

BEAST DB

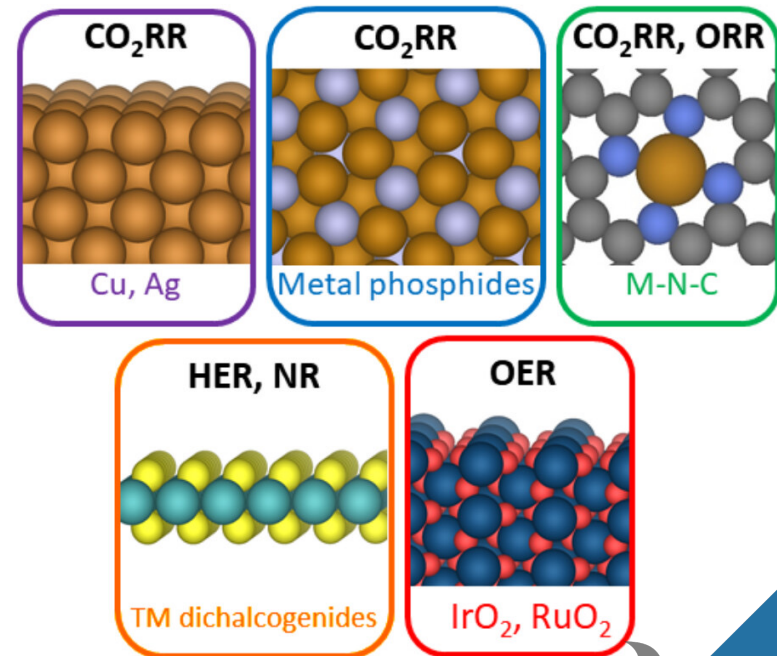
Derek Vigil-Fowler

2nd Annual BEAST workshop

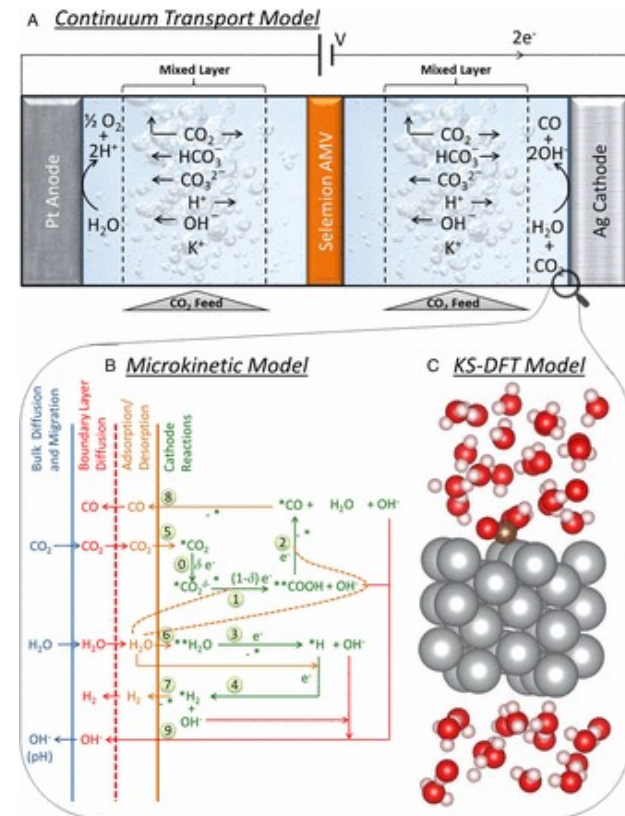
08/18/23



- Beyond-DFT Electrochemistry with Accelerated and Solvated Techniques Database
- Will have adsorption energies, other descriptors for range of common electrocatalysts, chemistries



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Electrochemistry is hard!



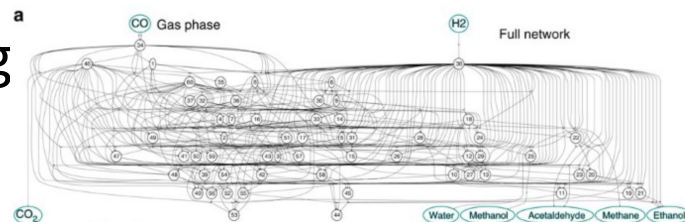
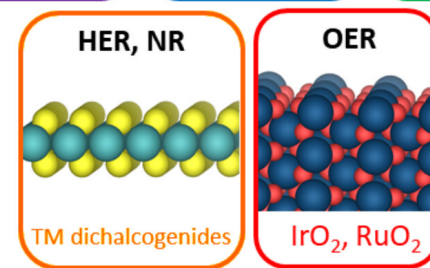
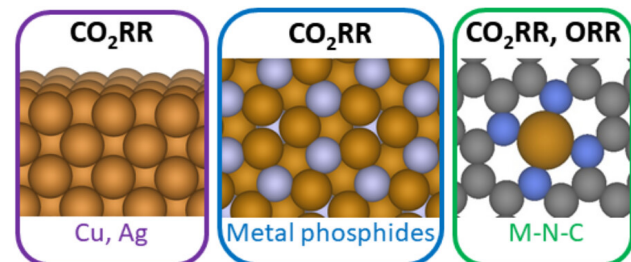
Electrochemistry is hard!

- Diversity and dynamics



Electrochemistry is hard!

- Diversity and dynamics
 - Combinatorial space of elements
 - Bulk vs reduced dimensional
 - Identity of active site, mechanism
 - Surface structure and reconstruction
 - Defects
 - Complex reaction pathways involving spectator species
 - Electrolyte identity, concentration
 - pH
 - Applied potential
 - Kinetic and thermodynamic considerations
 - Transport/morphology
 -



~100 species

~200 reactions

>2,000 pathways



Result: chemical intuition plays big role



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- Often unclear what governs choices



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- Often unclear what governs choices
- Example: CO₂ reduction on Sn/SnO

ACS Catal. 2015, 5, 5, 3148–3156

Conditions:

- Sn thin films;
- Potentials: –1 to –2.4 vs Ag/AgCl
- 0.1 M K₂SO₄ electrolyte
- pH = 4.4

ACS Catal. 2017, 7, 7, 4822–4827

Conditions:

- Metallic Sn foil; pretreatment removed excess oxide from the surface, leaving only a native oxide
- Potentials: –0.4 to –1.3 V vs. RHE
- 0.1 M KHCO₃ electrolyte
- pH = 6.8

ACS Catal. 2015, 5, 12, 7498–7502

Conditions:

- SnO₂ NPs on graphene oxide
- Potentials: –0.25 to –1.5 vs Ag/AgCl
- NaOH
- pH = 8.5 – 12 (alkaline conditions avoid SnO₂ reduction)

credit: Carrie Farberow



Experimental efforts on protocols



Suggested Testing Procedure for Round Robin Matrix

- Cell Assembly and Conditioning as specified. Test start.

- Measure CV and EIS in H_2/N_2 at BOT as specified.
 - Flow O_2 on cathode. Wait until OCV is stable (e.g. 5 min).
 - Measure activity in H_2/O_2 as specified.
 - Flow air on cathode. Wait until OCV is stable (e.g. 5 min).
 - Measure pol curve in H_2 /air as specified.
 - Change T, RH, and P conditions for next test. Wait at least 30 min for equilibration.
 - Measure pol curve in H_2 /air at 250 kPa, 75% RH, 95 °C as specified.
 - Flow N_2 on cathode and set back T = 80C, P=150 kPa, RH=100%. Wait until OCV decreases to ~0.1 V or for at least 20 min. If OCV does not decrease enough, reduce the residual O_2 adsorbed on the catalyst, in order to have a CV shape “centered” around 0 current.
 - Measure CV and EIS in H_2/N_2 at BOT again (CV and EIS may vary after measuring the first pol curve due to full hydration of CL and ionomer due to water generation).
 - Flow air on cathode. Wait until OCV is stable (e.g. 5 min).
-
- **Start AST cycling up to 100 cycles** (should take ~10 min → helpful to set cathode N_2 purge delay). When finished (or asap after finishing) start to flow N_2 on cathode in case the test is going to be paused for long time (e.g. overnight) to not leave the cell under OCV conditions for too long.
 - Flow N_2 on cathode. Wait until OCV decreases to ~0.1 V or for at least 20 min. If OCV does not decrease enough, reduce the residual O_2 adsorbed on the catalyst, in order to have a CV shape “centered” around 0 current.
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Why build a computational electrochemistry database?

- Quick access to structures, energies, properties of catalysts for researchers working on catalysts in database (or related)
 - Increases ability to quickly get started with confidence, speeds catalyst discovery
 - Properties: give fundamental understanding of what's driving observed catalytic activity and selectivity



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Materials databases



Materials databases

- Materials Project, NOMAD, OQMD, AFLOW, AiiDA
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Materials databases

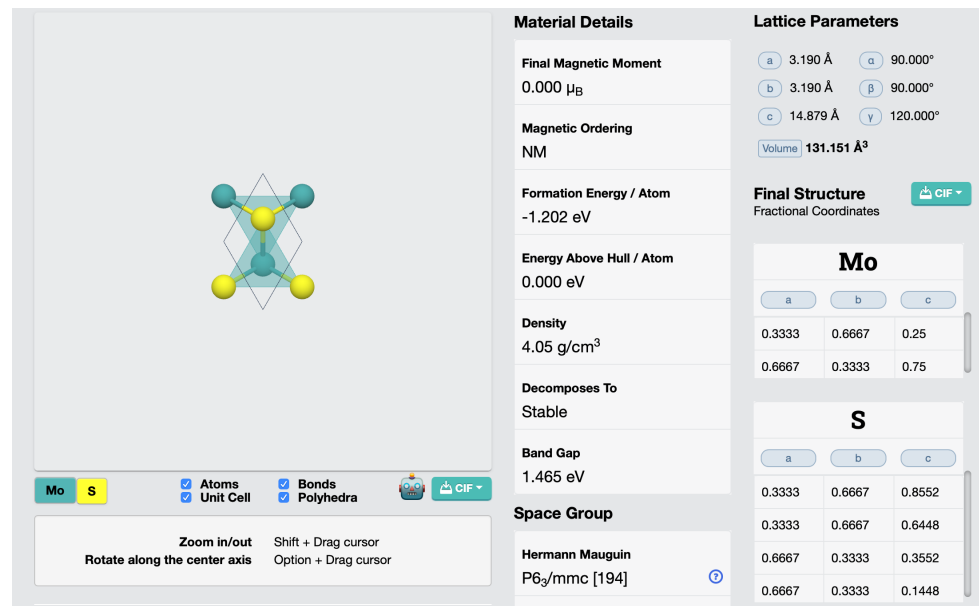
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Materials Id	Formula	Spacegroup	Formation Energy (eV)	E Above Hull (eV)	Band Gap (eV)	Volume	Nsites	Density (gm/cc)	
mp-2815	MoS ₂	P6 ₃ /mmc	-1.202	0	1.465	131.151	6	4.053	<input type="checkbox"/>
mp-1434	MoS ₂	R3m	-1.201	0	1.578	62.649	3	4.243	<input type="checkbox"/>
mp-1027525	MoS ₂	P $\bar{3}$ m1	-1.201	0.001	1.491	350.447	12	3.034	<input type="checkbox"/>
mp-1025874	MoS ₂	P $\bar{6}$ m2	-1.201	0.001	1.509	284.872	9	2.799	<input type="checkbox"/>
mp-1023939	MoS ₂	P $\bar{3}$ m1	-1.2	0.001	1.554	219.296	6	2.424	<input type="checkbox"/>
mp-1018809	MoS ₂	P6 ₃ /mmc	-1.2	0.001	1.336	123.452	6	4.306	<input type="checkbox"/>



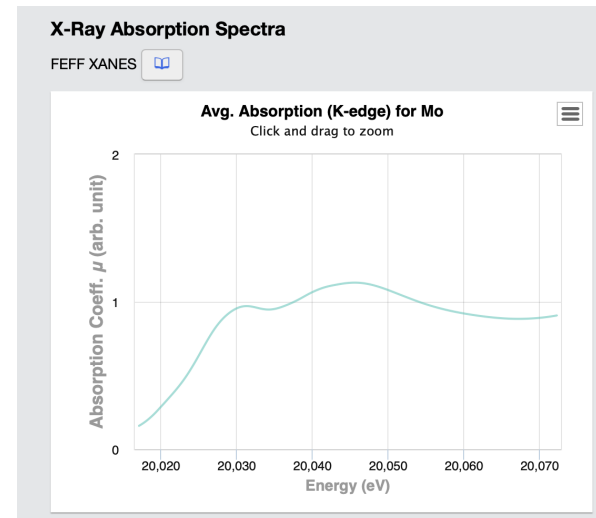
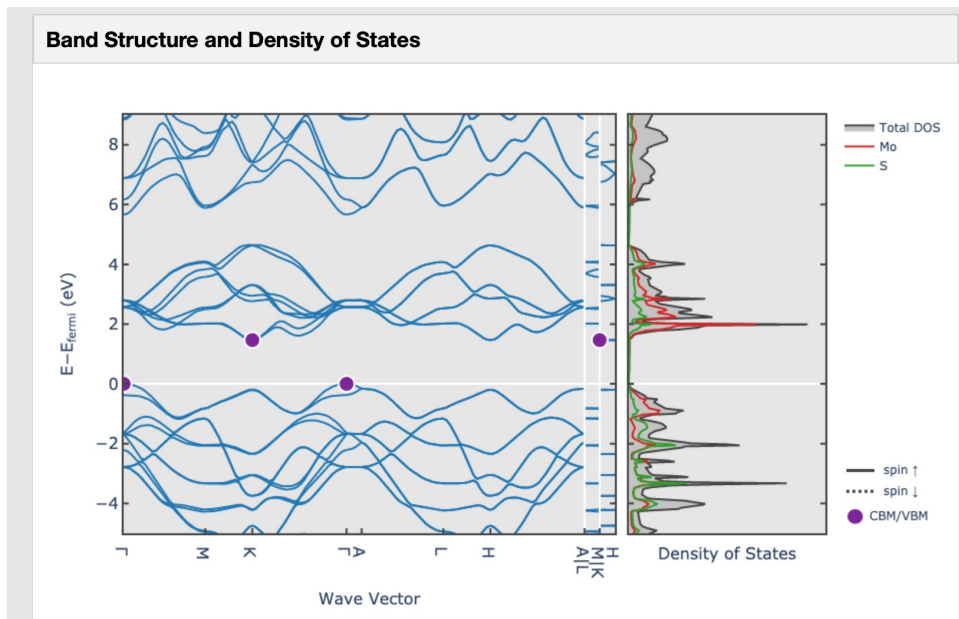
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Existing catalysis databases: Open Catalyst Project



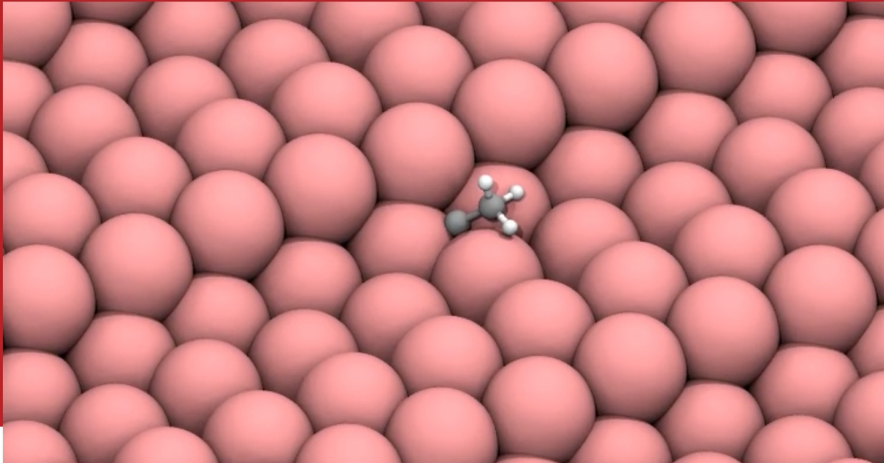
Existing catalysis databases: Open Catalyst Project

FACEBOOK AI Carnegie Mellon University

Home Leaderboard Dataset Challenge Discuss

Open Catalyst Project

Using AI to model and discover new catalysts to address the energy challenges posed by climate change.



Existing catalysis databases: Open Catalyst Project

- Team: Facebook, Carnegie Mellon
- Goal/philosophy: use machine learning to replace DFT relaxations that find adsorbed states of molecules on catalysts to speed up catalyst screening and discovery
- Points of interest
 - Big data, but not lowest energy states
 - No UI since meant for ML
 - No treatment of applied potential/solvent
 - Not based on promising catalysts



Existing catalysis databases: Catalyst Property Database

Catalyst Property Database



Existing catalysis databases: Catalyst Property Database

- Team: ChemCatBio EMN, NREL
- Goal: (1) reduce time searching literature for previously computed catalytic pathways by providing data in a central, searchable location, (2) enable accelerated discovery of catalyst descriptors, property correlations
- Points of interest
 - Less data in initial set, but labeled if lowest energy states
 - Excellent UI, metadata
 - No treatment of applied potential/solvent
 - Not based on promising catalysts



Existing catalysis databases: Catalysis Hub



Existing catalysis databases: Catalysis Hub

- Team: SUNCAT, Stanford
- Goal: (1) provide fully self-contained data for predicting experimental observations from electronic structure calculations (primary), (2) starting point for training and developing machine-learning based approaches accelerating quantum chemical simulations (secondary)
- Points of interest
 - Moderate data
 - Good UI, but so-so metadata and data standardization
 - Good post-processing tools (pourbaix diagrams, volcano plots, etc.)
 - No treatment of applied potential/solvent
 - Not based on promising catalysts



Existing catalysis databases: Summary

- Catalysis Hub (Stanford), Open Catalyst Project (CMU/Facebook), Catalyst Property Database (NREL)
- Less complete than materials databases because calculations are more expensive, catalysts are more diverse than materials
- Fewer properties, e.g. PDOS, bond orders, than materials databases
- Simpler or no APIs
 - Specifying catalytic reactions harder than specifying materials



BEAST DB



- Thrust 1: develop next generation of electrochemical solvation models with detailed double layer structure
- Thrust 2: beyond-DFT electrochemistry at exascale
- Thrust 3: apply techniques to electrocatalytic systems, forming BEAST-DB database for first-principles electrochemistry



BEAST DB

- Team: NREL, CU, RPI, LBNL, U. of S. Carolina
- Goal: (1) provide insight into how changes in electrochemical conditions lead to observed activity and selectivity, (2) give beyond-DFT accuracy for reaction energetics and electronic structuring using ML
- Points of interest
 - Allow variation of electrolyte, applied potential and easy comparison of variations with these knobs
 - Variety of electronic descriptors (PDOS, Bader charges, bond orders, wavefunction localization), electrolyte descriptors for rationalizing changes in behavior with different conditions
 - Base catalyst models on which are already promising
 - Beyond-DFT accuracy for reaction energetics (planned)
 - Smaller range of catalysts (targeted)



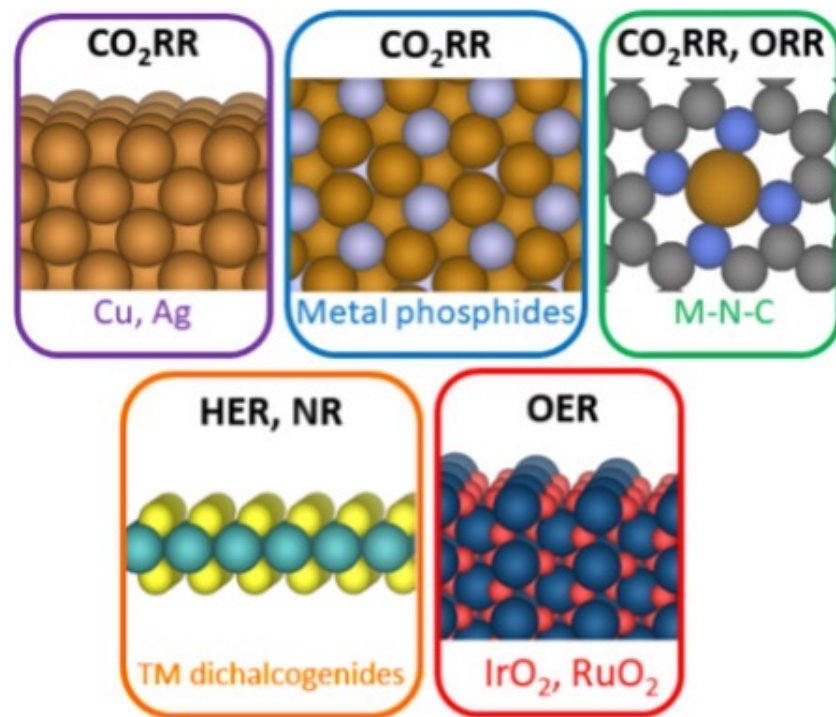


Fig. 14. Promising electrocatalysts and chemistries proposed as a starting point for our electrochemical database, BEAST DB.



- Reaction energetics, and electrolyte and electronic properties (e.g. PDOS)
- Start with most promising facets, electrolytes, electrolyte concentrations, and potential ranges for chosen catalysts
- Use ML to obtain beyond-DFT accuracy

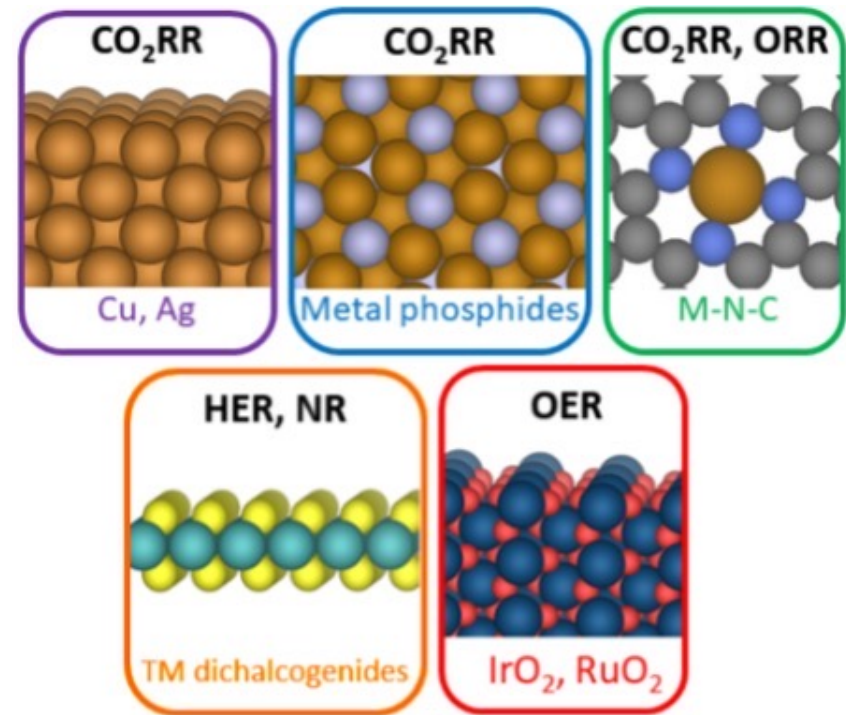


Fig. 14. Promising electrocatalysts and chemistries proposed as a starting point for our electrochemical database, BEAST DB.



BEAST DB: current status

- BEAST dataset contains 80 unique bulk compositions, 120 surface facets, over 3000 adsorbate calculations



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- **Chemistries:** HER, OER, CO₂R, NRR, ORR, partial methane oxidization reaction (pMOR) to methanol
- **Materials classes:** d-block metals, metal oxides, 2D chalcogenides, single-atom alloys (SAAs), 2D metalnitrogen-carbon surfaces (MNCs), and multinary Chevrel phase (CP) chalcogenides
- **Properties:** adsorption energies, converged structure geometries, Δ electrons, orbital-projected density of states (pDOS), surface-adsorbate pDOS overlap, Bader charge distributions



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- BEAST DB backend is close to being completed and UI is currently being implemented



BEAST DB: UI development

[Home](#) [Web Application](#) [API](#) [User Guide](#)

Search Materials

Select adsorbate ▾

Select catalyst ▾

[+ Add criterion](#)

*Results in Beast DB Standard ⓘ

Unique ID	Plot E	Adsorbate	Reference Species	Canonical Eads(eV)	Gads(eV)	Oxidation State

First Previous 1 2 3 Next Last



BEAST DB: UI development

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Search Materials

Select adsorbate ▾

Select catalyst ▾

+ Add criterion

Select criterion ▾

Unique ID >

Facet >

Reference Species >

Canonical E_{ads} >

*Results in Beast DB Standard ⓘ

Adsorbate	Reference Species	Canonical E _{ads} (eV)	G _{ads} (eV)	Oxidation State
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BEAST DB: UI development

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Search Materials

H

Zn

+ Add criterion

*Results in Beast DB Standard ⓘ

Unique ID	Plot E	Adsorbate	Reference Species	Canonical Eads(eV)	Gads(eV)	Oxidation State
123445	<input type="checkbox"/>	H	H2	-0.4	0.12	0.68
444112	<input type="checkbox"/>	H2	H2O	-0.55	0.4	0.78
444112	<input type="checkbox"/>	H2	H2O	-0.55	0.4	0.78
444112	<input type="checkbox"/>	H2	H2O	-0.55	0.4	0.78

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BEAST DB: current status

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Catalyst

Facet

Adsorbate

Calculation settings

Solvation method

Notes table

ID	Plot ads. energy	Plot Struct.	Adsorbate	Reference species	Canonical Eads (eV)	Gads (eV) # V vs SHE	Oxid. St. # V vs SHE
1155	<input checked="" type="checkbox"/>	<input type="checkbox"/>	1H	H3O+-H2O	-0.40	0.13	+0.68
1158	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	1O2	O2	-1.02	-0.50	-0.21
1161	<input type="checkbox"/>	<input type="checkbox"/>	1OH	OH-	-1.23	-1.63	-0.34

GCDFT Potential

Applied potential (V vs SHE)	Gads (eV) for 1H	Gads (eV) for 1O
-1.0	-0.25	-0.9
-0.5	-0.15	-0.75
0.0	0.0	-0.5
0.5	0.25	-0.25
1.0	0.5	-0.2

Applied potential ☒ V vs SHE ☐ V vs RHE

Export structure

Density of states

E - EF (eV)	DOS (C)	DOS (N)	DOS (Fe)
-6	0.5	0.1	0.1
-4	0.5	0.1	0.1
-2	0.5	0.1	0.5
0	0.5	0.1	0.1
2	0.5	0.1	0.1



BEAST DB: UI development

Home Web Application API User Guide

About BEAST

Disclaimers and Legal

Using the Web Application

Searching

Visualizations

Uploading Data

Downloading Data

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About BEAST

Computational modeling of electrochemistry is limited in accuracy by

1) the lack of a universal framework that efficiently treats arbitrary electrolytes, solvents and applied potentials with sufficient detail and fidelity to realistically and accurately model electrochemical systems, and
2) the deficiencies of density functional theory (DFT), the primary computational tool for reaction modeling, in describing charge transfer and reaction barriers. Accurate reaction barriers are crucial for connecting predictions to measured rates of chemical reactions, and quantum chemical techniques that may be accurate enough are not yet practicable for heterogeneous and electrocatalyst systems involving solid-liquid interfaces.

Funded by the DoE Computational Chemical Sciences program under DE-SC0022247 starting October 2021, the BEAST collaboration will address both challenges above by developing accurate and efficient exascale-ready solvated beyond-DFT methods.

The first ingredient in these methods are [accurate atomic-scale electrolyte solvation models](#) that capture the equilibrium effect of electrolyte in a single electronic structure calculation.

The second ingredient is the incorporation of GW many-body perturbation theory and the [random phase approximation \(RPA\) total energy](#), which are accurate methods beyond DFT, into solvated and grand-canonical techniques to make them practicable for electrochemistry including solvation and bias effects.

Finally, in addition to optimizing these combined techniques for exascale computing, we will also make them more widely applicable using machine learning (ML) approaches trained to a beyond-DFT electrochemical database to [make RPA-quality predictions at DFT cost](#).



BEAST DB: community involvement

- Want contributions from researchers in the electrocatalysis community, like YOU



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- Planning on having public workshop in the next year to solicit ideas further development of BEAST DB



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- Want contributions from researchers in the electrocatalysis community, like YOU
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- BEAST team has developed uniform set of computational parameters for database that will be required and checked for community upload



BEAST DB

