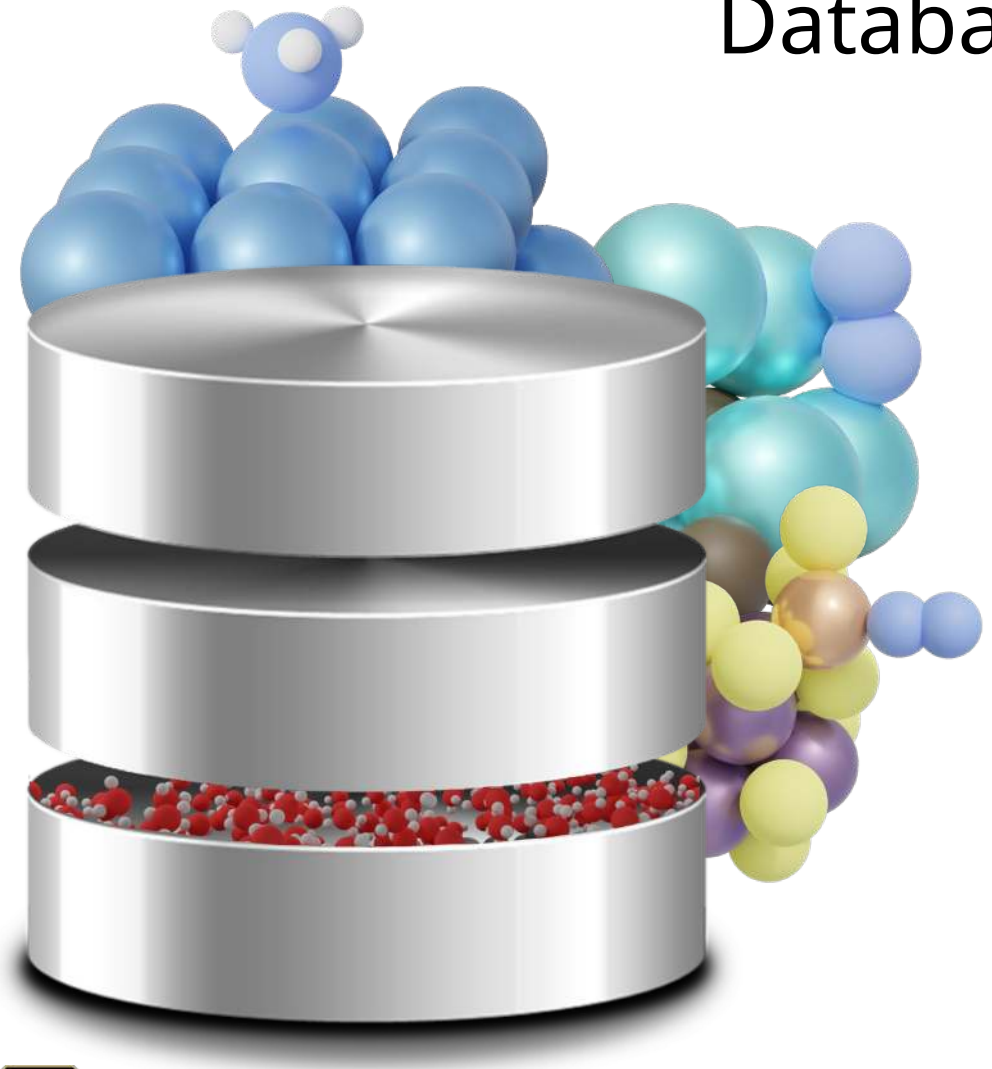
BEAST Database: Ab-Initio Electrochemistry Database to Accelerate Electrocatalyst Development



ACS, August 22-23, 2024

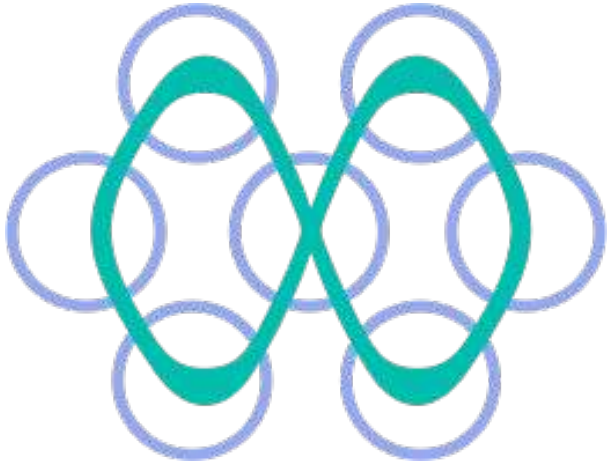
Cooper Tezak

BEAST Workshop

University of Colorado **Boulder**

U.S. DEPARTMENT OF ENERGY | Office of Science

Materials databases have accelerated materials science



MATERIALS CLOUD

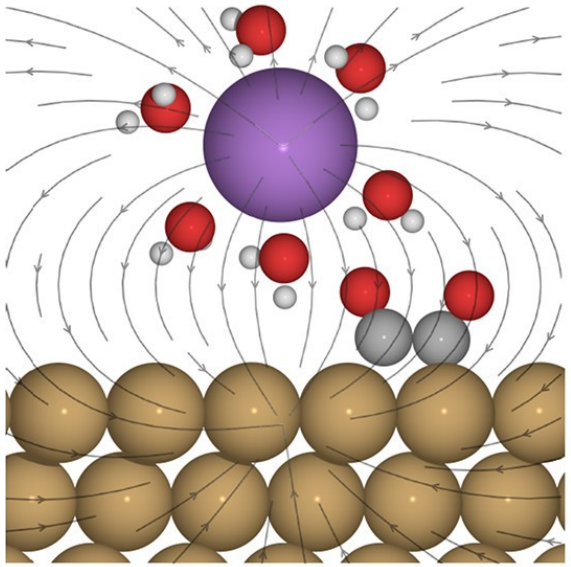
Materials Project
- Optoelectronic properties
- Stability analysis
- OC20 catalysis dataset
- XAFS data

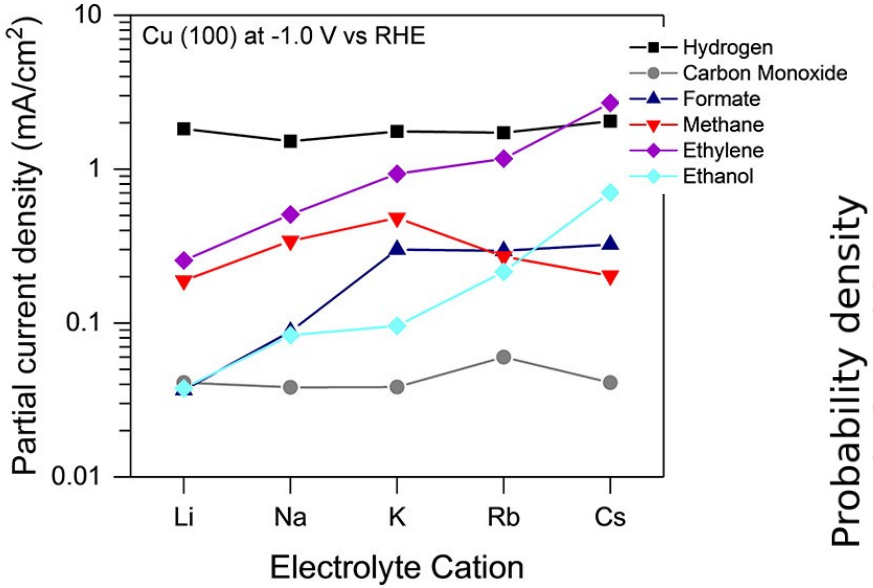
Catalysis-hub
- Adsorption energies
- Barriers
- Some electrocatalysis

Materials Cloud
- Specific material space archives
- Standardized workflow codes
- Quantum code benchmarks

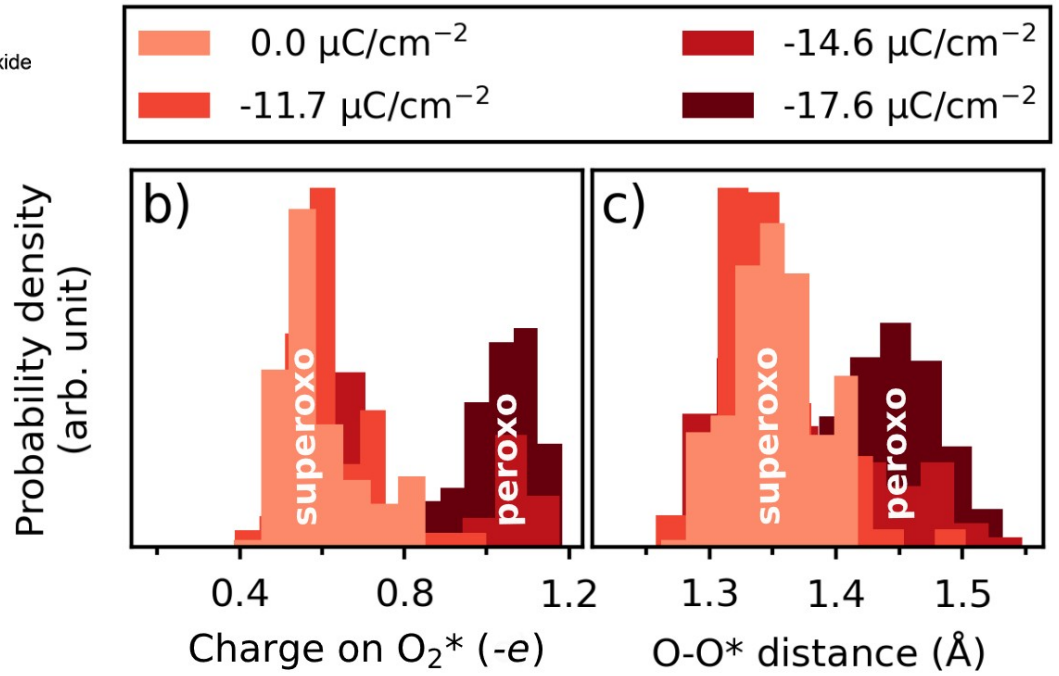
Electrocatalysis has unique complexity

Solvation environment





Applied potential



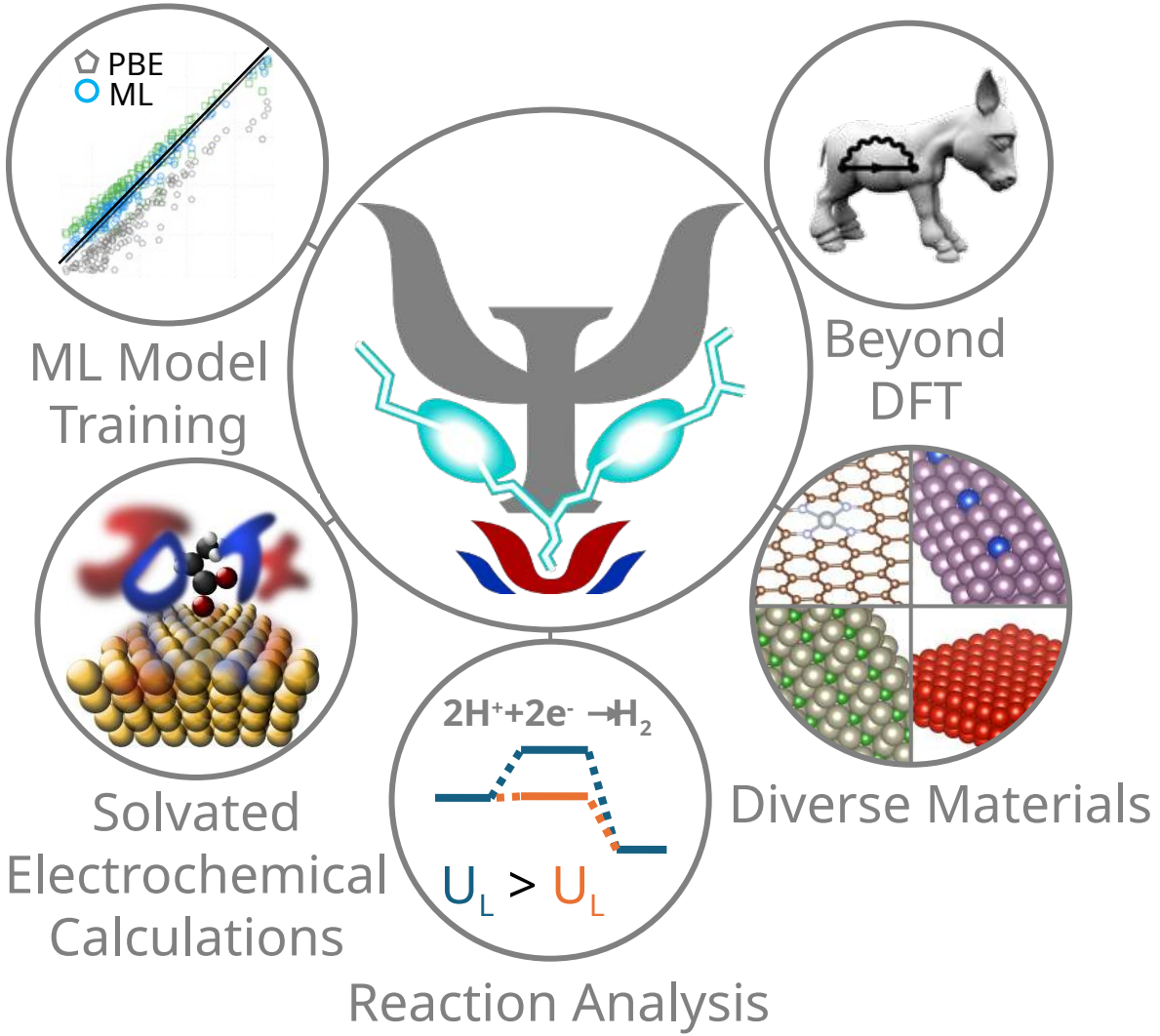
The computational community must embrace the complexities of electrocatalysis to accelerate development

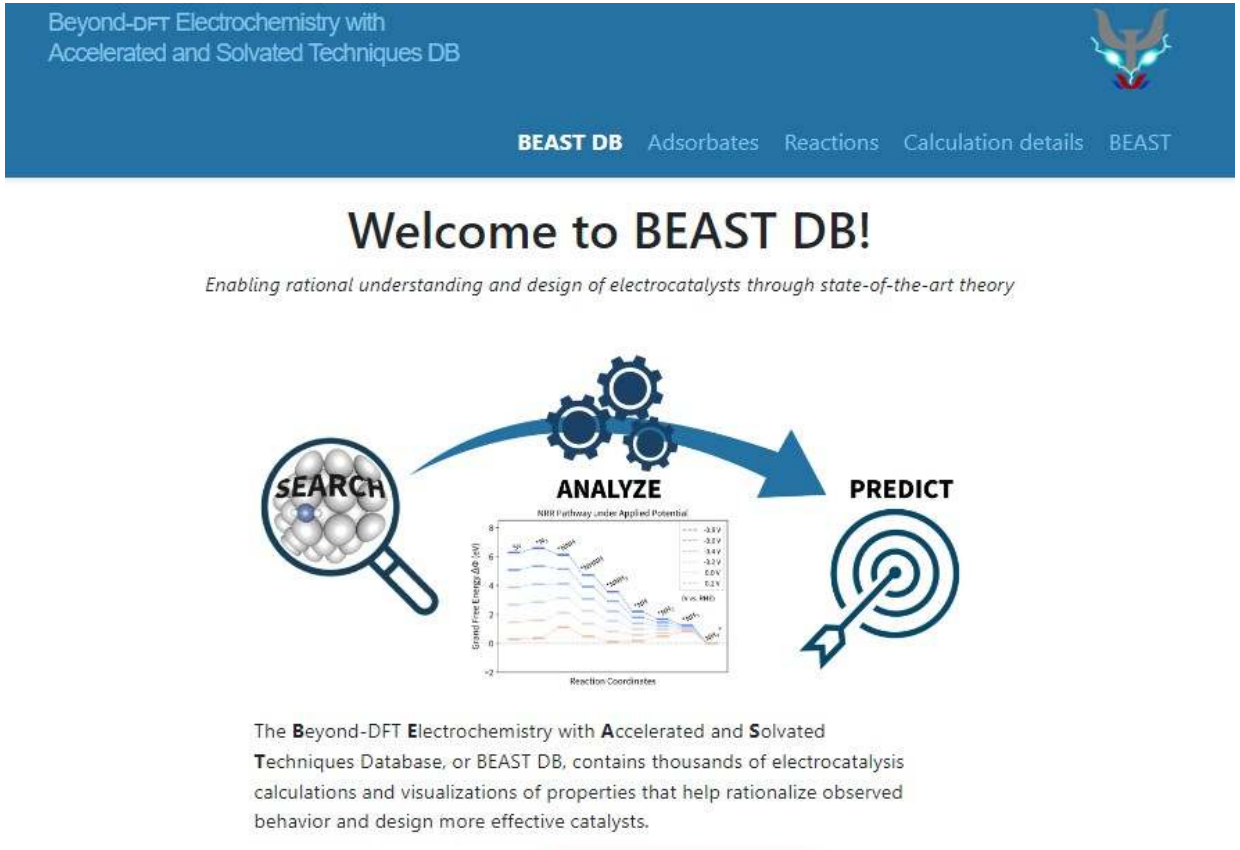
Resasco, J.; et al. Promoter Effects of Alkali Metal Cations on the Electrochemical Reduction of Carbon Dioxide.

Dudzinski, A. M.; et al. First Step of the Oxygen Reduction Reaction on Au(111): A Computational Study of O2 Adsorption at the Electrified Metal/Water Interface

Introducing BEAST DB

https://beastdb.nrel.gov/

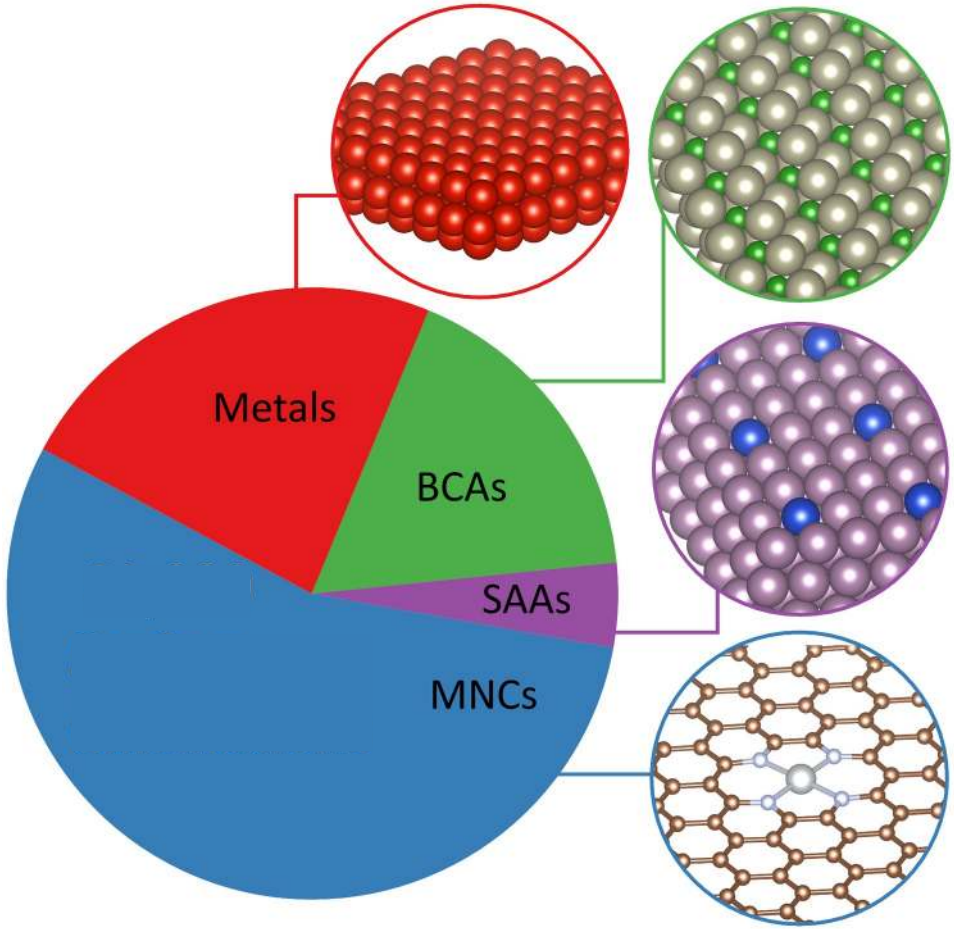




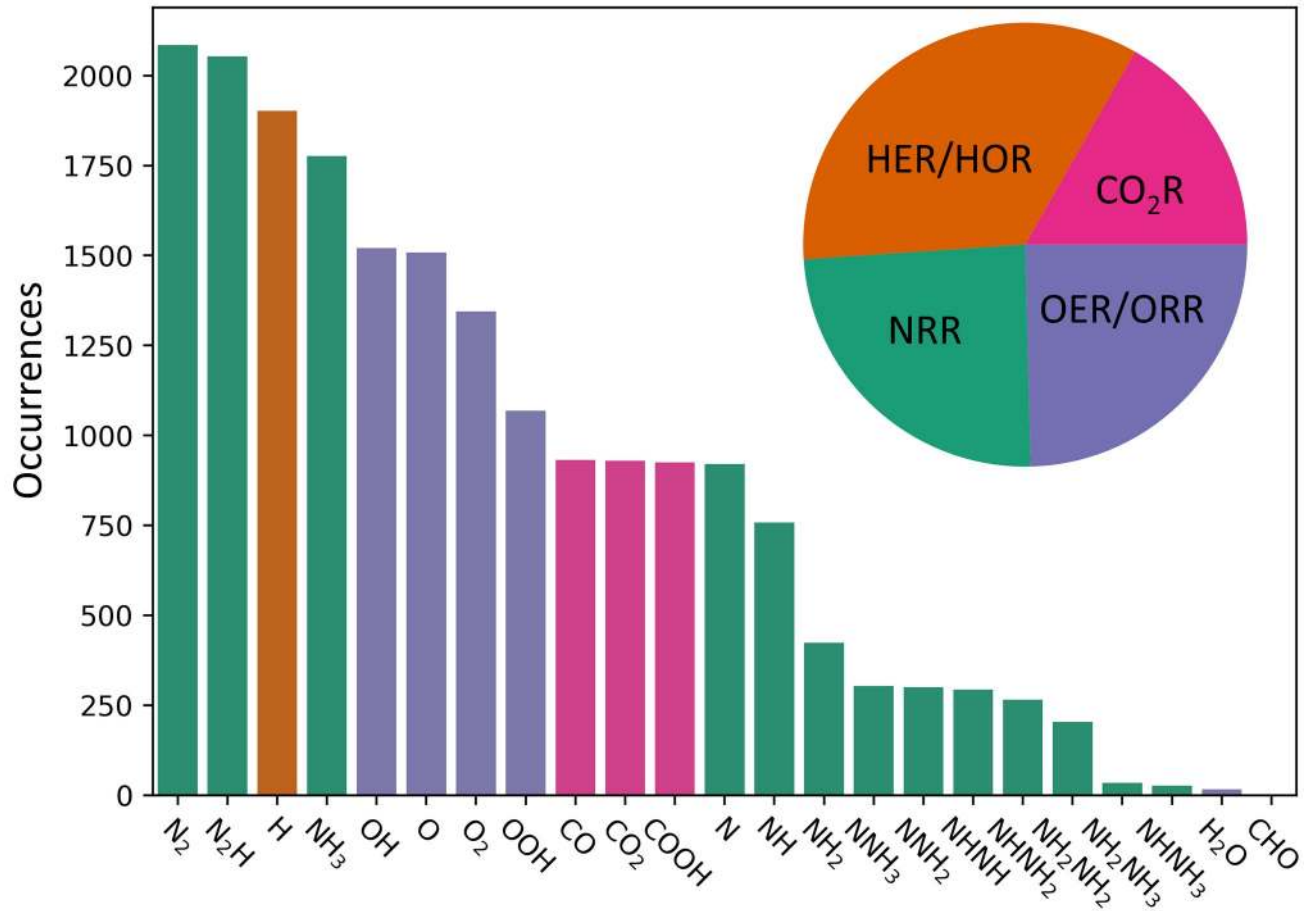
Open access user interface

Tezak, C. et al. BEAST DB: Grand-Canonical Database of Electrocatalyst Properties. https://doi.org/10.48550/arXiv.2405.20239. *Under review*

Database overview



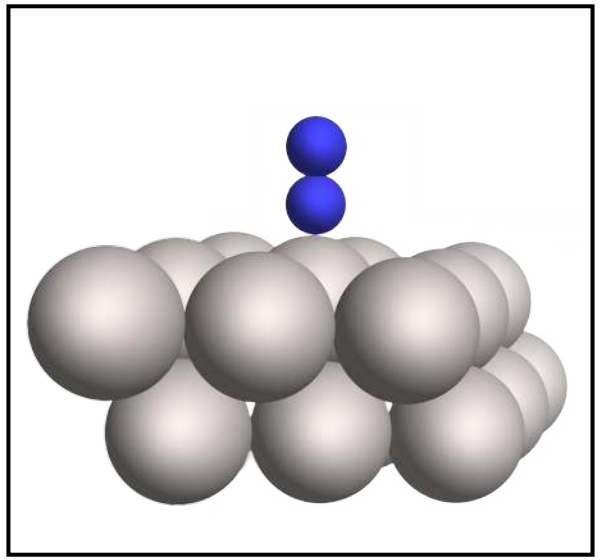
20,000 Surface Calculations



Common reactions coverage

We use electrochemical methodology

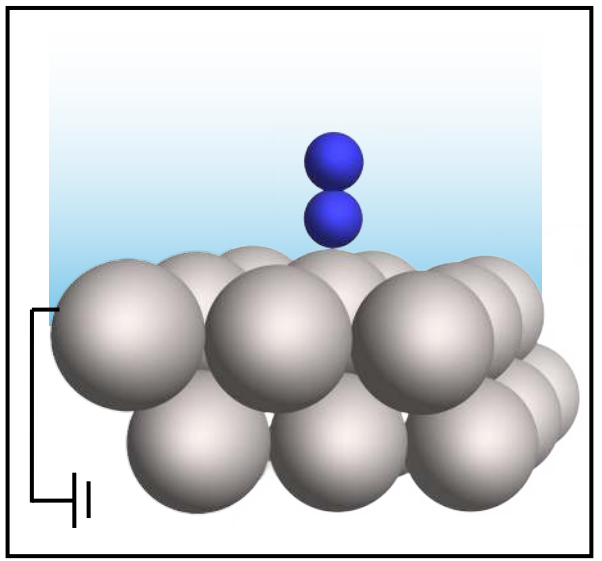
Canonical DFT



- Fixed charge
- Potential correction schemes

$$F_{DFT}$$

Grand Canonical DFT



- Fixed potential
- Self-consistent free energies

$$\Phi = F_{DFT} - \mu N_e$$

Accurate solvation models

Solvation Model	pKw
CANDLE	17.2
VaspSol	34.0

Consistent calculation parameters

Parameter	Value
Functional	PBE
Solvent	Water
Electrolyte	1M NaF
Cutoff energy	20 Ha

Our methodology facilitates electrocatalytic mechanisms

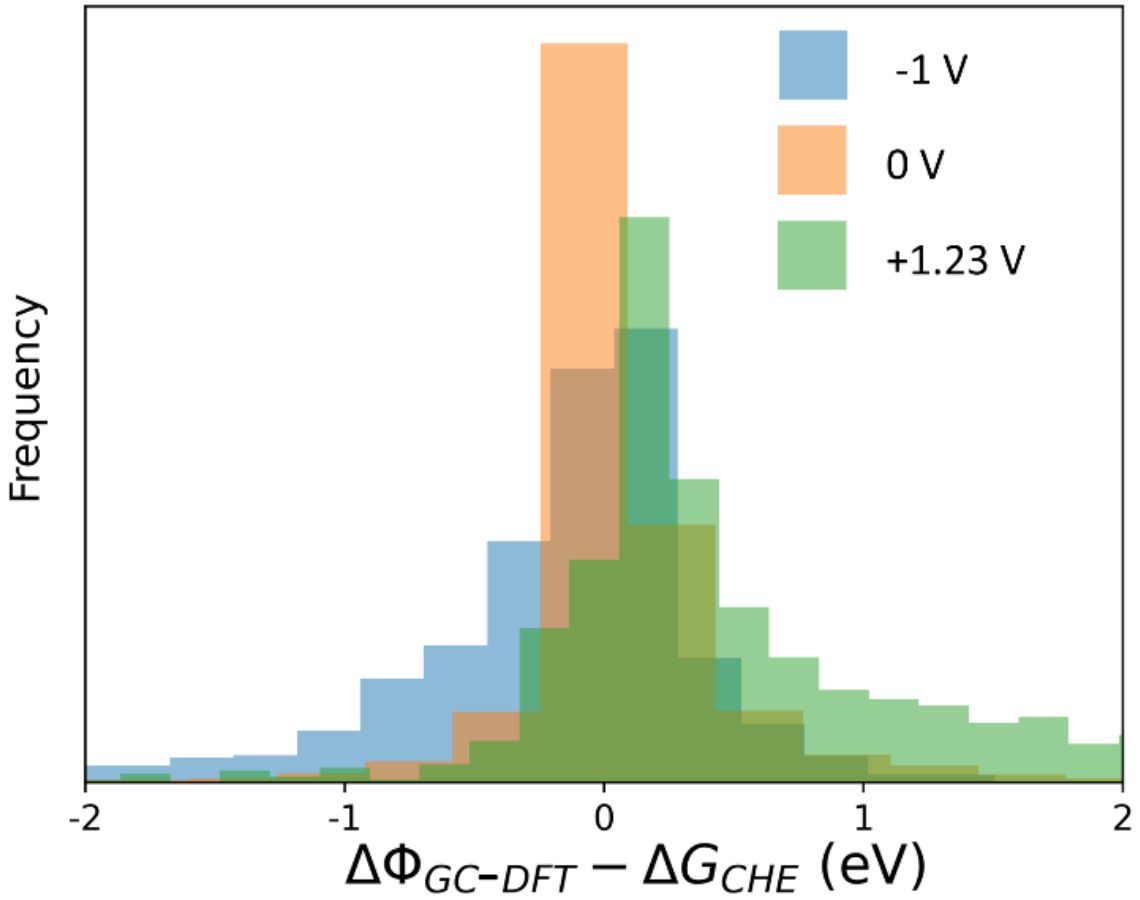
Canonical DFT relies on the computational hydrogen electrode (CHE)

$$G_{H^+ + e^-} = \frac{1}{2} G_{H_2} - U_{SHE}$$

Limitations:

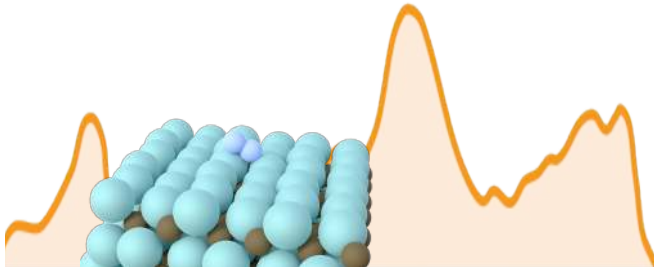
- Proton-coupled electron transfers only
- Linear potential dependence

GC-DFT places no step restrictions and makes no assumptions about potential dependence

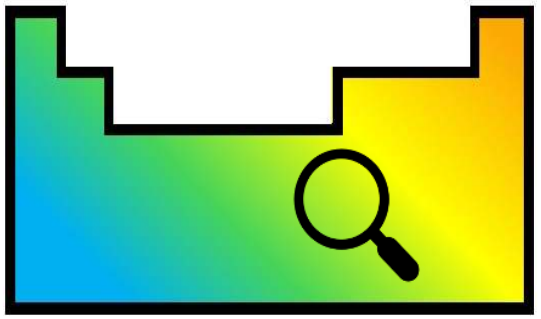


Capabilities

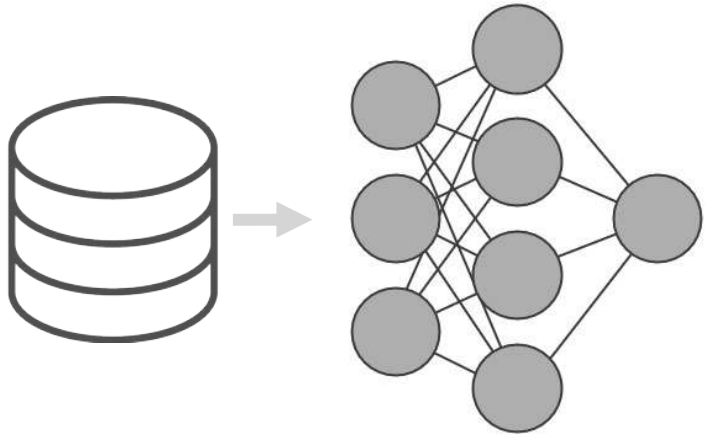
1. Individual system studies



2. Material space screening

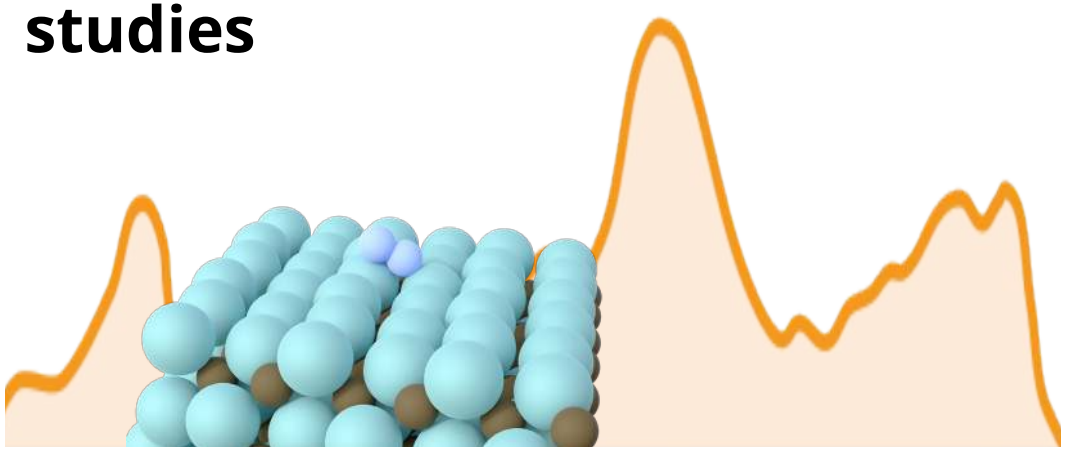


3. Statistical model training



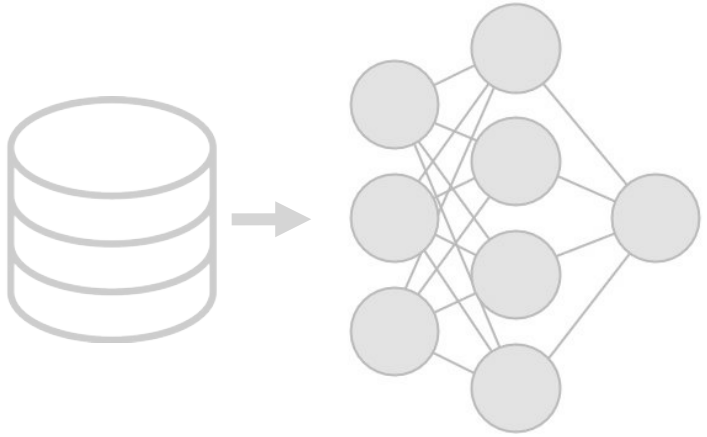
Capabilities

1. Individual system studies

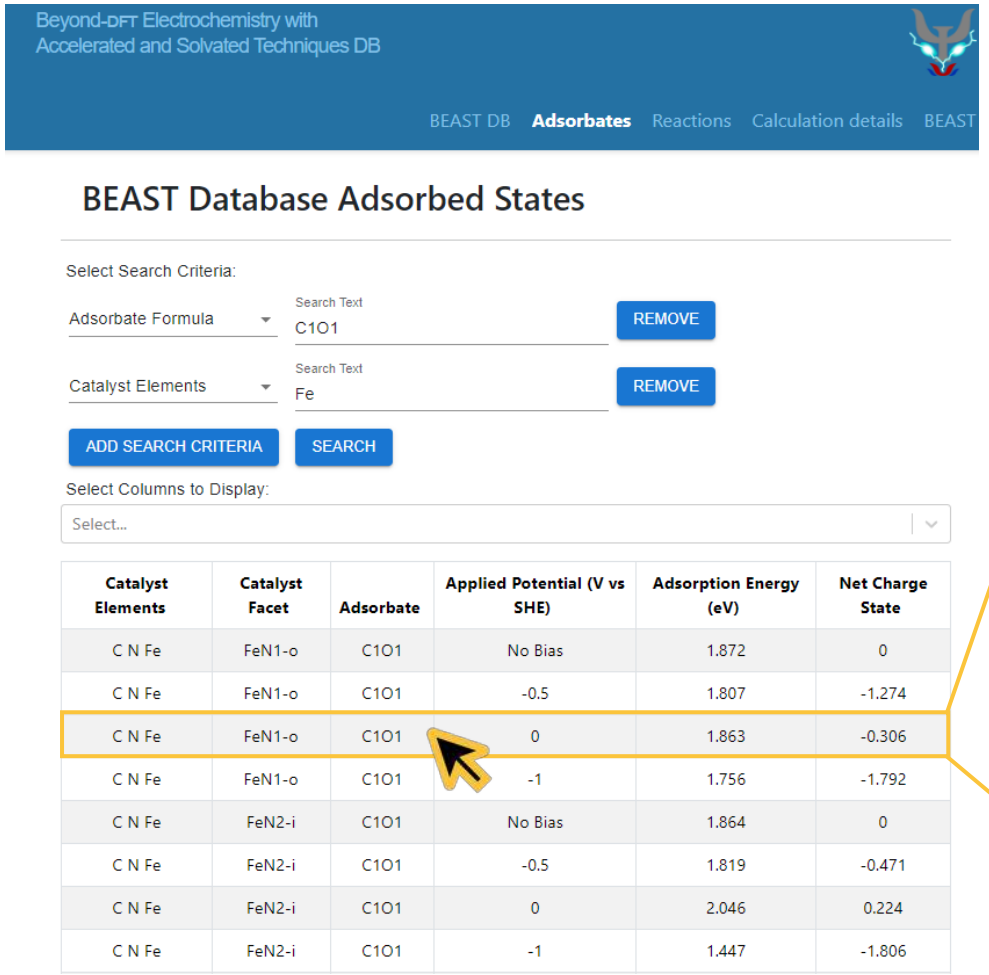


2. Material space screening

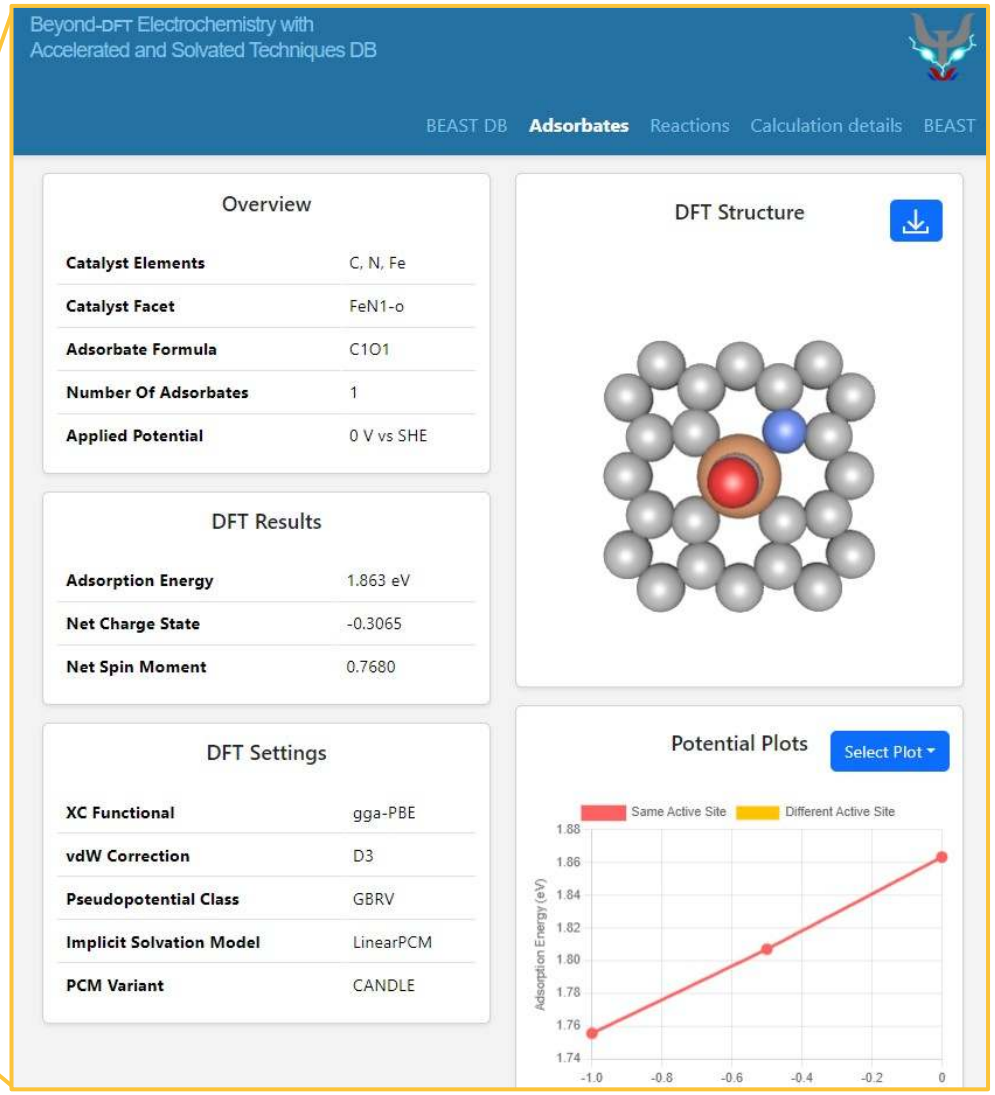
3. Statistical model training



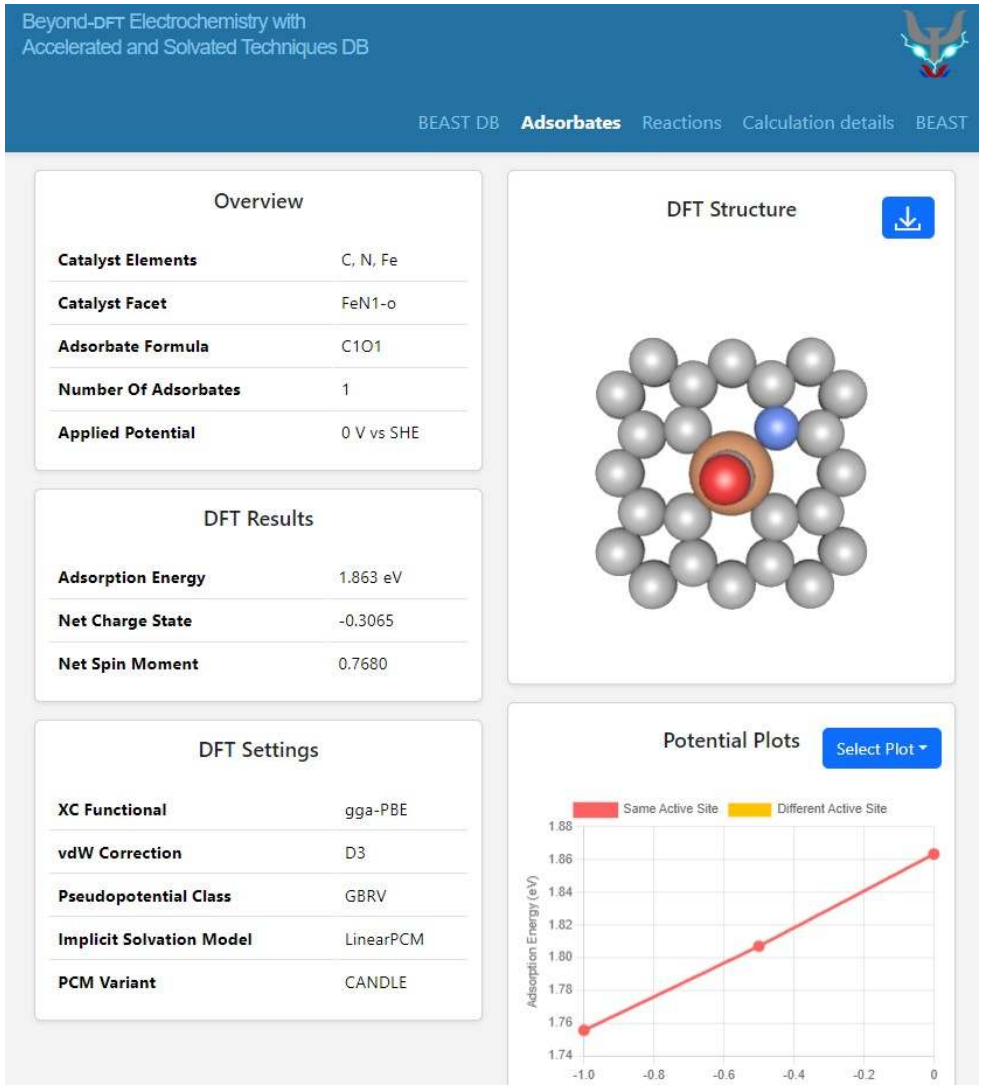
UI facilitates granular system understanding



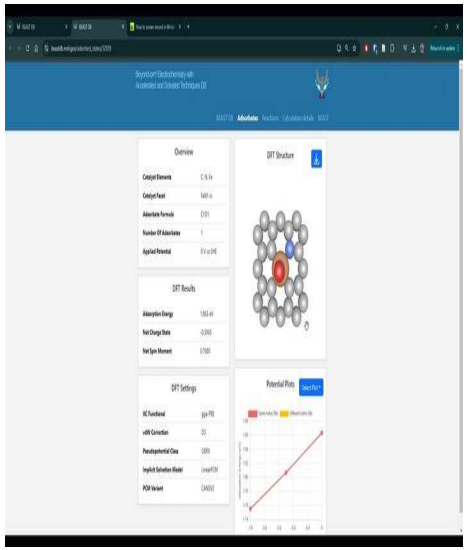
Catalyst Elements	Catalyst Facet	Adsorbate	Applied Potential (V vs SHE)	Adsorption Energy (eV)	Net Charge State
C N Fe	FeN1-o	C1O1	No Bias	1.872	0
C N Fe	FeN1-o	C1O1	-0.5	1.807	-1.274
C N Fe	FeN1-o	C1O1	0	1.863	-0.306
C N Fe	FeN1-o	C1O1	-1	1.756	-1.792
C N Fe	FeN2-i	C1O1	No Bias	1.864	0
C N Fe	FeN2-i	C1O1	-0.5	1.819	-0.471
C N Fe	FeN2-i	C1O1	0	2.046	0.224
C N Fe	FeN2-i	C1O1	-1	1.447	-1.806

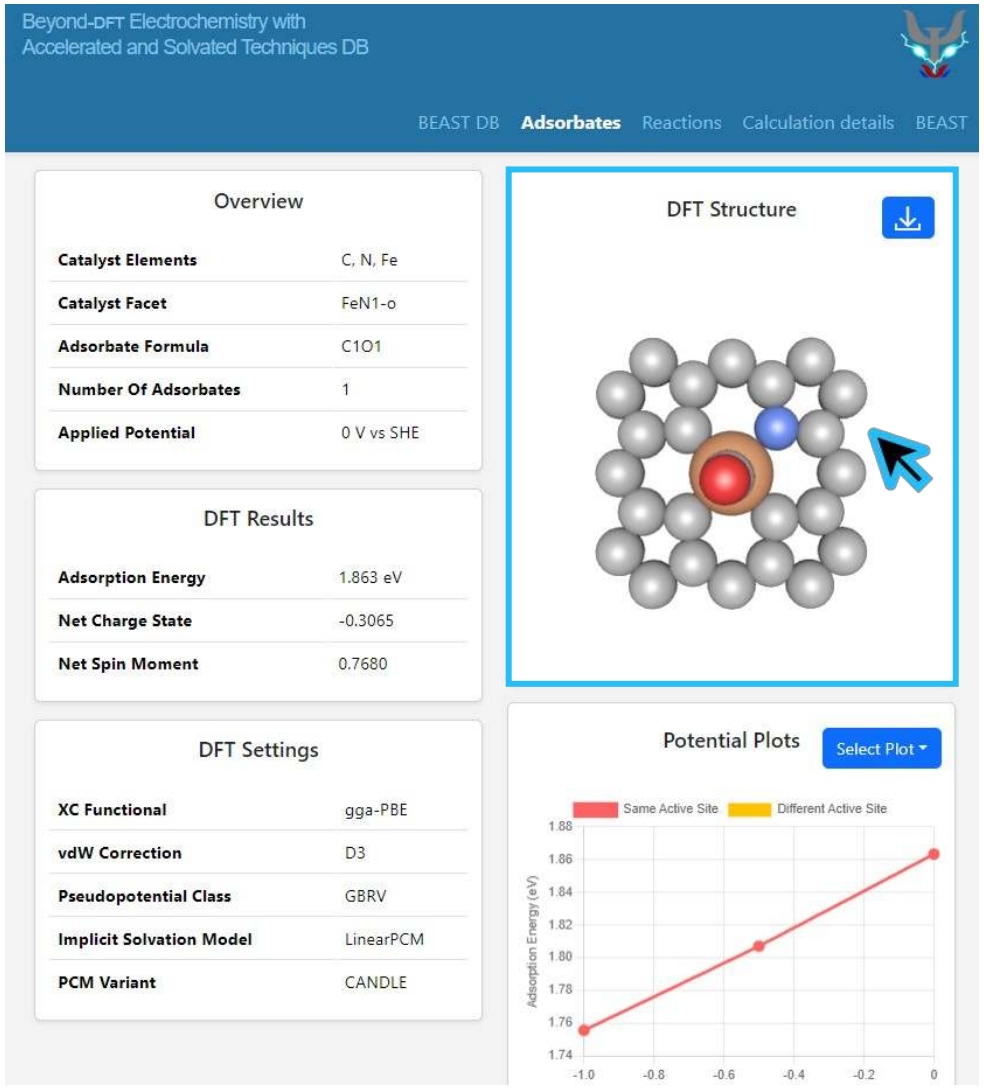


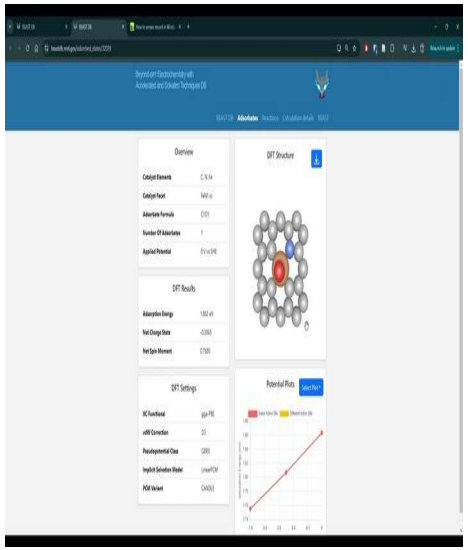
UI facilitates granular system understanding



UI facilitates granular system understanding





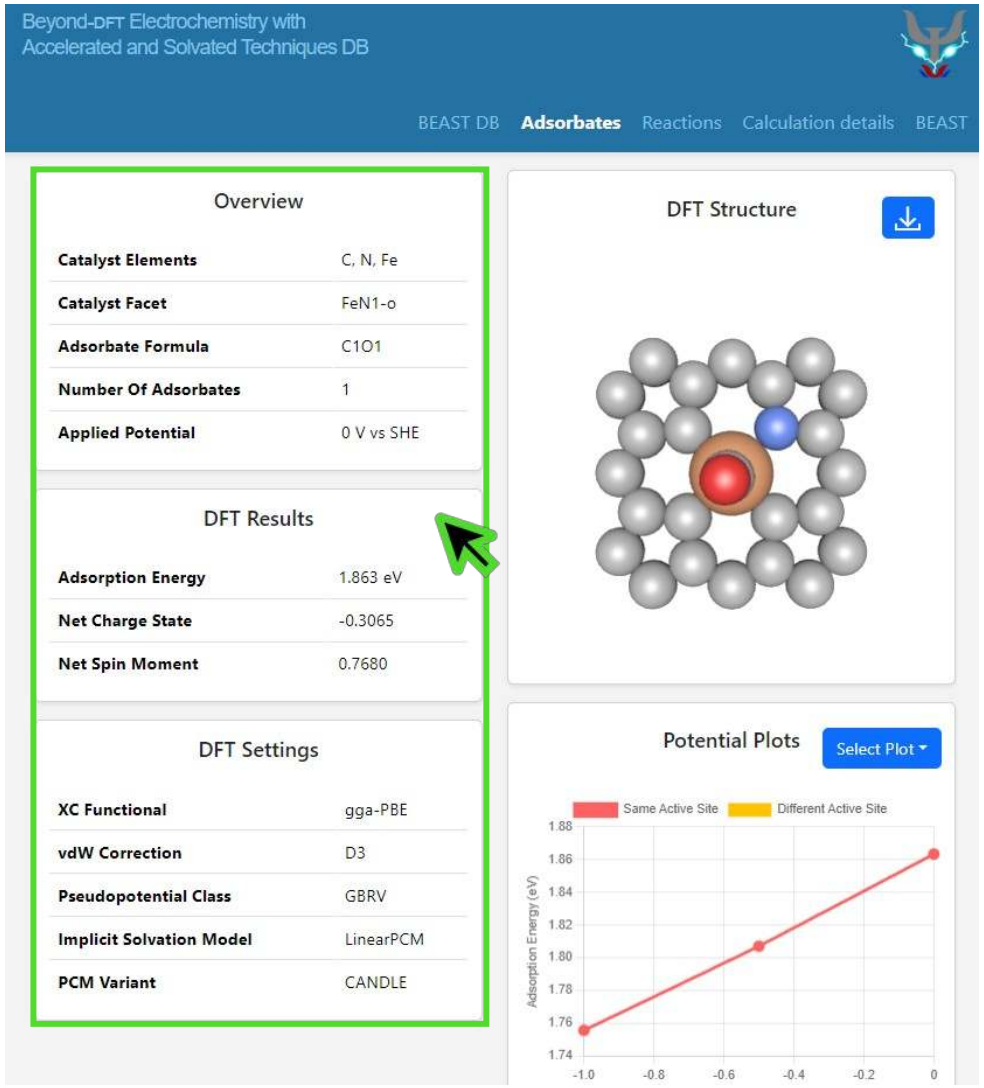


Overview	
Catalyst Elements	C, N, Fe
Catalyst Facet	FeN1-o
Adsorbate Formula	C1O1
Number Of Adsorbates	1
Applied Potential	0 V vs SHE

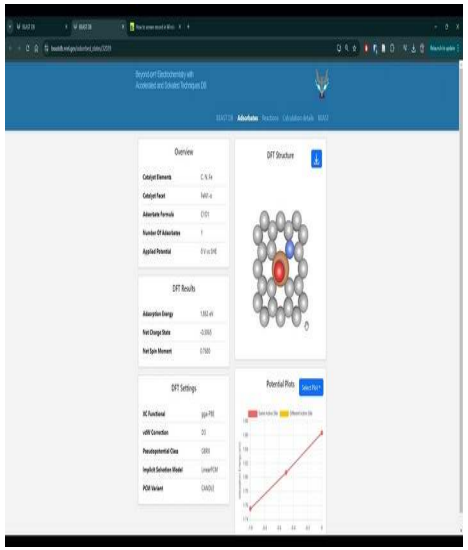
DFT Results	
Adsorption Energy	1.863 eV
Net Charge State	-0.3065
Net Spin Moment	0.7680

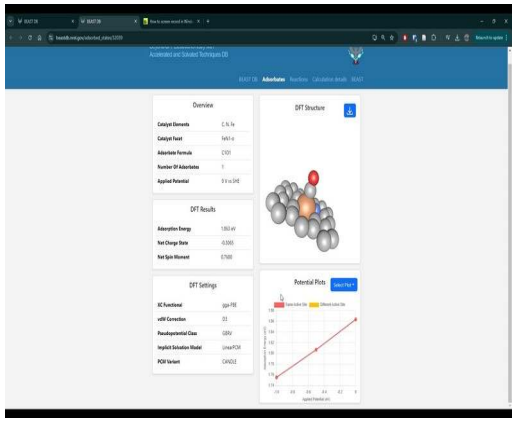
DFT Settings	
XC Functional	gga-PBE
vdW Correction	D3
Pseudopotential Class	GBRV
Implicit Solvation Model	LinearPCM
PCM Variant	CANDLE

UI facilitates granular system understanding



UI facilitates granular system understanding





Overview

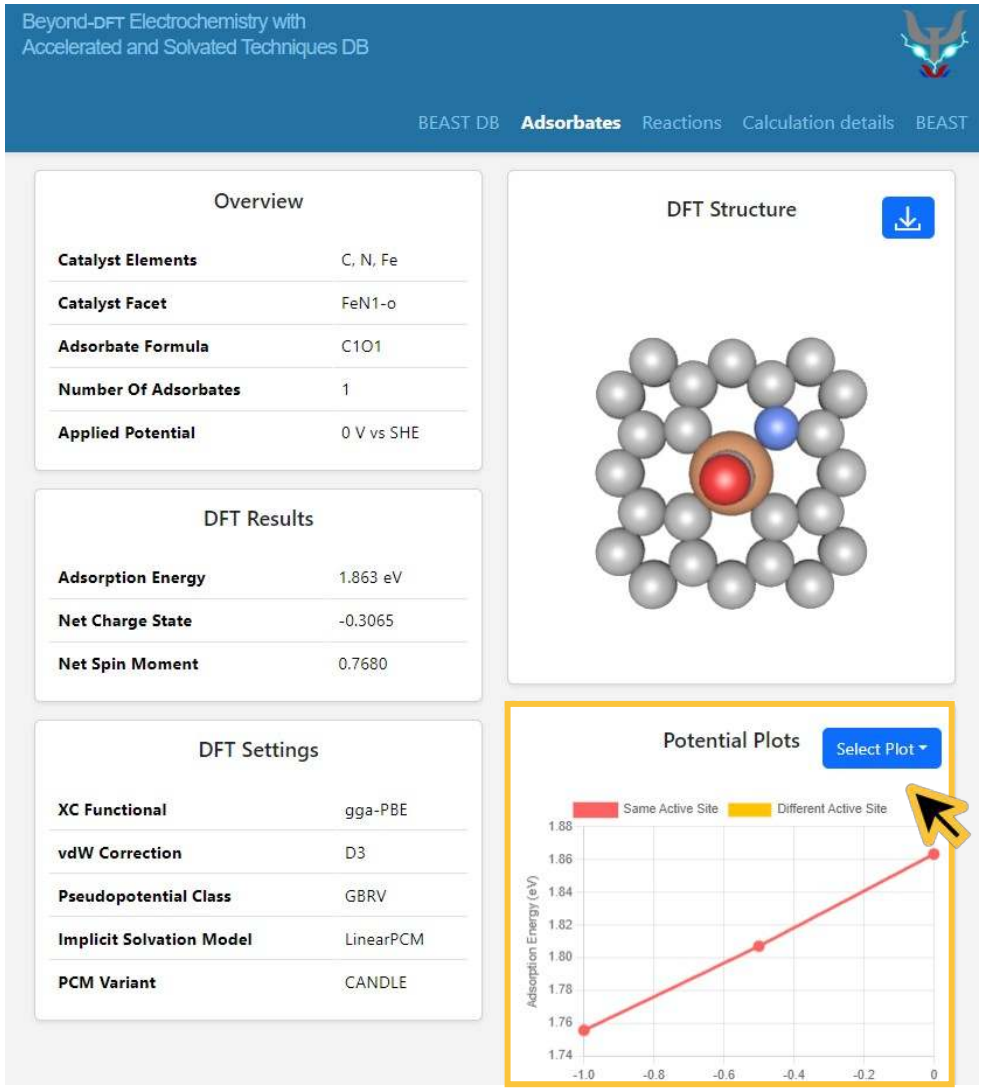
Catalyst Elements	C, N, Fe
Catalyst Facet	FeN1-o
Adsorbate Formula	C1O1
Number Of Adsorbates	1
Applied Potential	0 V vs SHE

DFT Results

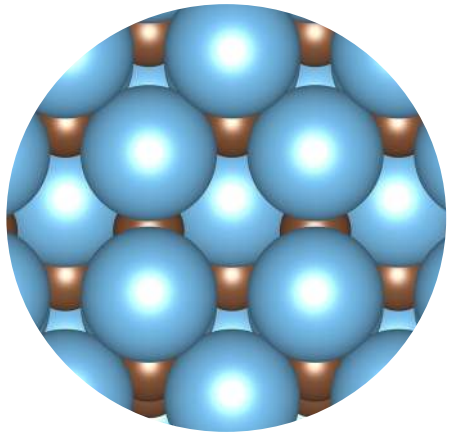
Adsorption Energy	1.863 eV
Net Charge State	-0.3065
Net Spin Moment	0.7680

DFT Settings

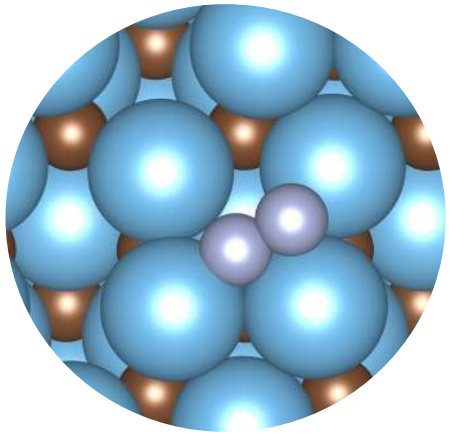
XC Functional	gga-PBE
vdW Correction	D3
Pseudopotential Class	GBRV
Implicit Solvation Model	LinearPCM
PCM Variant	CANDLE



Electronic structure shows potential-dependent N_2 adsorption

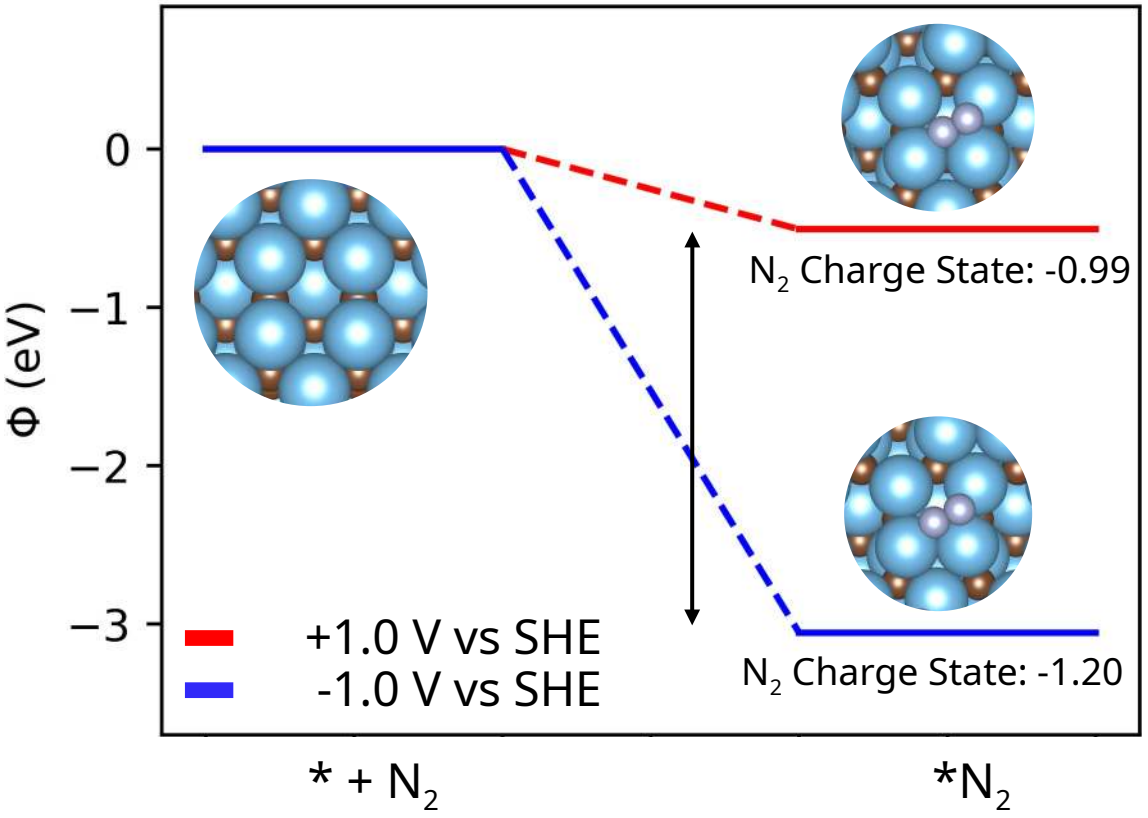


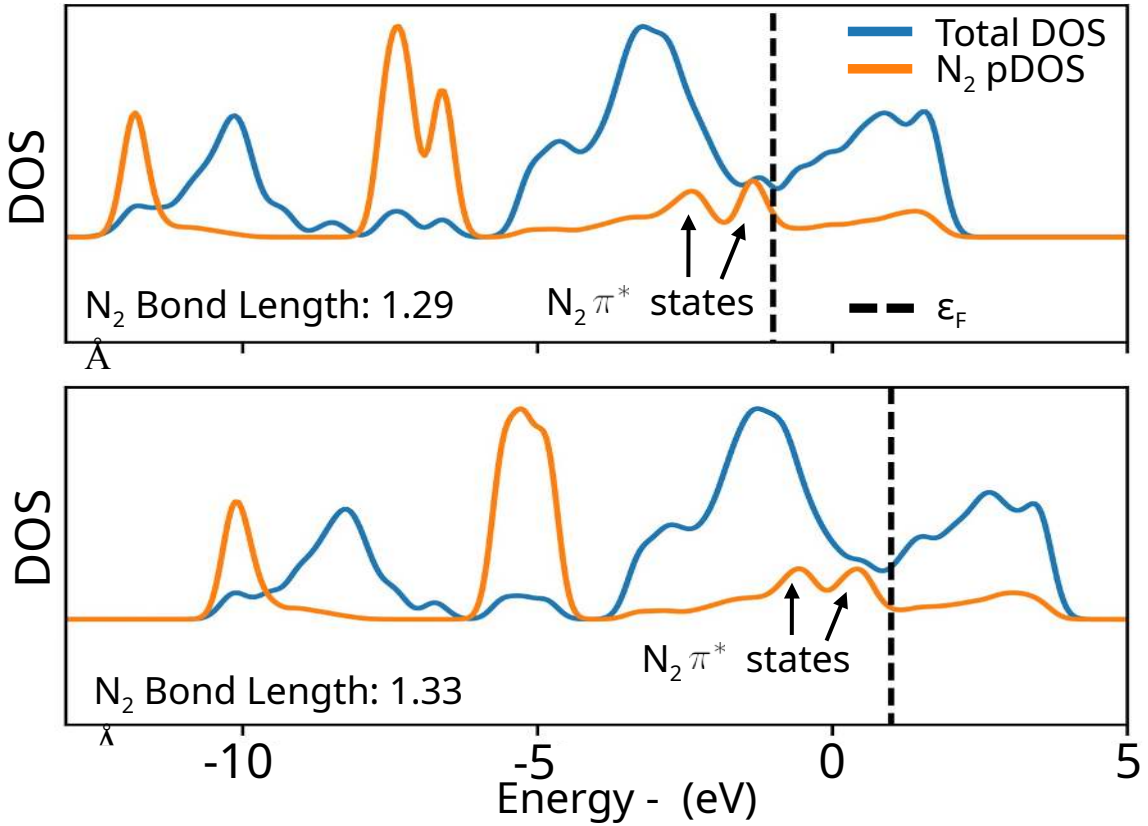
$* + N_2$



$*N_2$

Electronic structure shows potential-dependent N_2 adsorption



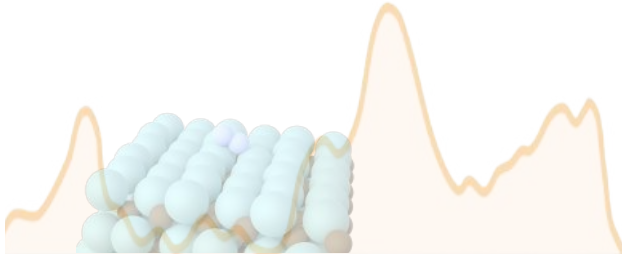


Our model predicts **potential-dependent N_2 adsorption** with partial charge transfer

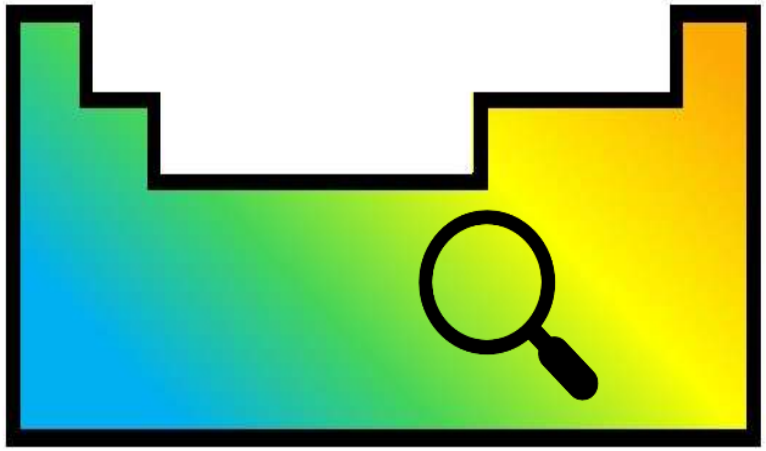
Database facilitates **rationalization** of fundamental **electrochemical processes**

Capabilities

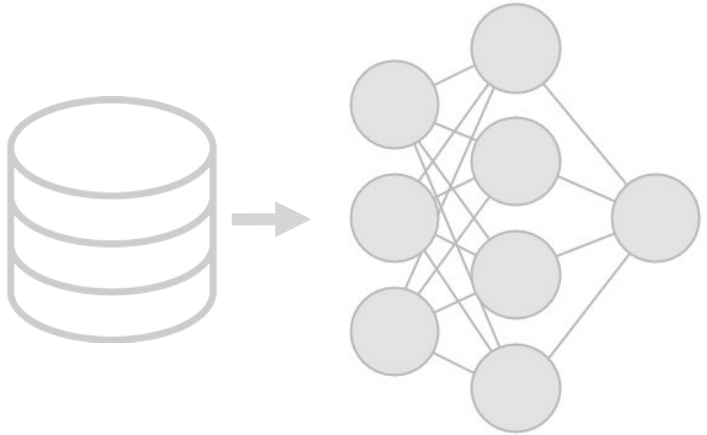
1. Individual system studies



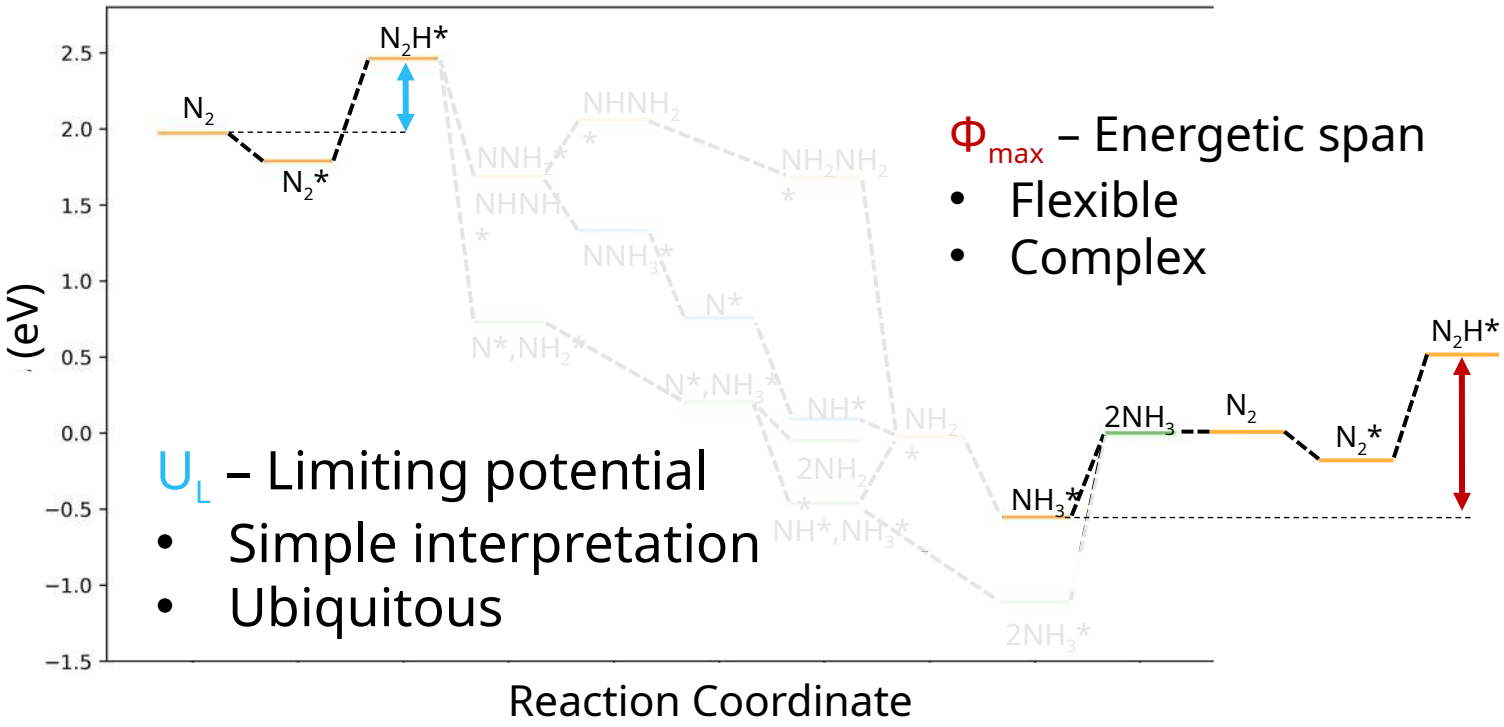
2. Material space screening



3. Statistical model training

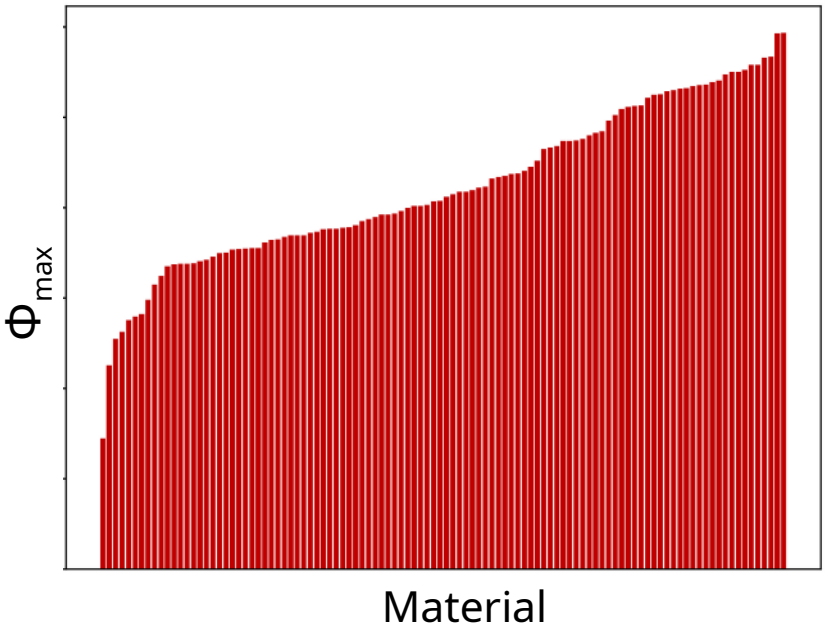


Thermodynamic activity descriptors



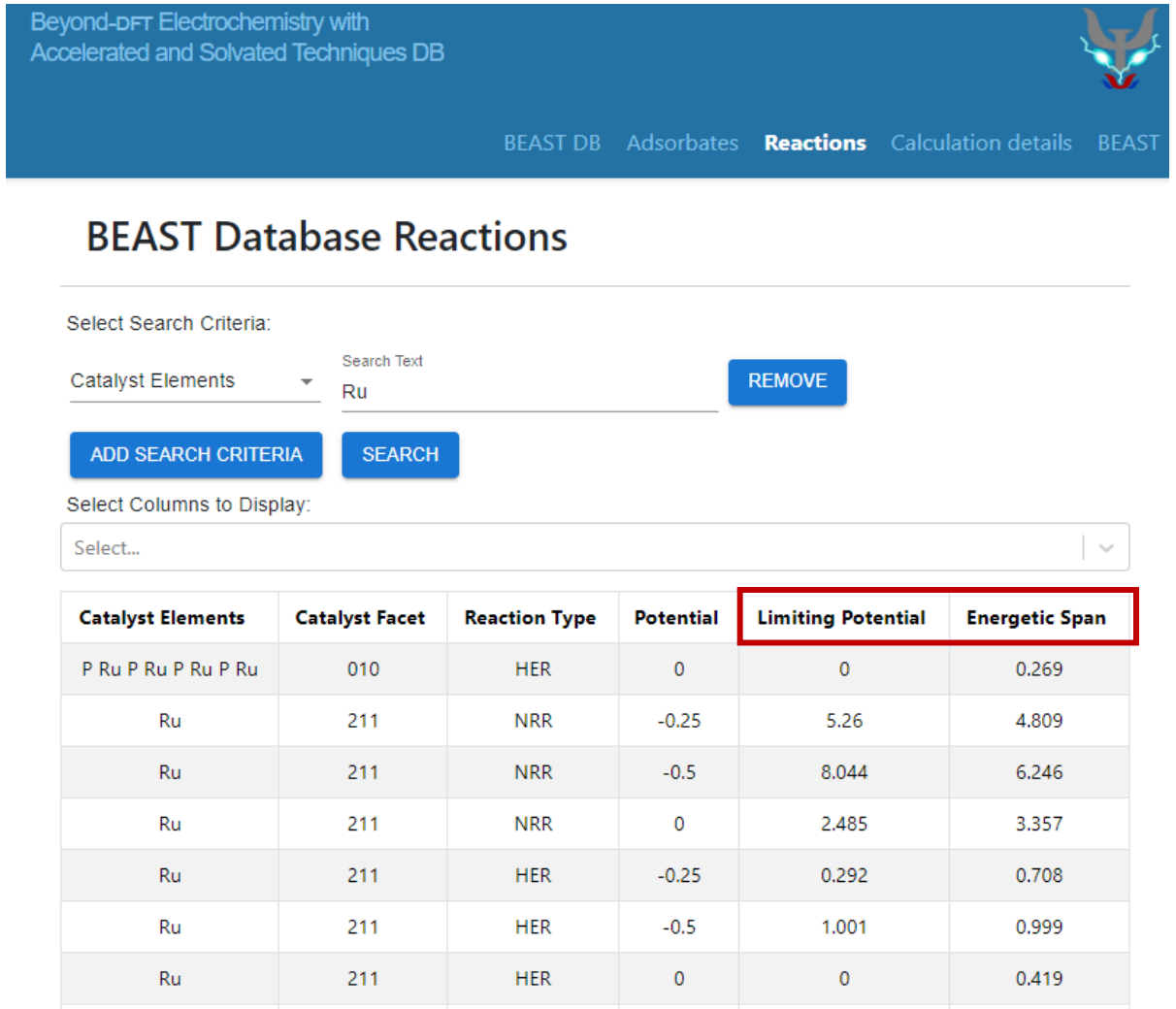
Φ_{max} – Energetic span
- Flexible
- Complex

U_L – Limiting potential
- Simple interpretation
- Ubiquitous



Complex mechanisms require cheap descriptors to sort large material spaces

Reaction pathway visualization



Users can sort materials by descriptors

Reaction pathway visualization

Beyond-DFT Electrochemistry with
Accelerated and Solvated Techniques DB

BEAST DB | Adsorbates | **Reactions** | Calculation details | BEAST

BEAST Database Reactions

Select Search Criteria:

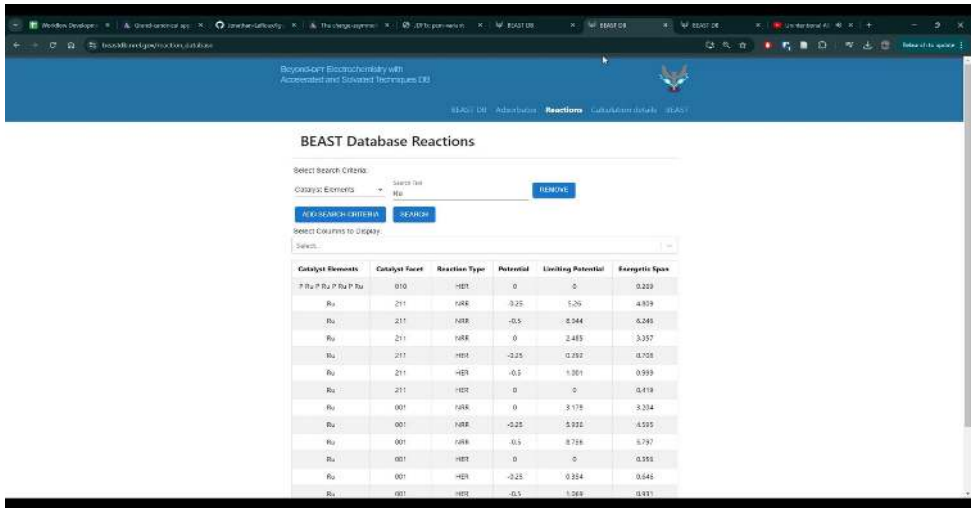
Catalyst Elements | Search Text: Ru | REMOVE

ADD SEARCH CRITERIA | SEARCH

Select Columns to Display:

Select...

Catalyst Elements	Catalyst Facet	Reaction Type	Potential	Limiting Potential	Energetic Span
P Ru P Ru P Ru P Ru	010	HER	0	0	0.269
Ru	211	NRR	-0.25	5.26	4.809
Ru	211	NRR	-0.5	8.044	6.246
Ru	211	NRR	0	2.485	3.357
Ru	211	HER	-0.25	0.292	0.708
Ru	211	HER	-0.5	1.001	0.999
Ru	211	HER	0	0	0.419



Reaction pathway visualization

Beyond-DFT Electrochemistry with
Accelerated and Solvated Techniques DB

BEAST DB | Adsorbates | **Reactions** | Calculation details | BEAST

BEAST Database Reactions

Select Search Criteria:

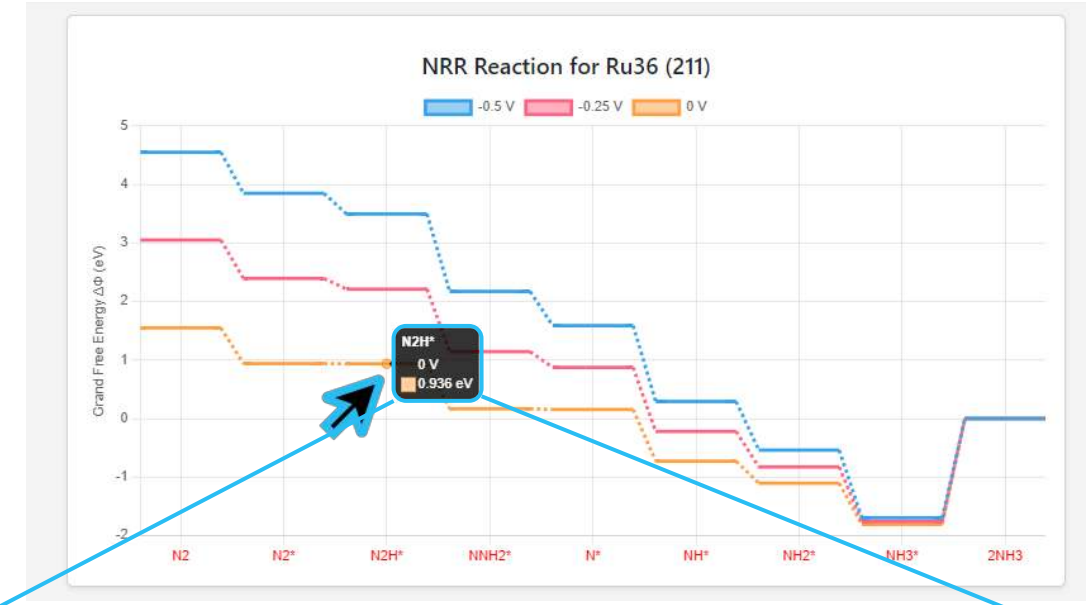
Catalyst Elements | Search Text: Ru | REMOVE

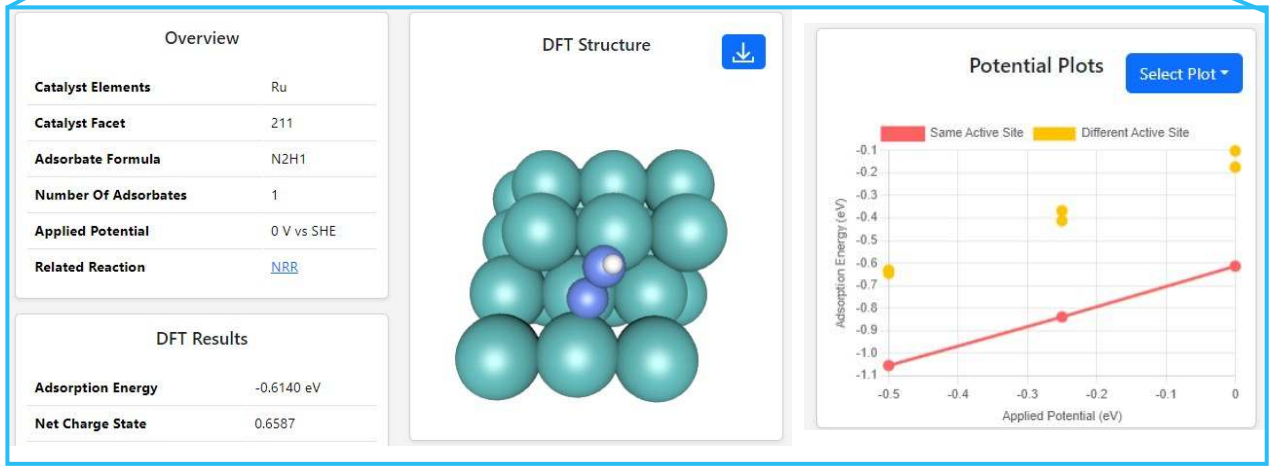
ADD SEARCH CRITERIA | SEARCH

Select Columns to Display:

Select...

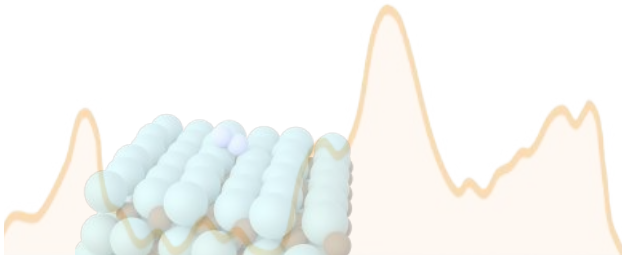
Catalyst Elements	Catalyst Facet	Reaction Type	Potential	Limiting Potential	Energetic Span
P Ru P Ru P Ru P Ru	010	HER	0	0	0.269
Ru	211	NRR	-0.25	5.26	4.809
Ru	211	NRR	-0.5	8.044	6.246
Ru	211	NRR	0	2.485	3.357
Ru	211	HER	-0.25	0.292	0.708
Ru	211	HER	-0.5	1.001	0.999
Ru	211	HER	0	0	0.419





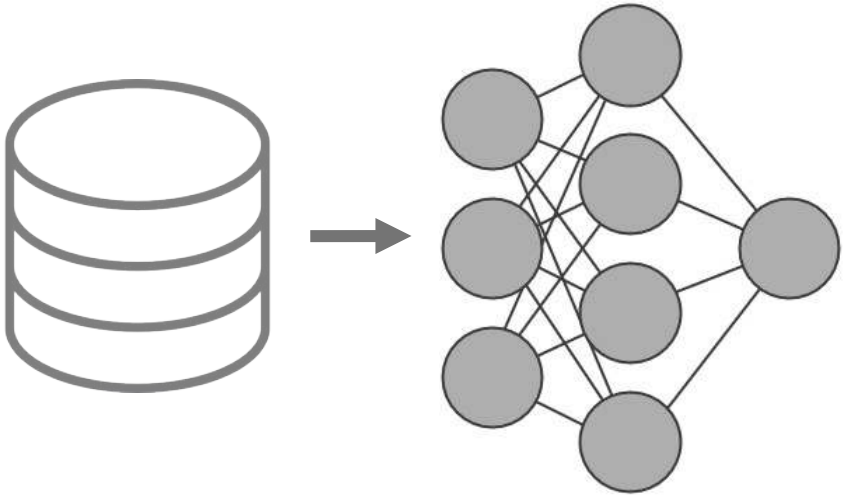
Capabilities

1. Individual system studies



2. Material space screening

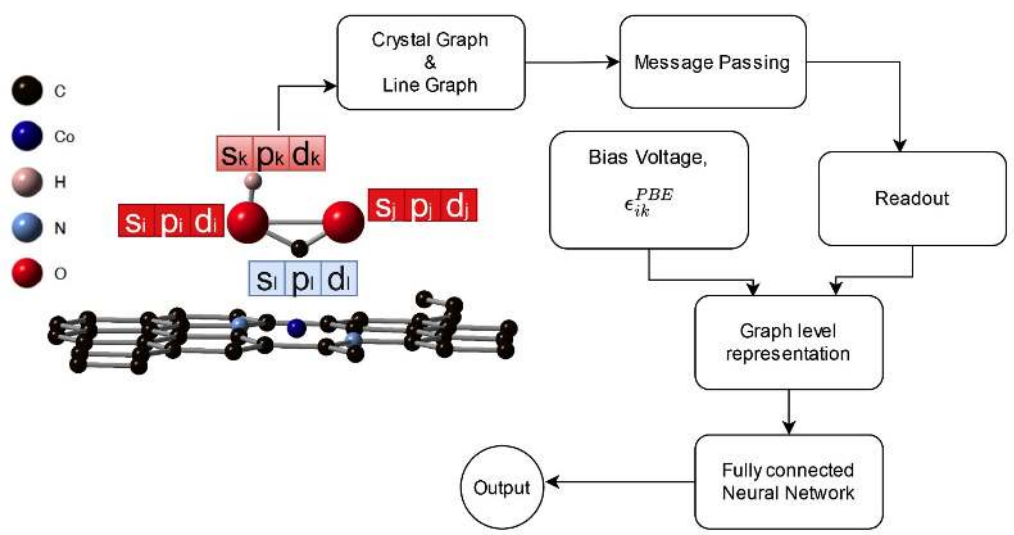
3. Statistical model training

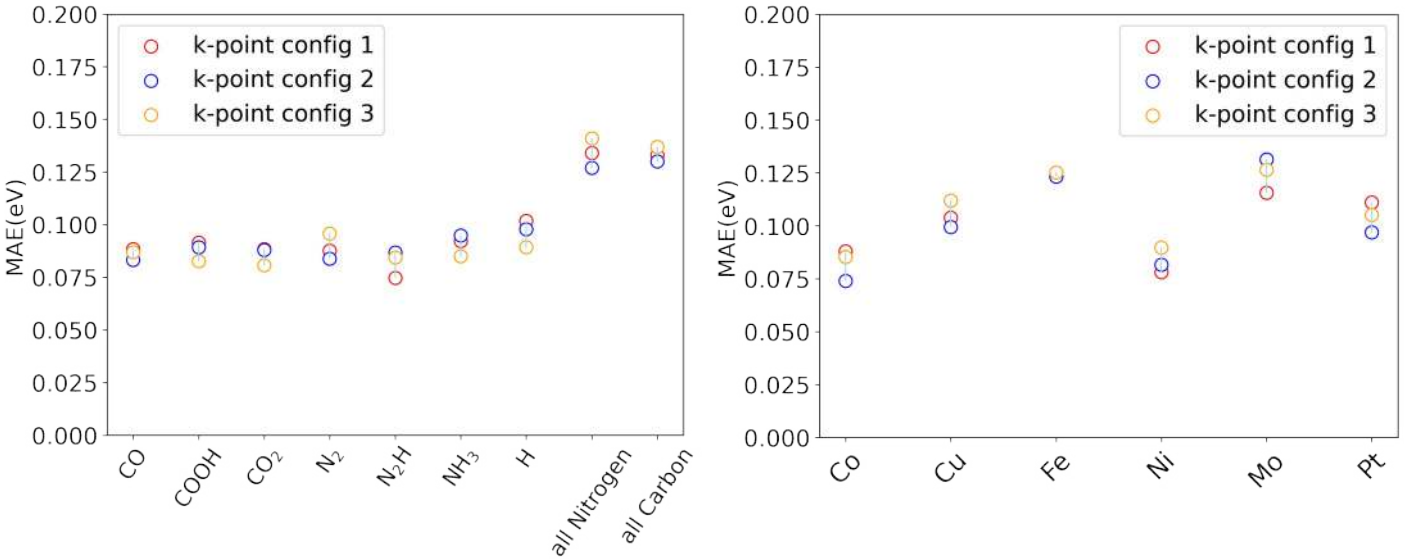


Learned HSE eigenvalues with PBE data at constant voltage

ALIGNN graph neural network architecture
- PBE atomic band projections are node features
- Voltage and PBE eigenvalues are global features
- Model output is HSE eigenvalues

	Default ALIGNN	Only orbital projections	orbital projections, eigenvalues and voltage
Average MAE (eV)	3.67	0.32	0.09





Choudhary, K.; et al. Atomistic Line Graph Neural Network for Improved Materials Property Predictions.

Model generalizes well outside of adsorbates/metals seen in training set

Future database developments

Adoption of pymatgen ecosystem of workflow code (immediate priority)

Jobflow pymatgen Custodian

User submission acceptance (1 year)

Greater material space and reactions coverage (ongoing)

- Surface speciation data (immediate priority)
- More complex mechanisms of existing data (1 year)

Acknowledgements

U.S. DEPARTMENT OF ENERGY | Office of Science

NREL
NATIONAL RENEWABLE ENERGY LABORATORY

Database Developers:

Joshua Quinton – RPI

Rachel Hurst – NREL

Struan Clark – NREL

Jacob Clary – NREL

Advisors:

Charles Musgrave – CU Boulder

Derek Vigil-Fowler – NREL

Ravishankar Sundararaman – RPI

Colleagues:

Sophie Gerits – CU Boulder

Benjamin Rich– CU Boulder