

ACS, August 22-23, 2024 **Cooper Tezak** 

**BEAST Workshop** 



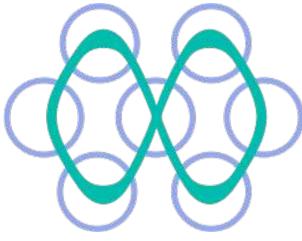
University of Colorado **Boulder** 

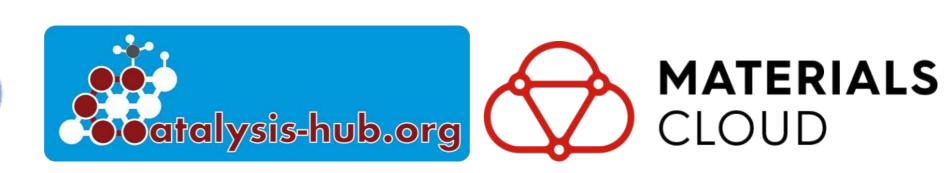






## Materials databases have accelerated materials science





#### **Materials Project**

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

#### **Catalysis-hub**

- Adsorption energies
- Barriers
- Some electrocatalysis

#### **Materials Cloud**

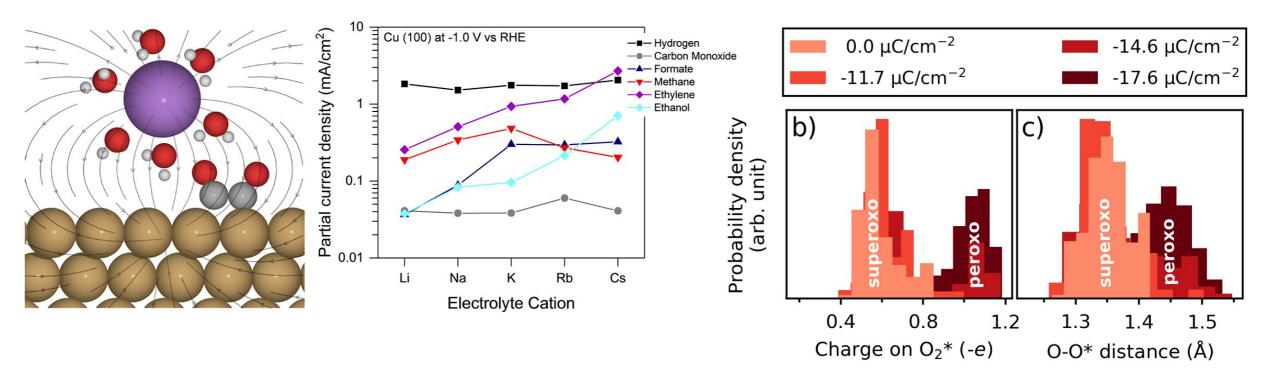
- Specific material space archives
- Standardized workflow codes
- Quantum code benchmarks



## Electrocatalysis has unique complexity

#### **Solvation environment**

**Applied potential** 

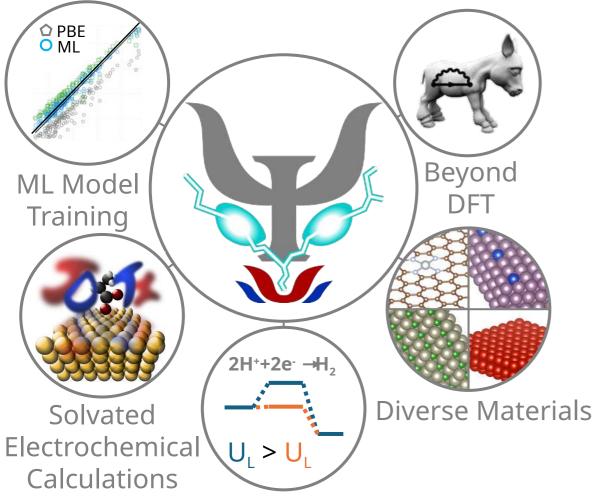


## The computational community must embrace the complexities of electrocatalysis to accelerate development

Resasco, J.; et al. Promoter Effects of Alkali Metal Cations on the Dudzinski, A. M.; et al. First Step of the Oxygen Reduction Reaction on Electrochemical Reduction of Carbon Dioxide. Metal/Water Interface

## Introducing BEAST DB





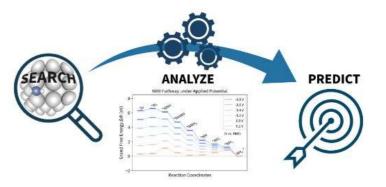
Beyond-DFT Electrochemistry with Accelerated and Solvated Techniques DB



BEAST DB Adsorbates Reactions Calculation details BEAST

#### Welcome to BEAST DB!

Enabling rational understanding and design of electrocatalysts through state-of-the-art theory



The Beyond-DFT Electrochemistry with Accelerated and Solvated Techniques Database, or BEAST DB, contains thousands of electrocatalysis calculations and visualizations of properties that help rationalize observed behavior and design more effective catalysts.

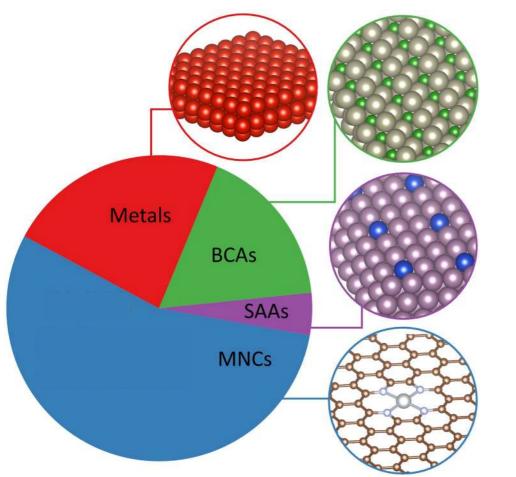
### Open access user interface

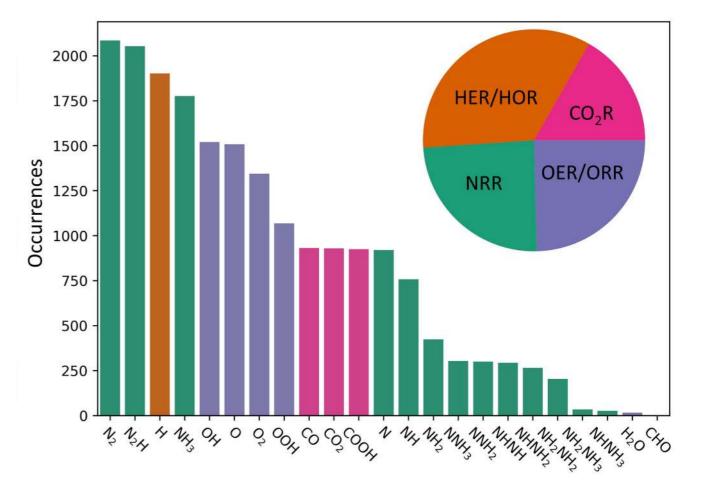
#### **Reaction Analysis**



Chemical and Biological Tezak, C. et al. BEAST DB: Grand-Canonical Database of Electrocatalyst Engineering Properties. <u>https://doi.org/10.48550/arXiv.2405.20239</u>. *Under review* 

## Database overview





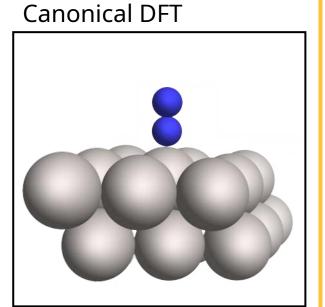
20,000 Surface Calculations

Common reactions coverage



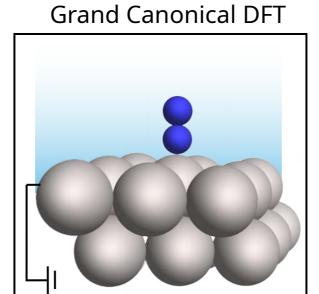
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## We use electrochemical methodology



- Fixed charge
- Potential correction schemes

 $F_{DFT}$ 



- Fixed potential
- Self-consistent free energies

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

#### Accurate solvation models

Solvation Model	рКw
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

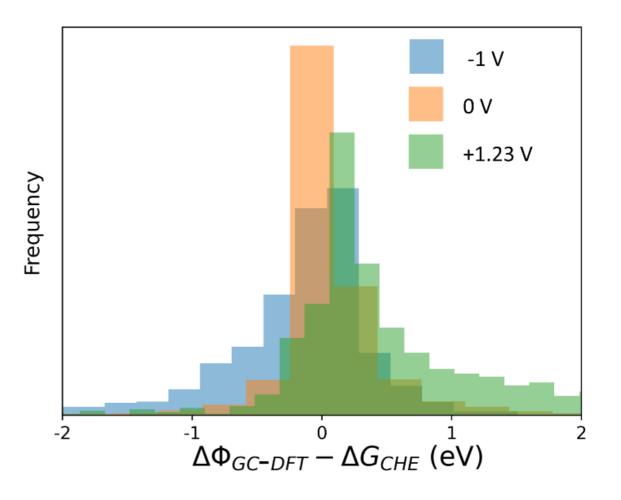
Canonical DFT relies on the computational hydrogen electrode (CHE)

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

Limitations:

- Proton-coupled electron transfers only
- Linear potential dependence

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

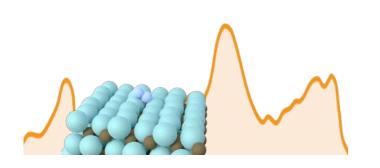




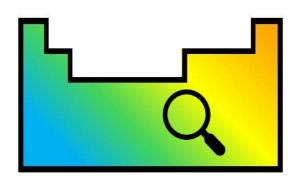
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## Capabilities

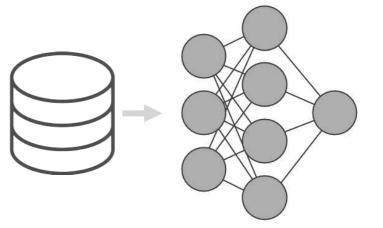
1. Individual system studies



2. Material space screening



3. Statistical model training

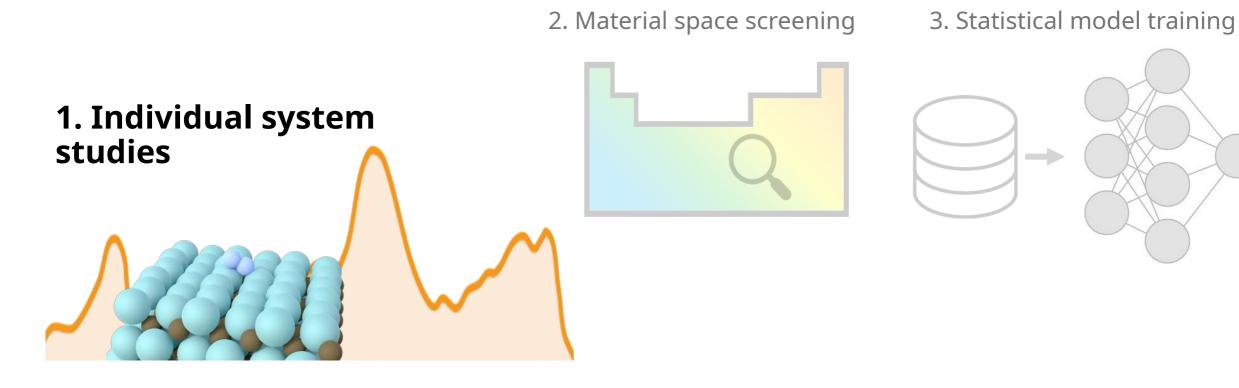




Chemical and Biological Engineering

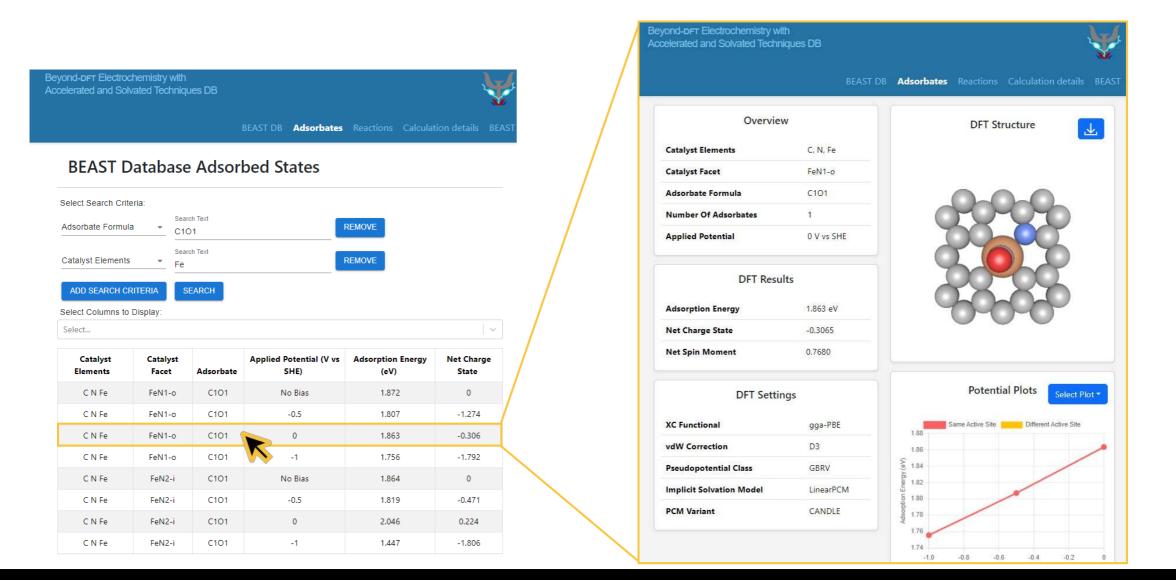
Capabilities

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Engineering

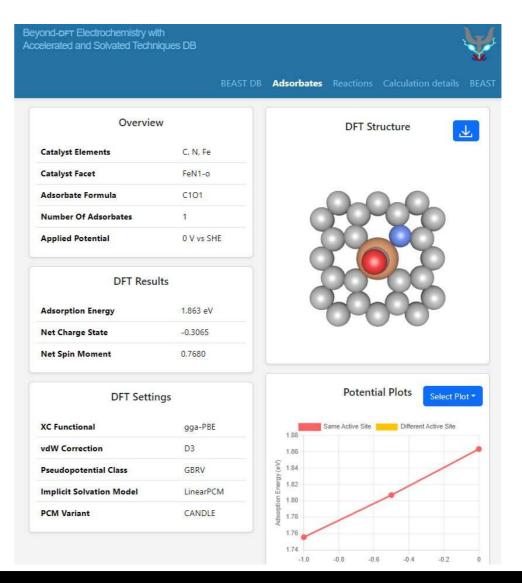
#### UI facilitates granular system understanding

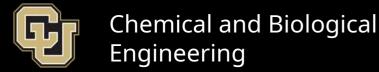




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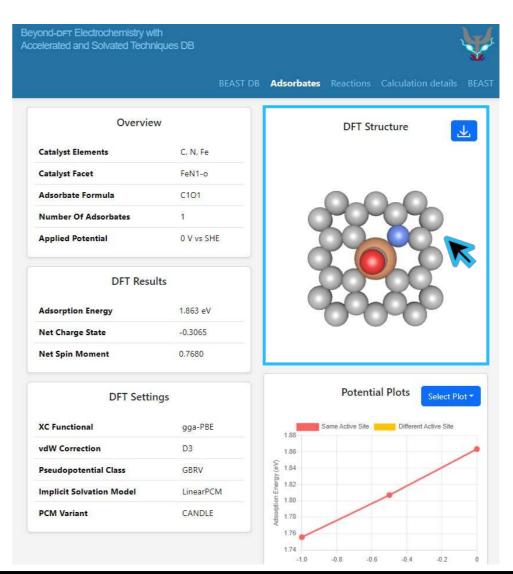
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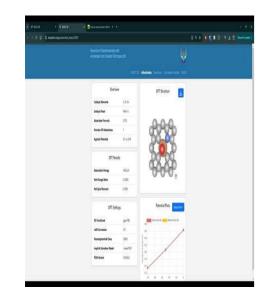


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#### UI facilitates granular system understanding



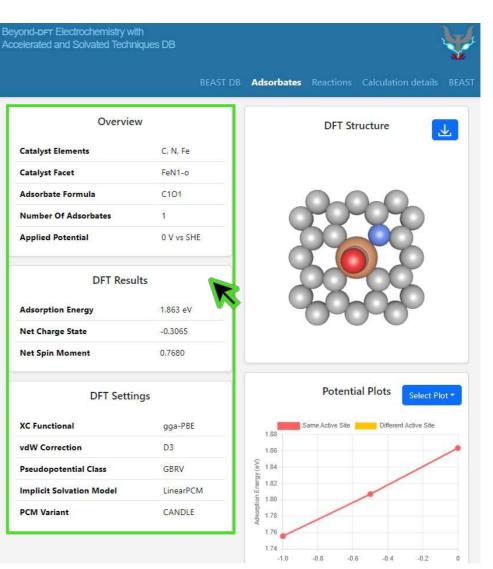
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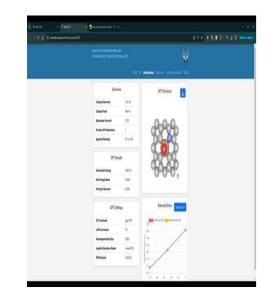
Overview	,
Catalyst Elements	C, N, Fe
Catalyst Facet	FeN1-o
Adsorbate Formula	C1O1
Number Of Adsorbates	1
Applied Potential	0 V vs SHE
DFT Result	ts
Adsorption Energy	1.863 eV
Net Charge State	-0.3065
Net Spin Moment	0.7680
DFT Setting	gs
XC Functional	gga-PBE
vdW Correction	D3
Pseudopotential Class	GBRV
Implicit Solvation Model	LinearPCM
PCM Variant	CANDLE

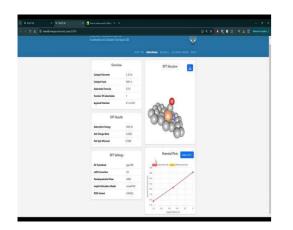
Overview

#### UI facilitates granular system understanding





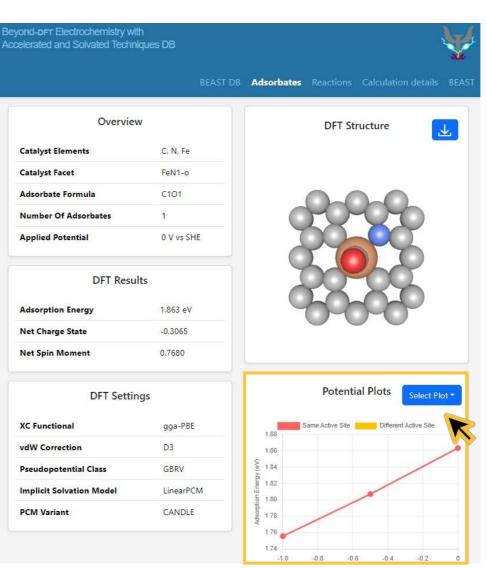




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Pseudopotential Class Implicit Solvation Model	GBRV LinearPCM

Overview

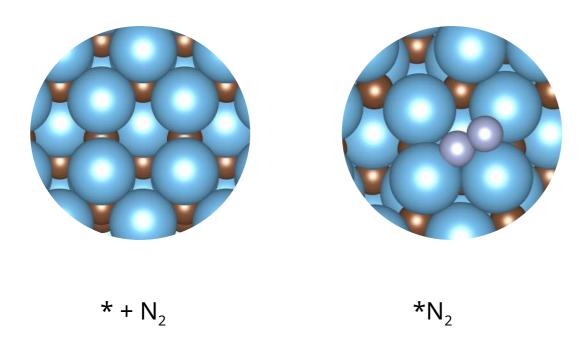
#### UI facilitates granular system understanding



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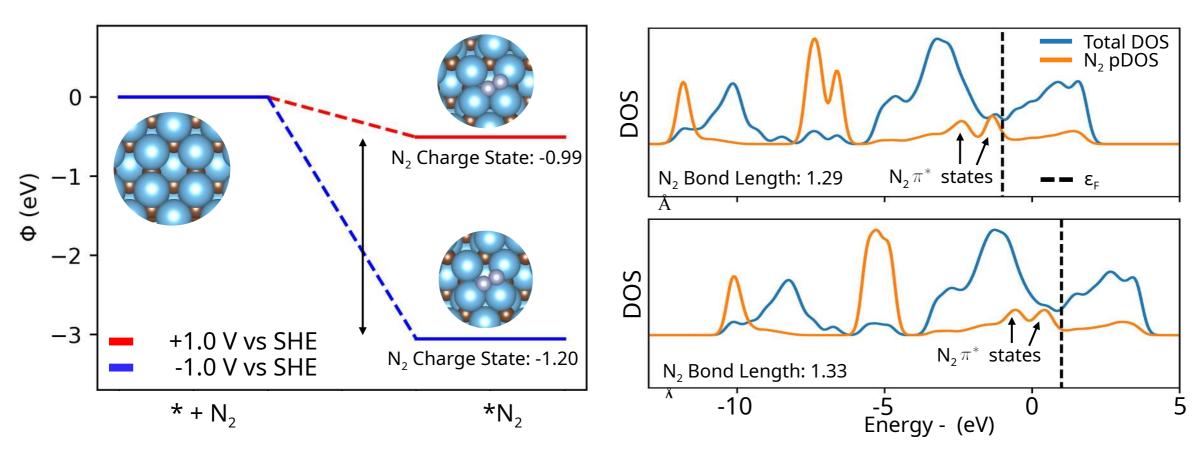
5,

## Electronic structure shows potential-dependent N<sub>2</sub> adsorption





Electronic structure shows potential-dependent N<sub>2</sub> adsorption



Our model predicts **potential-dependent** N<sub>2</sub> **adsorption** with partial charge transfer Database facilitates **rationalization** of fundamental **electrochemical processes** 

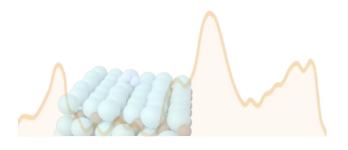


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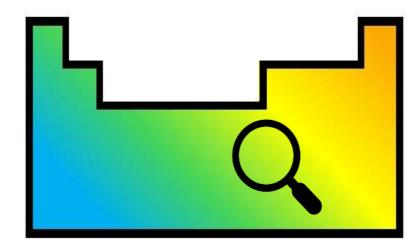
## Capabilities

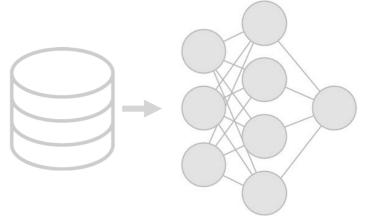
1. Individual system studies

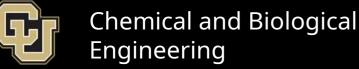
3. Statistical model training



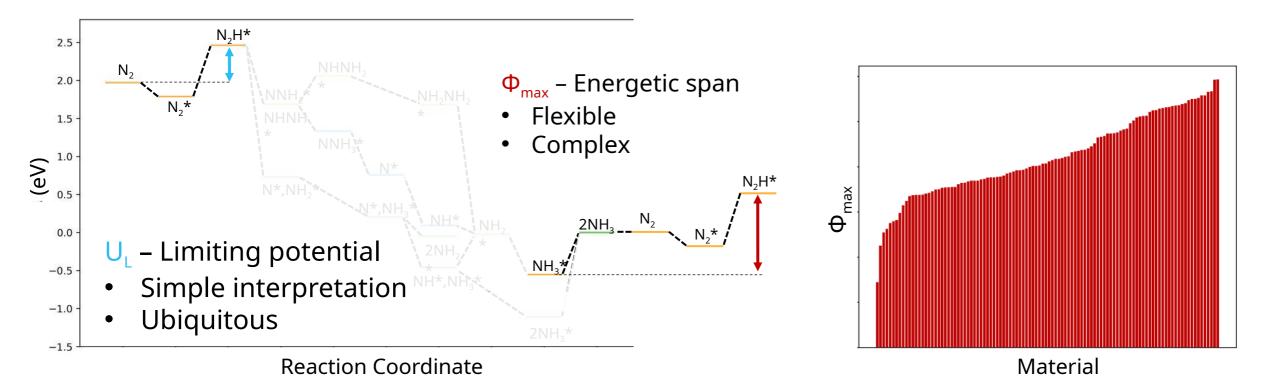
2. Material space screening







## Thermodynamic activity descriptors



Complex mechanisms require cheap descriptors to sort large material spaces



## Reaction pathway visualization

Beyond-DFT Electrochemistry with Accelerated and Solvated Techniques DB BEAST DB Adsorbates Reactions Calculation details BEAST

#### **BEAST Database Reactions**

elect Search Criteria: Catalyst Elements ADD SEARCH CRITER elect Columns to Disp Select				REMOVE	~
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

Users can sort materials by descriptors



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## Reaction pathway visualization

Beyond-DFT Electrochemistry with Accelerated and Solvated Techniques DB

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BEAST DB Adsorbates Reactions Calculation details BEAST

#### **BEAST** Database Reactions

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	Ra	211	NR	-0.5	2.944	6.245					
	Pro	211	1485	0	2.485	3.357					
	Bu	211	HER	-2.35	0.292	41708					
	Ð <sub>2</sub>	211	HER	-0.5	1.001	0.999					
	Ru.	211	127	0	0	0,418					
	Bu	001	NAR.	0	3.178	3.204					
	Ru	001	NR	-0.25	200	4.595					
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	Ra	021	HER	D	۰	0.551					
	Ro	001	HER	-3.25	0354	0.646					
	Ra	001	HER	-0.5	1.049	11.913					



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## Reaction pathway visualization

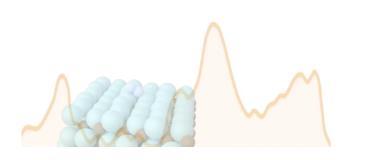
NRR Reaction for Ru36 (211) Beyond-DFT Electrochemistry with -0.5 V \_\_\_\_\_ -0.25 V \_\_\_\_\_ 0 V Accelerated and Solvated Techniques DB Reactions Calculation details BEAST BEAST DB Adsorbates (No) ΦØ **BEAST Database Reactions** N2H\* 80 0 V nd F 0.936 eV Select Search Criteria: ö Search Text REMOVE Catalyst Elements -1 Ru ADD SEARCH CRITERIA SEARCH N2 N2\* N2H<sup>a</sup> NNH2<sup>1</sup> NH2\* 2NH3 Select Columns to Display: Select...  $\sim$ Overview **DFT Structure** 1 **Catalyst Elements Catalyst Facet Limiting Potential Energetic Span** Reaction Type Potential **Potential Plots** Select Plot 🔻 **Catalyst Elements** Ru P Ru P Ru P Ru P Ru 010 HER 0 0 0.269 **Catalyst Facet** 211 Same Active Site Different Active Site -0 1 Adsorbate Formula N2H1 Ru 211 NRR -0.25 5.26 4.809 -0.2 Number Of Adsorbates 1 -0.3 -0.4 Ru 211 NRR -0.5 8.044 6.246 **Applied** Potential 0 V vs SHE -0.5 -0.6 **Related Reaction** NRR Ru 211 NRR 0 2.485 3.357 -0.7 -0.8 211 HER -0.25 0.292 Ru 0.708 -0.9 **DFT Results** -1.0 -1.1 HER Ru 211 -0.5 1.001 0.999 -0.6140 eV **Adsorption Energy** -0.5 -0.4 -0.3 -0.2 -0.1 Applied Potential (eV) 0.6587 Net Charge State Ru 211 HER 0 0 0.419



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## Capabilities

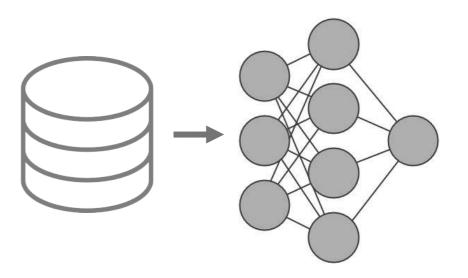
1. Individual system studies



2. Material space screening



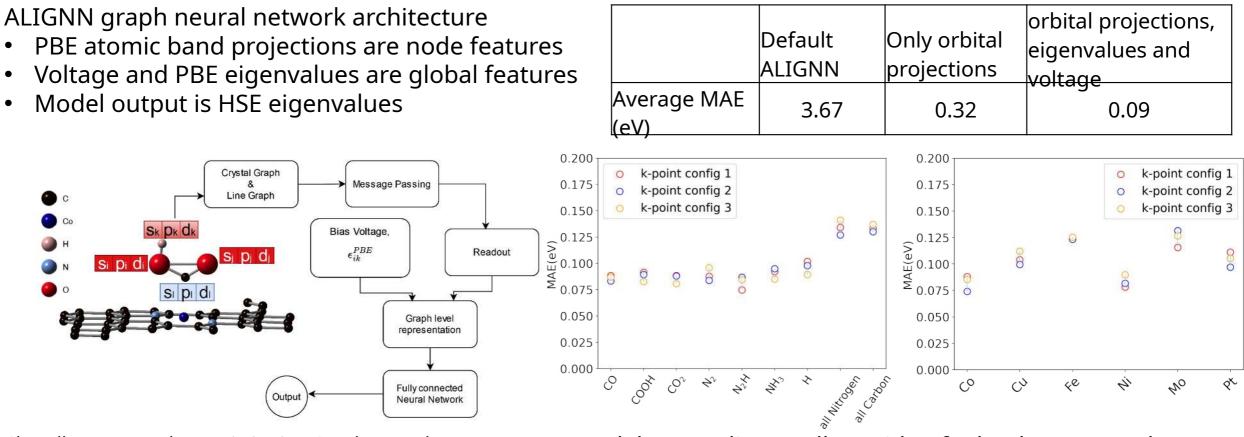
#### 3. Statistical model training





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## Learned HSE eigenvalues with PBE data at constant voltage



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 pymolgen 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





#### **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

