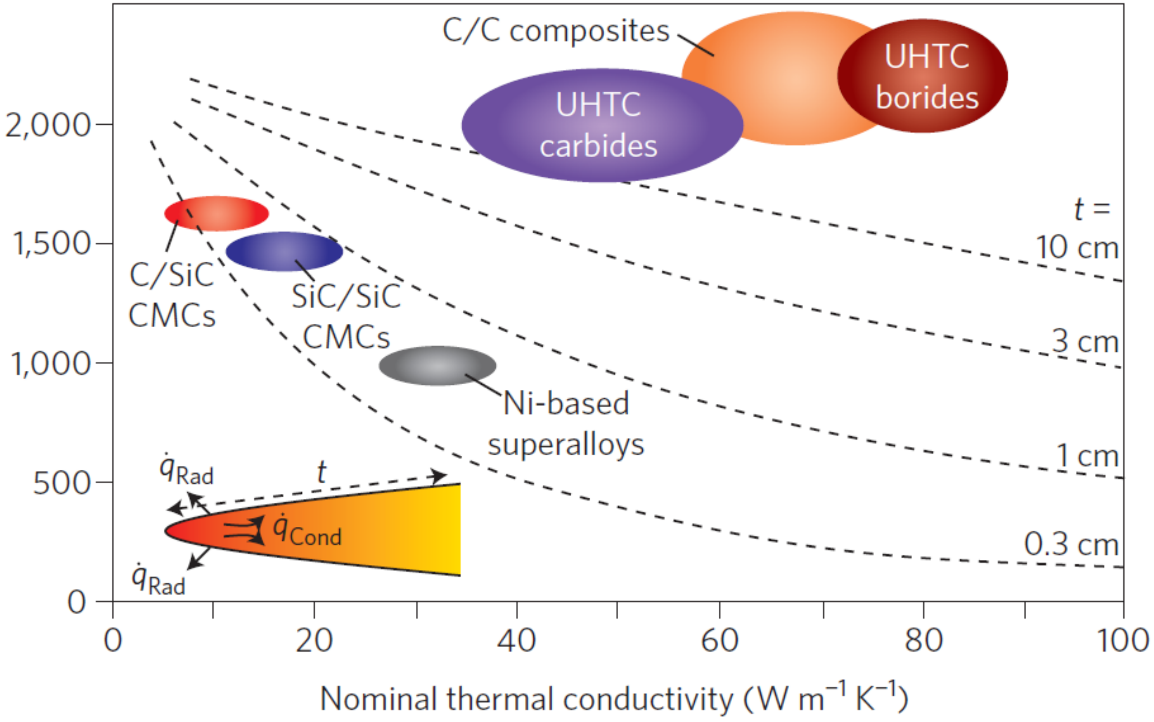
Classification and Properties of UHTCs



Material	Crystal structure	Melting temperature (℃)	Density ($g \cdot cm^{-3}$)	CTE, α (10^{-6} K^{-1})	Thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)	Electrical resistivity ($\mu\Omega \cdot cm$)	Elastic modulus (GPa) Cal.	Elastic modulus (GPa) Exp.	Hardness (GPa)
TaB_2	HCP	3040	12.5	8.2–8.8	10.9–16.0	33	497	551	19.6
TiB_2	HCP	3225	4.5	7.6–8.6	64.4	16–28.4	583	575	24.0
ZrB_2	HCP	3245	6.1	5.5–8.3	57.9	9.2	523	489	23.0
HfB_2	HCP	3380	11.2	6.3–7.6	51.6	8.8–11	535	451	28.0
TiC	FCC	3100	4.9	7.5–7.7	17–21	52.5	455	437	30.0
ZrC	FCC	3530	6.6	6.82	20.61	68.0	436	387	25.0
TaC	FCC	3800	14.5	6.6–8.4	22.2	30–42.1	550	537	17.0
HfC	FCC	3900	12.8	6.3	22.2	45.0	537	461	24.2
TaN	FCC	2900	13.4	3.2	8.3	128–135	490	490	10.8
TiN	FCC	2950	5.4	9.35	29.1	21.7	463	400	18.6
ZrN	FCC	2950	7.3	7.24	20.9	13.6	390	384	15
HfN	FCC	3385	13.9	6.5	21.6	33	411	398	16.1

Why Borides?

Materials	Advantages	Disadvantages
Carbon based	• Short duration	• Non- Reusable • Low oxidation resistance
Borides	• Reusable • Oxidation resistance	• Brittle • Reinforcements required
Carbides	• Relatively high melting points	• Lower oxidation resistance



Temperature capability vs. thermal conductivity for superalloys, CMCs, UHTCs

Nature Mater 15, 804–809 (2016)

Literature review

- Thermal conductivity
  - SiC/ZrC
  - $ZrSi_2/MoSi_2$
- Oxidation resistance
  - Monolithic
  - SiC

Thermal conductivity : Carbides and Silicide's

Author	Effect on Thermal Conductivity	Attributed Factor
Loehman et al	Decreasing thermal conductivity with increasing SiC content (5vol% to 20vol%)	Inverse relationship between SiC content and thermal conductivity
Wang et al	Relatively lower thermal conductivity for ZrB_2-SiC composites	Average particle size of the SiC
Andrievskii et al	Overall reduction in thermal conductivity with increasing ZrC content	Content of ZrC
Fridlender et al	Decreasing thermal conductivity with increasing ZrC content	Content of ZrC
Hassan et al	Better thermal conductivity compared with HfB_2–SiC composite	Better sintering of ZrB_2-SiC composite
Monteverde et al	Increased thermal conductivity	$MoSi_2$ content
Guo et al	Decreased thermal conductivity with increasing $MoSi_2$ content	$MoSi_2$ content, interfacial thermal resistance at $ZrB_2/MoSi_2$ grain boundaries
Guo et al	Decreased thermal conductivity with increasing $ZrSi_2$ content	$ZrSi_2$ content, interfacial thermal resistance

Oxidation resistance

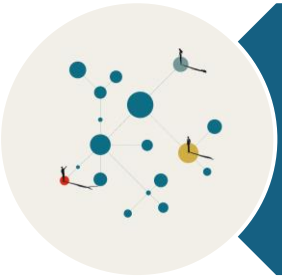
Author	Effect on oxidation resistance	Attributed Factor
Fahrenholtz et al	Increases and then it decreases(727°C - 2227°C)	B_2O_3 initially dominates vaporization, then it is disrupted at 1527°C and at 2227°C no protective scale of B_2O_3
Tripp et al	800°C-1400°C Increases till 1100°C (decreasing time dependence) post that temperature decreases	B_2O_3(l) limits O2 diffusion
Clougherty et al.	Conversion depths five times smaller than monolithic ZrB_2 (20 vol% SiC).	SiC addition and SiC depletion zone formation
Rezaie et al	Absence of boria in outer glass layer at 1500°C (air) (30 vol% SiC)	Selective volatilization of boria
Tripp et al	Significant improvements up to at least 1400°C (20 vol% SiC)	Borosilicate formation due to SiC inclusion
Fahrenholtz et al	Porous ZrO_2 scale without boria/silica/borosilicate in low pO_2 environment (30 vol% SiC)	Low pO_2 favoring active SiC oxidation and leaving ZrB_2 unaffected
Shugart et al	Three distinct layers above 1627°C: borosilicate glass, ZrO_2 with borosilicate, SiC depleted zone (30 vol%)	Dependence on free energy of formation of reactions and SiC content

Gap in the literature

- **Thermal conductivity dependency on various parameters of powder purity, powder geometry, initial composition, synthesizing methods and microstructural parameters such as porosity and mean grain size**
- **Equal emphasis needed to put on computational approaches for better design of application specific design**
- **Lack of synergistic approach, computational and experimental.**

Objectives

Build Machine Learning models to forecast the thermal conductivity and oxide scale thickness of ZrB_2 composites.



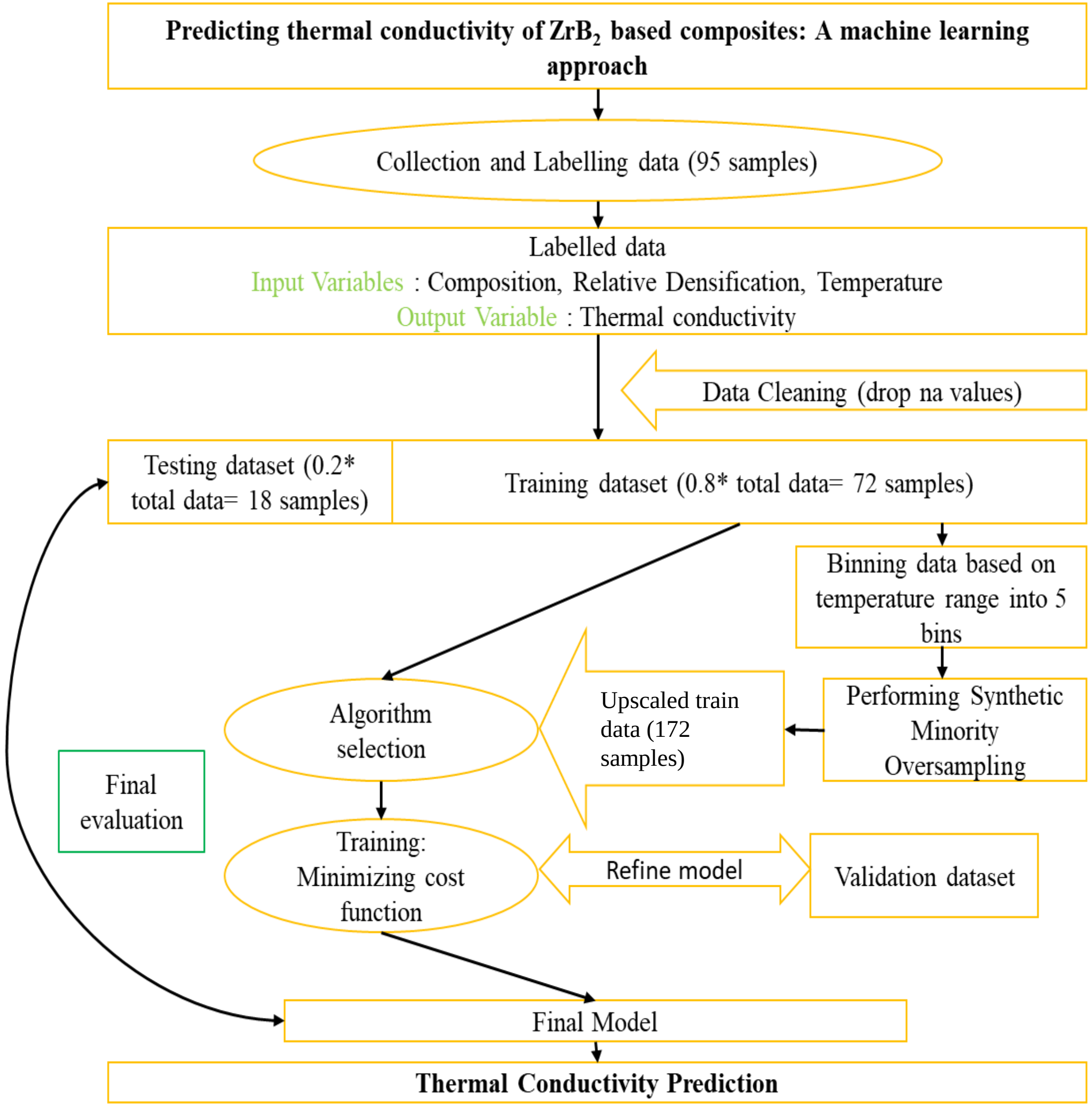
Understand the correlation between compositions, microstructure properties and thermal conductivity & oxide scale thickness.

Deduce which parameters from above mentioned affect the thermal conductivity and oxide scale the most statistically.

Machine Learning : Tenth hour hero

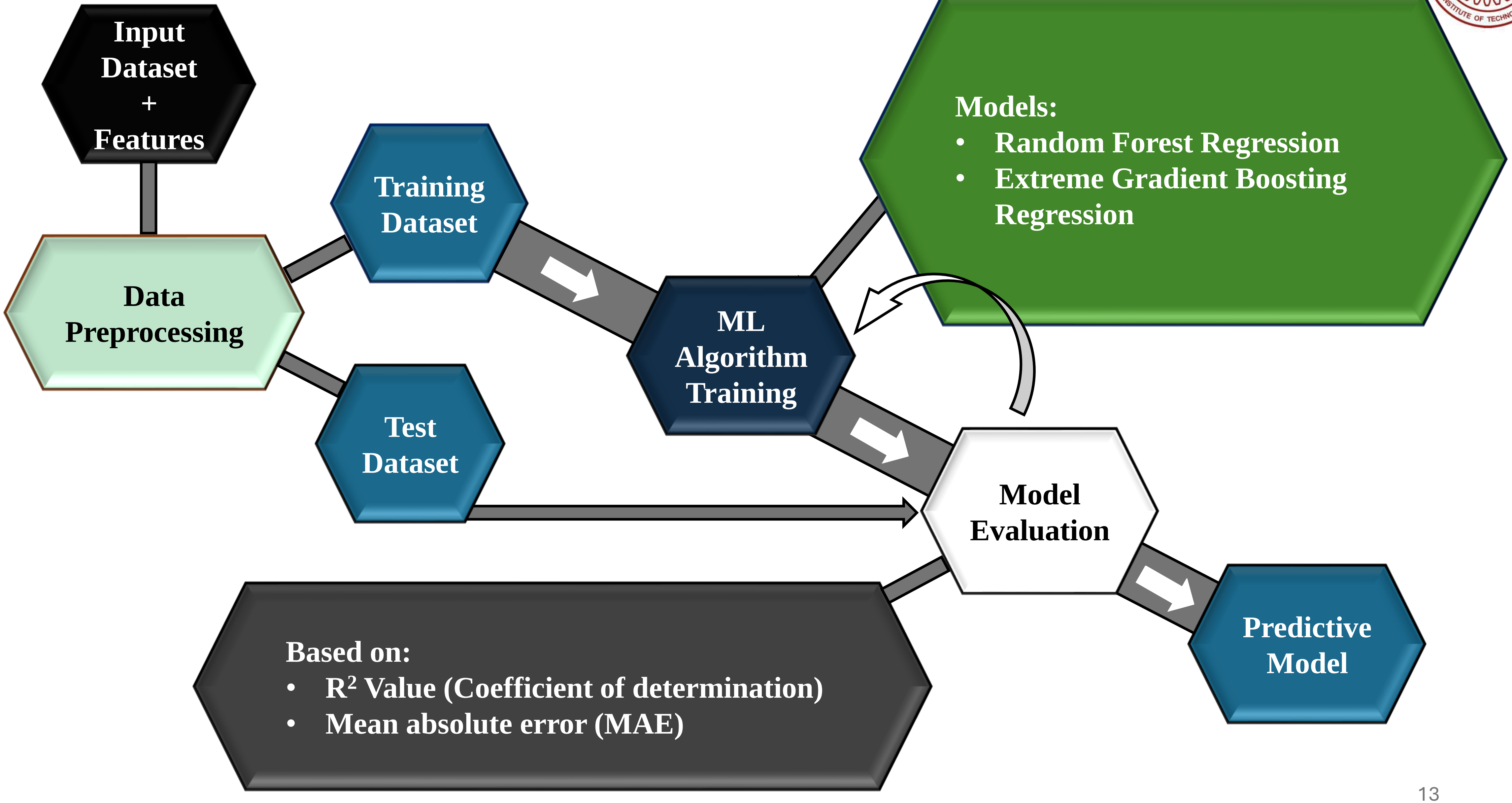
Author	Property predicted	Model used	Description
Antonio Vinci et al	Flexural Strength	Random Forest, Regression Tree	Key independent variables: SiC content, fiber volume, SiC/fiber ratio, ceramic matrix volume, geometrical porosity
Antonio Vinci et al	Fracture Toughness	Random Forest	Most influential variable: SiC/fiber ratio
Han et al	Young's Modulus, Flexural Strength, Fracture Toughness	Random Forest, Multilayer Perceptron	Material properties predicted based on chemical composition, sintering parameters, testing temperature, relative density, grain size
Bianco et al	Oxide Scale Thickness	Random Forest Regressor	Design elements and simulated environmental factors

Work Plan



Predicting thermal conductivity of ZrB_2 based composites: A machine learning approach
Collection and Labelling data (95 samples)
Labelled data
Input Variables : Composition, Relative Densification, Temperature
Output Variable : Thermal conductivity
Data Cleaning (drop na values)
Testing dataset (0.2* total data= 18 samples)
Training dataset (0.8* total data= 72 samples)
Binning data based on temperature range into 5 bins
Performing Synthetic Minority Oversampling
Upscaled train data (172 samples)
Algorithm selection
Final evaluation
Training: Minimizing cost function
Refine model
Validation dataset
Final Model
Thermal Conductivity Prediction

Machine Learning Methodologies



Input feature list

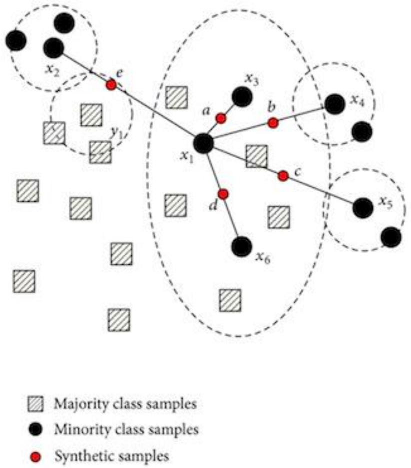
Feature list

- **Compositional features**
  - ZrB_2,SiC, ZrC, $MoSi_2$, $TaSi_2$, $ZrSi_2$
- **Microstructure features**
  - Grain size, Relative density
- **Testing parameters**
  - Temperature, time

Construction of dataset for thermal conductivity

INITIAL DATASET (90 compositions)

Constituents	Vol % Fractions	Relative Density %	Temperature °C	Thermal Conductivity $Wm^{-1}K^{-1}$
ZrB_2	100 0 0 0 0	85	25	25
ZrB_2, $MoSi_2$	70 30	98.8	25	18.421
ZrB_2, ZrC	50 50	100	900	81
ZrB_2, SiC	70 30	99.6	1330	51
ZrB_2, SiC, ZrC	55 20 20	98	25	82
ZrB_2, SiC, ZrC	51 33 16	96.5	25	60



Testing temperature is divided into 4 - bins for each composition.

51 points → T < 100 °C
6 points → 500 °C < T <=1000 °C
9 points → 1000 °C < T <=1500 °C
24 points → T > 1500 °C

IMBALANCED DATASET

51 + 6 + 9 + 24 = 90 Compositions

https://www.datacamp.com/tutorial/diving-deep-imbalanced-data

Construction of dataset for oxide scale thickness

INITIAL DATASET (84 compositions)

Constituents	Vol % fractions	Relative Density %	Mean Grain Size µm	Temperature °C	Time mins	Oxide thickness µm
ZrB_2 SiC	70 30	97.9	7	1200	720	25
ZrB_2 SiC	100 0	67.6	5.84	1300	33.33	69
ZrB_2 SiC	70 30	97.5	3.88	1700	300	229
ZrB_2 SiC	80 20	98.3	2.10	1100	60	15

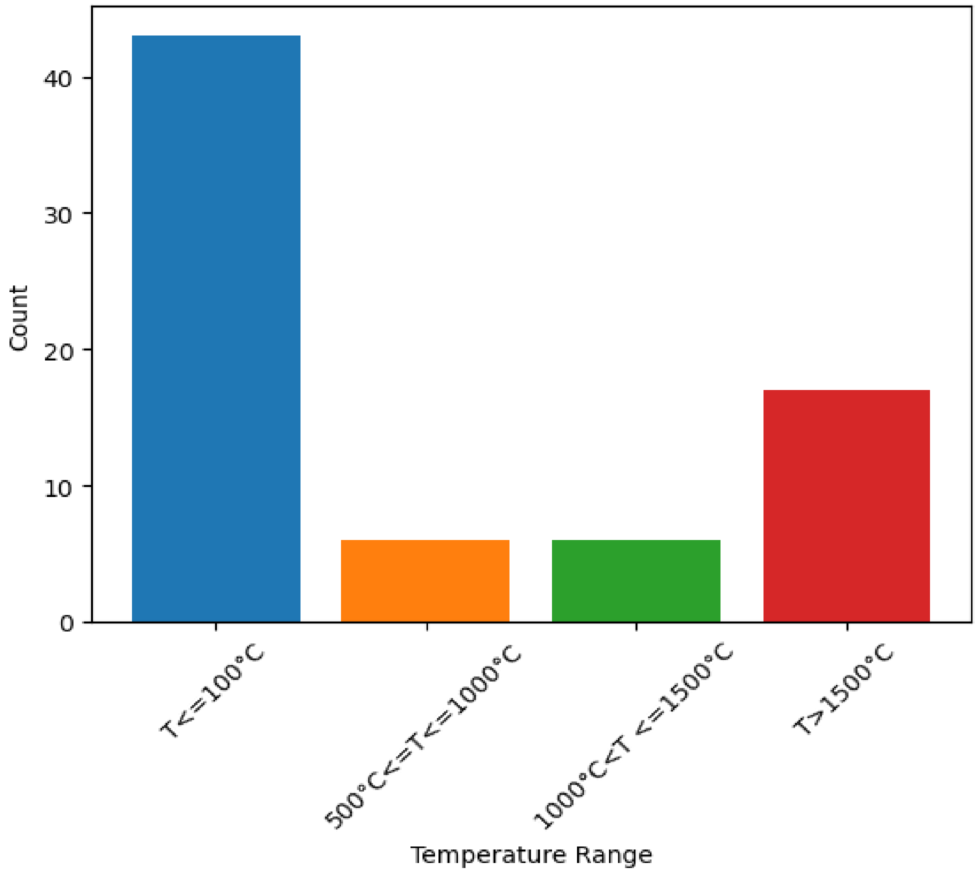
Testing temperature is divided into 4 - bins for each composition.

44 points → 800<=T < 1200 °C
14 points → 1200 °C < T <=1600 °C
9 points → T > 1600 °C

IMBALANCED DATASET

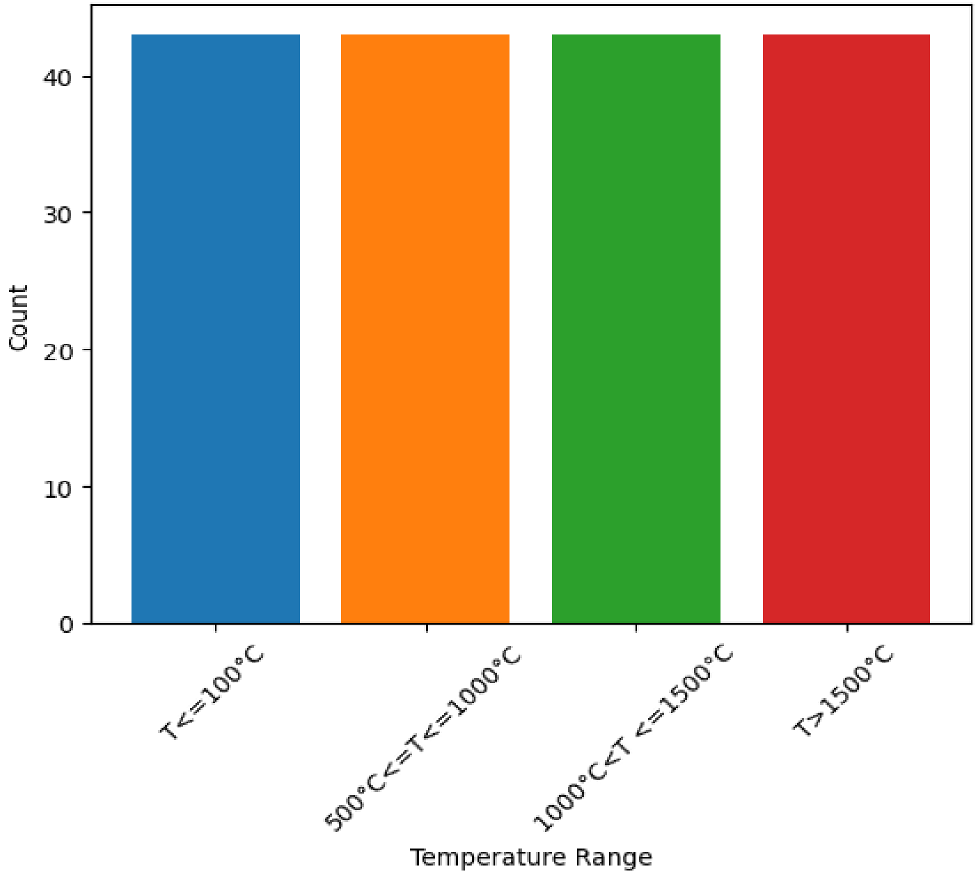
51 + 6 + 9 + 24 = 90 Compositions

Balancing the dataset for thermal conductivity



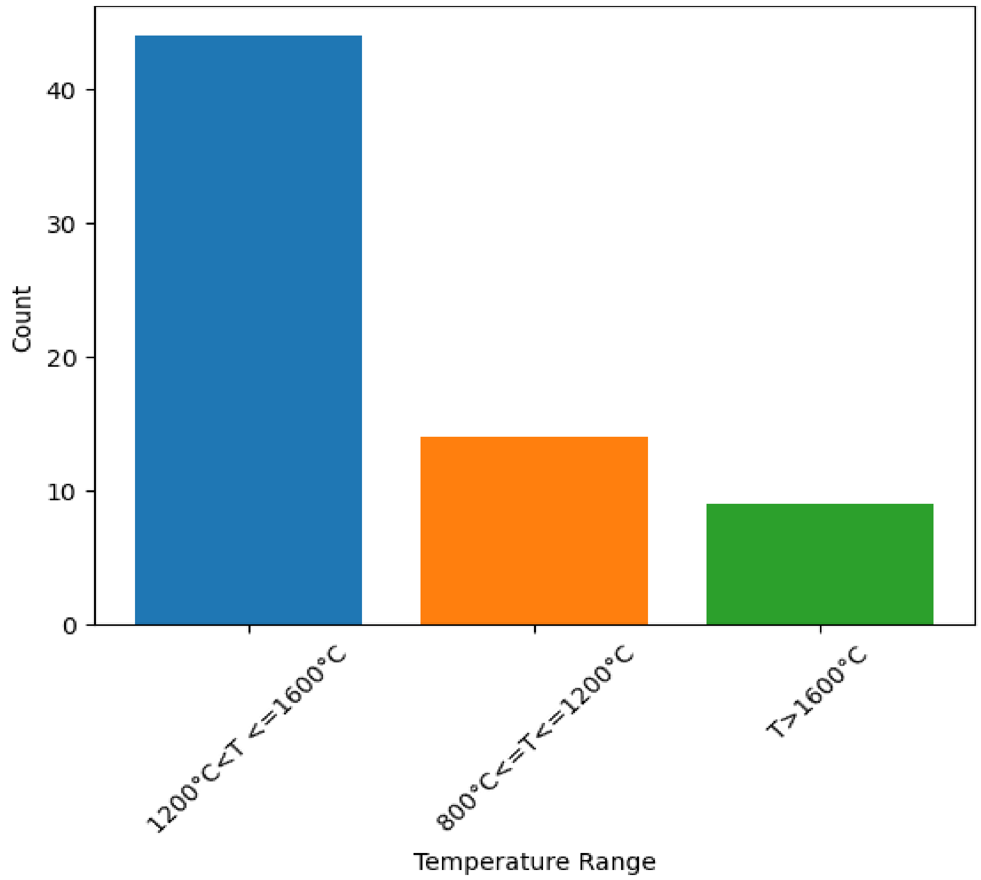
72 compositions

SMOTE



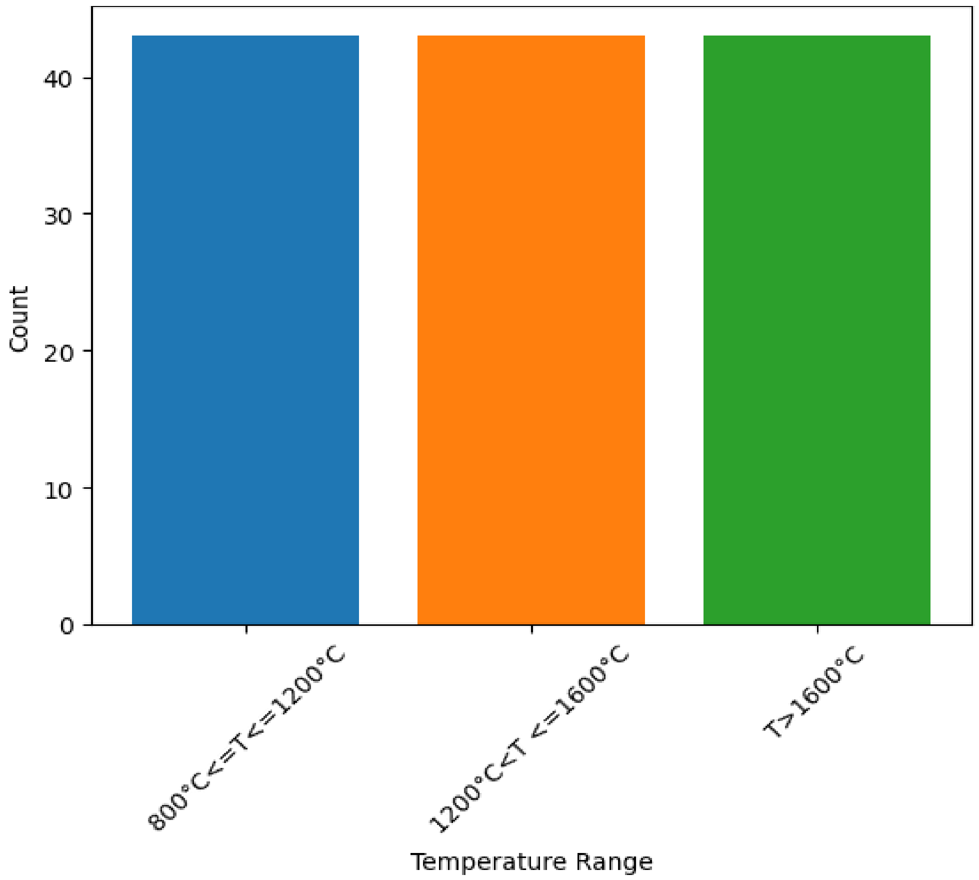
172 compositions

Balancing the dataset for oxide scale thickness



67 compositions

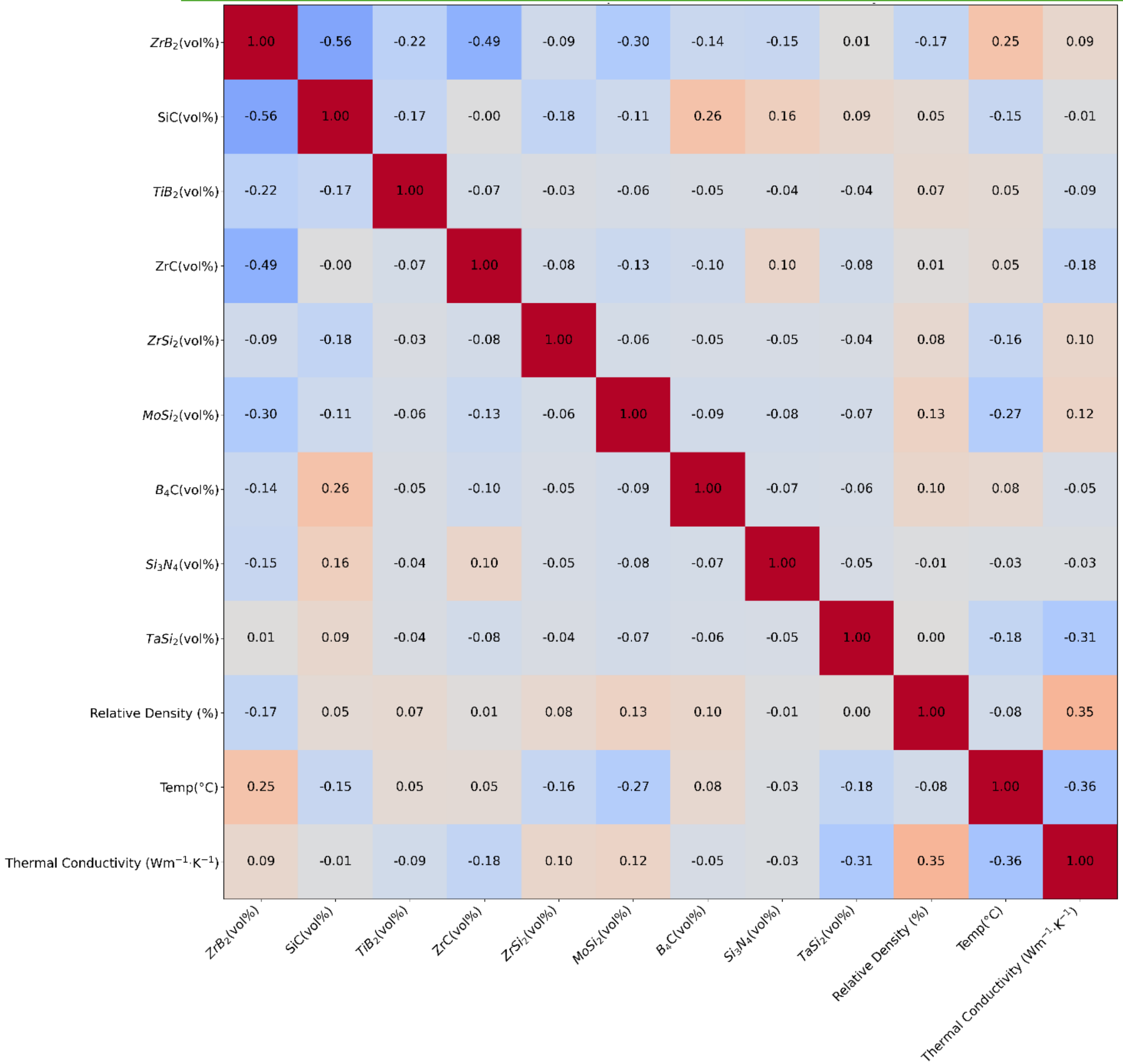
SMOTE



129 compositions

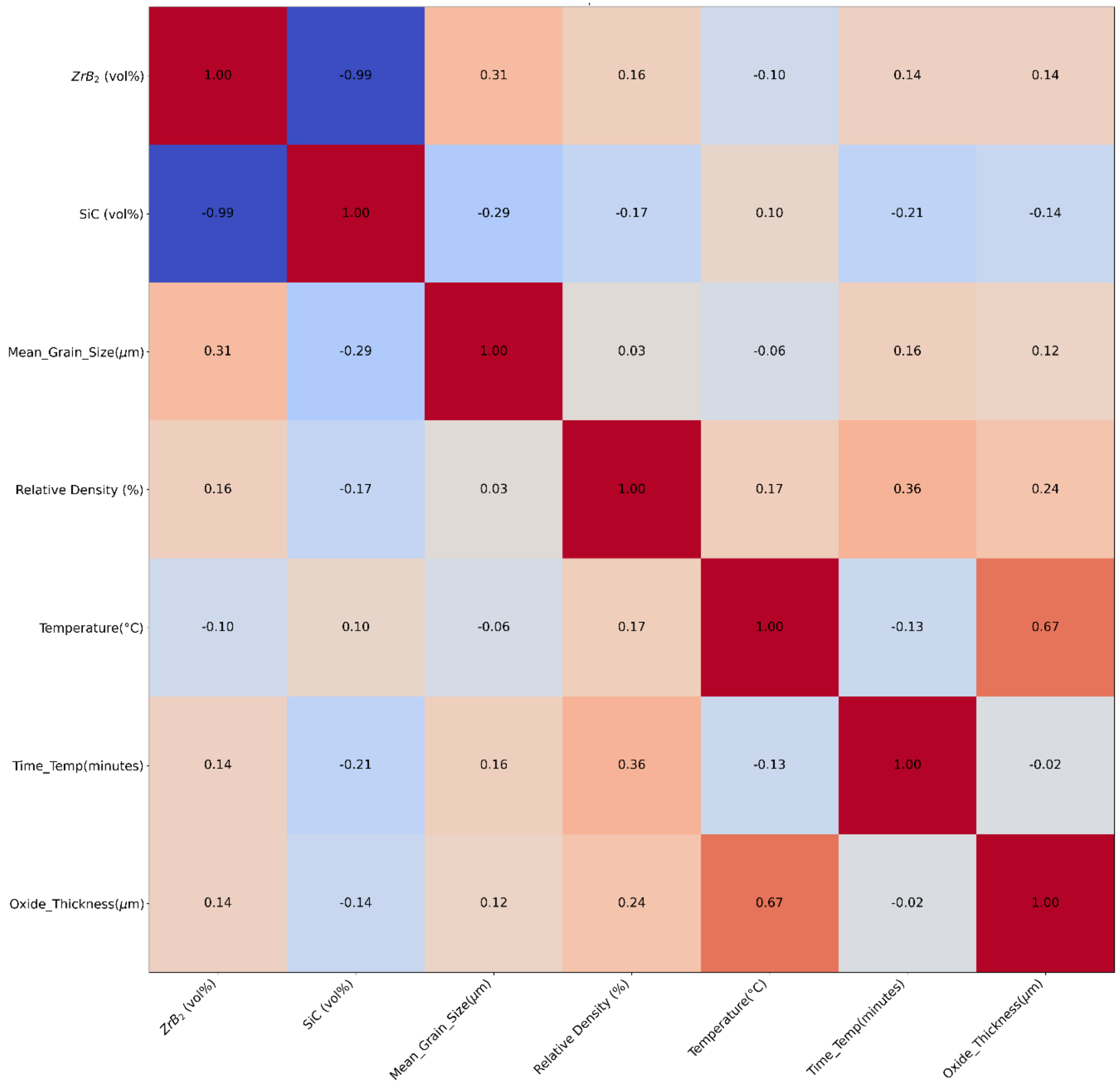
Results : Machine Learning Findings and Analysis

Correlations : Thermal conductivity



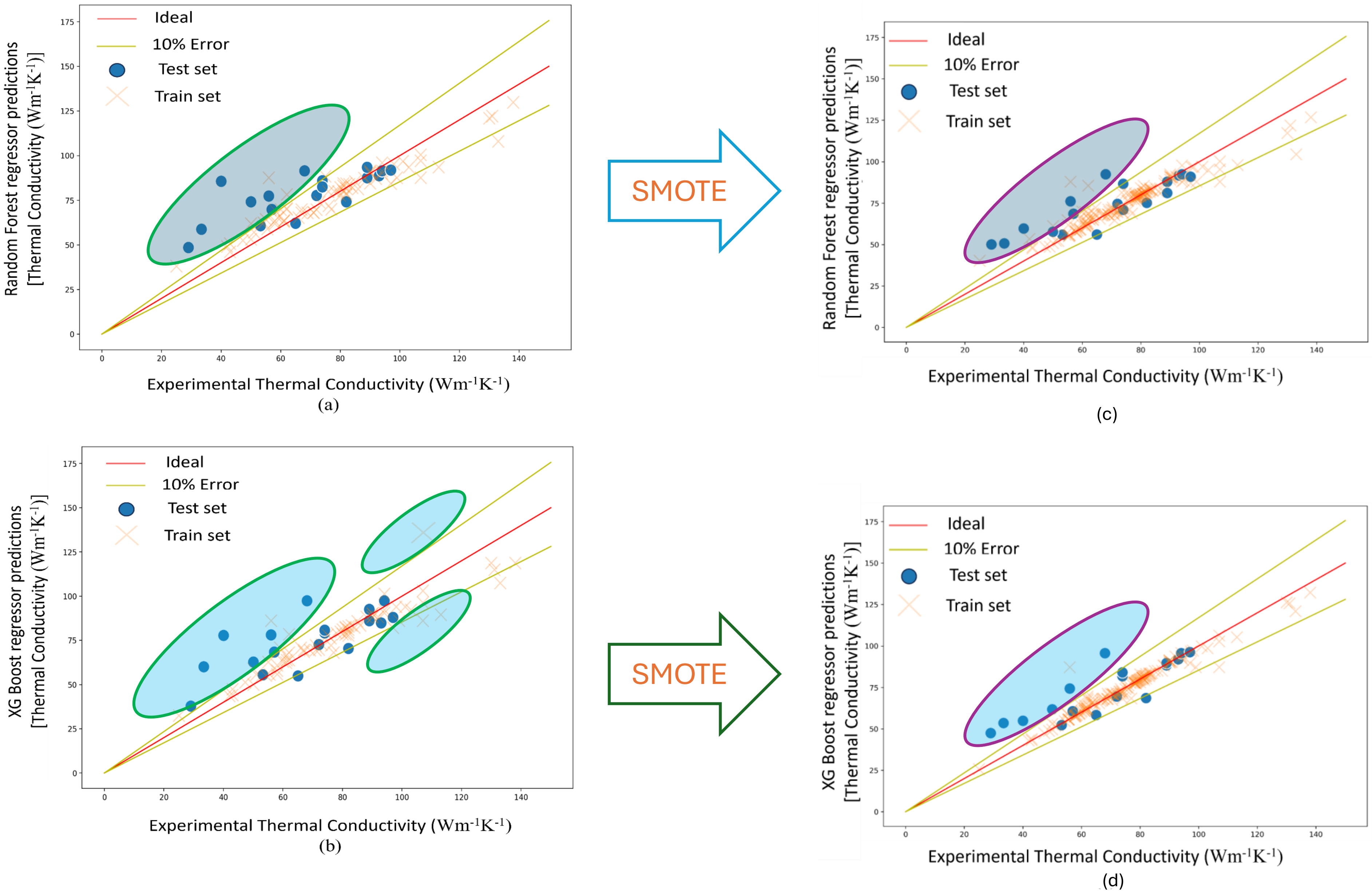
Parameters	Correlation with Thermal Conductivity
Temperature	-0.36
$TaSi_2$	-0.31
ZrC	-0.18
TiB_2	-0.09
B_4C	-0.05
Si_3N_4	-0.03
SiC	-0.01
ZrB_2	0.09
$ZrSi_2$	0.09
$MoSi_2$	0.12
Relative Density	0.35

Correlations : Oxide scale thickness

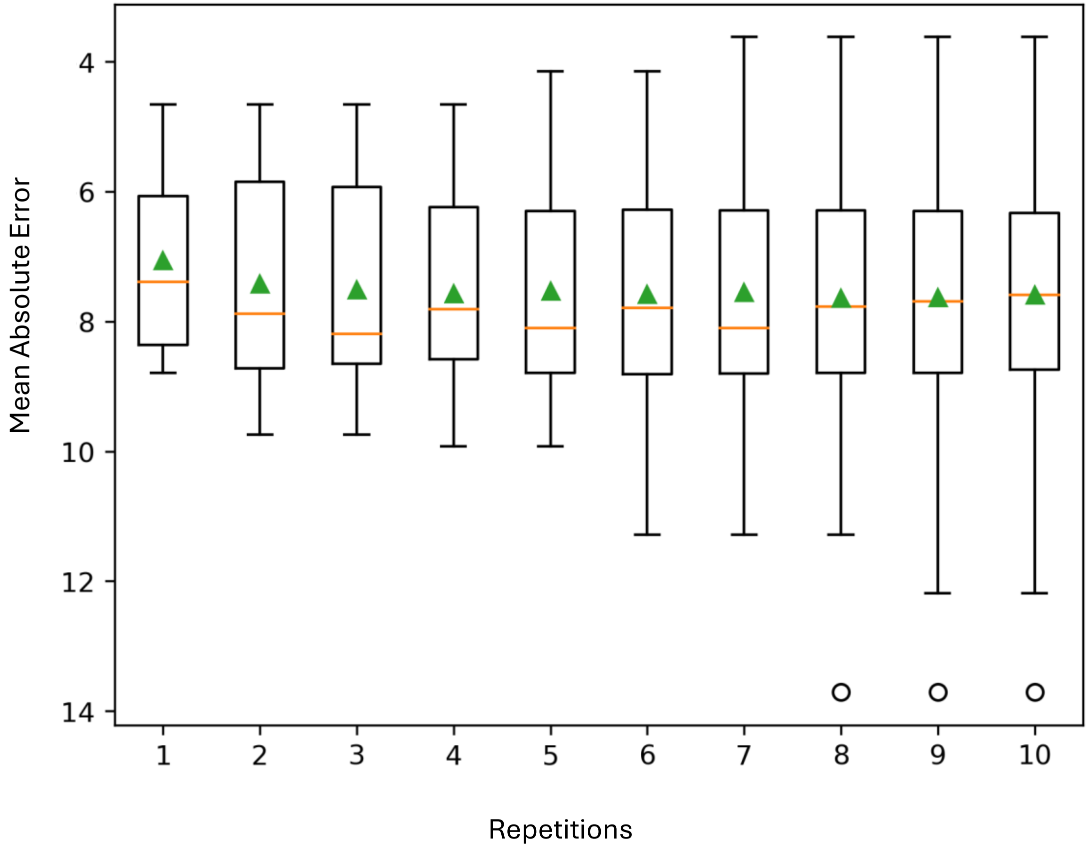


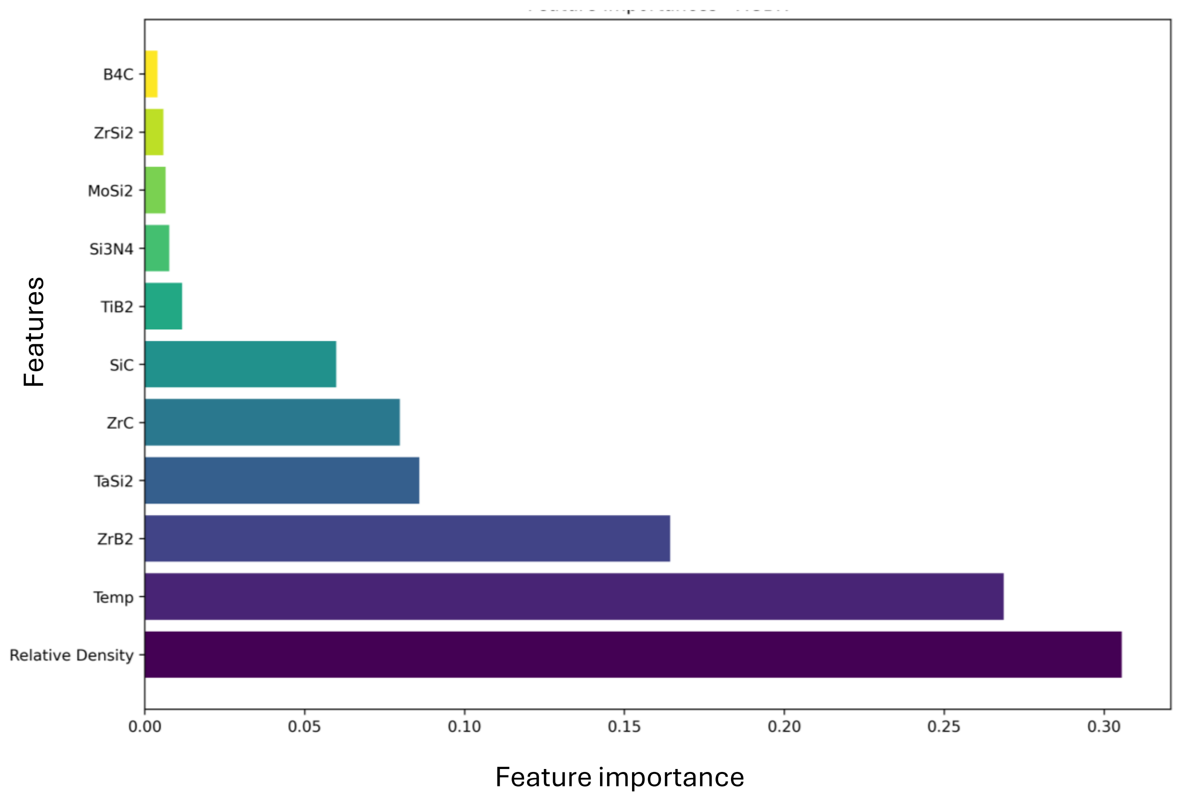
Parameters	Correlation with oxide scale thickness
Temperature	0.67
Relative Density	0.24
ZrB_2	0.14
Mean Grain Size	0.12
Time Temp	-0.02
SiC	-0.14

Thermal conductivity model results

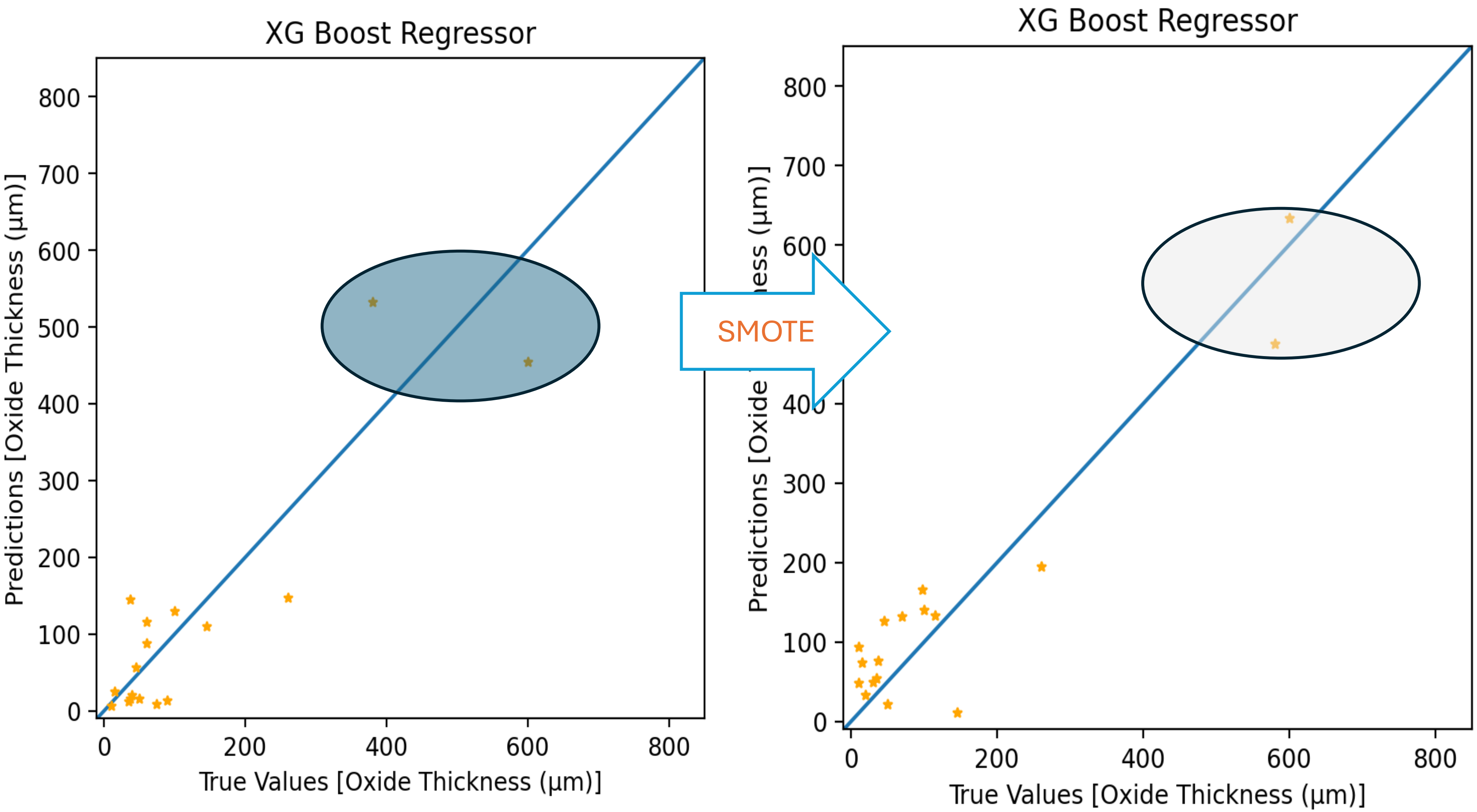


Thermal conductivity model results

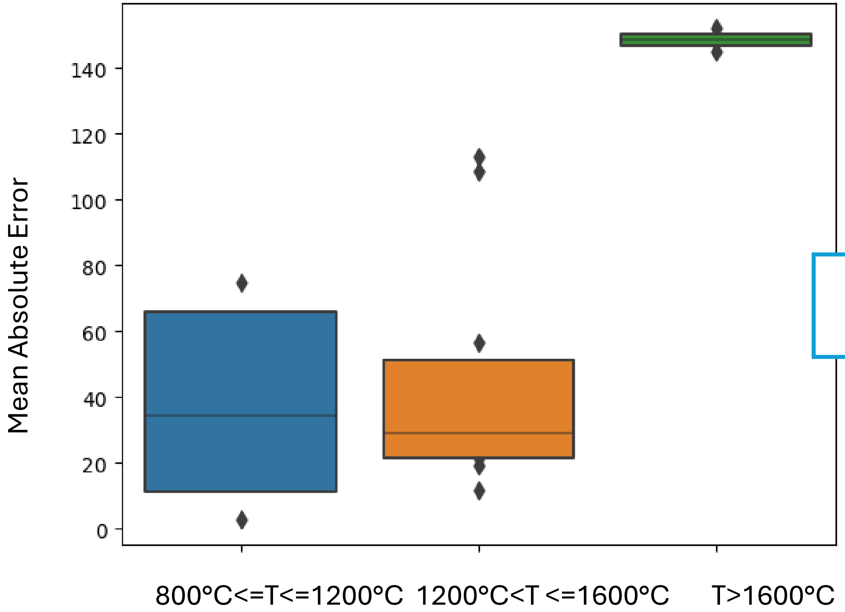




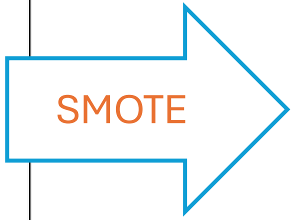
Oxide scale thickness model results

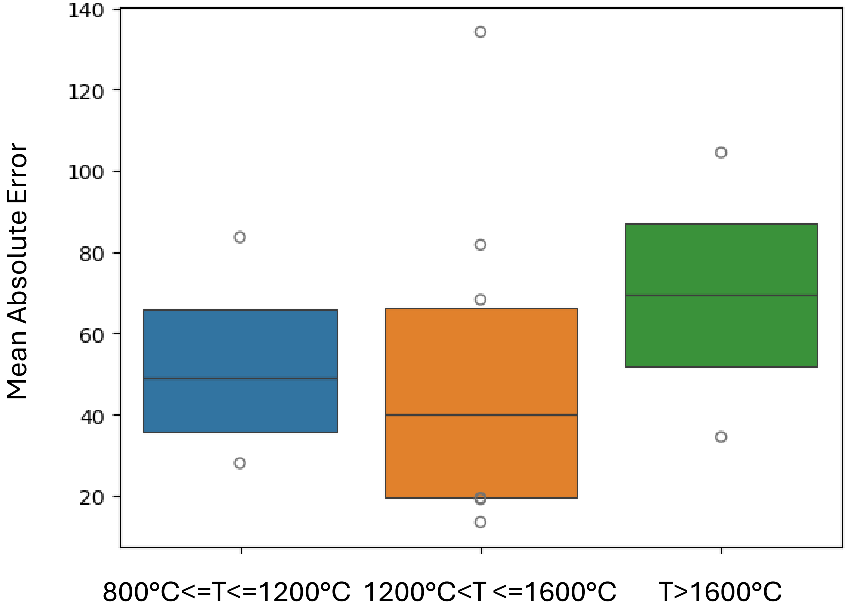


Oxide scale thickness model results



Box plots for mean absolute error from XGBR model for oxide thickness prediction before SMOTE



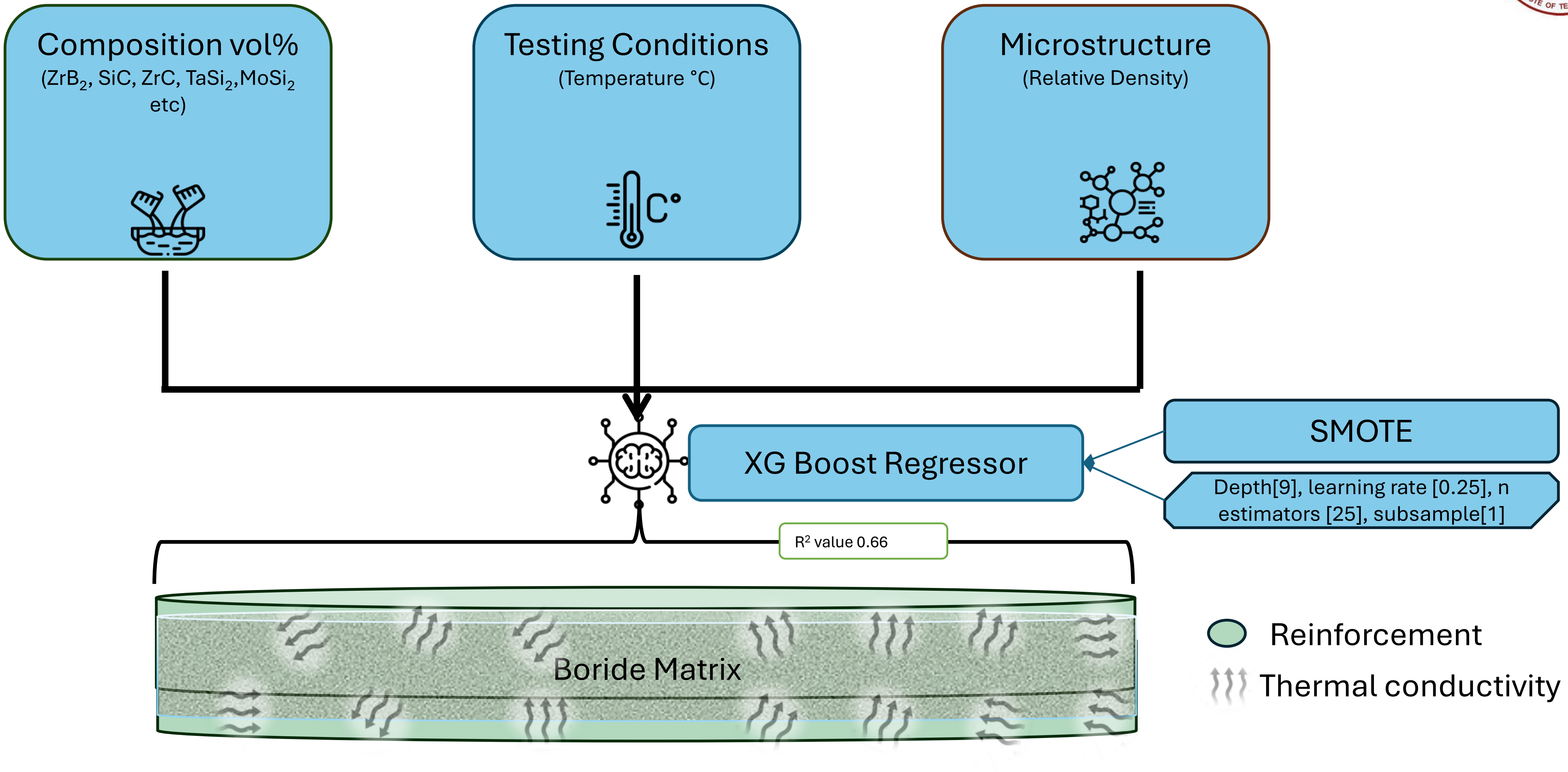


Box plots for mean absolute error from XGBR model for oxide thickness prediction after SMOTE

Range	MAE µm
800°C<=T<=1200°C	37.70
1200°C<T <=1600°C	44.51
T > 1600°C	148.52

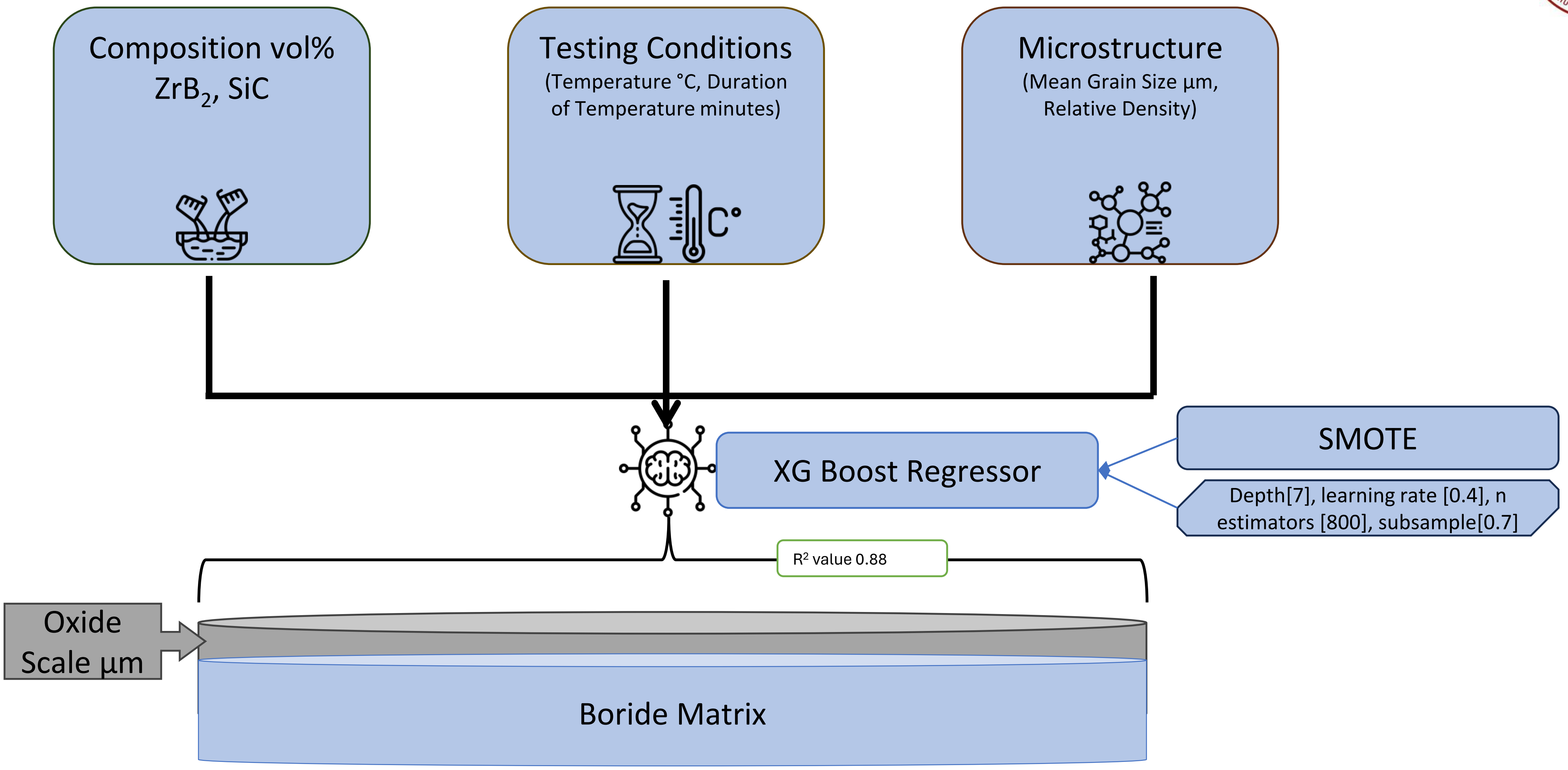
Range	MAE µm
800°C<=T<=1200°C	52.47
1200°C<T <=1600°C	51.11
T > 1600°C	69.42

Summary



Schematic illustration summarizing the overview of thermal conductivity

Summary



Schematic illustration summarizing the overview of oxide scale thickness

Conclusion

- In this work we established a data driven model to predict thermal conductivity and oxide scale thickness in ZrB_2 composites.

- The model achieved reasonable accuracy (MAE of 8.95 $Wm^{-1}K^{-1}$ and R-squared of 0.66 for thermal conductivity) despite limitations in the data.

- Key factors affecting thermal conductivity are relative density, temperature and ZrB_2 content.

- Like thermal conductivity, oxide scale thickness prediction is improved using the XGBoost model (R-squared value increased from 0.83 to 0.88).

- Key factors affecting oxide scale thickness are temperature and SiC content (positive and negative correlation respectively).

Future scope

Due to limited time, some extensions of the current work is suggested:

- Current models utilization of experimentally derived parameters, modeling argument is weakened due to this gap, especially evident in predicting oxide scale thickness using mean grain size.
- First step in synergizing the experimental and computational modelling has yet to be explored in UHTC space.
- Due to its efficiency, combining, experimental design with Bayesian optimization (ML model built on initial experimental data and acquisition function which drives the search for exploring and exploiting) can benefit on designing the target property materials.

Adv. Mater.2024,36, 2304269

Acknowledgement

I want to extend my sincere thanks to:

- Prof. Kantesh Balani (for giving me another chance)

- Lab members and staff of Biomaterials Processing and Characterization lab

- Members of the Materials Science and Engineering (MSE) Department and PK Kelkar Library staff for their all-round help and support.

- Special thanks to Dr. Rubia Hassan for helping me with initial problem formulation, thesis structuring and Dr. Shipra Bajpai for thesis writing.

- To my incredible friends and my ever-supportive mom for unwavering faith in me.

90 **Th** Thorium 232.03806	79 **Au** Gold 196.966569	7 **N** Nitrogen 14.0067	19 **K** Potassium 39.0983

39 **Y** Yttrium 88.90585	8 **O** Oxygen 15.9994	92 **U** Uranium 238.02891