BEAST DB

Derek Vigil-Fowler

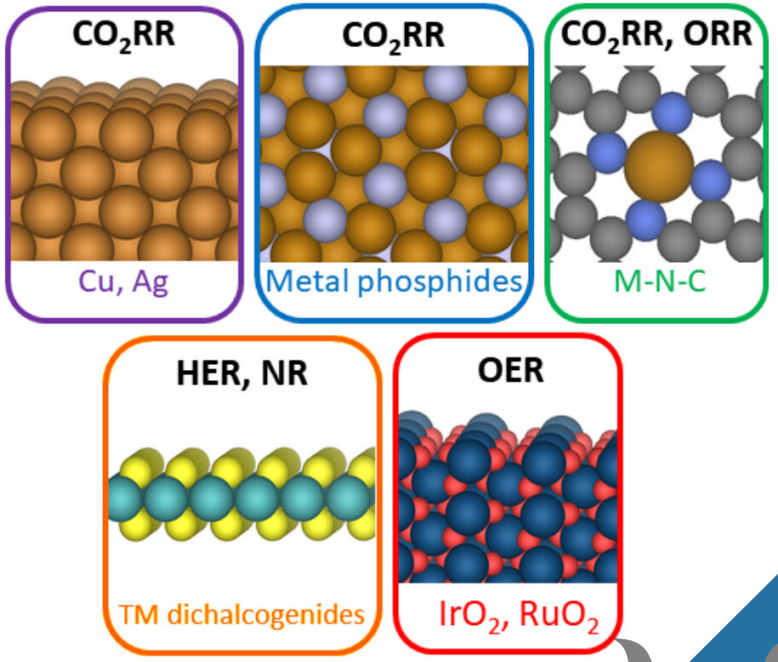
2nd Annual BEAST workshop

08/18/23

BEAST DB

- 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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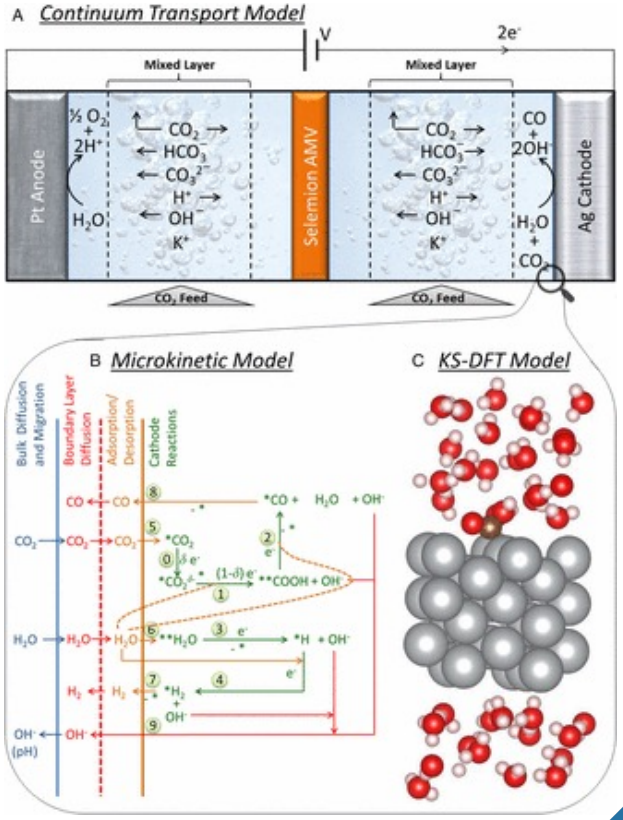

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PNAS 2017 114 (42) E8812-E8821

Electrochemistry is hard!

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- Diversity and dynamics

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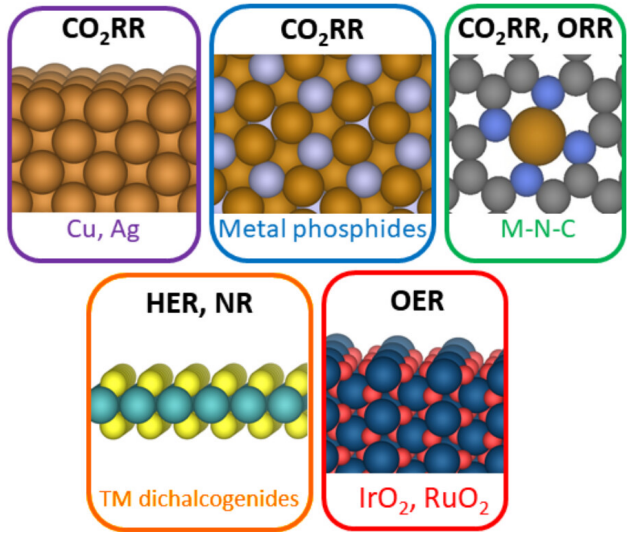
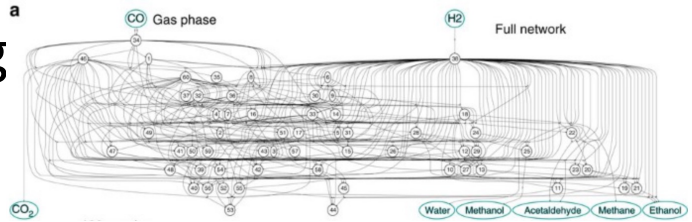
- Diversity and dynamics
  - Combinatorial space of elements
  - Bulk vs reduced dimensional
  - Identity of active site, mechanism
  - Surface structure and reconstruction
  - Defects
  - Complex reaction pathways involving spectator species
  - Electrolyte identity, concentration
  - pH
  - Applied 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.
- 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.
- Measure CV and EIS in H_2/N_2 as described before.
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- Measure activity in H_2/O_2 as specified.
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- Measure pol curve in H_2/air as described before.
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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

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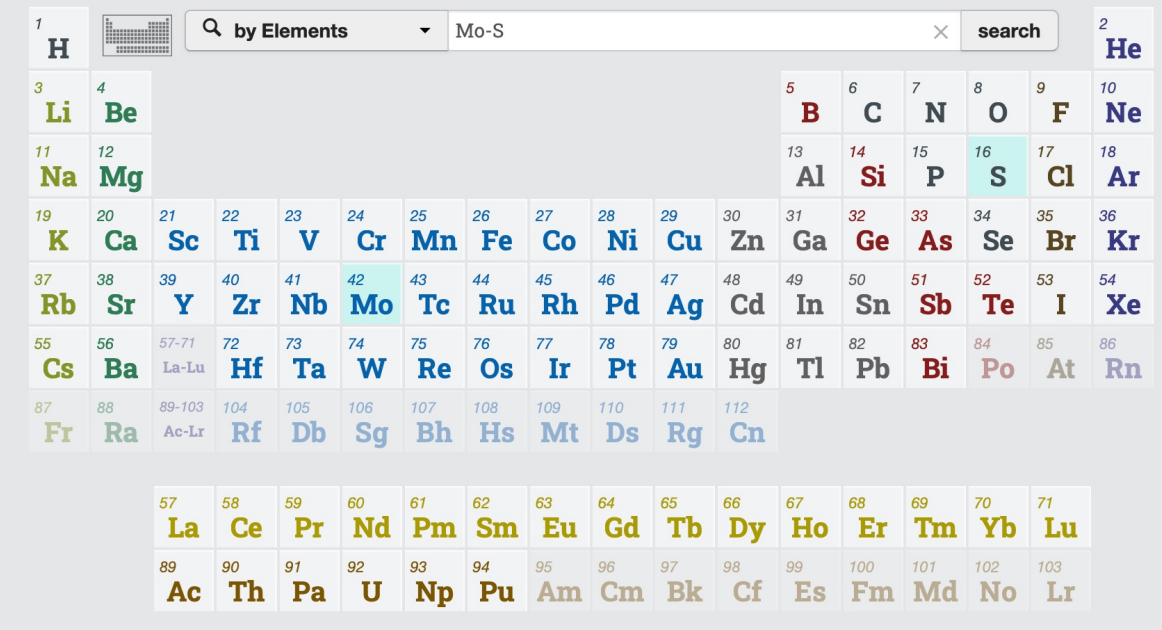

https://materialsproject.org/

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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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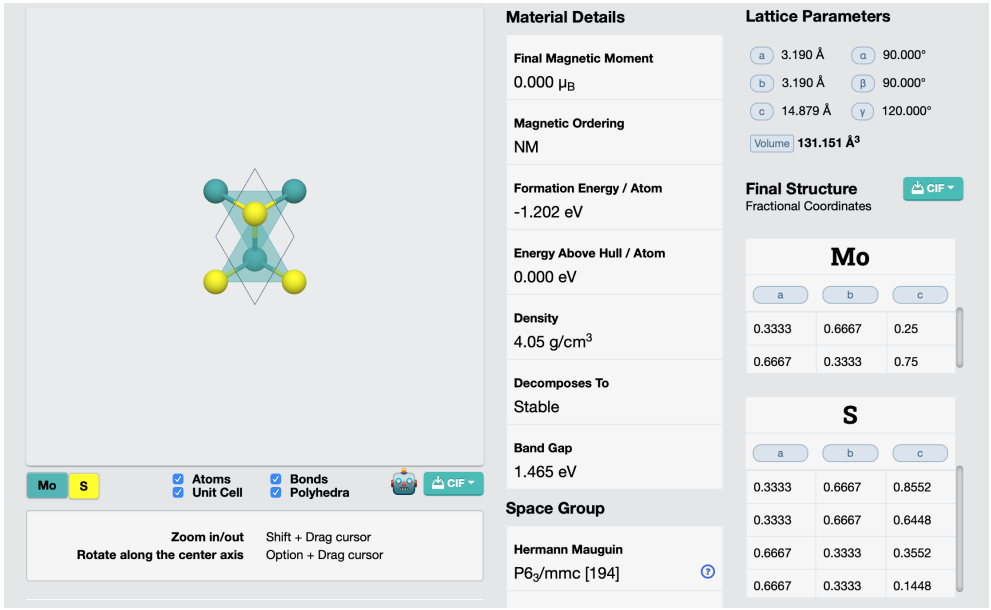

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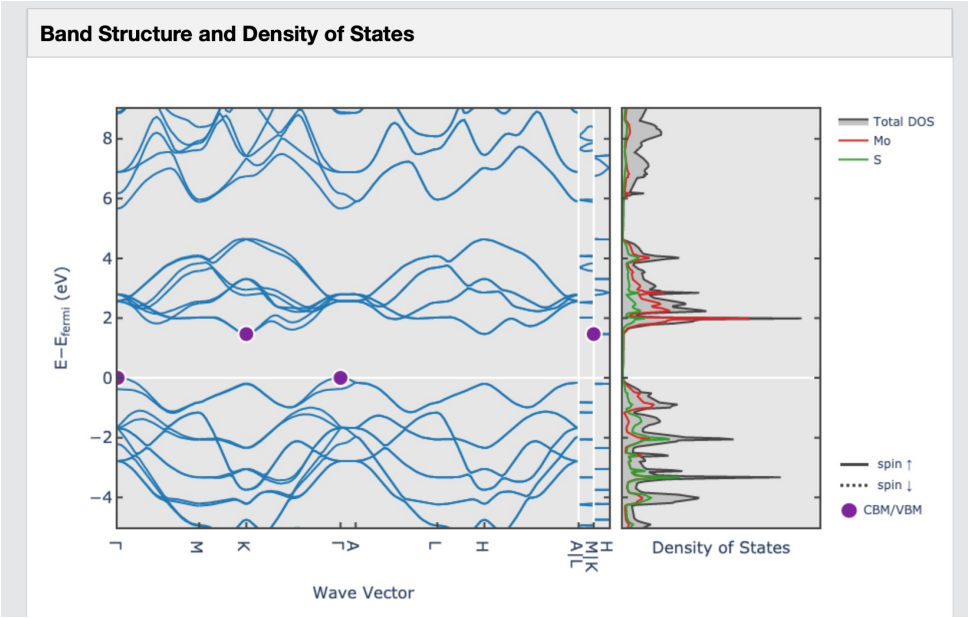

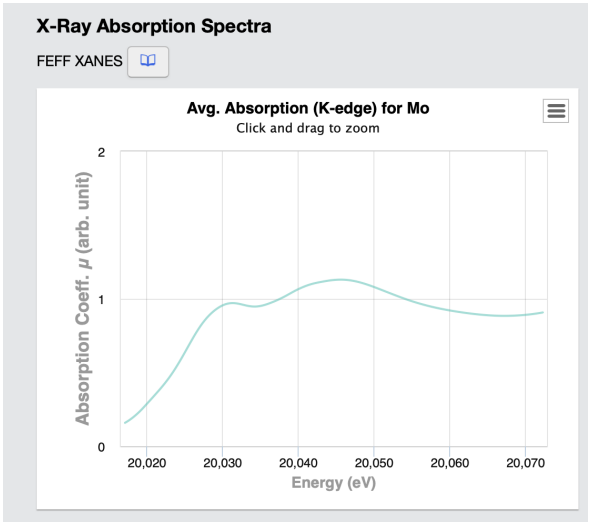

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Existing catalysis databases: Open Catalyst Project

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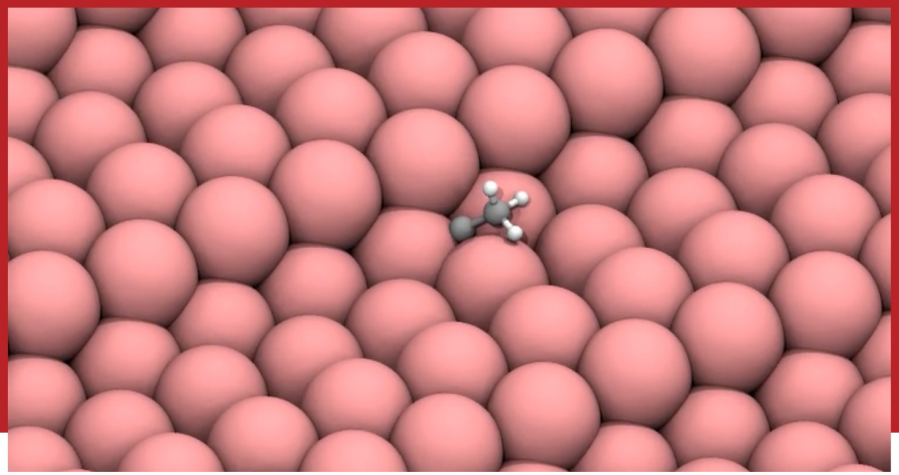
FACEBOOK AI Carnegie Mellon University

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

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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, 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

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)

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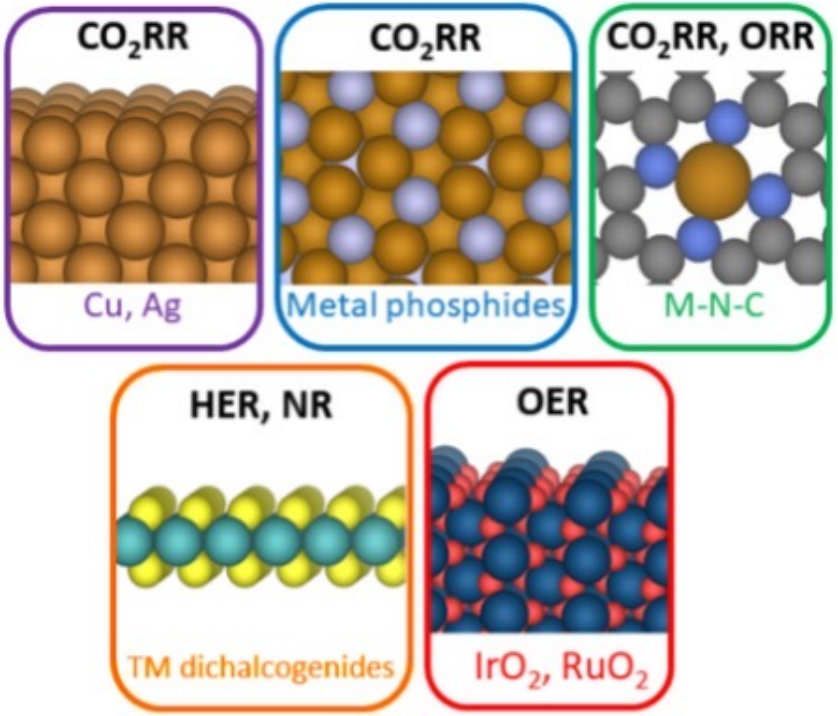
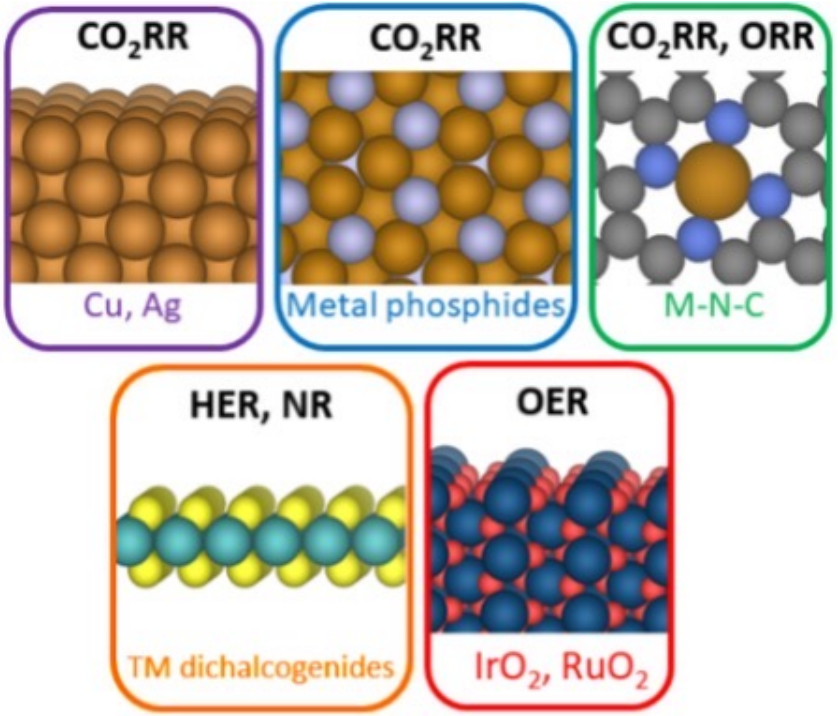


Fig. 14. Promising electrocatalysts and chemistries proposed as a starting point for our electrochemical database, BEAST DB.

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



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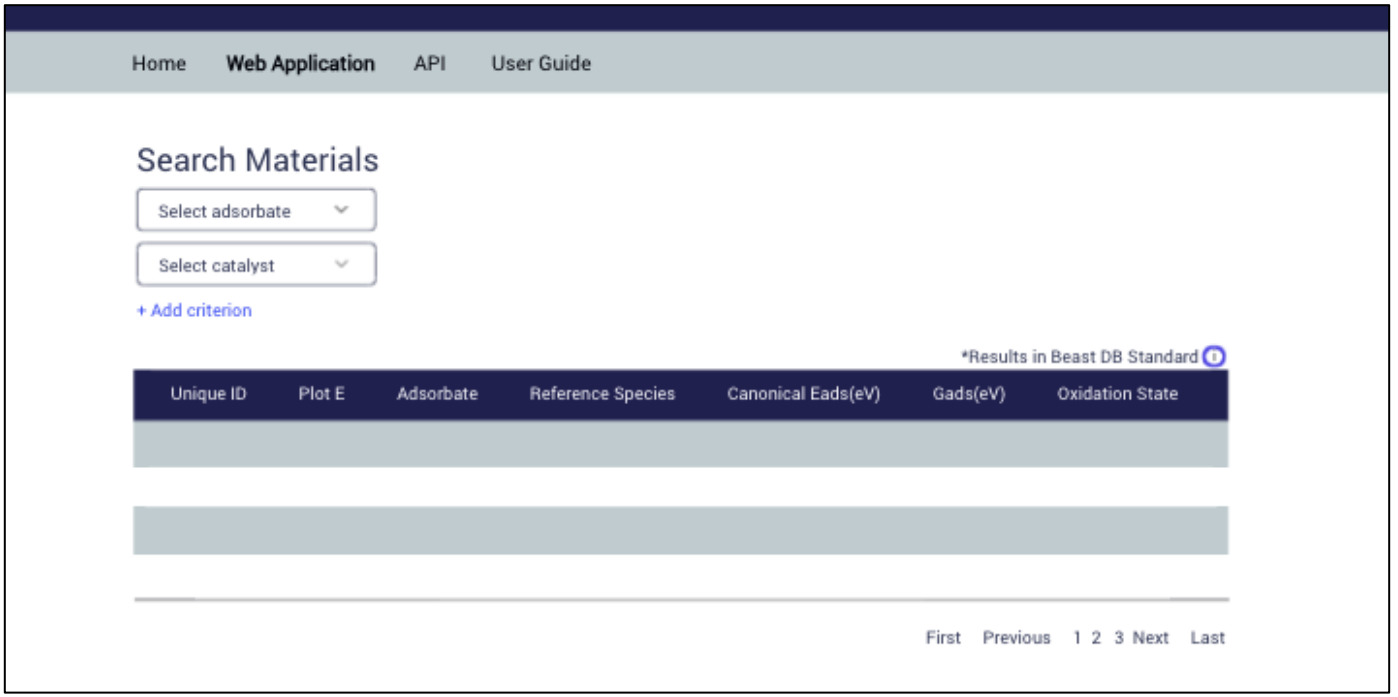
- BEAST DB backend is close to being completed and UI is currently being implemented

BEAST DB: UI development



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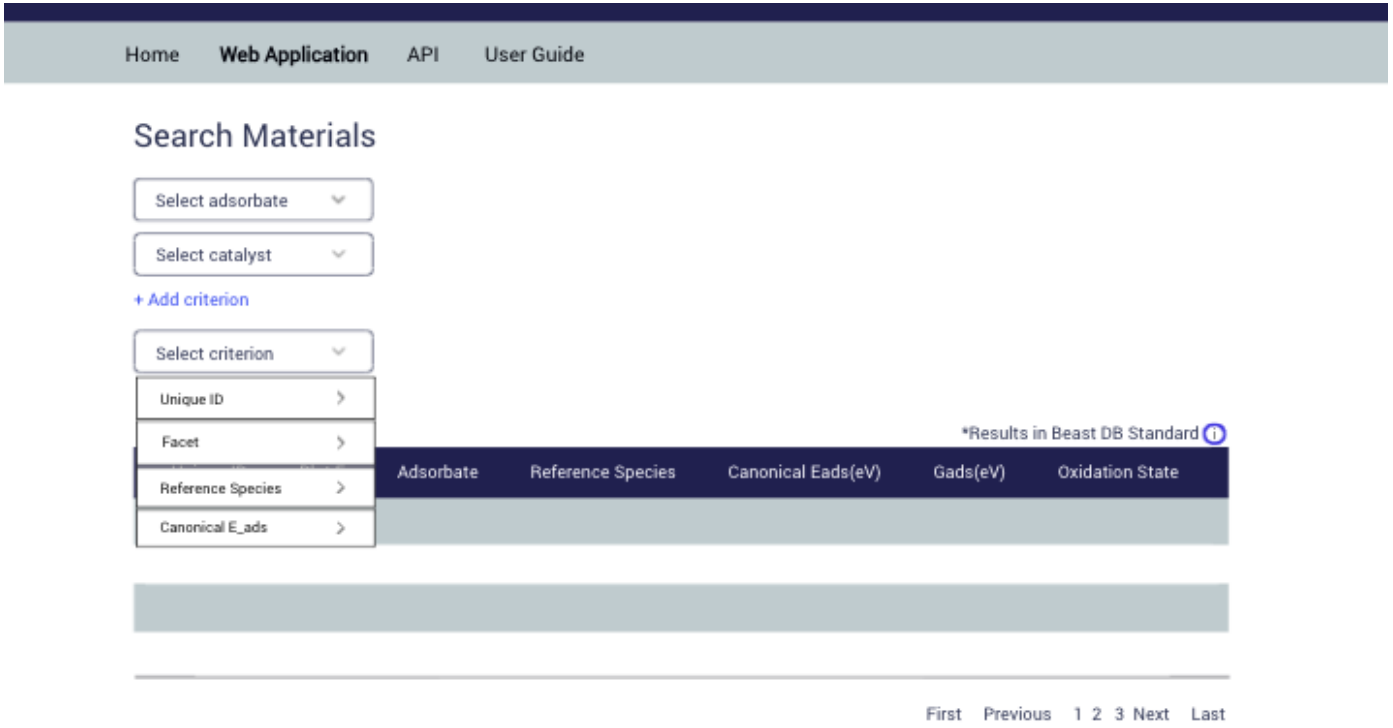

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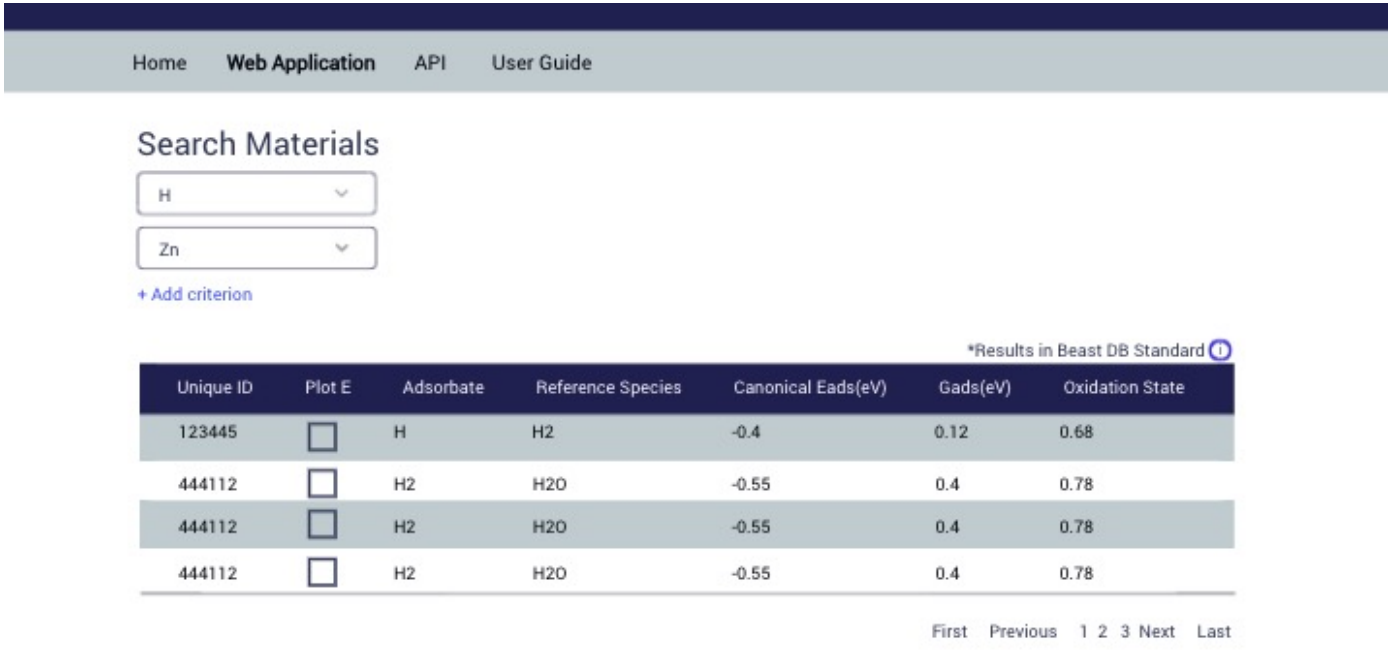

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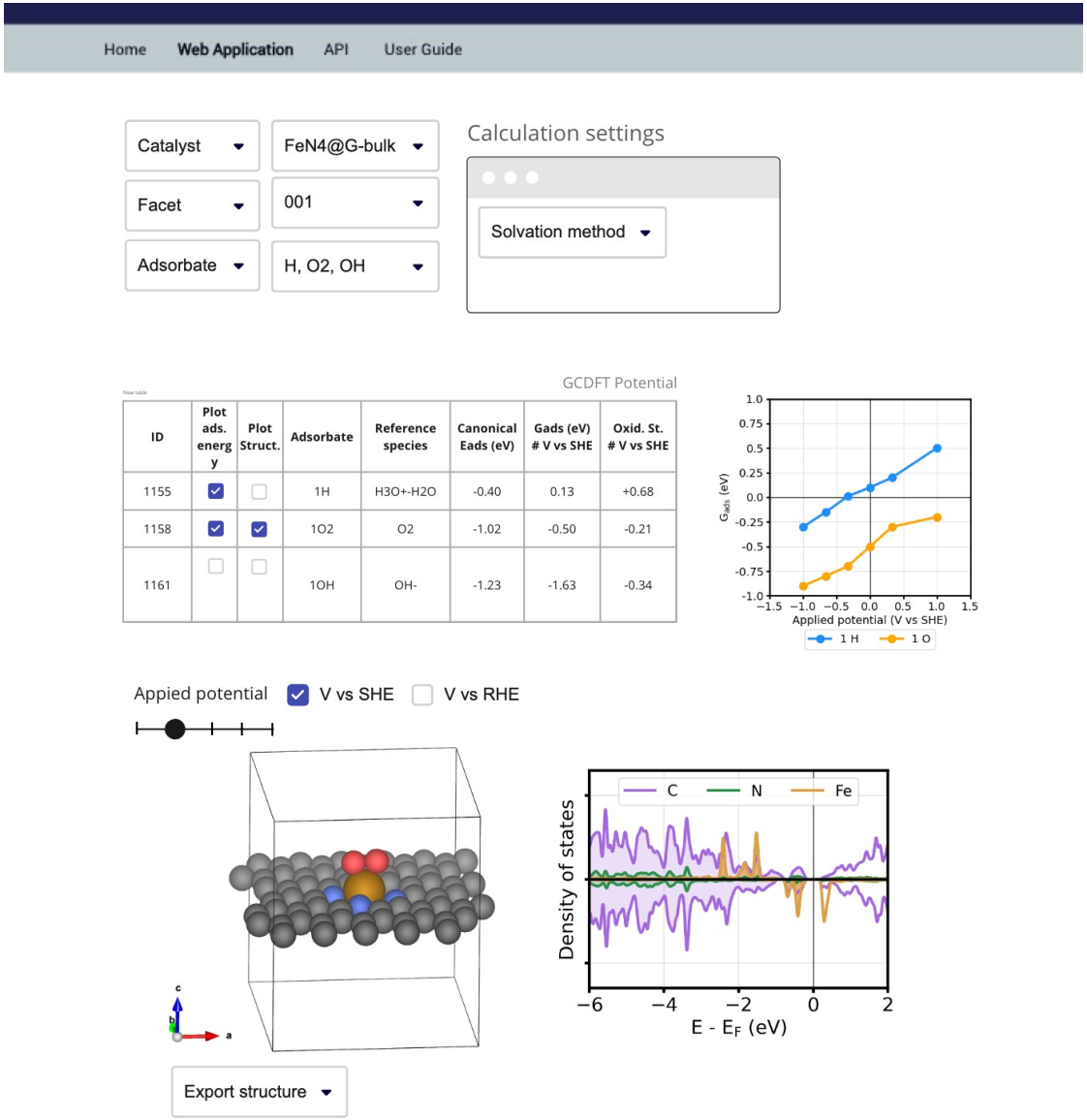

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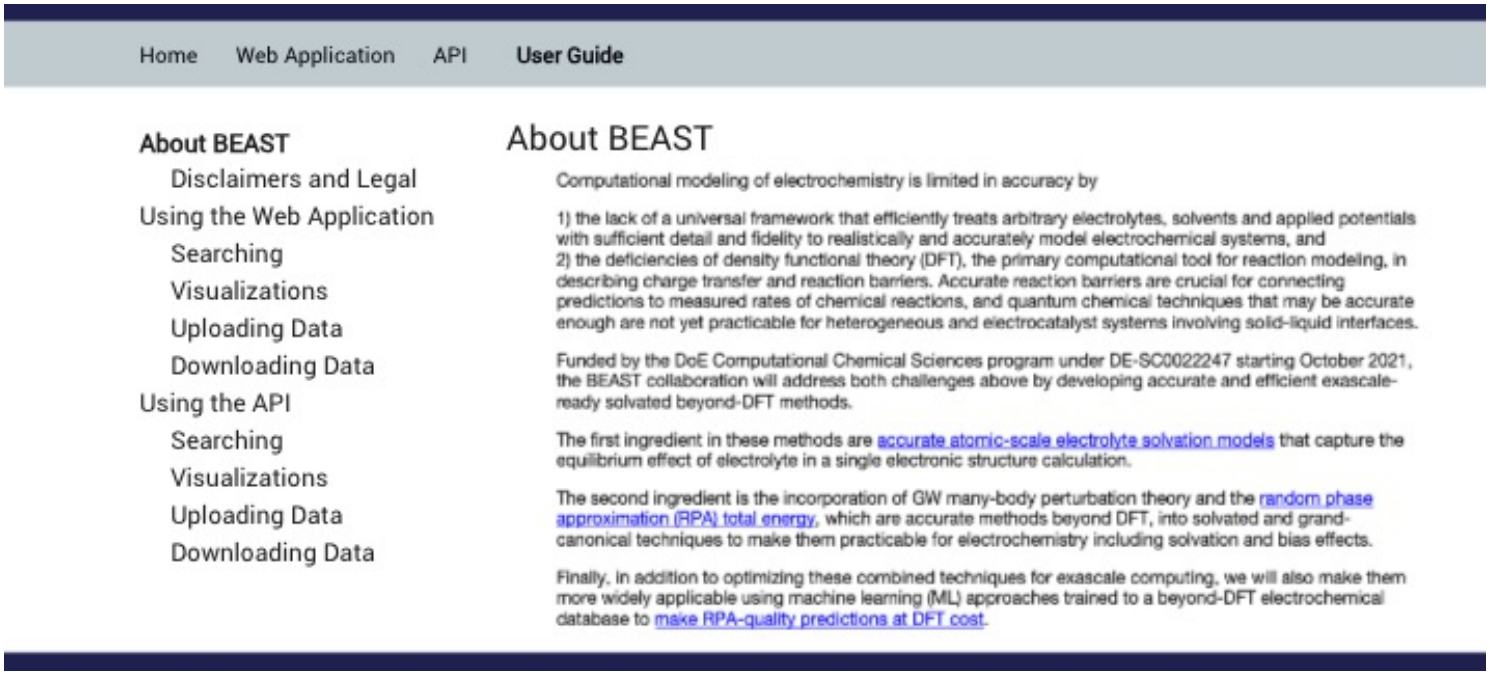

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- Planning on having public workshop in the next year to solicit ideas further development of BEAST DB

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- Planning on having public workshop in the next year to solicit ideas further development of BEAST DB
- BEAST team has developed uniform set of computational parameters for database that will be required and checked for community upload

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