# BEAST DB

Derek Vigil-Fowler

1st Annual BEAST workshop

08/16/22

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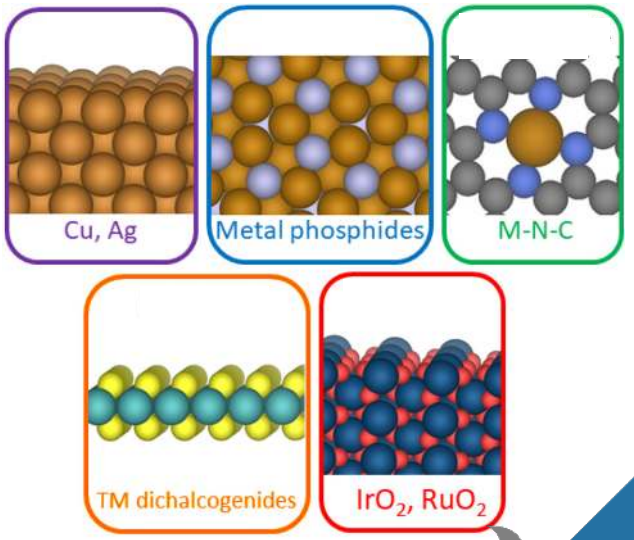
- Beyond-DFT Electrochemistry with Accelerated and Solvated Techniques Database

- Will have adsorption energies, other descriptors for range of common electrocatalysts, chemistries, use ML to obtain beyond-DFT accuracy



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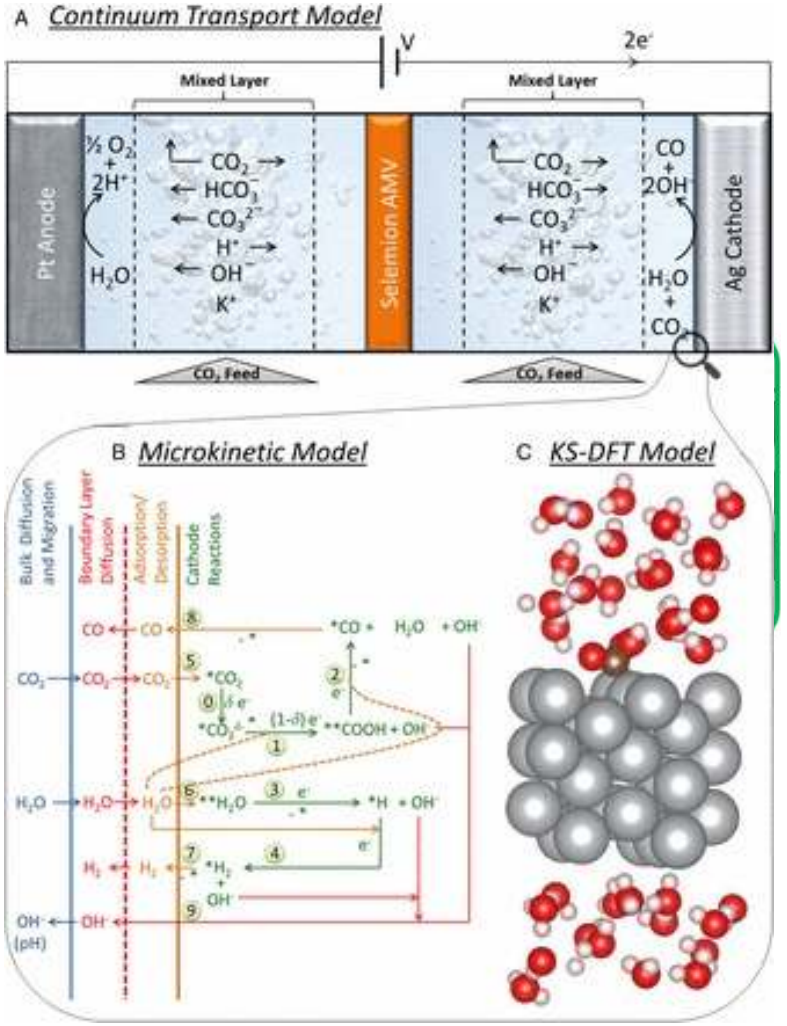


PNAS 2017 114 (42) E8812-E8821

# Electrochemistry is hard!

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- Diversity and dynamics

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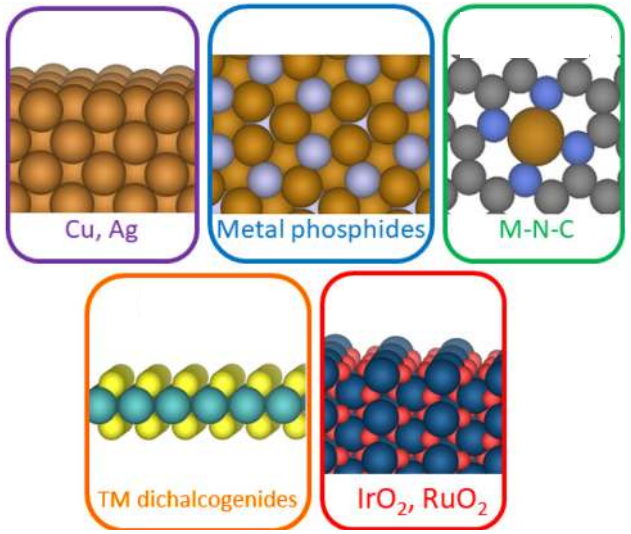
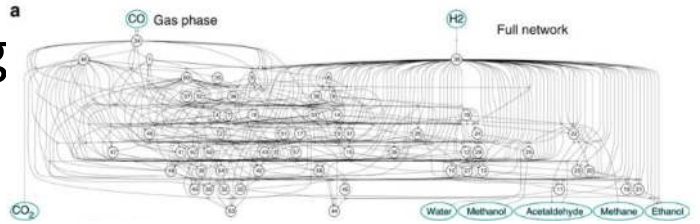
- Diversity and dynamics
  - Combinatorial space of elements
  - Bulk vs reduced dimensional
  - Identity of active site, mechanism
  - Surface structure and reconstruction
  - Defects
  - Complex reaction pathways involving intermediates, spectator species
  - Electrolyte identity, concentration
  -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




~100 species

~200 reactions

>2,000 pathways

*Nat. Comm.* **8**, 14621 (2017)

# 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: $CO_2$ 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 $KHCO_3$ 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.
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# Materials databases

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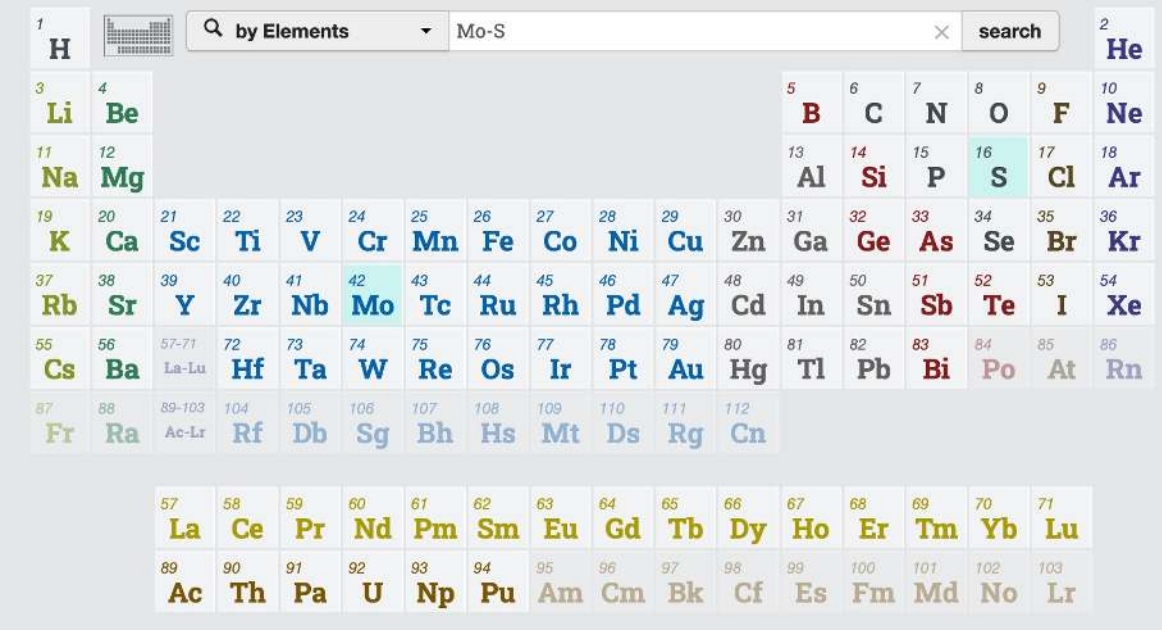


https://materialsproject.org/

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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 |
| mp-1434 | $MoS_2$ | R3m | -1.201 | 0 | 1.578 | 62.649 | 3 | 4.243 |
| mp-1027525 | $MoS_2$ | $P\bar{3}m1$ | -1.201 | 0.001 | 1.491 | 350.447 | 12 | 3.034 |
| mp-1025874 | $MoS_2$ | $P\bar{6}m2$ | -1.201 | 0.001 | 1.509 | 284.872 | 9 | 2.799 |
| mp-1023939 | $MoS_2$ | $P\bar{3}m1$ | -1.2 | 0.001 | 1.554 | 219.296 | 6 | 2.424 |
| mp-1018809 | $MoS_2$ | $P6_3/mmc$ | -1.2 | 0.001 | 1.336 | 123.452 | 6 | 4.306 |

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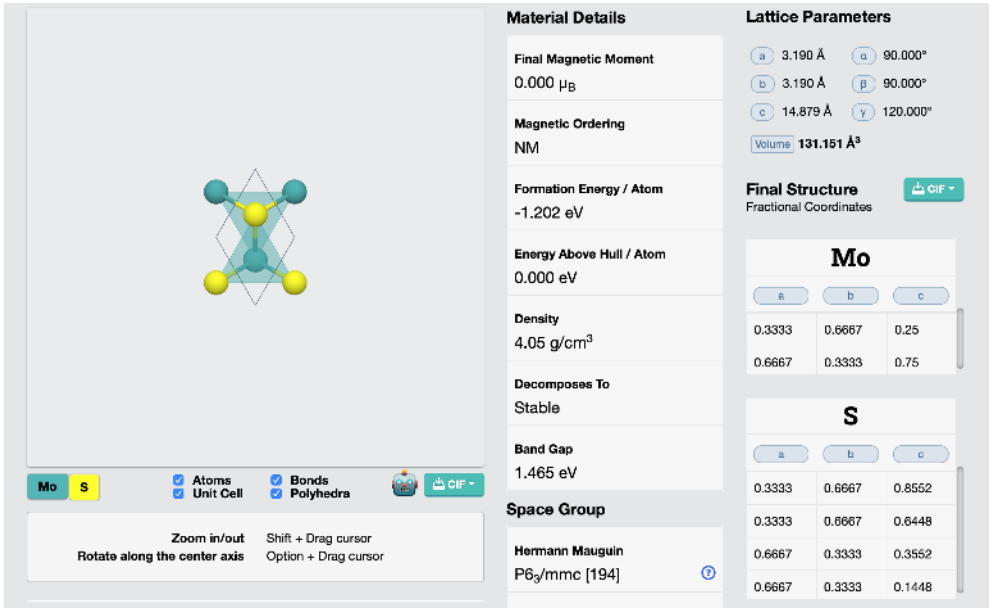


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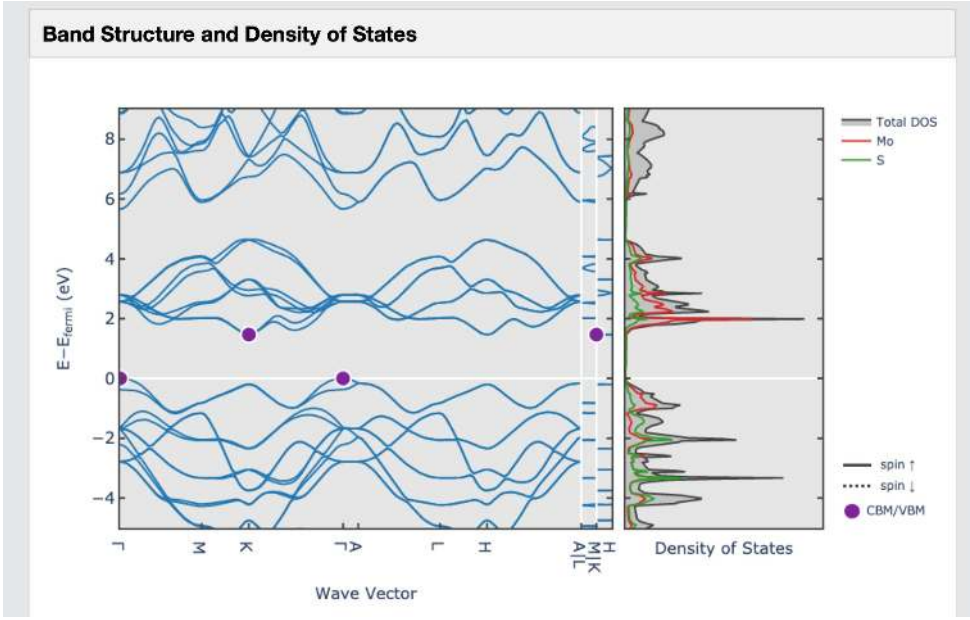


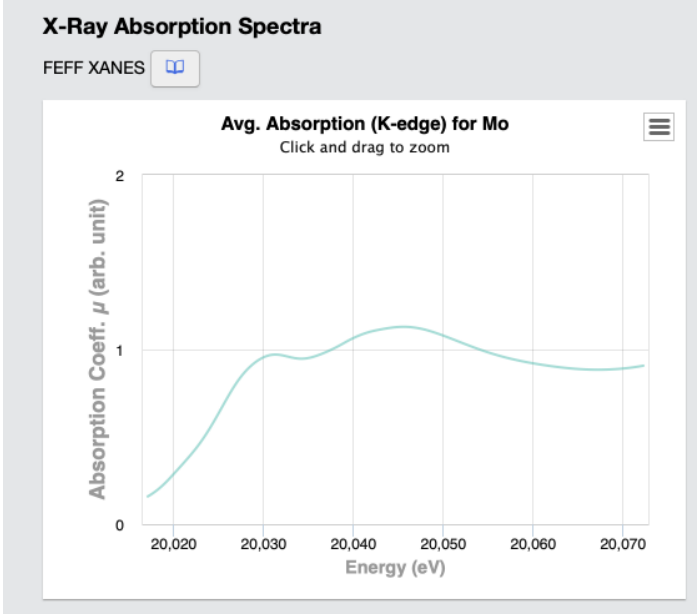


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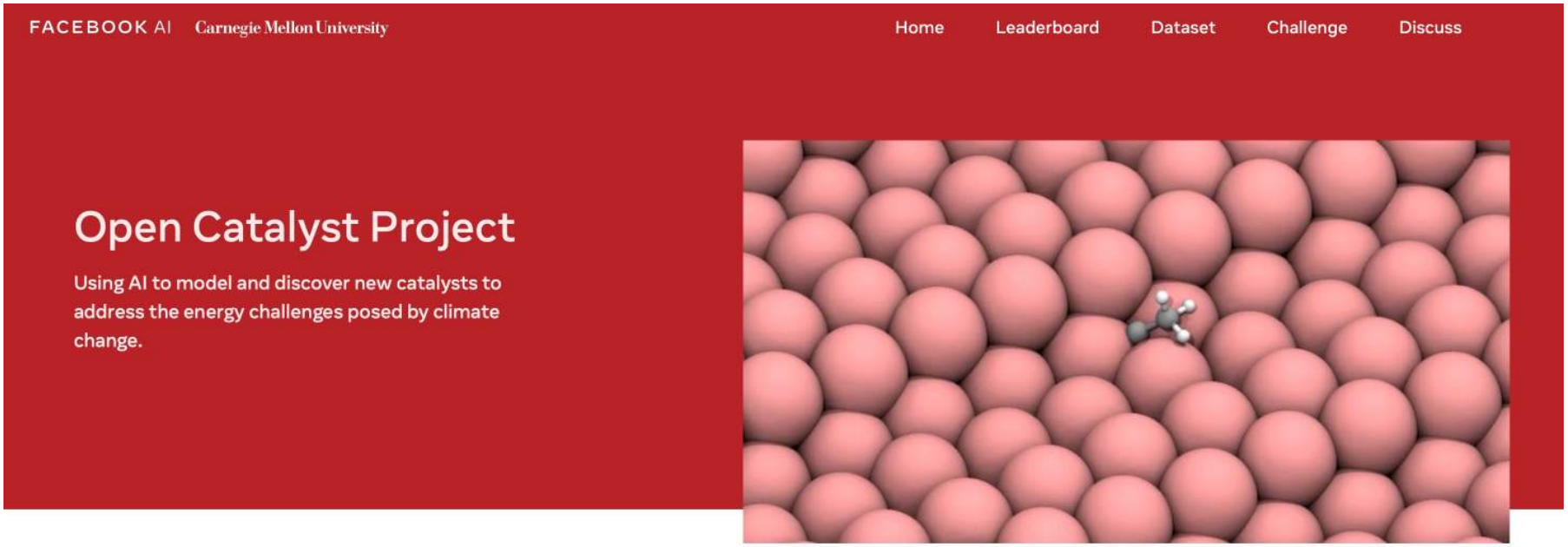

# Existing catalysis databases: Open Catalyst Project

- 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

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- 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

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- 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
  - Good post-processing tools (pourbaix diagrams, volcano plots, etc.)
  - No treatment of applied potential/solvent
  - Not based on promising catalysts

# Existing catalysis databases: Summary

- Don’t target promising catalysts, but do broader search
- 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

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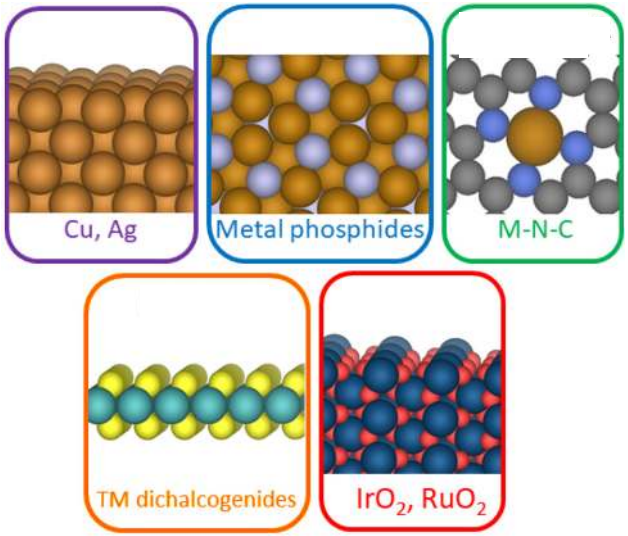
- Thrust 1: develop next generation of electrochemical solvation models with detailed double layer structure
- Thrust 2: beyond-DFT electrochemistry at exascale
- Thrust 3: apply techniques to electrocatalytic systems, forming BEAST-DB database for first-principles electrochemistry

# BEAST DB

- 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



# BEAST DB “demonstration”

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- Search feature similar to Catalyst Property Database, Catalysis Hub

# BEAST DB “demonstration”

- Fe-N-C

0 V 1 V

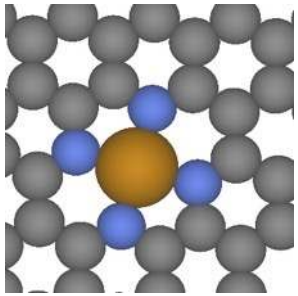
Potential:

$10^{-2}$ M 10 M

Concentration:

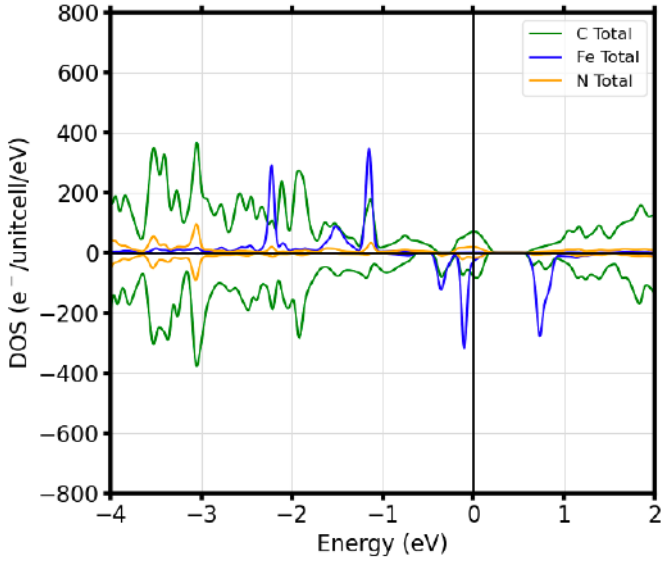
0.5

Electrolyte: ☒ $H_2SO_4$ ☐ $HClO_4$ ☐ HCl ☐ NaF



Total energy: -11526.2912 eV

| AtomType | TotalNetCharge |
|---|---|
| C | 3.676 |
| N | -0.970 |
| Fe | 2.736 |



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- Fe-N-C

0 V 1 V

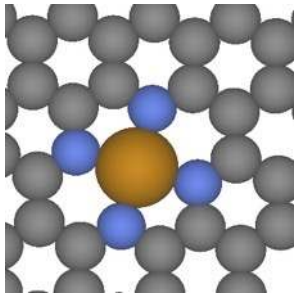
Potential:

Electrolyte: ☒ $H_2SO_4$ ☐ $HClO_4$ ☐ HCL ☐ NaF

$10^{-2}$ M 10 M

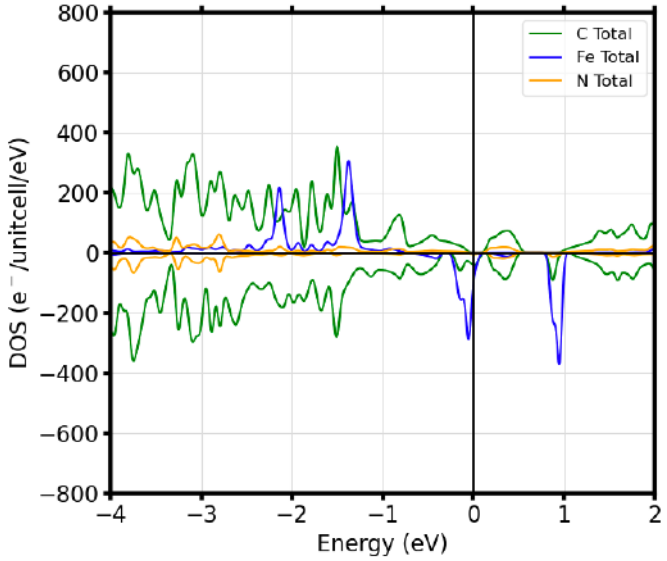
Concentration:

0.5



Total energy: -11275.8951 eV

| AtomType | TotalNetCharge |
|---|---|
| C | 4.992 |
| N | -0.778 |
| Fe | 2.841 |



# 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)

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

- Chemistries: HER, OER, $CO_2R$, NRR, ORR, partial methane oxidization reaction (pMOR) to methanol.

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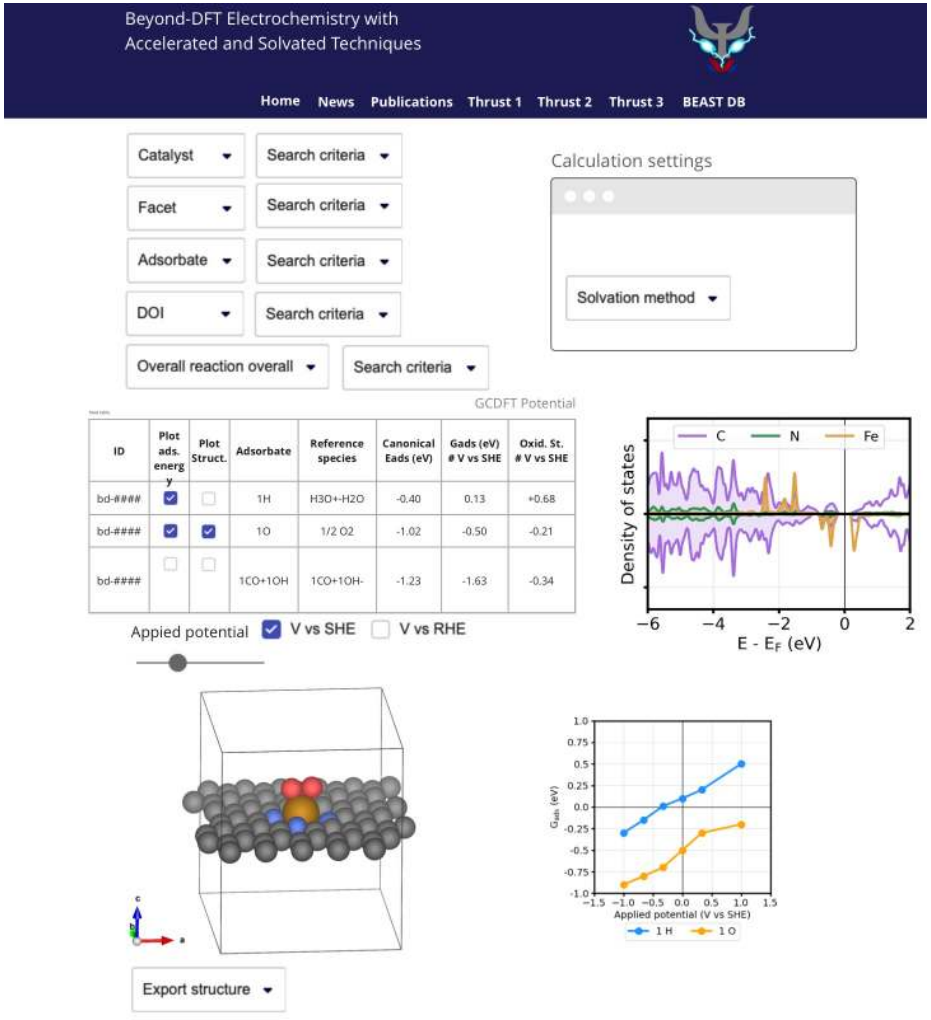
- Chemistries: HER, OER, $CO_2R$, NRR, ORR, partial methane oxidization reaction (pMOR) to methanol.
- Materials classes: d-block metals, metal oxides, 2D chalcogenides, single-atom alloys (SAAs), 2D metal-nitrogen-carbon surfaces (MNCs), and multinary Chevrel phase (CP) chalcogenides.
- Properties: adsorption energies, converged structure geometries, Δ electrons, orbital-projected density of states (pDOS), surface-adsorbate pDOS overlap, Bader charge distributions.

# BEAST DB: current status

- BEAST DB backend and UI under construction in upcoming year



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- BEAST team has developed uniform set of computational parameters for database that will be required and checked for community upload

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