Predicting Magnetic Saturation (Bs) and Curie Temperature (Tc) in Magnetic Alloys

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Outline

- Data Utilization: Composition and processing parameters of soft magnetic alloys
- Machine Learning Models: Ridge Regression, SVM, CatBoost, GPR, KNN, Dense Neural Networks
- Prediction Results: Model performance, RMSE metric
- Multi-Objective Optimization: Pareto front analysis
- Bayesian Optimization: Inverse design and property maximization
- Uncertainty Quantification: Bayesian strategy, posterior distribution analysis
- Constrained Optimization: Refined optimization, Pareto front with constraint

Inverse design workflow

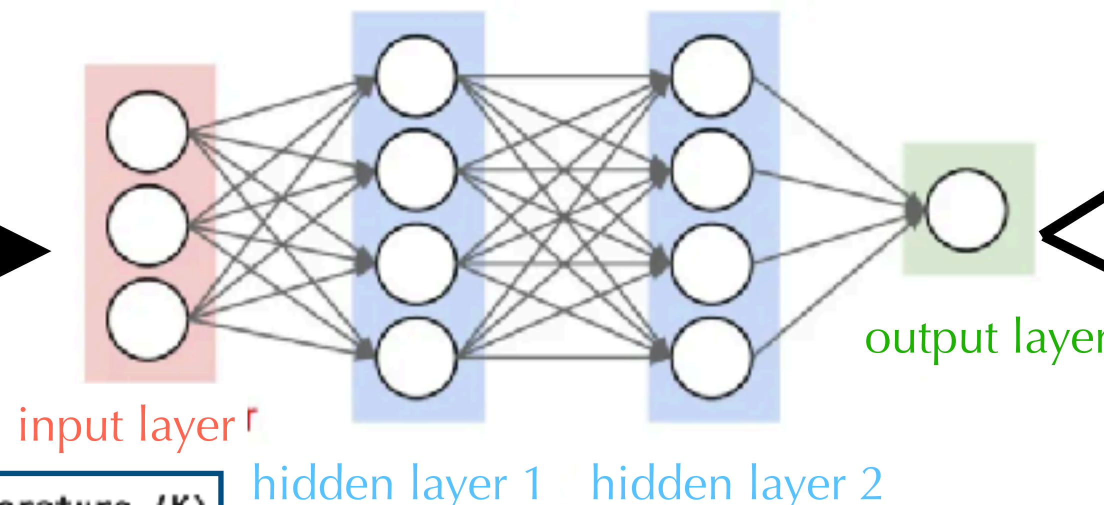
Al	B	C	Co	Cr	...
0.0	15.0	0.0	0.5	0.0	...
0.0	4.0	0.0	0.0	0.0	...
0.0	3.0	0.0	0.0	0.0	...
0.0	15.0	0.0	0.5	0.0	...

elemental compositions

processing parameters

Annealing Time (s)	Annealing Temperature (K)
0.0	0.00000
0.0	0.00000
3600.0	873.25952
1800.0	723.00000

Machine Learning (ML)

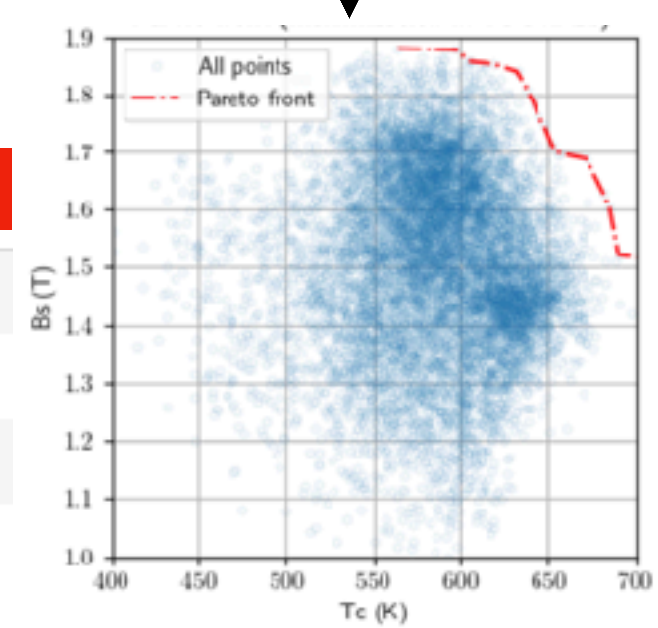


Magnetic Saturation (BS)

Predict Magnetic Properties

Curie Temperature (TC)

Bayesian Optimizer



optimized alloys

B11.8Cu0.7Fe81.7Si5.5
B9.0Cu1.4Fe83.0P1.0Si5.4
B7.5Co19.5Cu1.0Fe67.2P1.0Si4.0

- Combine ML and Bayesian optimization:
  Inverse design of magnetic alloys

Inverse design workflow

Contents lists available at ScienceDirect

Materials & Design

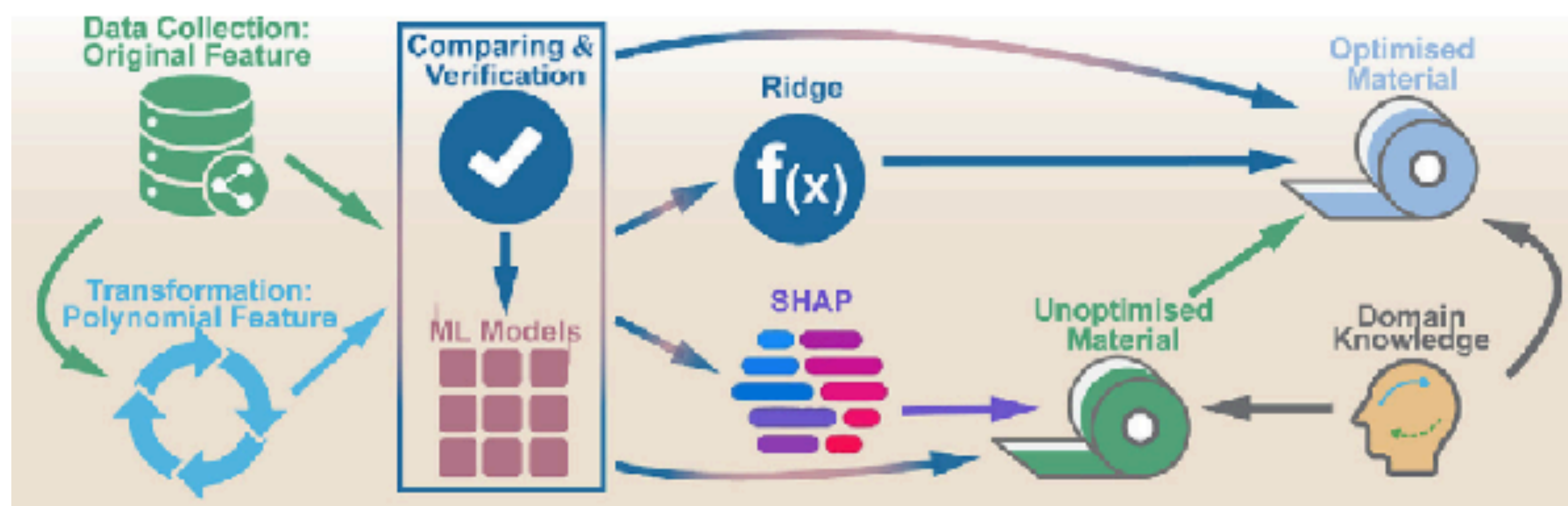
journal homepage: www.elsevier.com/locate/matdes

Accelerated discovery of Fe-based amorphous/nanocrystalline alloy through explicit expression and interpretable information based on machine learning

Bo Pang [a], Zhilin Long [b,c,*], Tao Long [a], Rong He [d], Xiaowei Liu [a], Mingwang Pan [d]

HIGHLIGHTS

- This work considers not only composition but also the effect of ribbon thickness and annealing on B_s and T_c.
- The prediction accuracy of ML models, especially Ridge regression, has been significantly improved by transforming original features to polynomial features.
- Interpretable information of ML models based on SHAP method and explicit expressions is of great aid to researchers in alloy design.
- The predicted values of B_s and T_c are in good agreement with the measured ones in the previous literatures and an alloy $Fe_{81.4}Si_{3.4}B_{11.4}Cu_{0.8}Nb_3$ with desired properties is designed.



Bs = 1.8 T
Tc = 662 K

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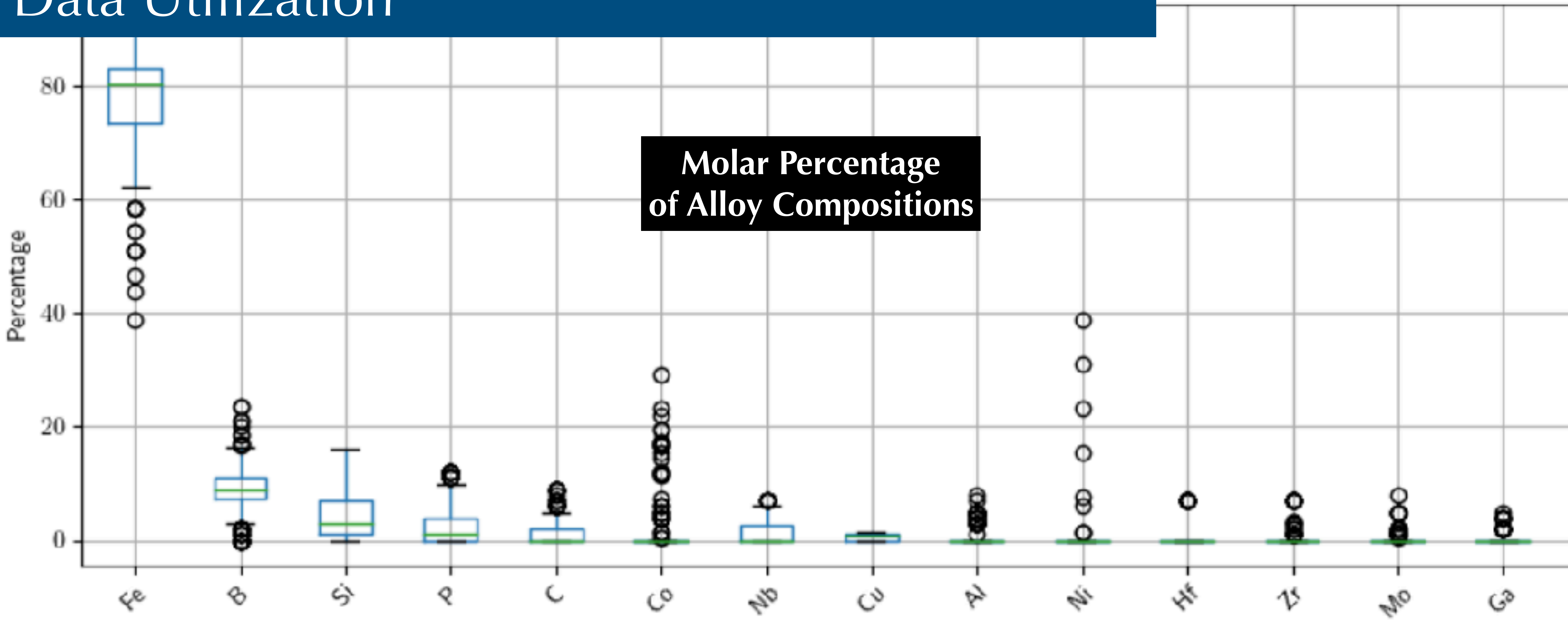
Data Utilization



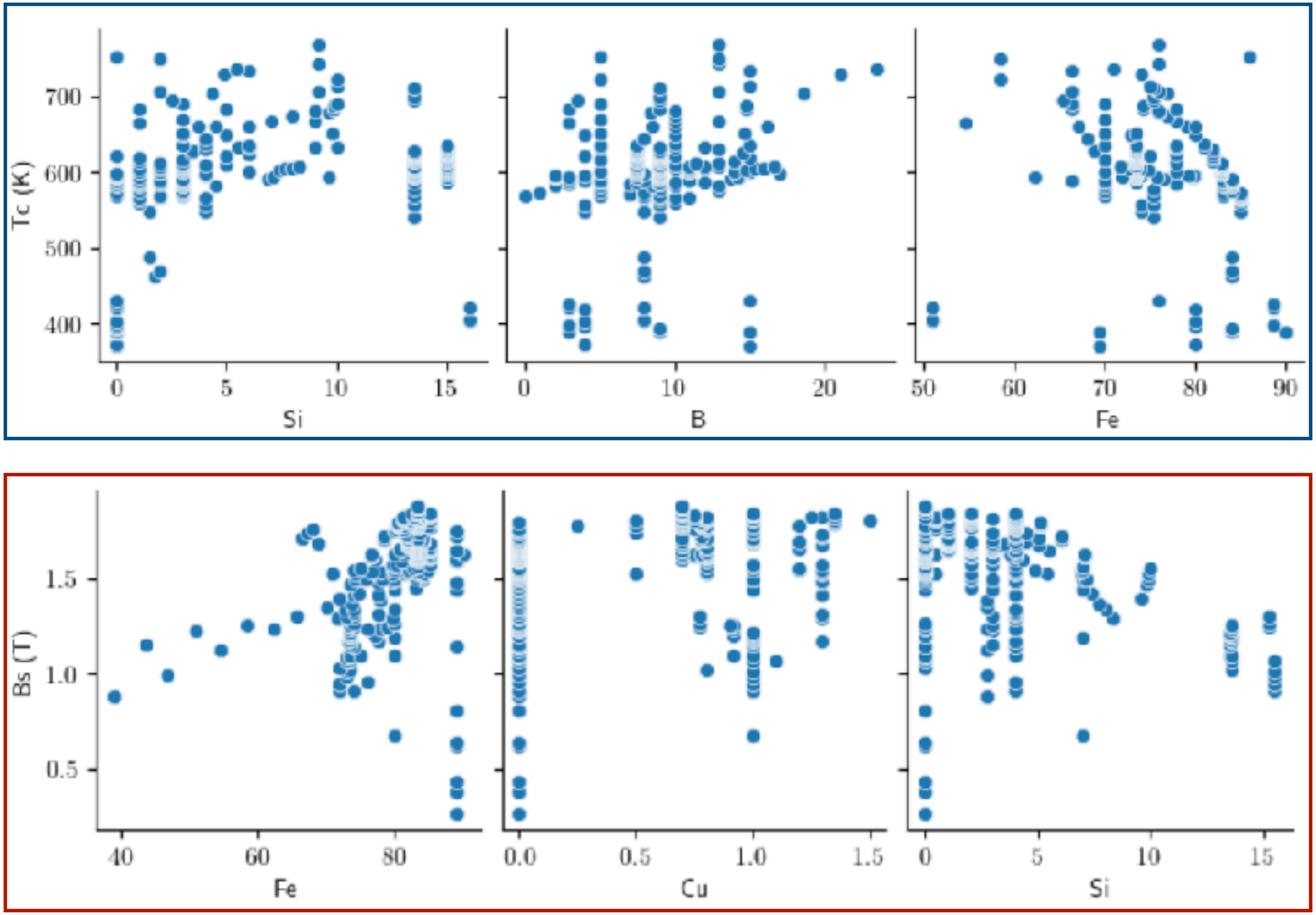
Dataset Details [1]

- Experimental Dataset (Fe-based magnetic alloys):
  514 data items, with 294 for Bs and 220 for Tc
- Features:
  Each data point includes 1 thickness, 2 annealing parameters, and compositional features

Composition	ta (s)	Ta (K)	Thickness (mu m)	Bs (T)
Fe89Hf7Zr1B3	1800	648.9	32.5	0.3
Fe89Hf7Zr1B3	1800	598.7	32.5	0.4
Fe89Hf7Zr1B3	1800	748.6	32.5	0.4
Fe89Hf7Zr1B3	1800	698.8	32.5	0.6
Fe89Hf7Zr1B3	0	0.0	32.5	0.6

[1] Pang, Bo, et al. *Materials & Design* 231 (2023): 112054.

Data Exploaration



Correlation features

- Opposite trends for Bs and Tc: increasing Fe enhances magnetic saturation but lowers Curie temperature, while increasing Si reduces Bs but improves Tc.

Features & Outputs

- **Features:** elemental compositions, processing parameters
- Targets: magnetic saturation (Bs) and Curie temperature (Tc)

$X =$

	29 Elemental features								3 Processing features		
	Al	B	C	Co	Cr	...	Pr	Sb	Annealing Time (s)	Annealing Temperature (K)	Thickness (mu m)
1	0.0	15.0	0.0	0.5	0.0	...	0.0	0.0	0.0	0.00000	20.0
2	0.0	4.0	0.0	0.0	0.0	...	0.0	0.0	0.0	0.00000	20.0
...	0.0	3.0	0.0	0.0	0.0	...	0.0	0.0	3600.0	873.25952	20.0
N	0.0	15.0	0.0	0.5	0.0	...	0.0	0.0	1800.0	723.00000	20.0

Feautures X **ML regressor** Targets $y =$

Tc (K)	Bs (T)
370.0	0.3
374.0	0.4
388.6	0.4
389.0	0.6

$$y = \mathcal{G}(X)$$

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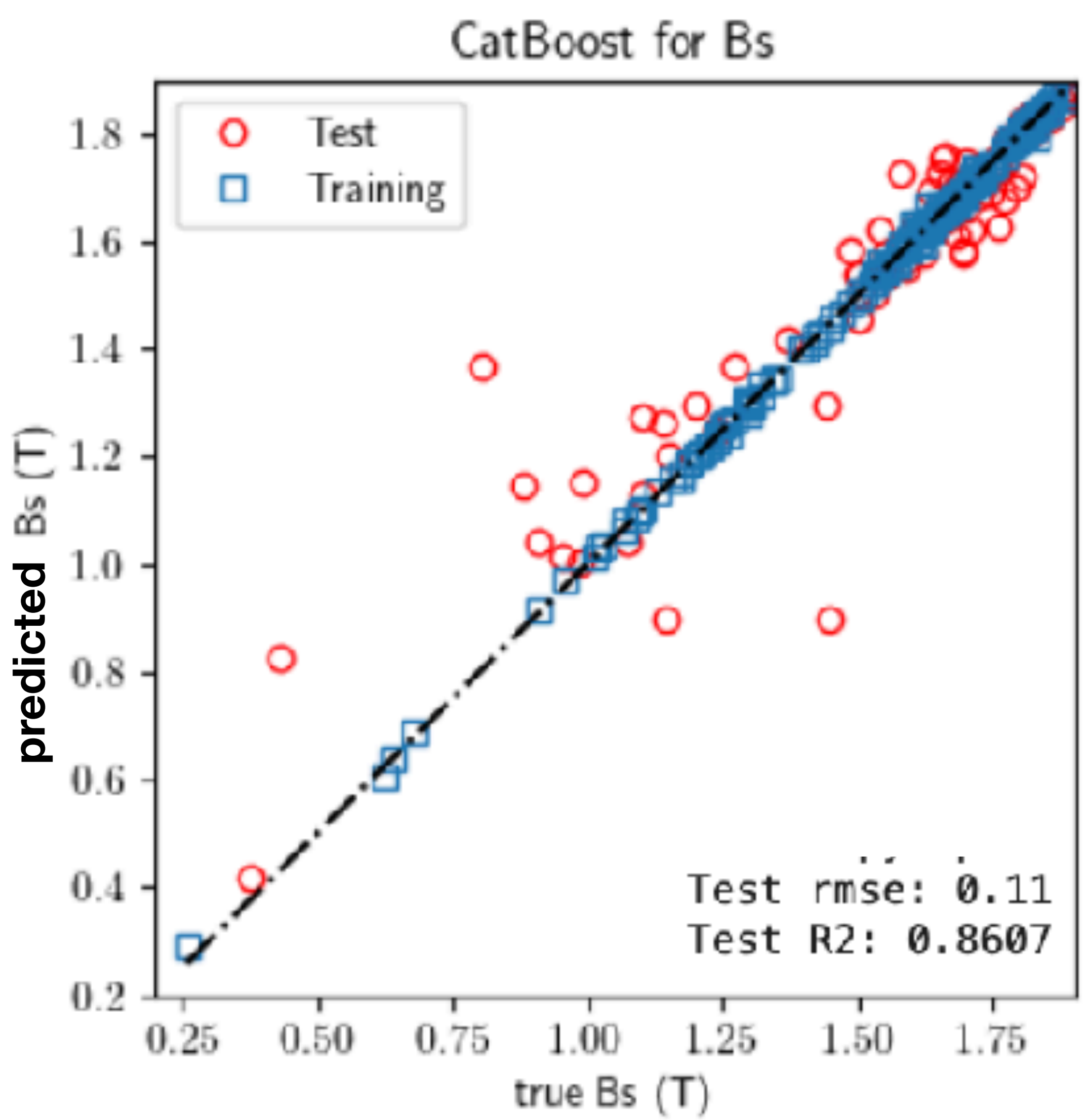
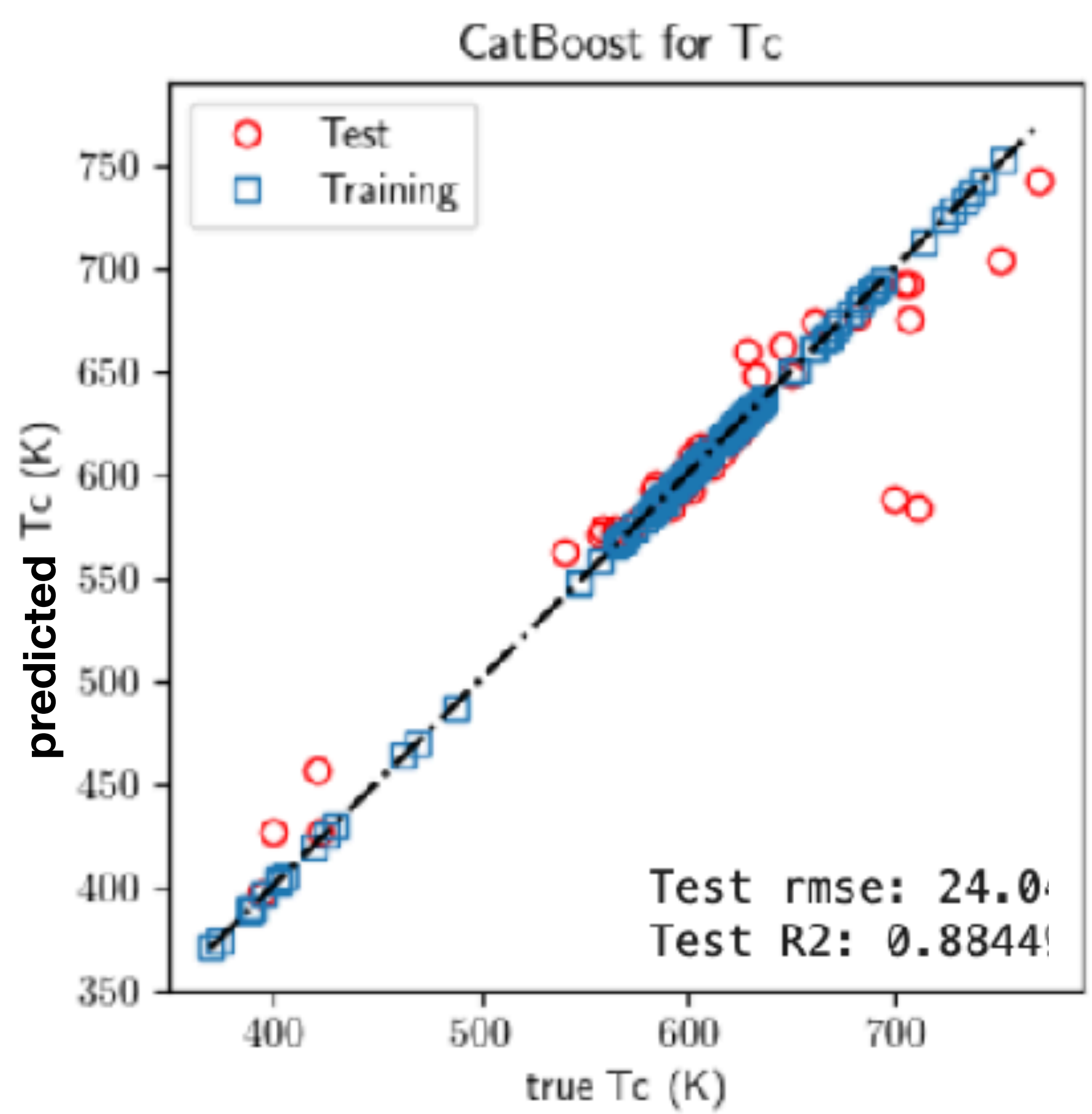
Machine Learning

- Models Used: Ridge, SVM, CatBoost, KNN, GPR, Neural Nets.
- Hyperparameter Tuning: GridSearchCV, cross-validation.
- Training Strategy: Train-test split, standardization, evaluation metrics.





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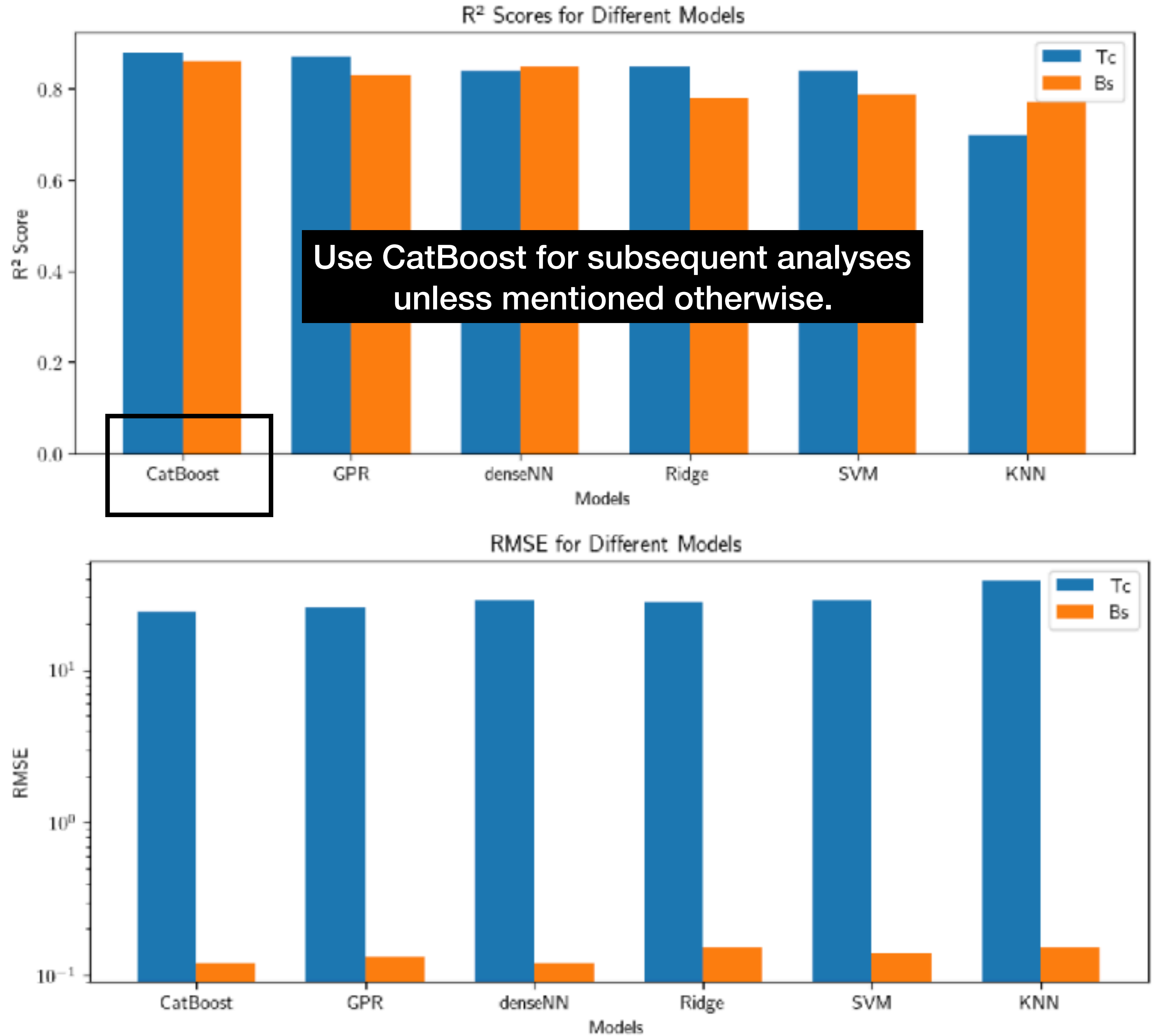
ML Performance

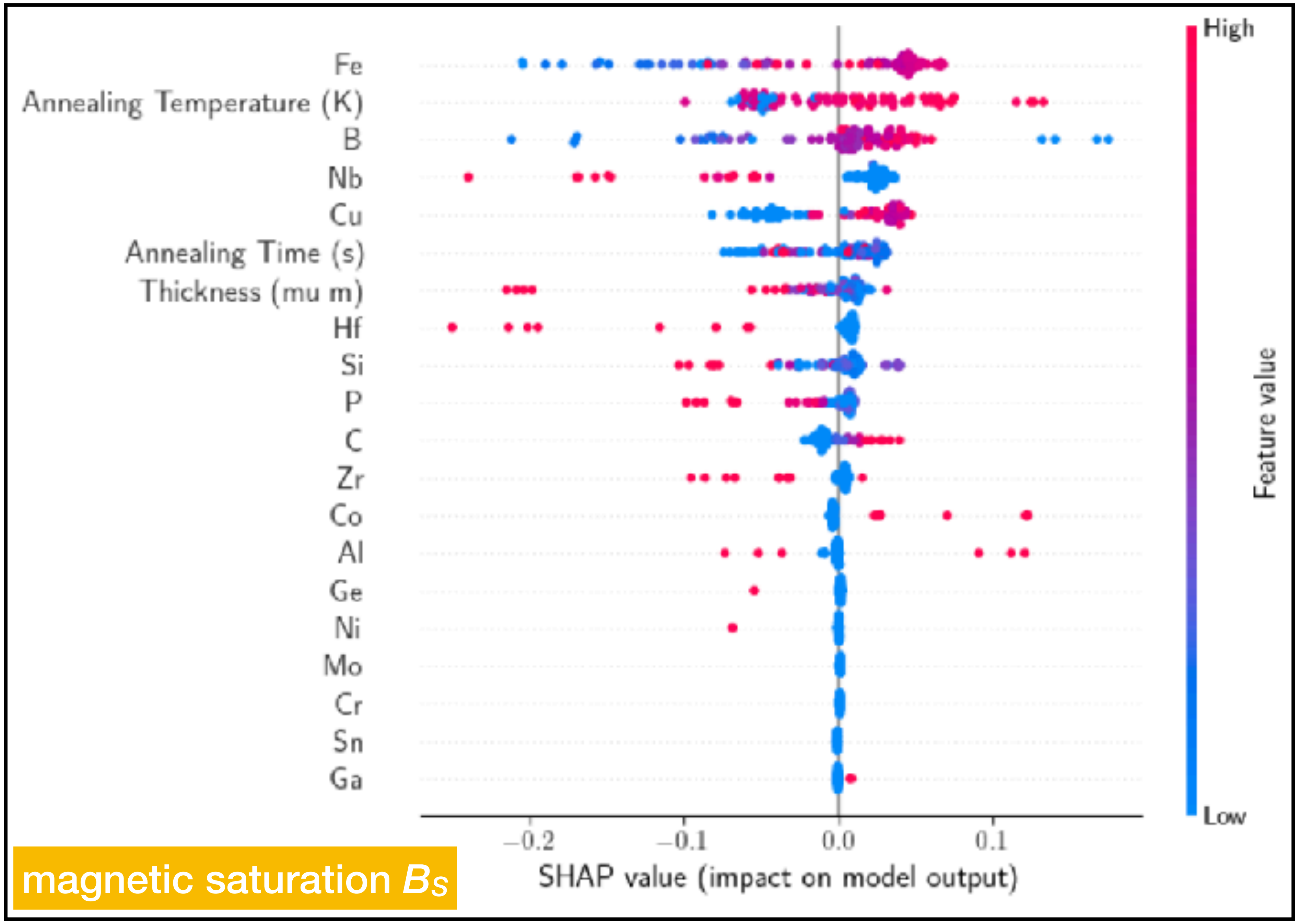


Use CatBoost for subsequent analyses unless mentioned otherwise.

Feature Importance

SHAP (SHapley Additive exPlanations) analysis

- Feature Importance: contribution of each feature to model predictions
- SHAP values: the direction and magnitude of influence



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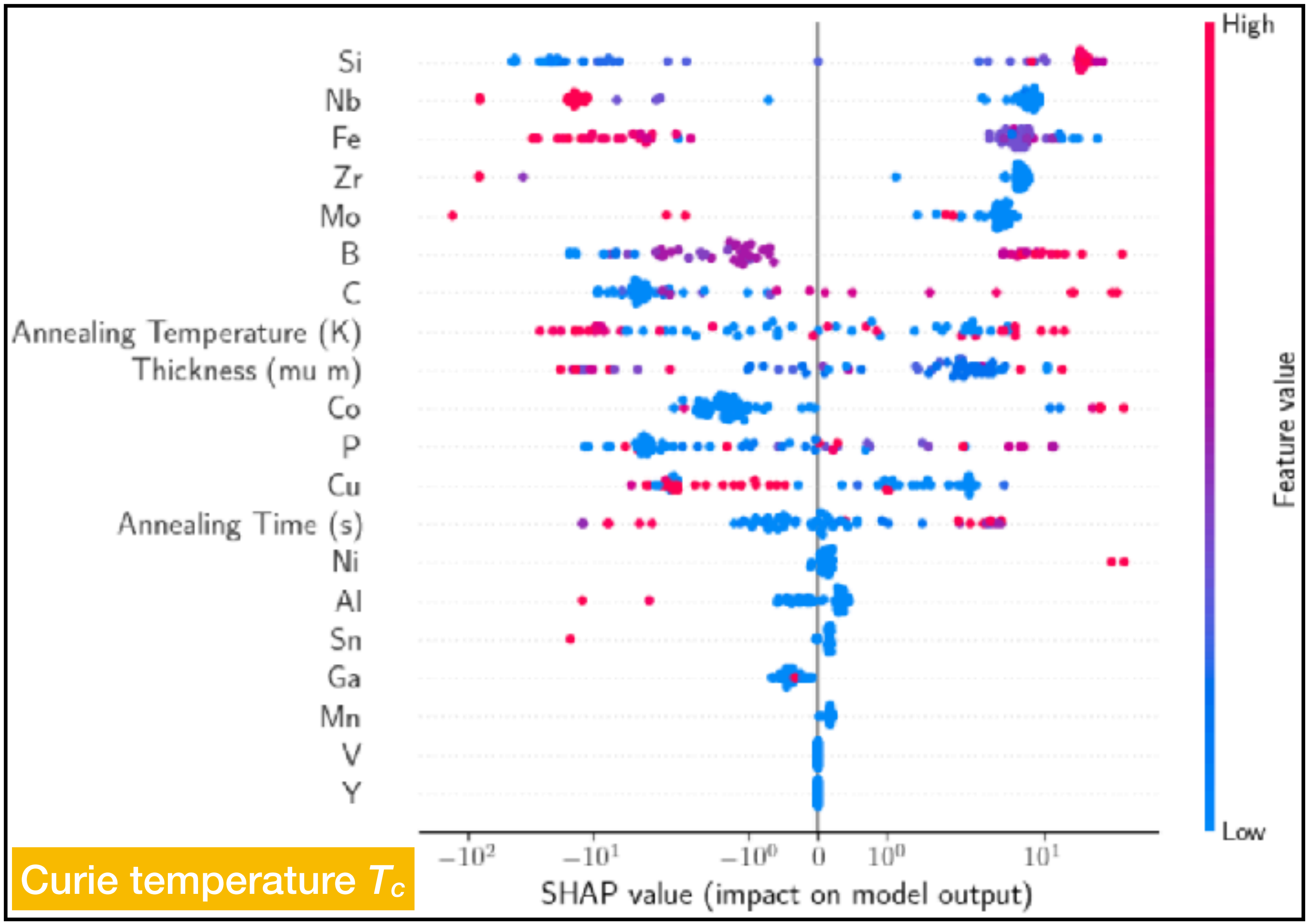

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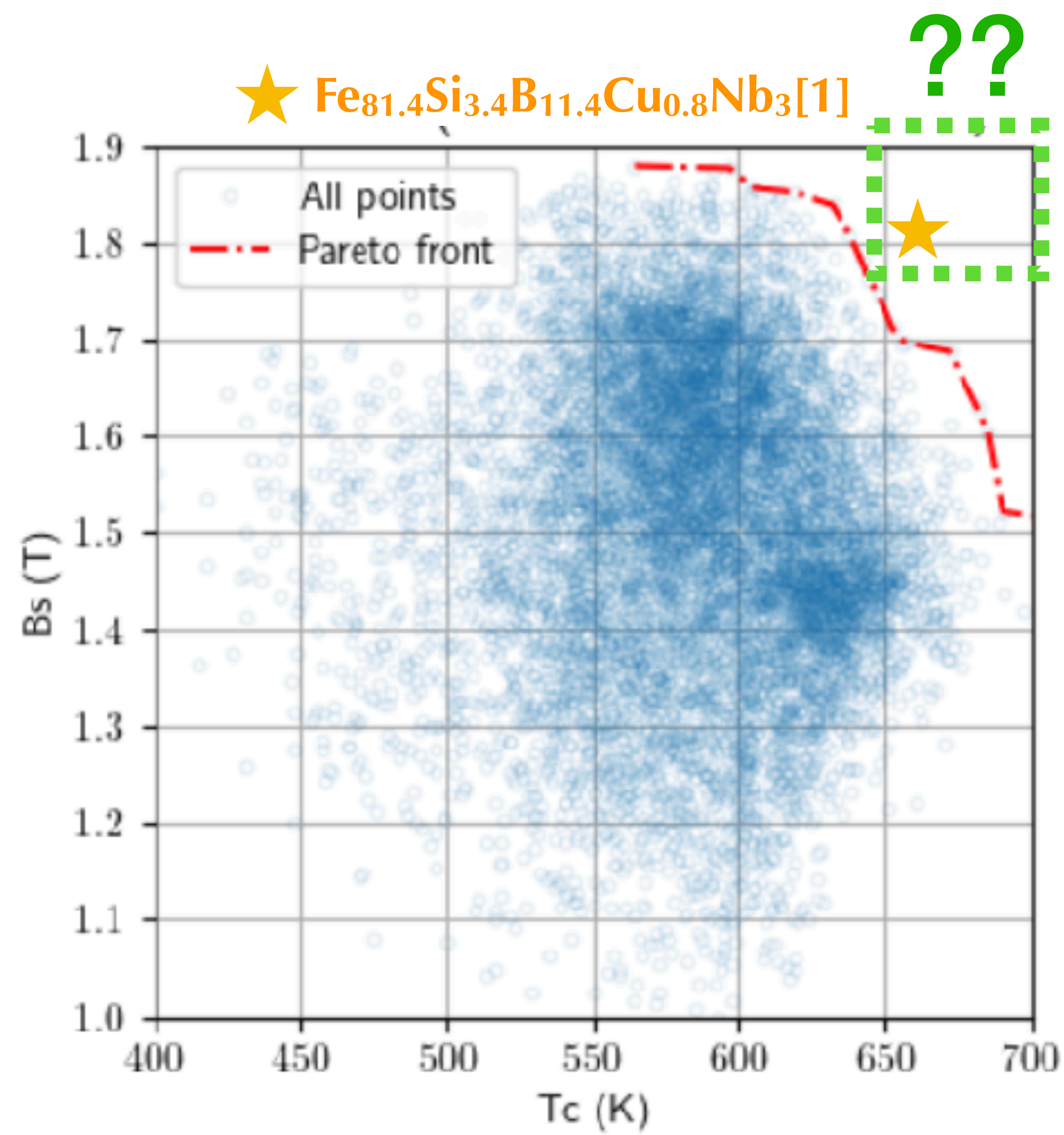
Pareto Front

compositions across the Pareto front	Tc (K)	Bs (T)
B9.0C4.0Fe83.0P1.0Si2.0	594.95	1.86
B13.0Fe82.0Si5.1	641.29	1.76
B7.5C5.0Cu1.0Fe82.7P4.0	559.67	1.86
B9.0C5.0Cu1.0Fe83.2P1.0Si1.0	574.75	1.86
B8.0C2.0Cu1.2Fe83.0P3.0Si3.0	607.87	1.85
B8.0Co4.0Fe82.8P1.0Si4.0	616.28	1.84
B9.0C5.0Cu1.3Fe85.0	551.88	1.90
B13.0Co5.0Fe73.5P3.0Si5.4	659.17	1.66
B11.8C2.0Fe83.0Si3.0	631.10	1.84
B9.0C4.0Cu1.0Fe83.3P3.0	559.17	1.89
B9.0Fe80.4Ga2.0Ni1.3P4.0Si3.4	655.58	1.71

- The Pareto front: improving one property (Bs or Tc) may compromise the other.
- Correlations: The correlation coefficient between predicted Tc and Bs is weak ~ -0.1



- Main Question: can we access the region marked by the green box within the phase space to maximize both Bs and Tc?

[1] Pang, Bo, et al. *Materials & Design* 231 (2023): 112054.

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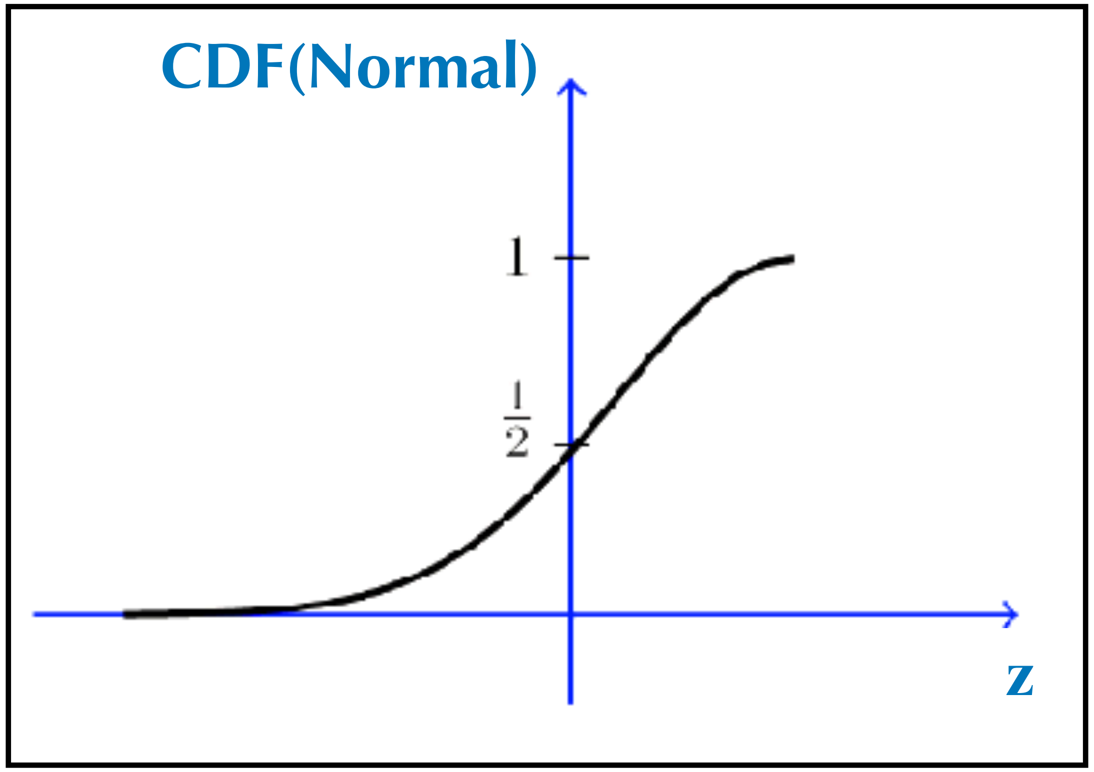
$$\boxed{x^* = \operatorname{argmax}_x f(x)}$$

Given observations $\mathcal{D}_{1:n} = \{(x_i, y_i)\}_{i=1:n}$, while $n \leq N$ repeat:

1. Update the surrogate model $f(x)$, i.e. the GP posterior $p(f) = \mathcal{GP}(f; \mu, \sigma)$, using all available data $\mathcal{D}_{1:n}$
2. Compute the acquisition function $u(x \mid \mathcal{D}_{1:n})$, using the current surrogate model
3. Let x_{n+1} be the maximizer of the acquisition function, i.e. $x_{n+1} = \operatorname{argmax}_x \; u(x \mid \mathcal{D}_{1:n})$
4. Evaluate $y_{n+1} = f(x_{n+1})$
5. Augment the data $\mathcal{D}_{1:n+1} = \{\mathcal{D}_{1:n}, (x_{n+1}, y_{n+1})\}$ and increment n

Acquisition function



Standardized sampled point

$$z = \frac{\mu(x_{n+1}) - \boxed{f^*}}{\sigma(x_{n+1})}$$

best observed value so far

Bayesian Optimization

Maximize {Tc, Bs} = f(Al, B, ..., Fe, ..., Zr)

Subject to:

0≤ Al ≤3, 0≤ B ≤15, ..., 0≤ Fe ≤85, 0≤ Zr ≤1 at %,

Al + B +...+ Fe +...+ Zr = 100 at %,

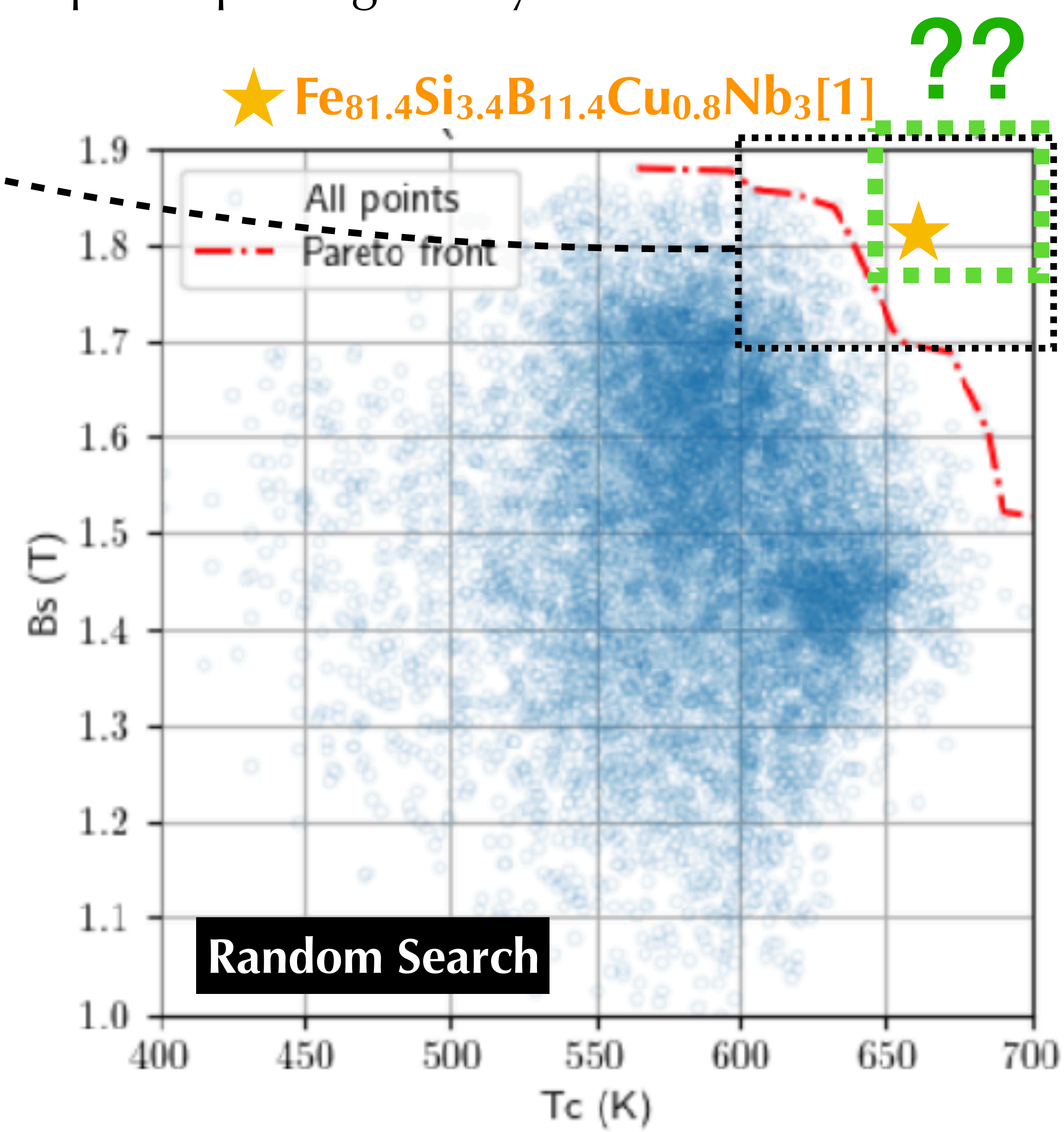
0 ≤ Annealing Time (s) ≤ 3600,

500 ≤ Annealing Temperature (K) ≤ 830,

20 ≤ Thickness (mu m) ≤ 30.

Optimization

- Objective: Suggest new alloys that maximize Bs and Tc.
- Methodology: Bayesian Optimization using the Optuna package in Python.



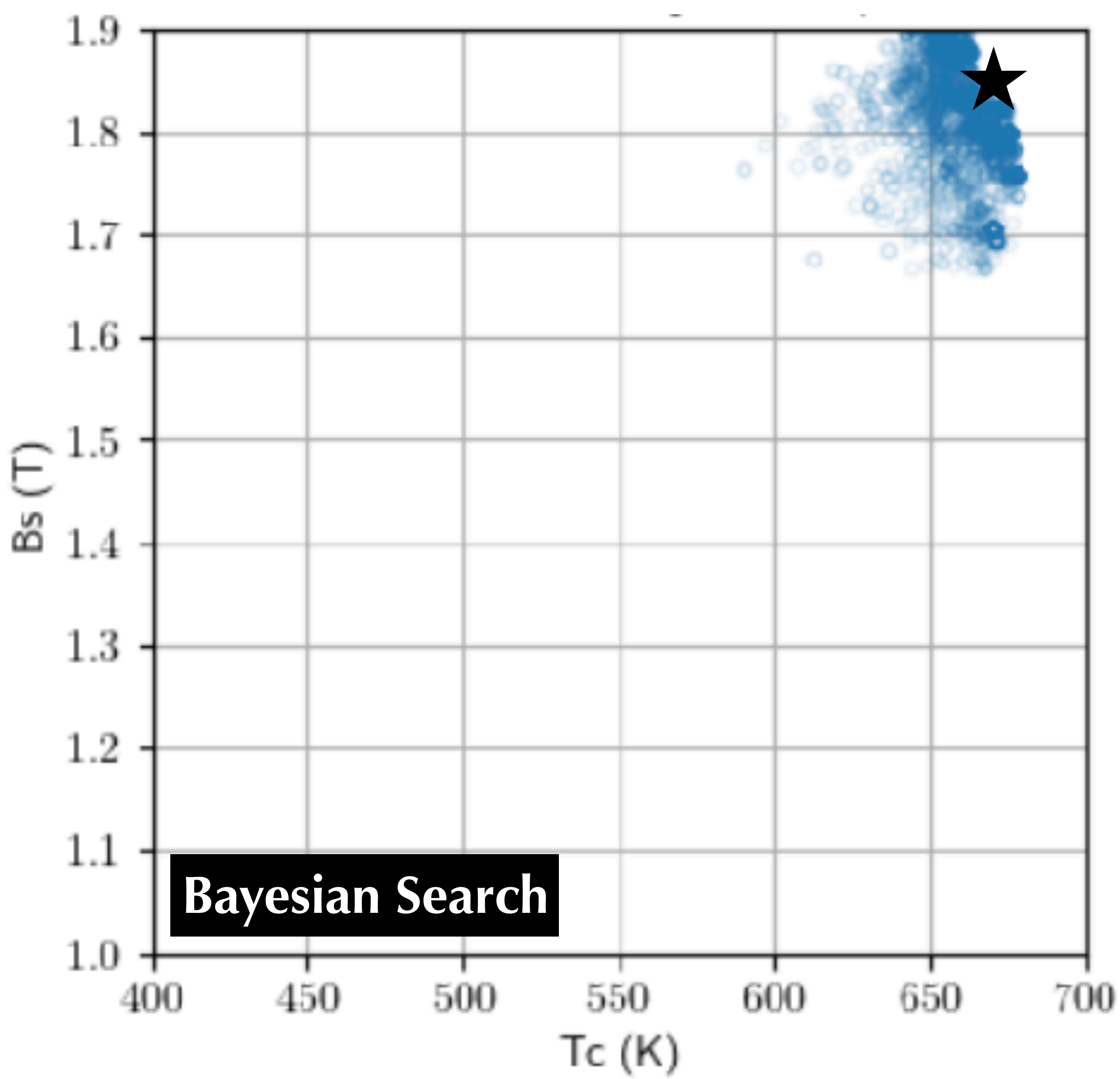
- Constrained optimization: Let's narrow down our search space based on our random search results.

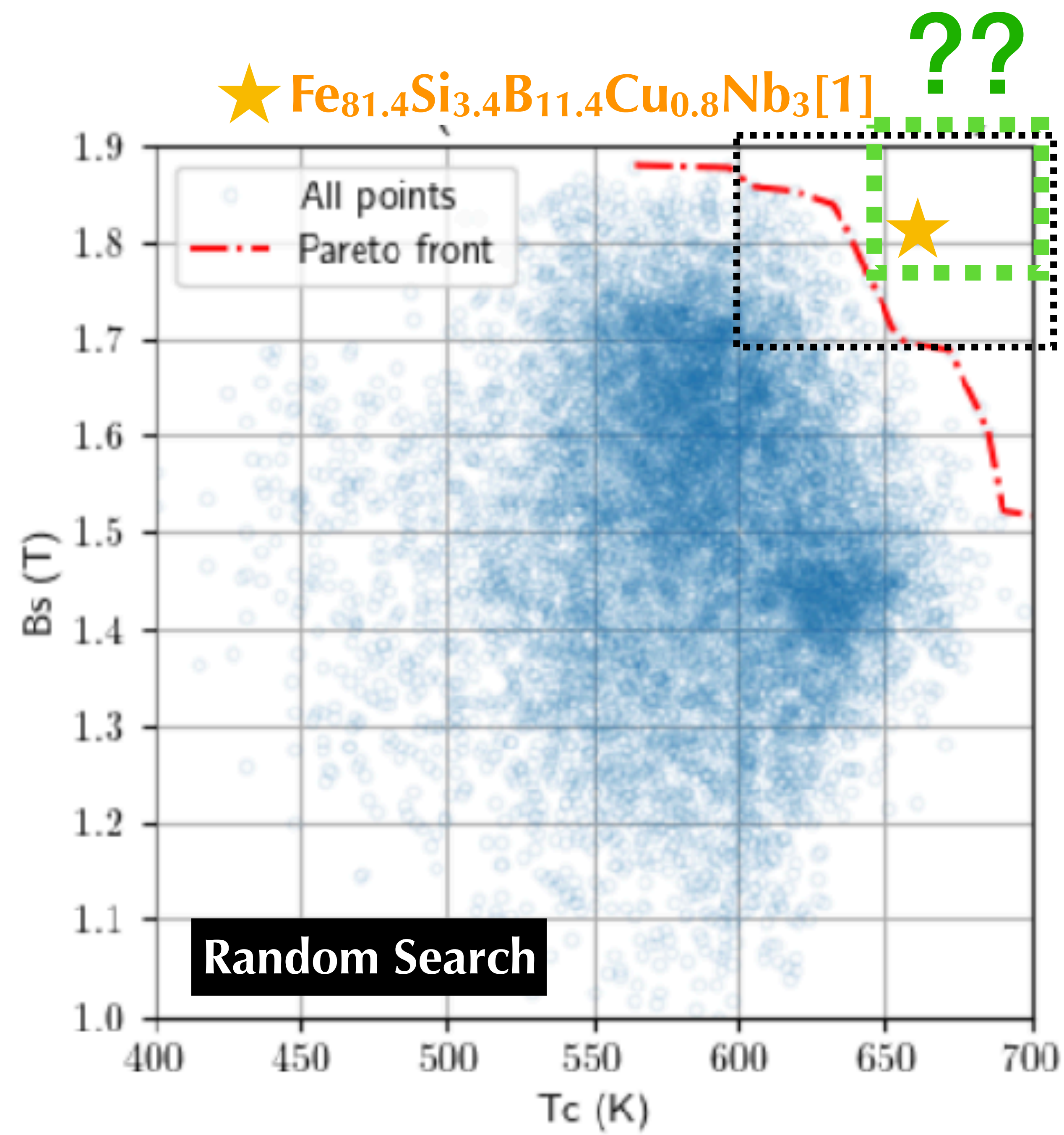
[1] Pang, Bo, et al. *Materials & Design* 231 (2023): 112054.

Bayesian Optimization

★	Annealing Time (s)	Annealing Temperature (K)	Thickness (mu m)	Tc (K)	Bs (T)
B11.9Co2.5Fe79.3Si4.7	831.78	789.98	18.21	670.32	1.85

- Main Question: can we access the region marked by the green box within the phase space to maximize both Bs and Tc?





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Uncertainty/sensitivity analysis

Bayes' theorem

$$\boxed{p(Fe|B_s^{obs})} \propto \boxed{p(B_s^{obs}|Fe)}\boxed{p(Fe)}$$

Posterior distribution — **Likelihood function** — **Prior distribution**

Bayesian framework

- Uncertainties in Bs: due to variations in Fe content within the alloy $Fe_{83}B_9Si_5P_3$ with Bs=1.68 T

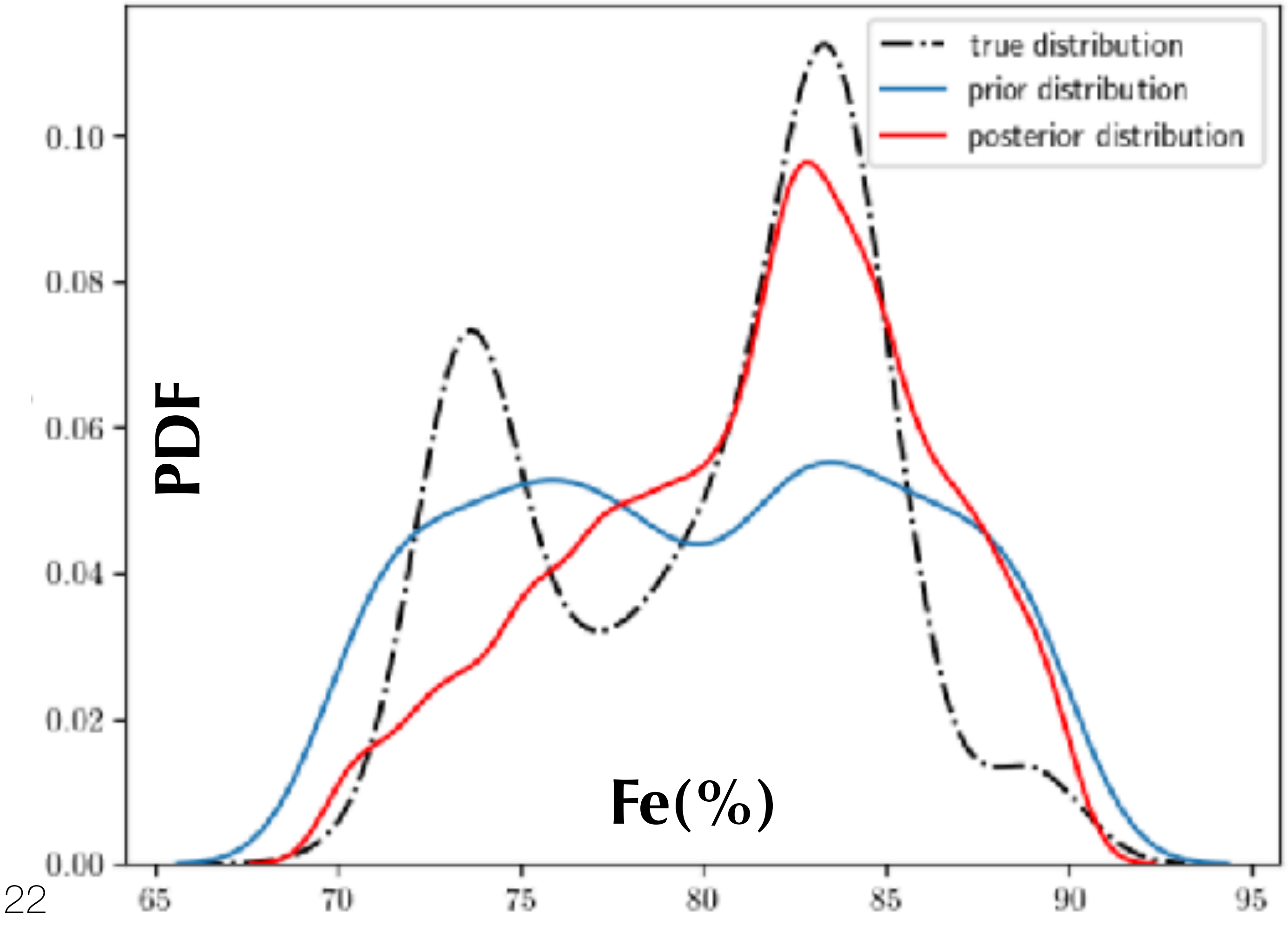
GPR's prediction — **GPR's variance**

$$p(B_s^{obs}|Fe) \propto exp[-\{B_s^{obs} - \boxed{\mu_{GPR}(Fe)}\}^2 \times \boxed{\sigma_{GPR}^{-2}(Fe)}]$$

- Define likelihood:
  the difference between observed Bs and GPR-predicted values, weighted by the GPR's variance

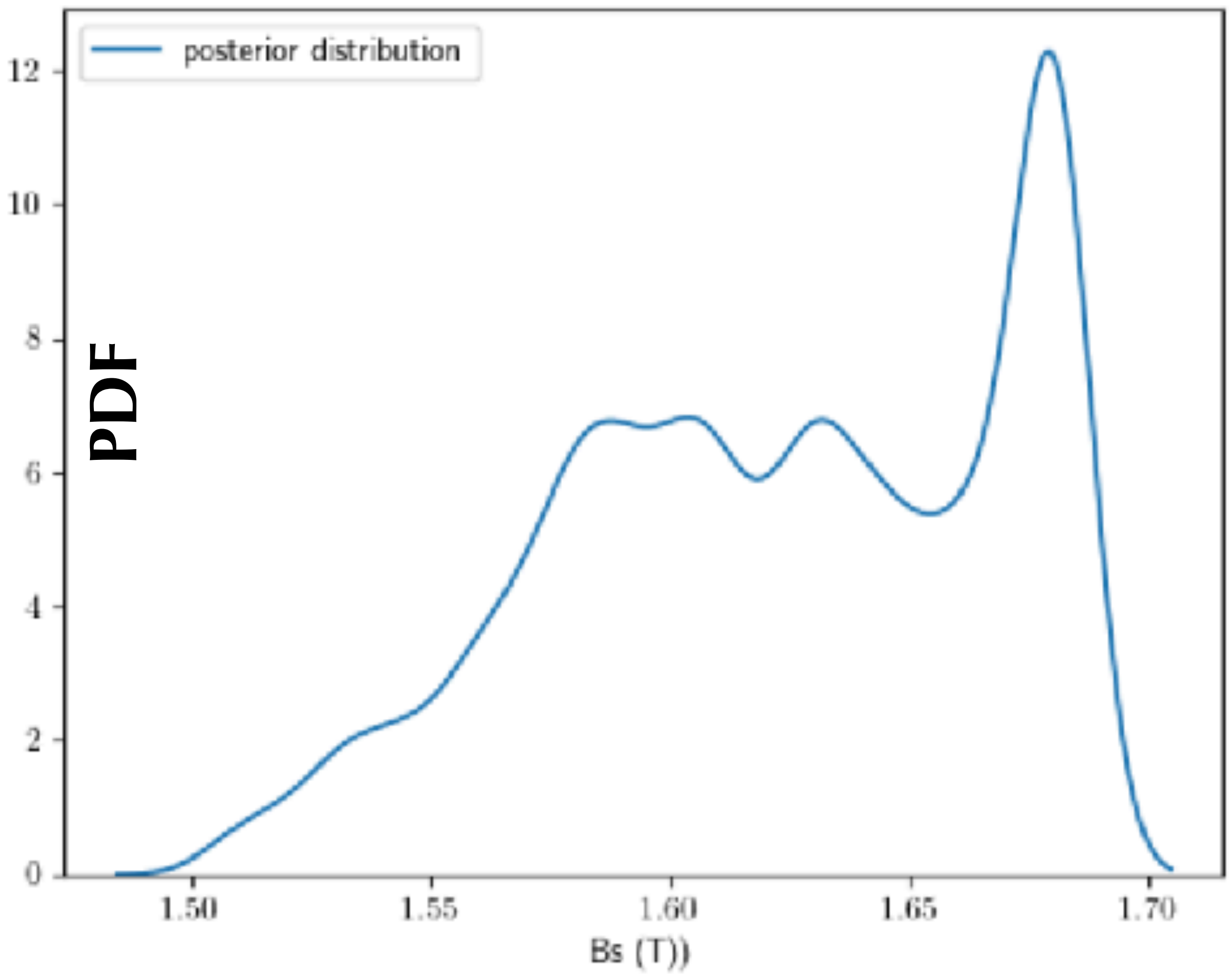
- Fe's prior distribution:
  uniform around 83%

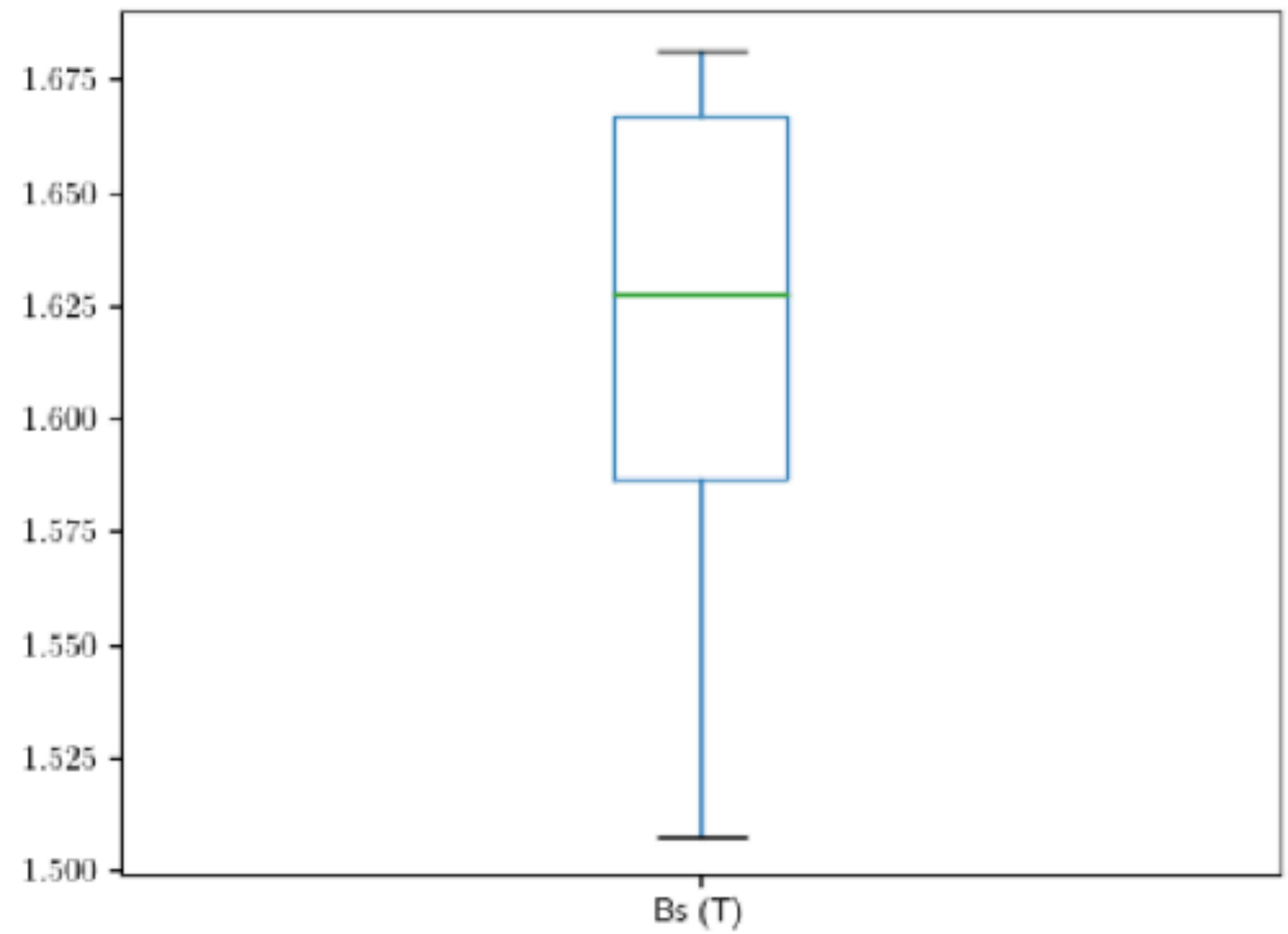
- Posterior distribution:
  combine the prior and the likelihood to obtain the posterior



Uncertainty/sensitivity analysis

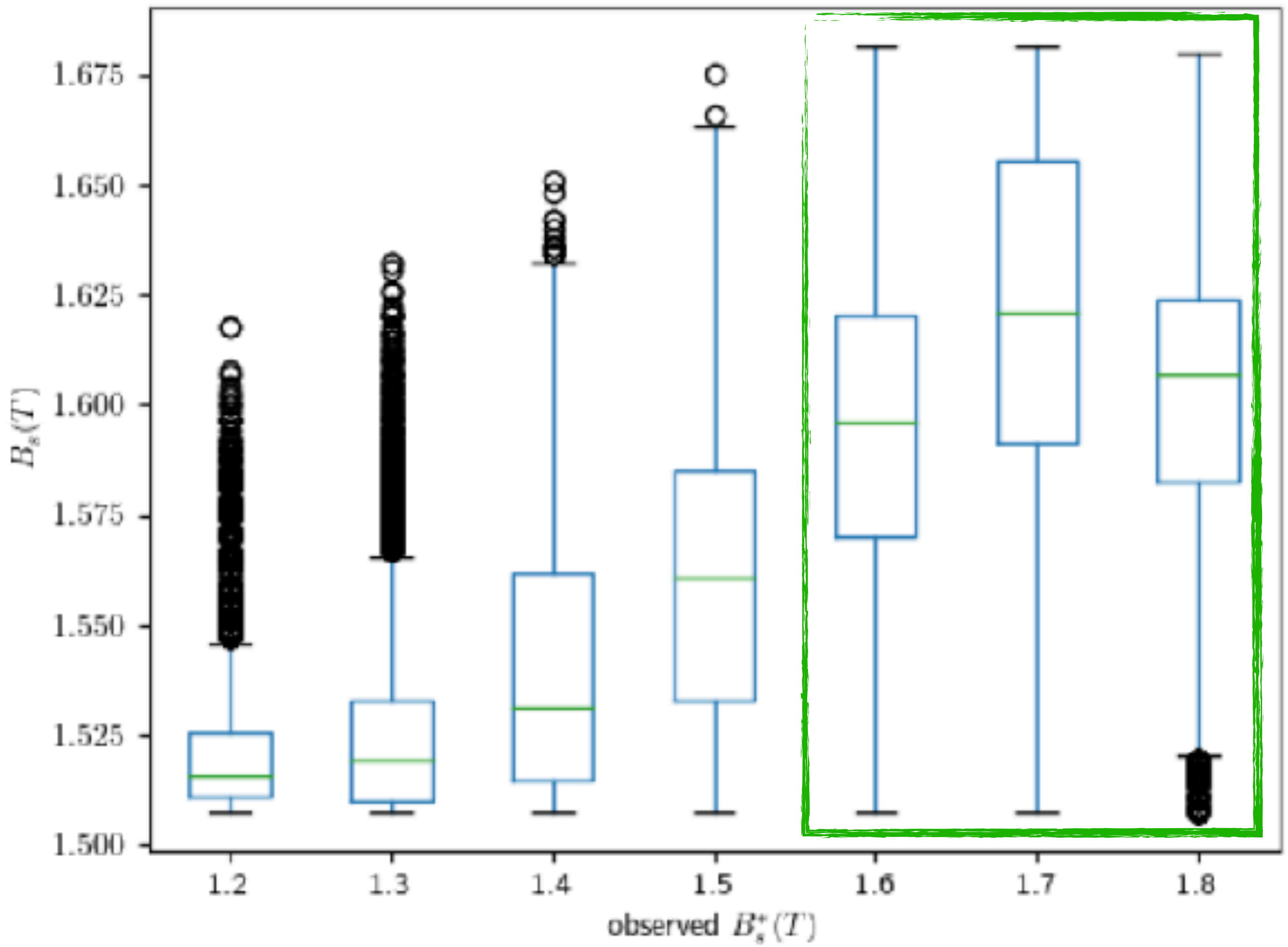
```
Mean BS: 1.6226832626630308
Standard Deviation of BS: 0.04634739613388416
90% Confidence Interval for BS: [1.53982611 1.68114307]
<Axes: >
```





Robustness

Robust confidence limit for Bs with respect to variations in the observed value B_s^{obs}



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Constrained Optimization

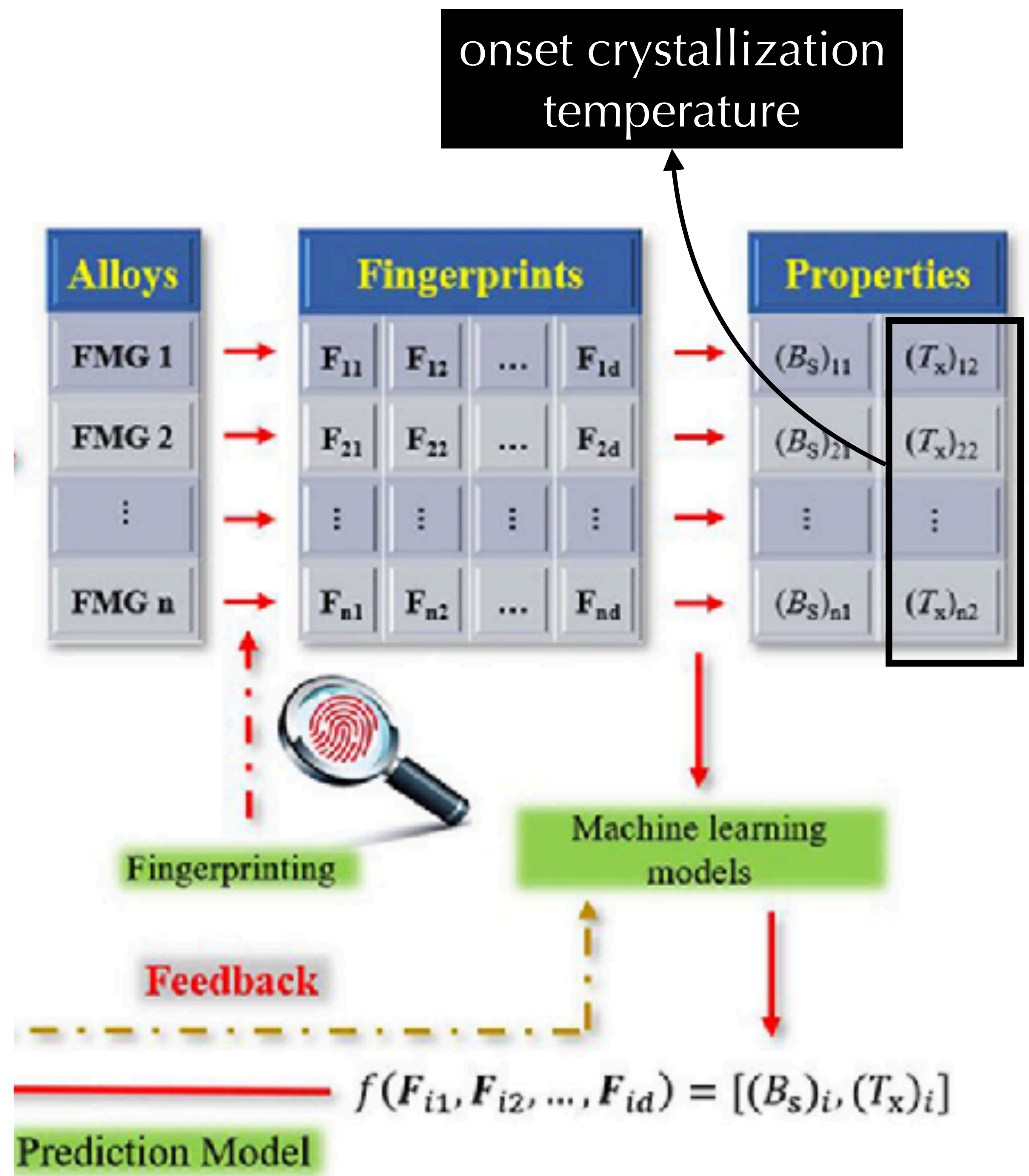
ARTICLE OPEN

Check for updates

Interpretable machine-learning strategy for soft-magnetic property and thermal stability in Fe-based metallic glasses

Zhichao Lu[1], Xin Chen[2], Xiongjun Liu[1]✉, Deye Lin[2,3], Yuan Wu[1], Yibo Zhang[1], Hui Wang[1], Suihe Jiang[1], Hongxiang Li[1], Xianzhen Wang[4] and Zhaoping Lu[1]✉

Composition (at. %)	Bs/T	Tx/K
Fe83Si2B11P3C1	1.67	727
Fe80P13C7	1.477	696
Fe80P11C9	1.37	720
Fe75B16.67Si8.33	1.57	839
Fe76C7.0Si3.3B5P8.7	1.52	795



Constrained Optimization

3. **Optimization with Constraints**:

- We are providing an additional dataset containing TX values. Can you refine your optimization code from Task 1 to include a constraint that TX must stay between 600 to 800 K?
  - Provide the optimized alloy compositions that satisfy this constraint.
  - Provide the Pareto front considering this new constraint.

Composition	ta (s)	Ta (K)	Thickness (mu m)	Tc (K)
Fe69.5Co0.5Mn10Mo5B15	0.0	0.0	20.0	370.0
Fe80Ni6Zr7Nb2B4Cu1	0.0	0.0	20.0	374.0
Fe90Zr7B3	3600.0	873.3	20.0	388.6
Fe69.5Co0.5Mn10Mo5B15	1800.0	723.0	20.0	389.0
Fe90Zr7B3	3600.0	822.9	20.0	389.7

Composition	ta (s)	Ta (K)	Thickness (mu m)	Bs (T)
Fe89Hf7Zr1B3	1800	648.9	32.5	0.3
Fe89Hf7Zr1B3	1800	598.7	32.5	0.4
Fe89Hf7Zr1B3	1800	748.6	32.5	0.4
Fe89Hf7Zr1B3	1800	698.8	32.5	0.6
Fe89Hf7Zr1B3	0	0.0	32.5	0.6

[2] Lu, Zhichao, et al. "Interpretable machine-learning strategy for soft-magnetic property and thermal stability in Fe-based metallic glasses." npj Computational Materials 6.1 (2020): 187.

Composition (at. %)	Bs/T	Tx/K
Fe83Si2B11P3C1	1.67	727
Fe80P13C7	1.477	696
Fe80P11C9	1.37	720
Fe75B16.67Si8.33	1.57	839
Fe76C7.0Si3.3B5P8.7	1.52	795

Combine these two datasets?

[1] Pang, Bo, et al. "Accelerated discovery of Fe-based amorphous/nanocrystalline alloy through explicit expression and interpretable information based on machine learning." *Materials & Design* 231 (2023): 112054.

Constrained Optimization

Maximize {`Tc`, `Bs`} = f(`Al`, `B`, ..., `Fe`, ..., `Zr`)

Subject to:

0≤`Al`≤3, 0≤`B`≤15, ..., 0≤`Fe`≤85, 0≤`Zr`≤1 at %,

`Al`+`B`+...+`Fe`+...+`Zr` = 100 at %,

`Annealing Time (s)` = 600,

`Annealing Temperature (K)` = 660,

`Thickness (mu m)` =21.

600 K ≤ `Tx` =g(`Al`, `B`, ..., `Fe`, ..., `Zr`)≤ 800 K

additional constraint

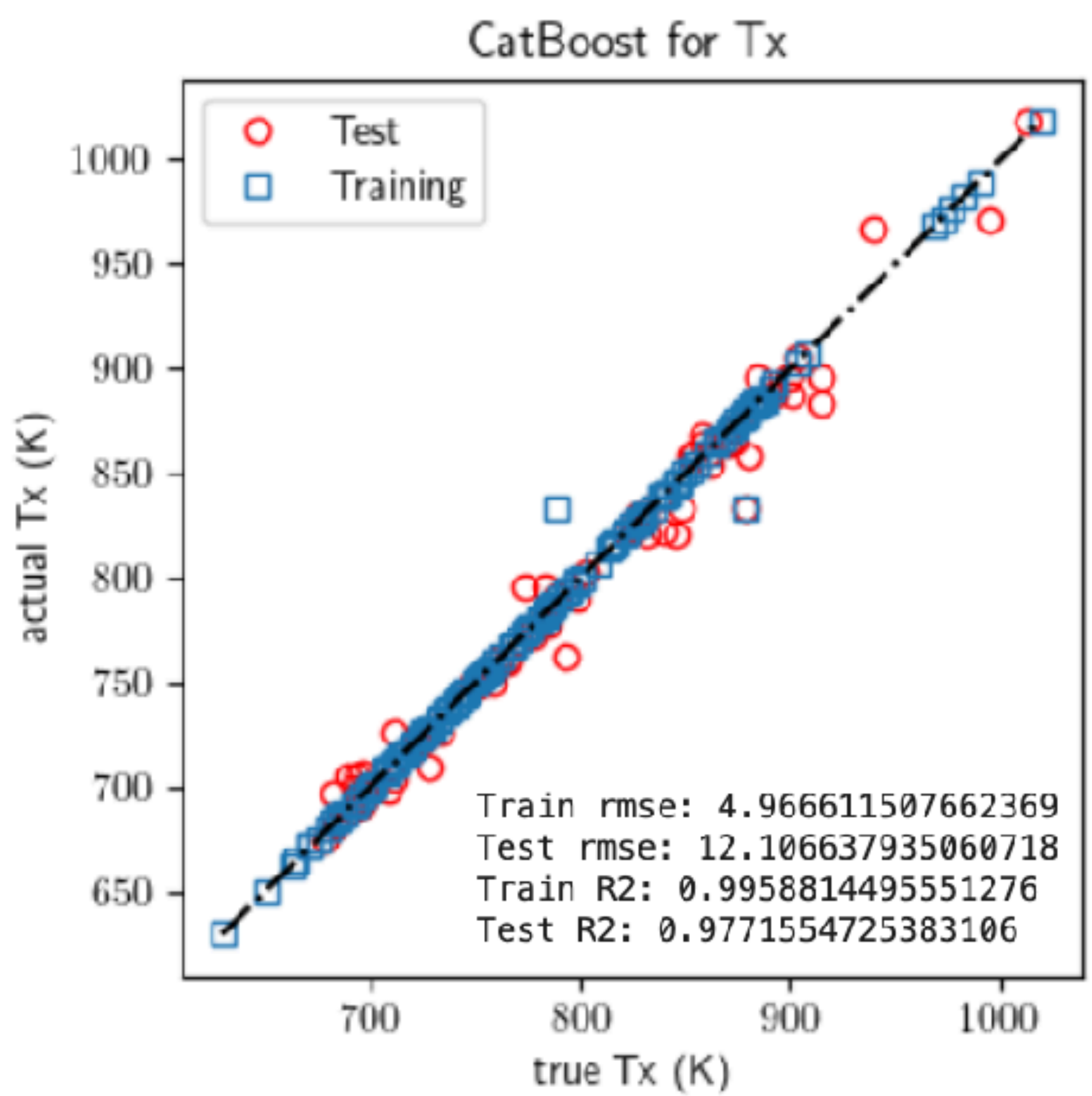
Let's follow the first approach

Two general approaches

- Train a third model:
  Tx=g(Al, B, …, Zr) based on the new data set and incorporate this trained Tx model as part of the constrained optimization
  - Make an assumption about the missing processing conditions
  - The propert map becomes 3-dimensional spanned by (Tc, Bs, Tx) but look at the 2-dimensional sub-scapce (Tc, Bs) where 600<Tx<800 K
- Use Tx as additional input feature:
  Re-train Tc=f1(x,Tx) and Bs=f2(x,Tx) with x being elemental/processing features
  - Combine datasets carefully

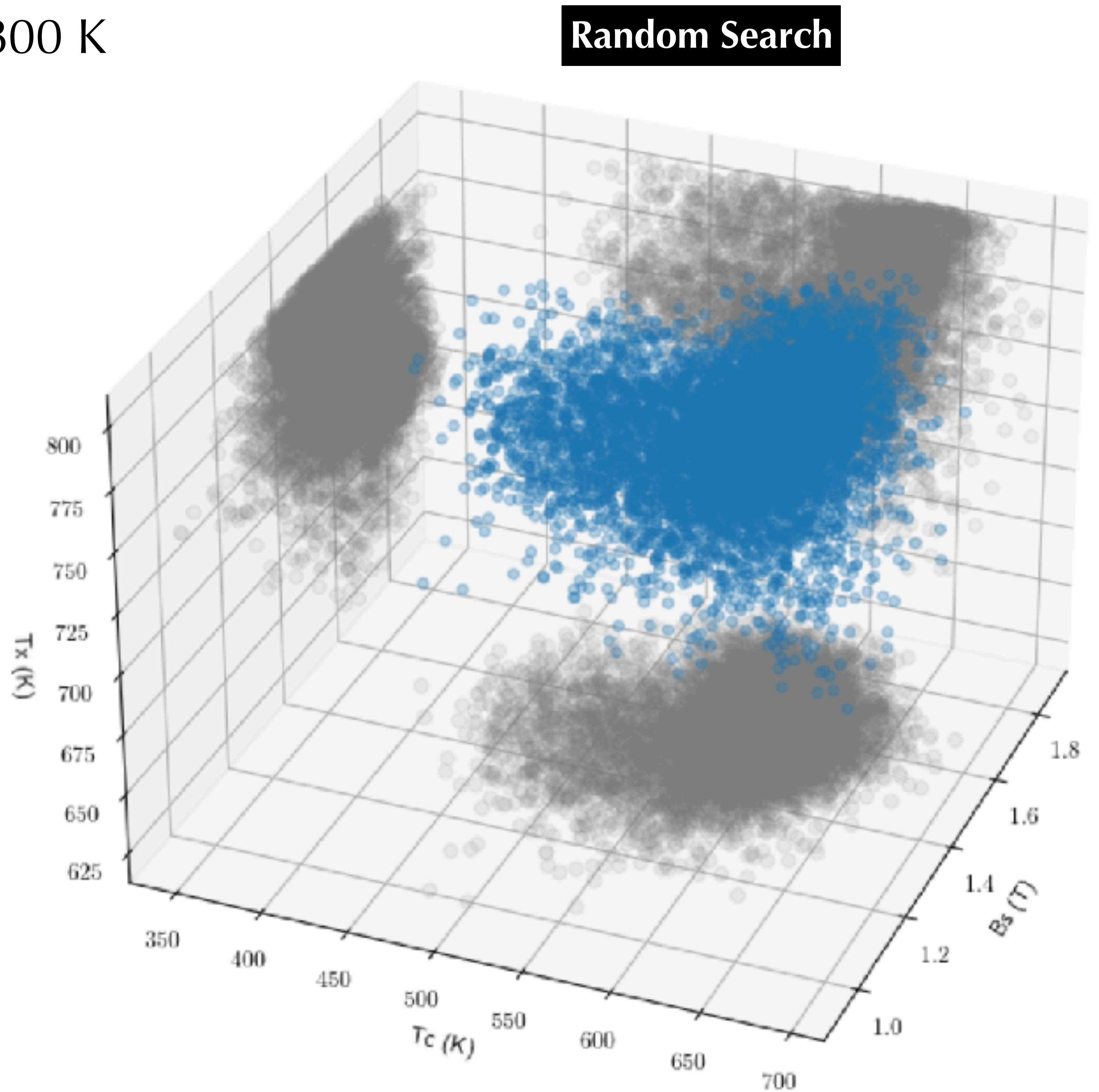
Machine Learning

- Models Used: CatBoost
- Hyperparameter Tuning: GridSearchCV, cross-validation
- Training Strategy: Train-test split, standardization, evaluation metrics



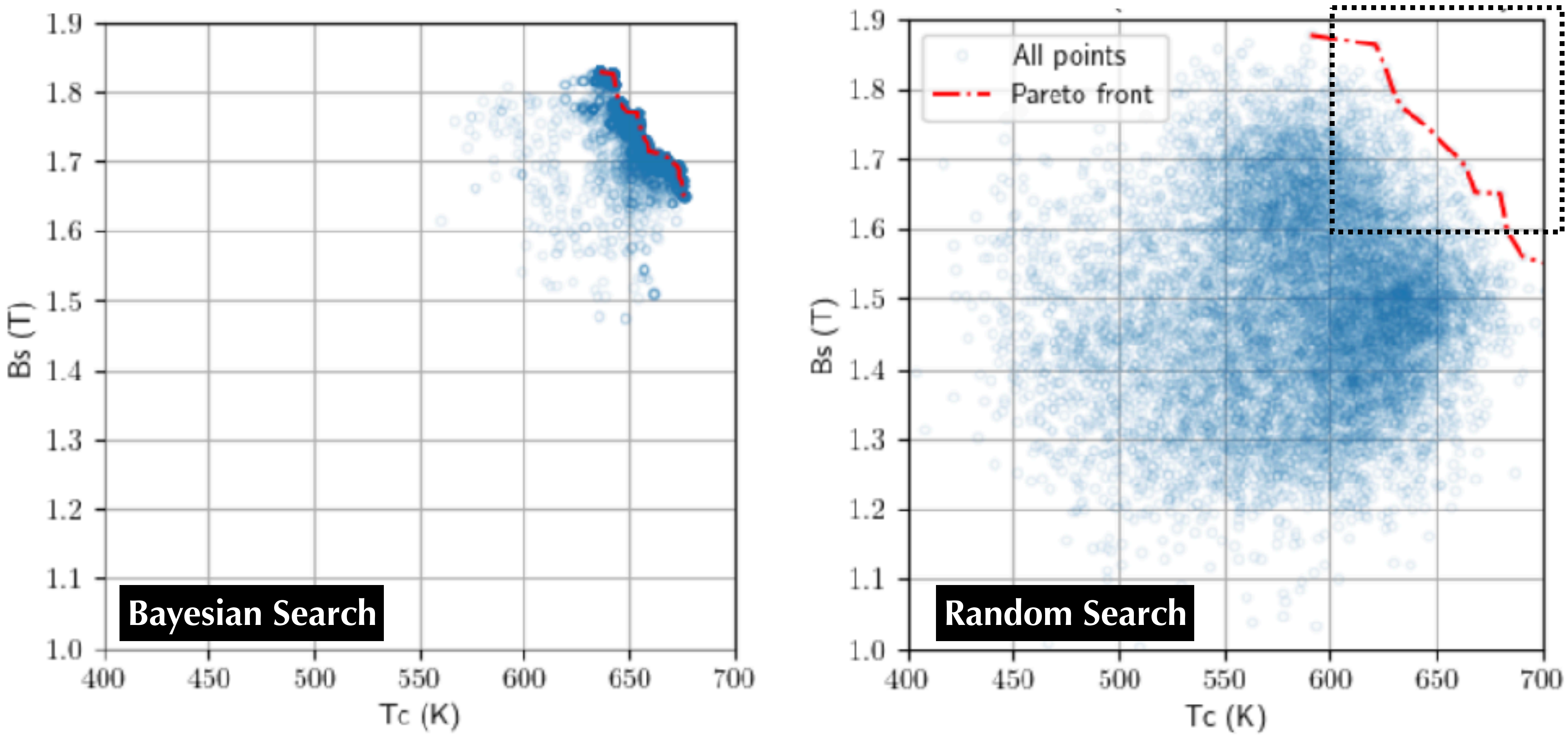
Pareto Front

600<Tx<800 K



Bayesian Optimization

Composition	Tc (K)	Bs (T)	Tx (K)
B11.5Co5.1Fe72.3P5.7Si5.2	687.7	1.5	775.8
B12.0C2.6Co2.0Fe78.3Si5.0	658.8	1.6	784.4
B11.5Co2.1Fe80.4Si5.2	645.7	1.7	753.8
B11.5Co4.8Fe72.4P5.0Si5.0	681.4	1.5	759.9



Conclusions

Key Findings & Insights:

- Optimization Results: The Pareto front analysis excluding the Tx constraint suggests a trade-off between maximizing Bs and Tc. Applying this constraint, it might be feasible to optimize both properties at the same time.
- Uncertainty Quantification: Bayesian analysis reveals the impact of fluctuations in Fe composition on the uncertainty of Bs, highlighting the importance of precise composition control.
- Additional Data: Need more comprehensive dataset to capture the interplay between elemental compositions and processing conditions and develop more comprehensive alloy designs.
- Model Refinement: Further refinement and fine-tuning of ML models during the optimization process could enhance prediction accuracy.
- Real-world Validation: Include experimental validation of the predicted alloy compositions to assess the model's effectiveness in practical applications.