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Sandia National Laboratories

A data-driven roadmap towards Pareto optimal hydrogen storage alloys

PRESENTED BY

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Collaborators & Acknowledgements

Experiments

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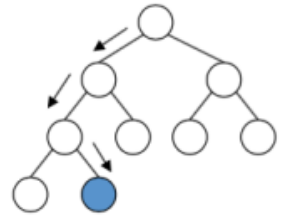
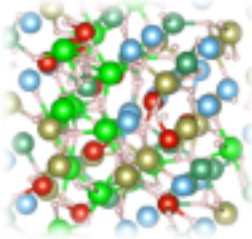
Seminar Organization

Approach: Using data-driven/machine learning techniques for materials discovery

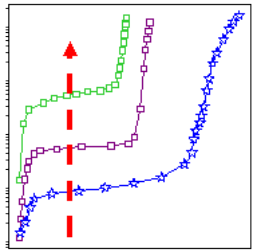


Application: Discovering more efficient materials-based hydrogen storage



Results: Success stories in data-driven discovery of hydrogen storage materials

- New technical capabilities/physical insights
- Towards Pareto optimal materials



Future Work: Overcoming current limitations in ML-driven discovery for hydrogen storage materials

$$v = \mathrm{avg}(v_1^{t0}, v_2^{t0})$$

$$v_1^{t1} = (v_1^{t0} \oplus v_2^{t0} \oplus \mathbf{e}_{12}) \cdot \mathbf{W} + \mathbf{b}$$

Approach:

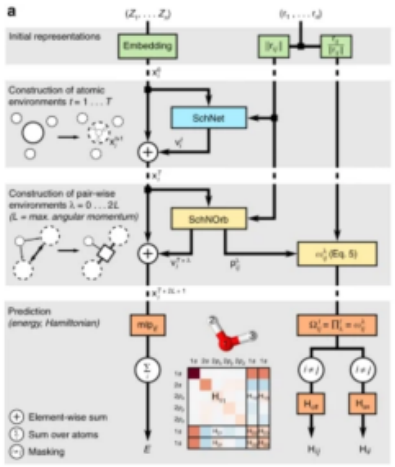
Why use data-driven/machine learning techniques for materials discovery?

Data science/ML models in materials/chemical sciences can be applied to problems of "all shapes and sizes"

Angstroms … to … **Acres**

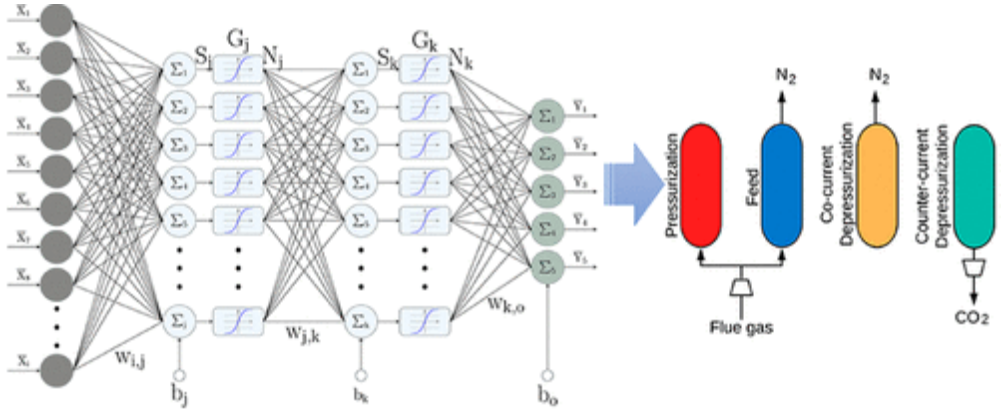
Small molecule property predictions[1]

Inputs
(***Z***, ***r***)



Outputs
(*E*)

Chemical plant performance prediction[2]



Inputs
(PSA unit parameters)

Outputs
(CO_2 purity of PSA units)

ML models **are not**: a magic box that solve all our problems, will always require "truth" data from experiments or first principles calculations (often difficult in materials science)

ML models **are**: *surrogates that execute **many orders of magnitude** faster than an experiment or first principles calculation to make task X tractable (high-throughput screening, optimization, active search, etc.)*

[1] Schutt, et al. *Nat. Comm.*10, **2019**
[2] Leperi, et al. *I&EC*. 58 (39), **2019**

Three main approaches to materials discovery

Experiments

Chemical intuition

↓

Synthesize material

↓

Characterize material

↓

Test Performance

Physics-based simulations

First-principles theory

↓

Run *simulation* & predict performance

Machine learning (ML)

Material property/training data

↓

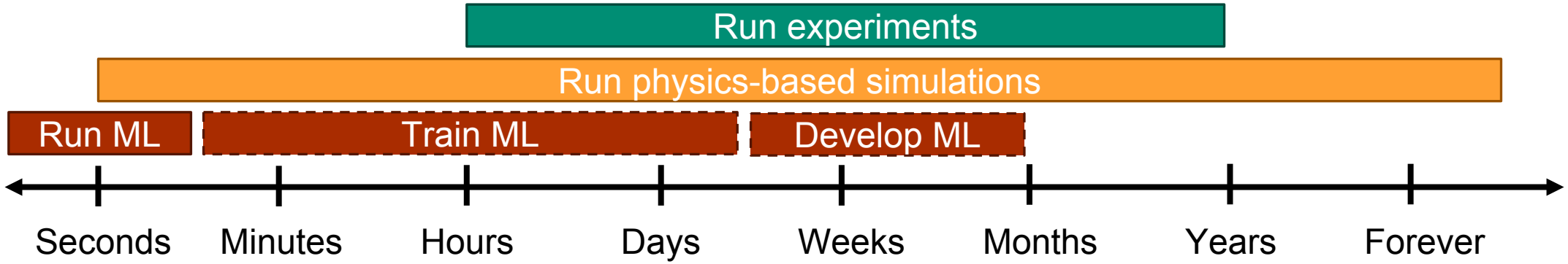
Convert materials to features

↓

Construct & train a model

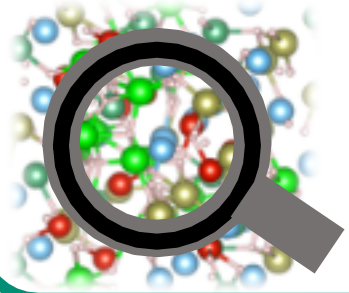
↓

Run *model* & predict performance



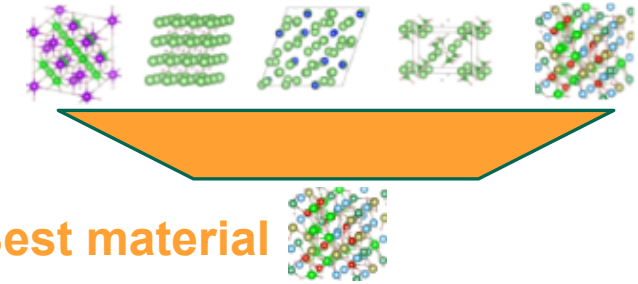
Generally speaking, data science and machine learning techniques can…

Predict properties and elucidate design rules for optimal materials

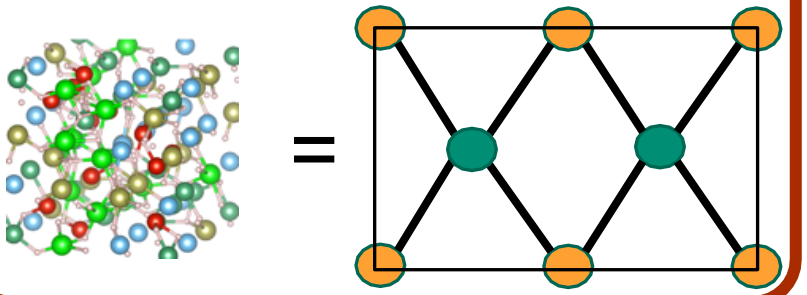


$$P_{eq} = m \quad + b$$

High-throughput screen materials orders of magnitude faster than experiments or simulations



Accelerate physics-based simulations when lacking experimental training data



Tools provide a roadmap for…

H_2 storage: Data-driven discovery of Pareto optimal hydrogen storage alloys

H_2 generation: Data-driven discovery of water-splitting materials (STCH, liquid metals, etc.)

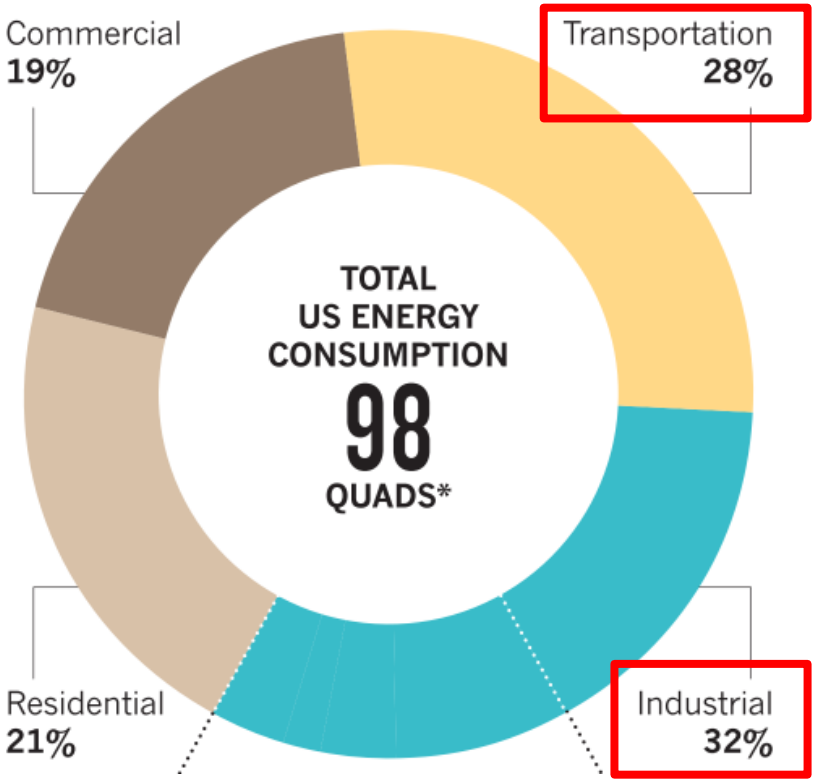
Application:

Discovering more efficient materials-based hydrogen storage

Discovering improved H_2 storage and generation techniques will facilitate progress towards a hydrogen economy

H_2 to play a critical role in…

Heavy duty (planes, trains, ships)



Sholl, Lively. Nature 532 (7600) **2016**.

High T applications

Personal transportation (Toyota/Hyundai)

Air Travel (Airbus)

Home energy storage (Lavo)

Utility-scale generation (Shell)

First Energy Earthshot Aims to Slash the Cost of Clean Hydrogen by 80% to $1 per Kilogram in One Decade

"Clean hydrogen is a game changer" -- Secretary Granholm

Are there undiscovered materials that could improve upon conventional H_2 storage and generation technologies?

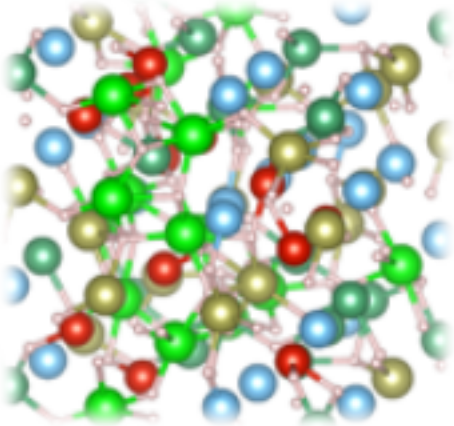
H_2 Storage objectives:

- High H_2 gravimetric and/or volumetric density
- Fast, reversible release near ambient T
- Practical/cost-effective
- Reduced infrastructure cost

700 bar

"Conventional" (Compressed gas)

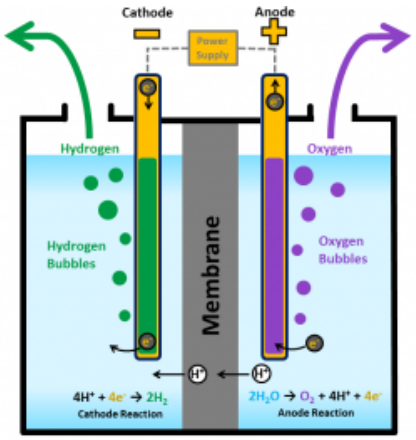
vs.



Material X ??

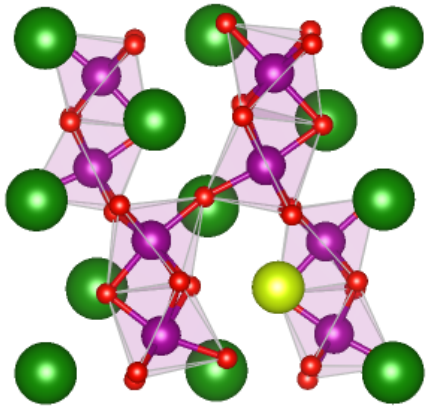
H_2 Generation objectives:

- Water-splitting using only renewable energy
- Practical/cost-effective



"Conventional"
1.2 V in theory
1.8 V in practice

vs.

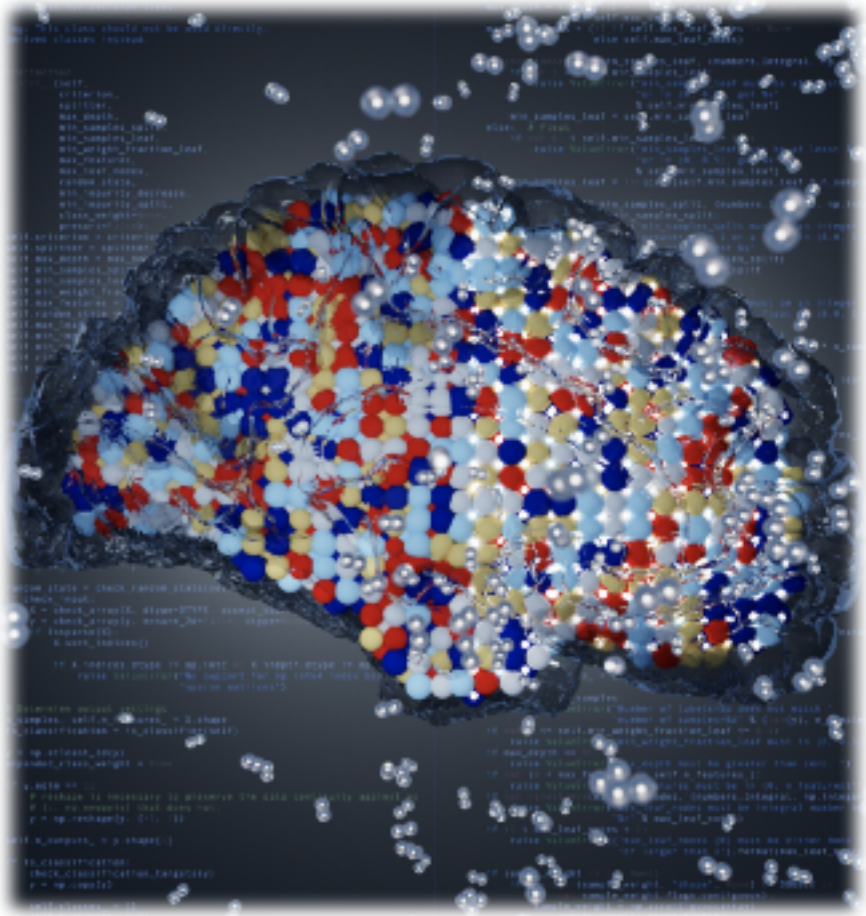


Material Y ??

Results: A data-driven roadmap towards Pareto optimal hydrogen storage alloys

- **Milestone #1** : Explainable ML models predict metal hydride thermodynamics
- **Milestone #2** : ML enables discovery of destabilized high entropy alloy (HEA) hydrides
- **Milestone #3** : ML screening & identification of *Pareto optimal* HEA hydrides

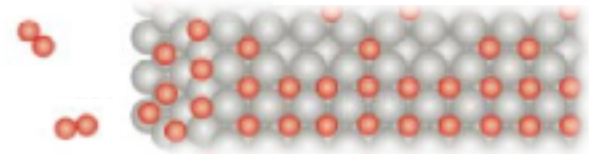
Milestone #1: Explainable ML models predict metal hydride thermodynamics



Milestone #1: Explainable ML models predict metal hydride thermodynamics

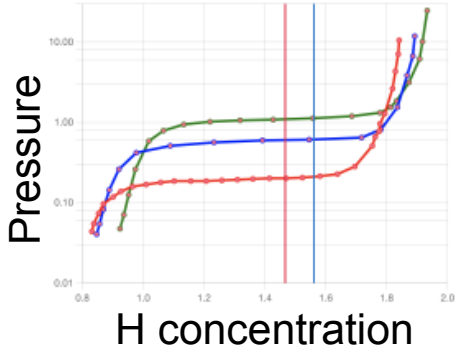
(1) $\ln(P^o_{eq}/P_o)$ target property

H_2 Metal Hydride



$$\ln(P^o_{eq}/P_o) = -\frac{\Delta H}{R(25^oC)} + \frac{\Delta S}{R}$$

Extracted from isotherms & van't Hoff



$\Delta H, \Delta S$ are tabulated in HydPARK.csv
- Missing data, errors, etc.
- ***Only*** 15% of 2500 entries usable

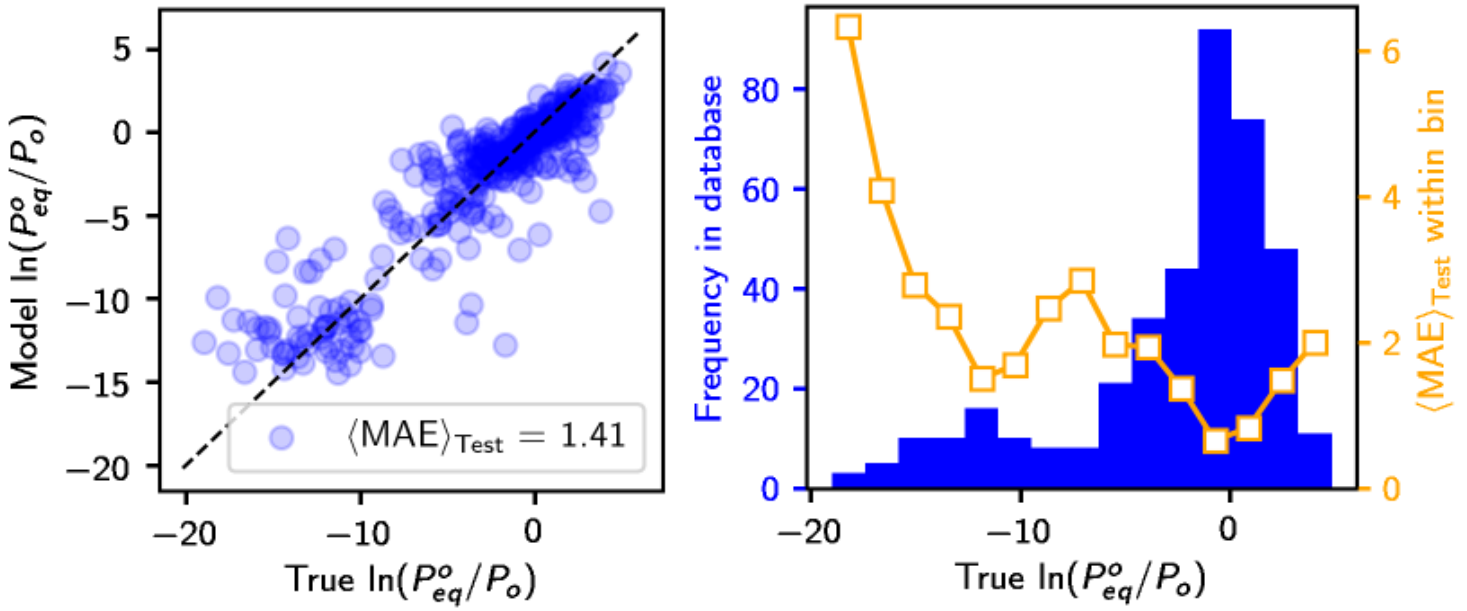
(2) Constructing a compositional ML model[1]

- Features derived from substituent elemental properties:
  $f_i \equiv$ fraction of element i, $p_i \equiv$ elemental property of i
- Simple operations map composition to scalar feature:
  $\bar{p} = \sum_i f_i p_i$, $\hat{p} = \sum_i f_i |p_i - \bar{p}|$, $\ddot{p} = \max(p_i) - \min(p_i)$, etc…
- Any composition mapped to same dimensional feature vector:
  $$\boldsymbol{x}_{\text{Magpie}} = \{\bar{\nu}_{pa}, \bar{r}_{cov}, \bar{\chi}, \ldots\} \in \mathbb{R}^{145}$$
- **Necessary if exact structure unknown** (e.g. $TiFe_{0.92}Nb_{0.08}$)
- Gradient boosting trees (ensemble of *m* trees):
  $y = F_m(\boldsymbol{x}) + h_m(\boldsymbol{x})$
  $h_m(\boldsymbol{x})$, a new estimator, corrects the previous model, $F_m(\boldsymbol{x})$
  i.e. is fit to minimize residual error, $y - F_m(\boldsymbol{x})$
- **Expressive, interpretable model if sufficient data**

[1] Ward, et al. *npj Comp. Mater.* 2016
Witman, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**

Milestone #1: Explainable ML models predict metal hydride thermodynamics

Thermodynamics (equilibrium plateau pressure) model validation

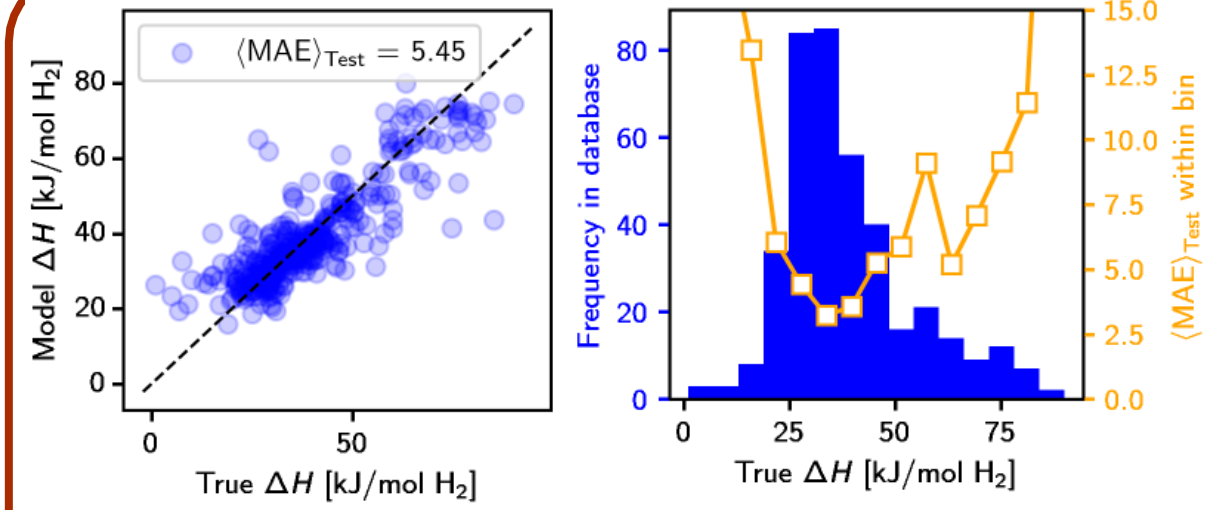


*ML model **can predict** $\hat{y} \equiv \ln(P^o_{eq}/P_o)$ with cross-validation test <MAE> = 1.4

Witman, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**

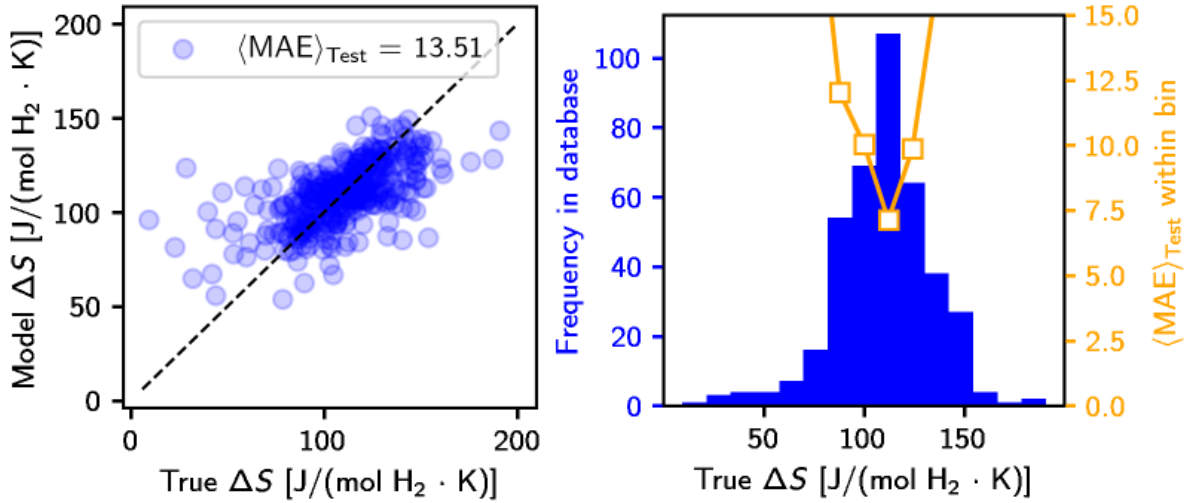
Milestone #1: Explainable ML models predict metal hydride thermodynamics

(3) Thermodynamics (individual ΔH and ΔS) model validation



Can predict $\hat{y} \equiv \Delta H$ with cross-validation test
<MAE> = 5.5 kJ/molH2

Can predict $\hat{y} \equiv \Delta S$ with cross-validation test
<MAE> = 14 J/(mol K)



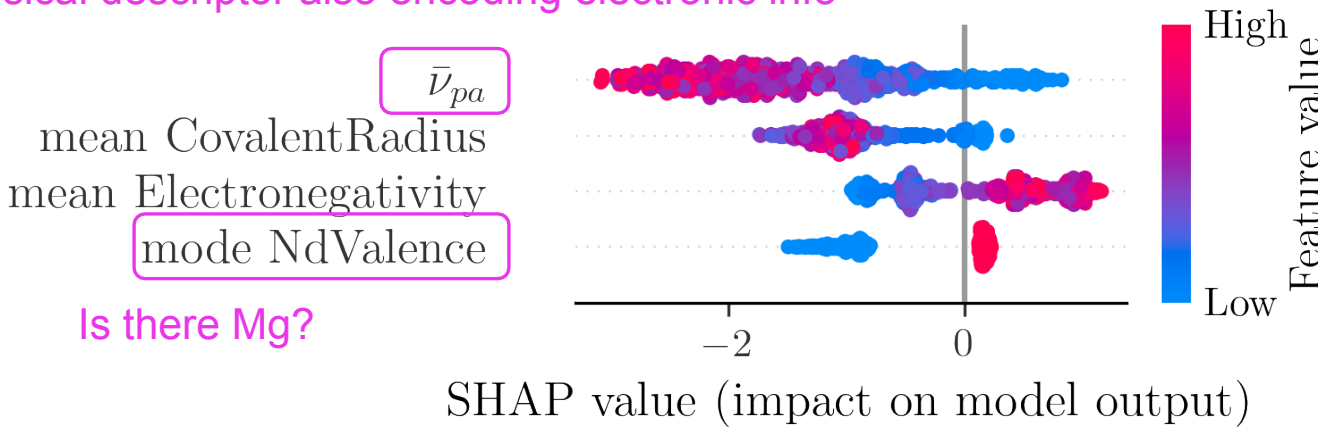
Witman, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**

Milestone #1: Explainable ML models predict metal hydride thermodynamics

Model interpretability with SHapely Additive Predictions (SHAP)[1]

How does a model's output depend on the value of a given feature?

"Elemental solid ground state volume/atom":
A physical descriptor also encoding electronic info



Is there Mg?

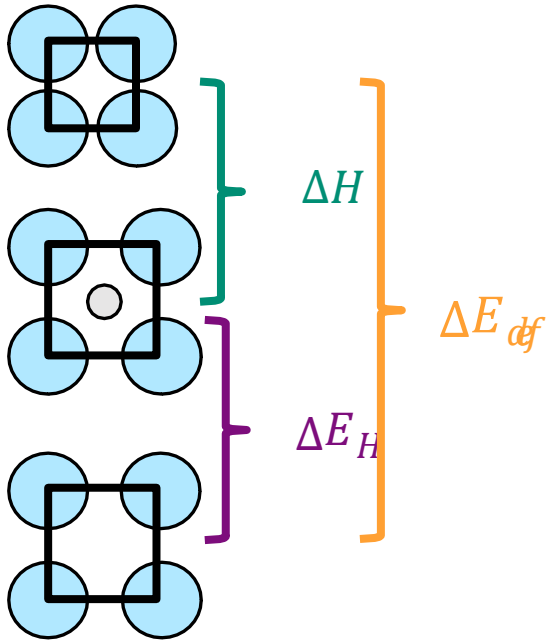
***Linear correlation** with $\bar{\nu}_{pa}$: $\ln\left(\frac{P^o_{eq}}{P_o}\right) \approx -m\,\bar{\nu}_{pa} + b$

[1] Lundberg, et al. *NIPS*, **2017**.
Witman, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**

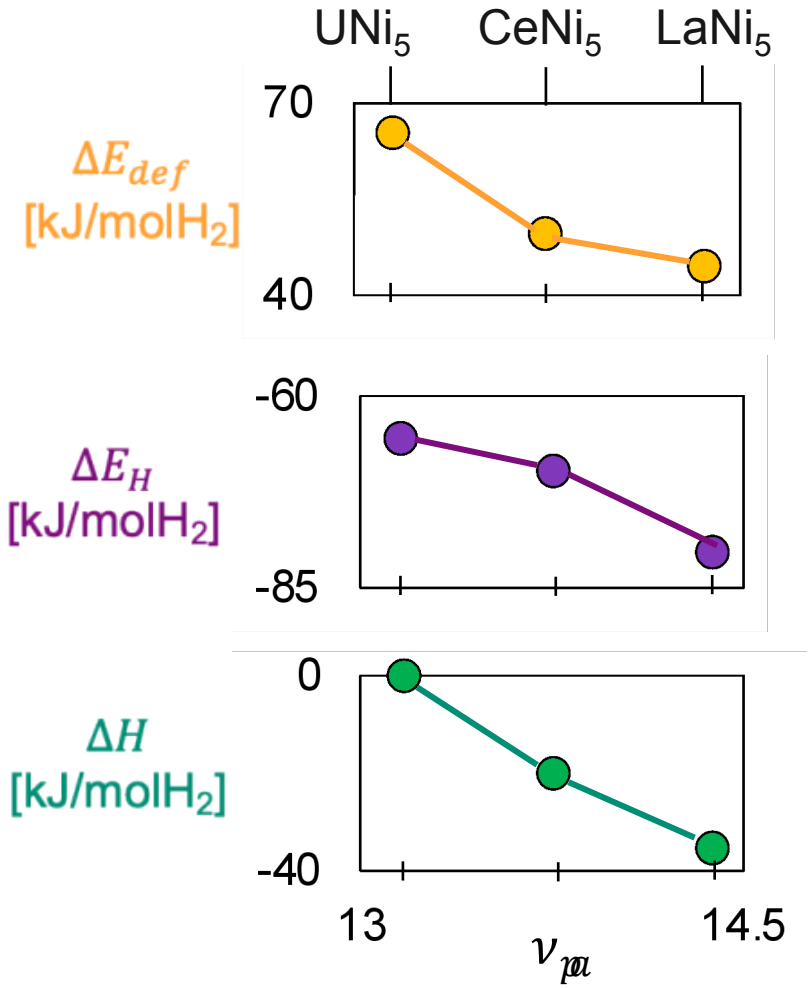
Milestone #1: Explainable ML models predict metal hydride thermodynamics

A DFT case study on correlation of hydride thermodynamics with $\bar{\nu}_{pa}$

Probe the reaction:
$AB_5 + 3.5H_2 \rightarrow AB_5H_7$



Lattice deformation penalty
H stabilization energy
Hydride enthalpy

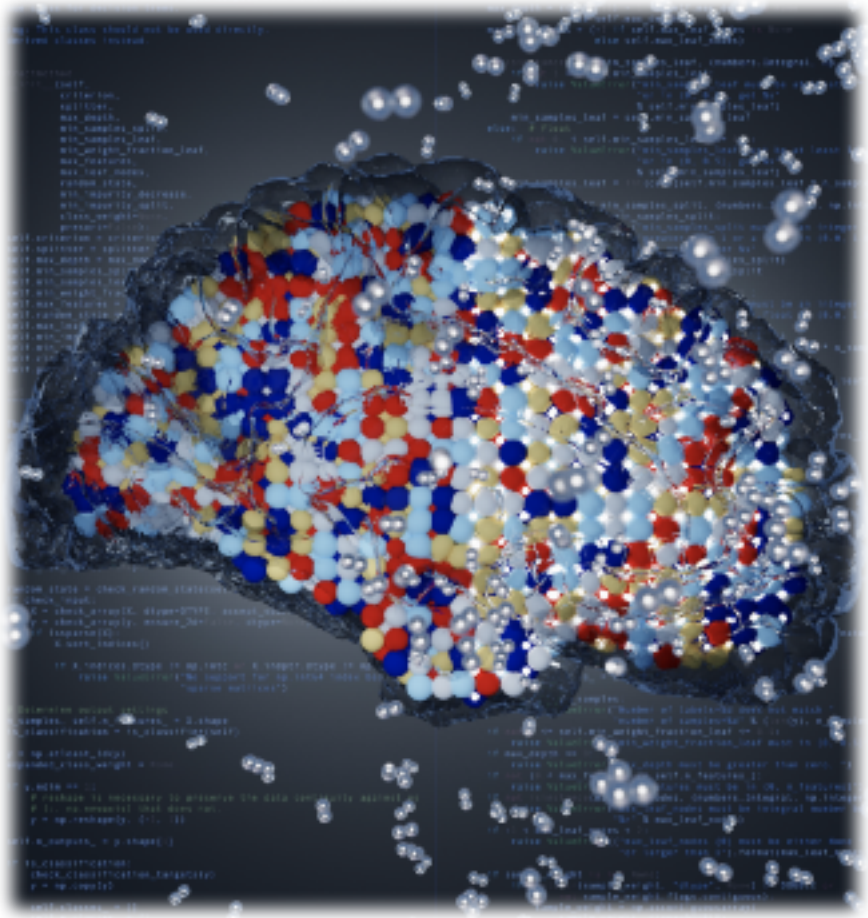


Decreases with $\bar{\nu}_{pa}$

Decreases with $\bar{\nu}_{pa}$

Net decreases with $\bar{\nu}_{pa}$

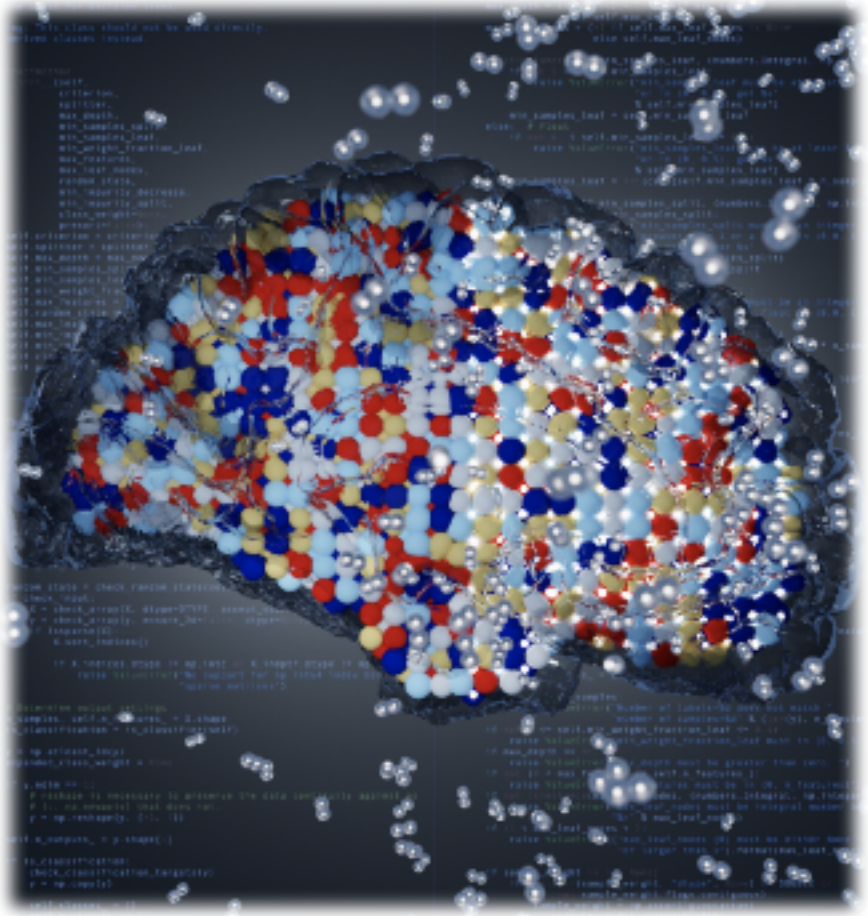
Witman, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**



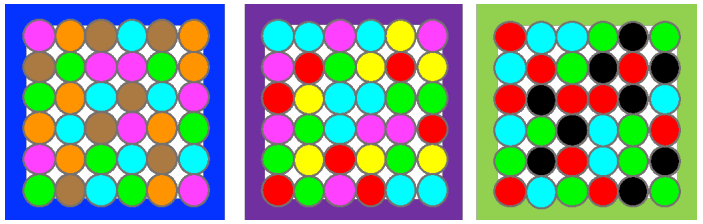
Conclusions:

- Compositional ML models can predict metal hydride thermodynamics
- Interpretability of ML models reveals design rules of increasing complexity
- DFT can validate ML-established design rules for specific hydride classes with limited throughput

Milestone #2: ML enables discovery of destabilized high entropy alloy (HEA) hydrides



Milestone #2: ML enables discovery of destabilized high entropy alloy (HEA) hydrides



1. Define chemical/materials exploration space

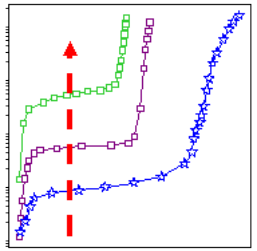
$\ln\left(\frac{P_{eq}}{P_o}\right)$ Model

2. High-throughput screening/property prediction

Phase Model

Manual Select

3. Apply necessary additional down-selection criteria

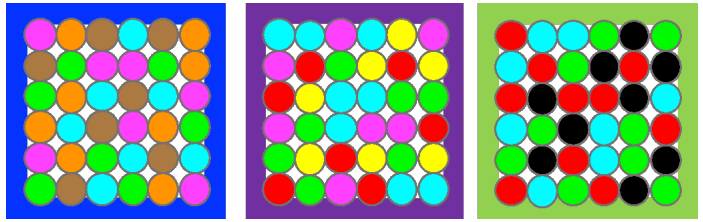


4. Experimental validation

Witman, Ek, Ling, Chames, Agarwal, Wong, Allendorf, Sahlberg, Stavila. *Chem. Mater.* 30 (11), **2021**

Milestone #2: ML-based discovery of destabilized high entropy alloy (HEA) hydrides

(1) HEA overview:



- > 4 elements, ~ equimolar
- Defined lattice type
- Solid solution character necessitates a compositional ML model

(2) Enumerating refractory HEA space

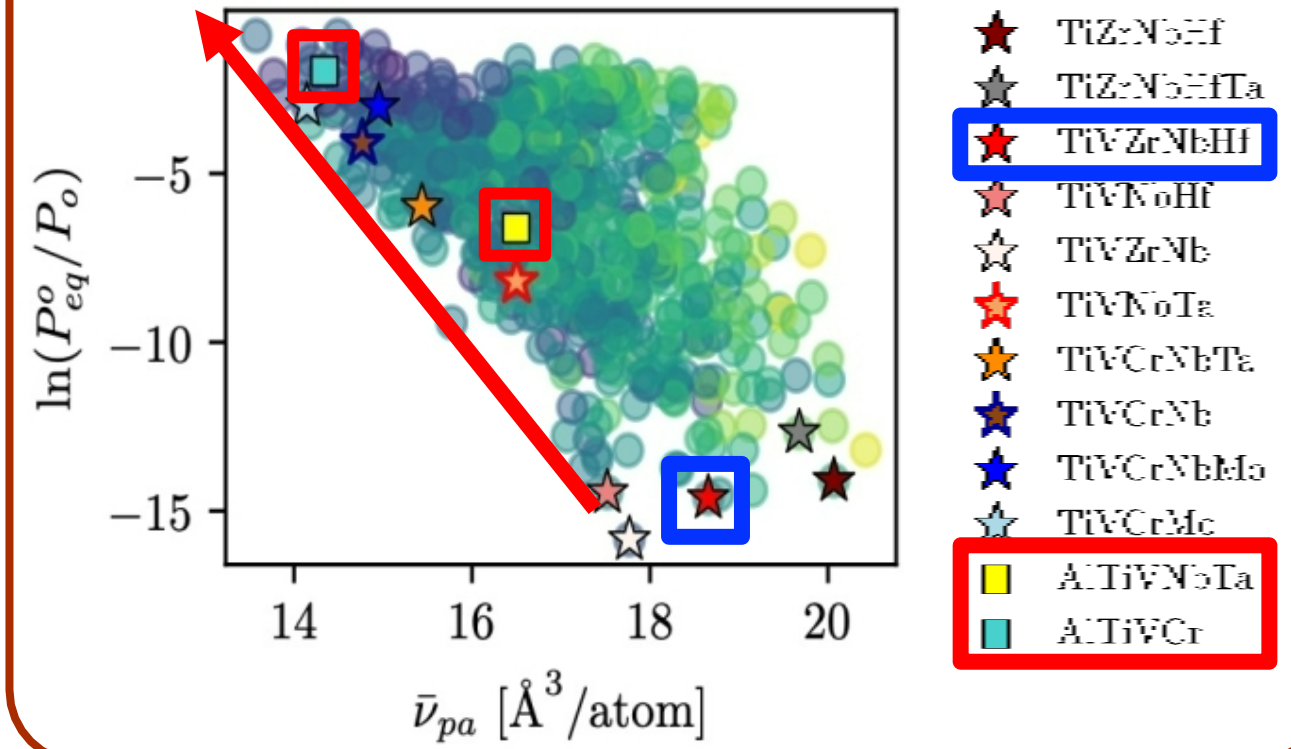
E = {Al, Ti, V, Cr, Zr, Nb, Mo, Pd, Hf, Ta}

$$\binom{E}{4} + \binom{E}{5} + \binom{E}{6} \rightarrow 672 \text{ compositions}$$

Far too many for experiments…

(3) Screening refractory HEA space

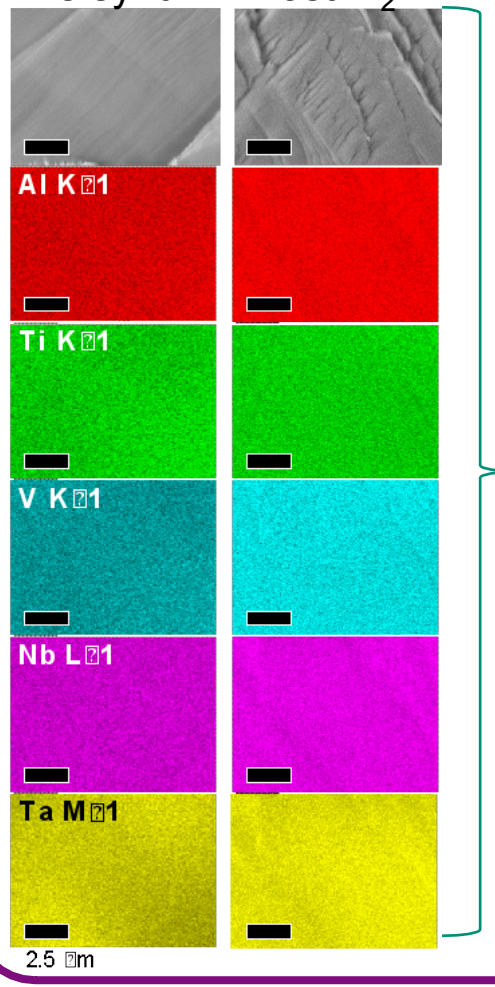
Destabilized hydrides experimentally confirmed!



Witman, Ek, Ling, Chames, Agarwal, Wong, Allendorf, Sahlberg, Stavila. *Chem. Mater.* 30 (11), **2021**

Milestone #2: ML enables discovery of destabilized high entropy alloy (HEA) hydrides

(1) AlTiVNbTa & AlTiVCr synthesis



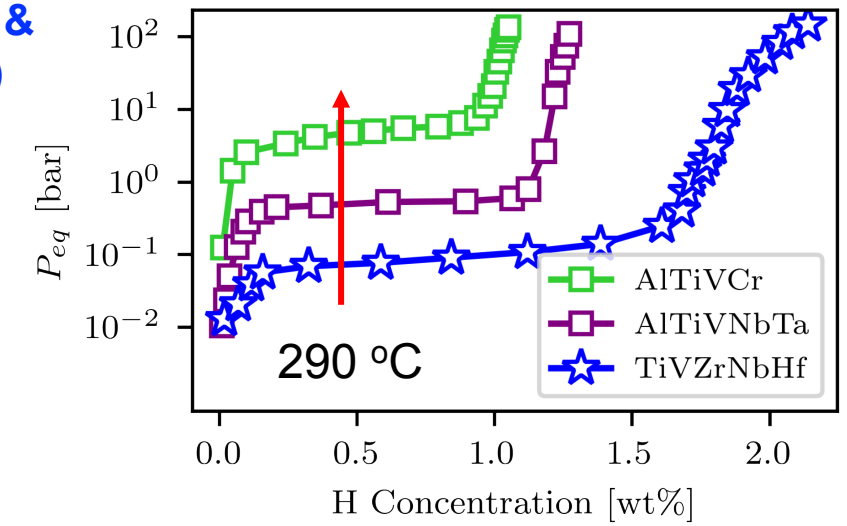
(Gustav, Martin, & Jeffrey)

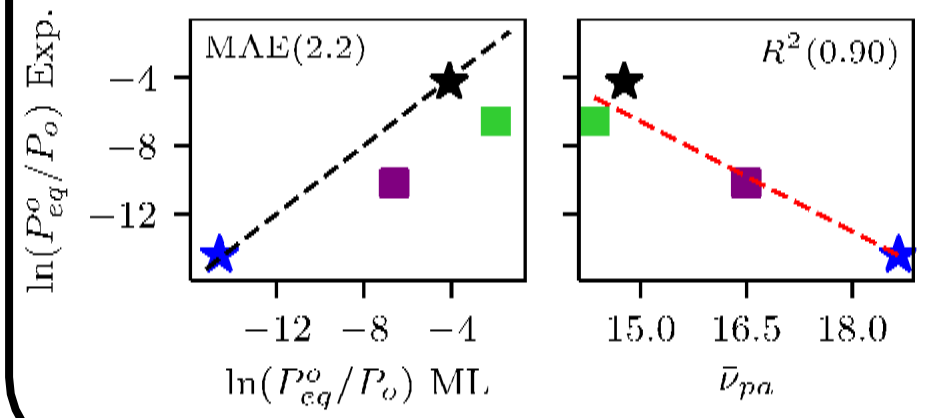
No elemental segregation

(2) ML model & design rule confirmed by PCT experiments

(Vitalie & Robert)

Successfully targeted destabilized hydrides (increase in P_{eq})





Validated ML model & design rule

Witman, Ek, Ling, Chames, Agarwal, Wong, Allendorf, Sahlberg, Stavila. *Chem. Mater.* 30 (11), **2021**

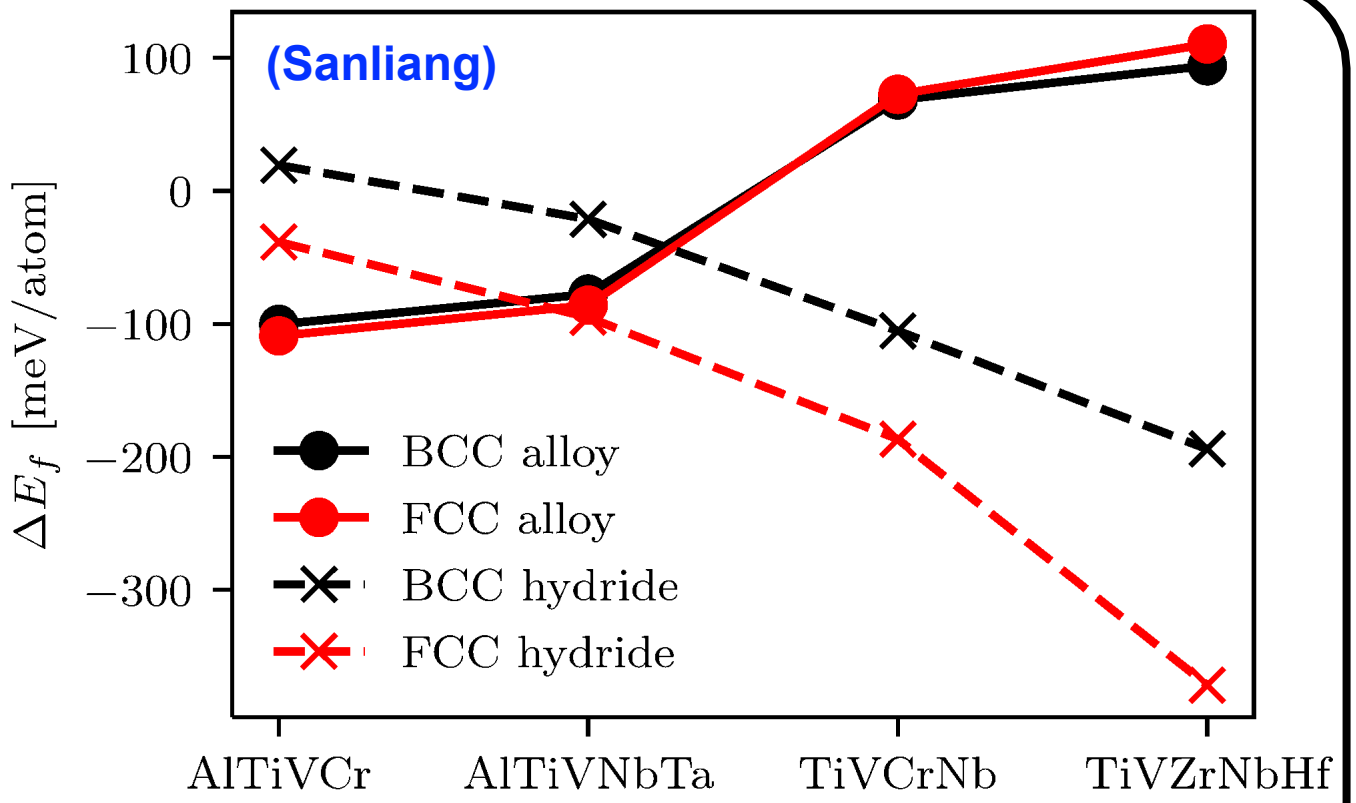
Milestone #2: ML enables discovery of destabilized high entropy alloy (HEA) hydrides

Investigating thermodynamic trends with Density Functional Theory

Compute 0K formation enthalpy of alloy and hydride for single HEA config.:

$$\Delta E_{f,\text{alloy}} = \frac{1}{N}\left(E_{\text{alloy}} - \sum_{i\in\{\text{alloy}\}} E_i\right)$$

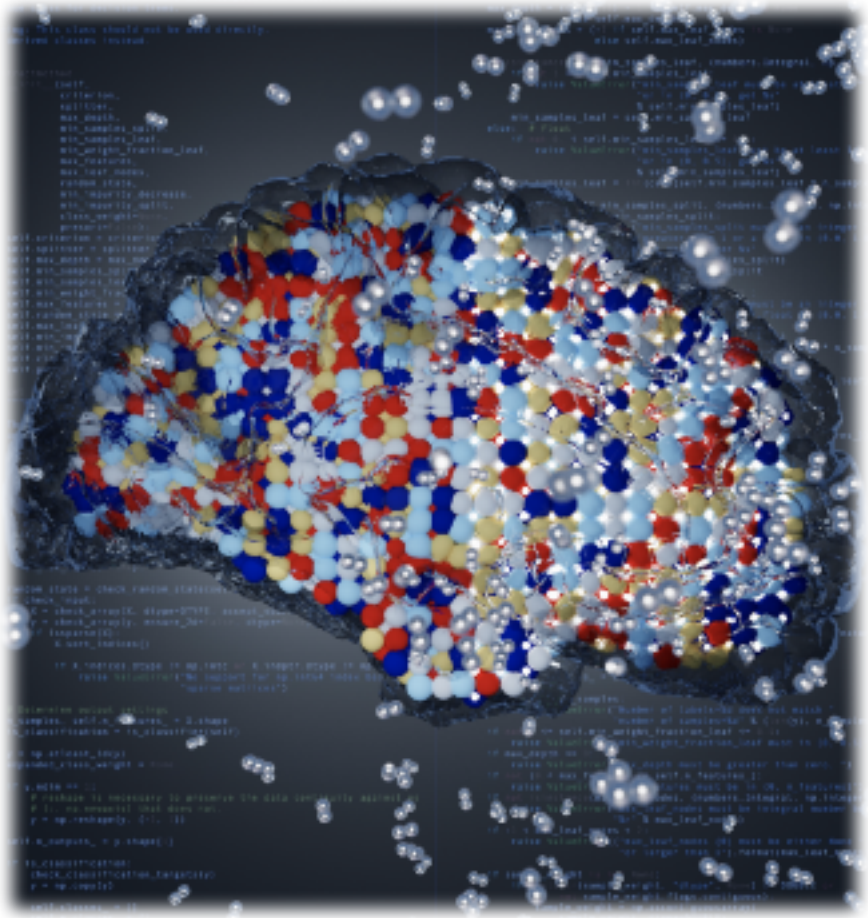
$$\Delta E_{f,\text{hydride}} = \frac{1}{N}\left(E_{\text{hydride}} - \left(\frac{N}{2}\right)E_{H_2} - \sum_{i\in\{\text{alloy}\}} E_i\right)$$



Analysis Reveals:

- Enthalpic driving force for BCC alloy -> FCC transition @ high H/M ratios
- The general correlation of less stable lattice = more stable hydride (similar to $\bar{\nu}_{pa}$ design rule)

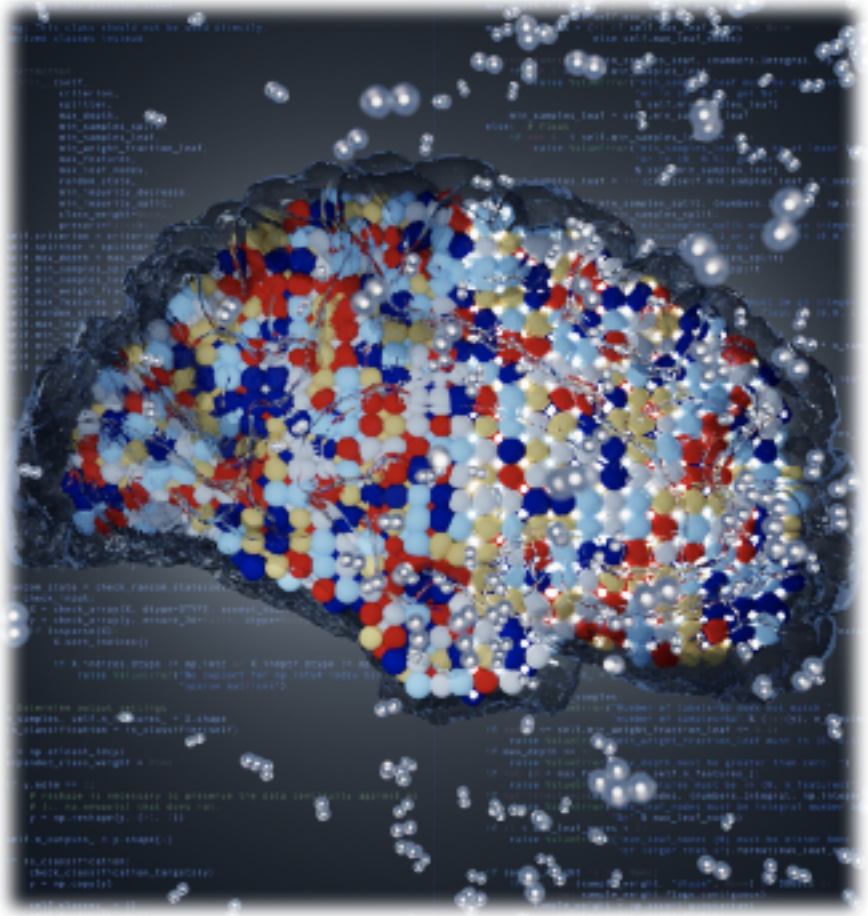
Witman, Ek, Ling, Chames, Agarwal, Wong, Allendorf, Sahlberg, Stavila. *Chem. Mater.* 30 (11), **2021**



Conclusions:

- Introduced a powerful ML capability for high-throughput screening
- ML-directed and experimentally validated synthesis of HEA hydrides
- DFT-enabled insights into thermodynamic trends

Milestone #3: ML screening & identification of *Pareto* optimal HEA hydrides



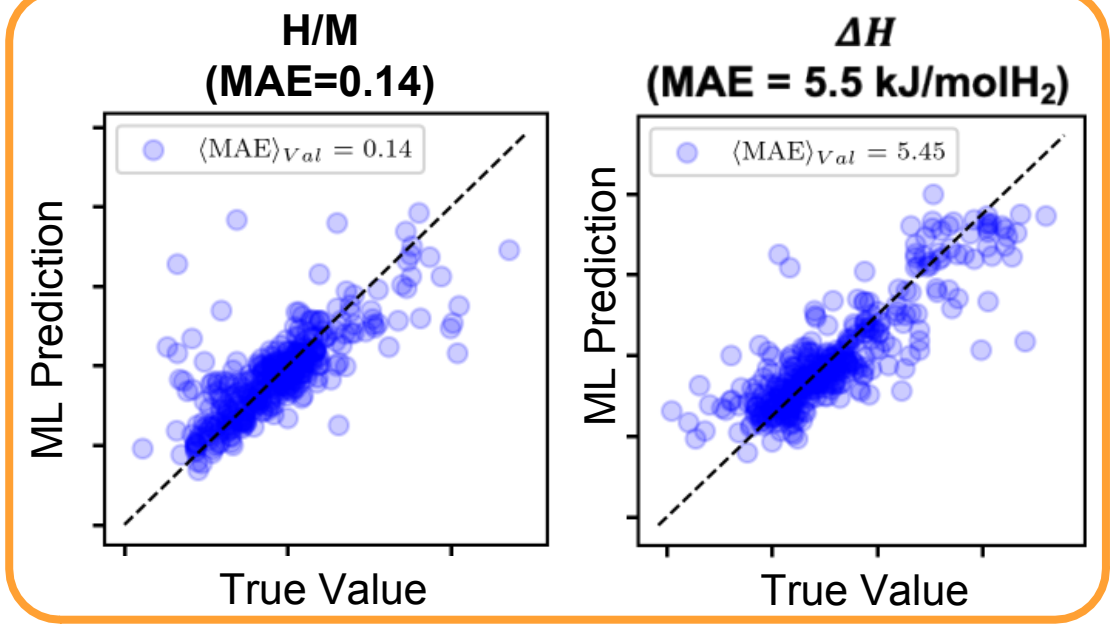
Milestone #3: ML screening & identification of Pareto optimal HEA hydrides

(1) Screening an expansive HEA space

E = {Mg, Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, Nb, Mo, Pd, Hf, Ta}

$$\binom{E}{4} + \binom{E}{5} + \binom{E}{6} \rightarrow 20{,}944 \text{ compositions}$$

(2) Multiple ML property predictions

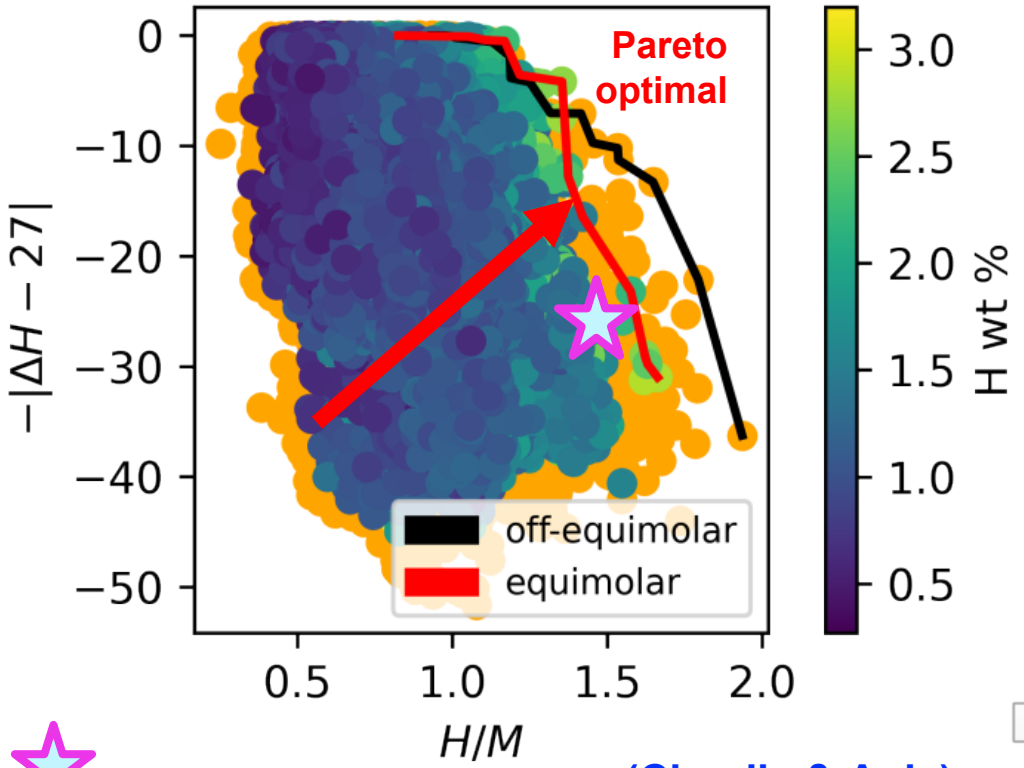


In preparation

(3) Identification of ~100 Pareto optimal materials for stationary storage

Objectives / Quantity to maximize:

- Optimal thermodynamics -> $-|\Delta H - 27|$
- High volumetric capacity -> H/M
- High gravimetric capacity -> Hwt%
- Raw material cost -> -cost



= Experimental validation (Claudia & Anis)

Milestone #3: ML screening & identification of Pareto optimal HEA hydrides

(1) Screening an expansive HEA space

E = {Mg, Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, Nb, Mo, Pd, Hf, Ta}

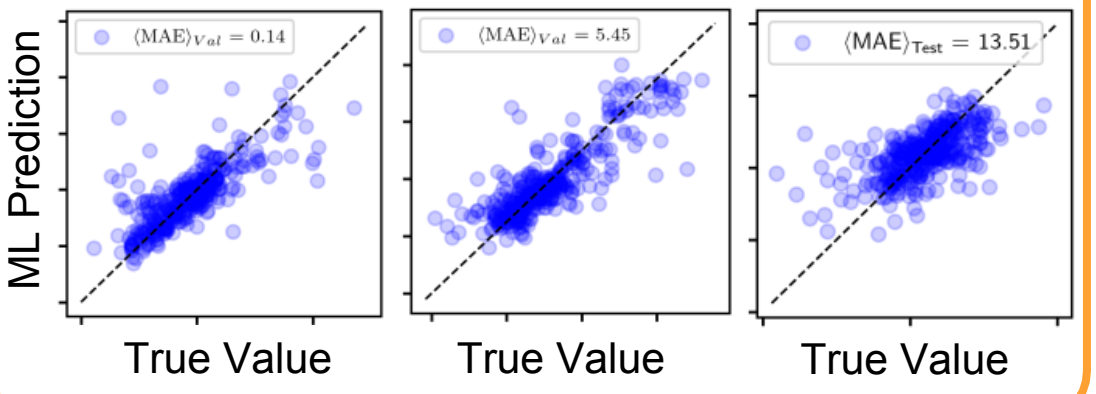
$$\binom{E}{4} + \binom{E}{5} + \binom{E}{6} \rightarrow 20{,}944 \text{ compositions}$$

(2) Multiple ML property predictions

H/M (MAE = 0.14)

ΔH (MAE = 5.5 kJ/molH$_2$)

ΔS (MAE = 14 J/molK)

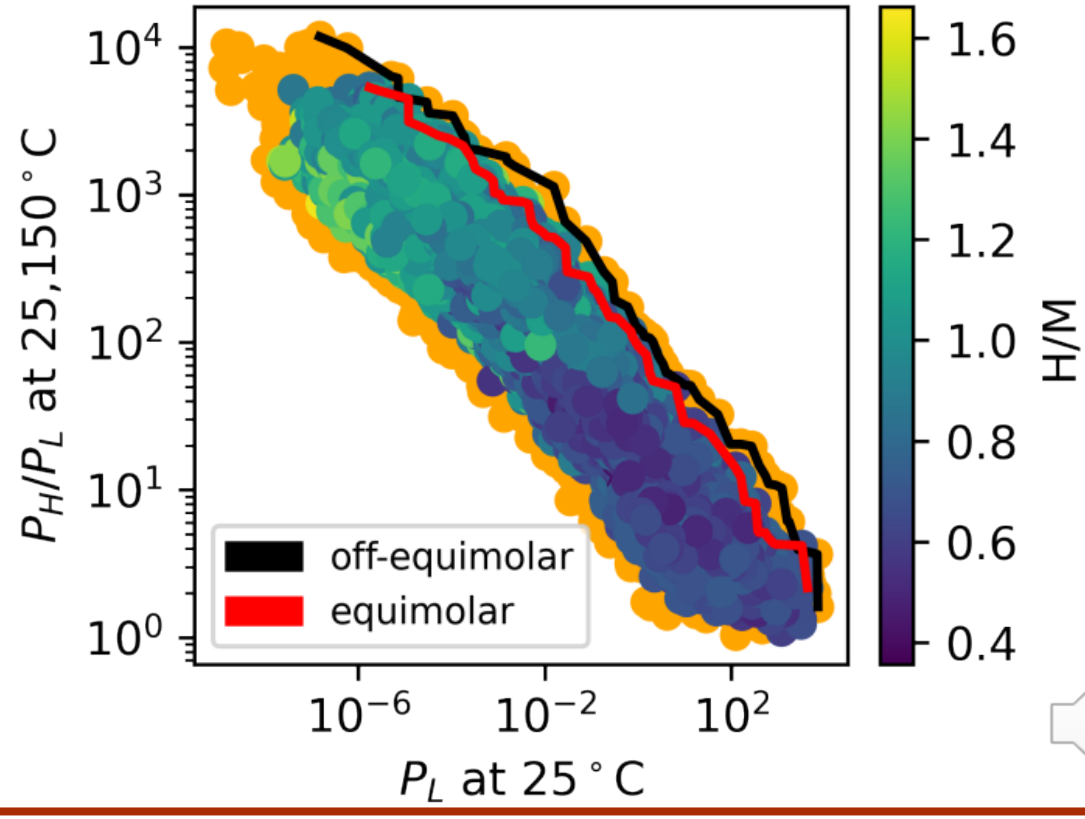


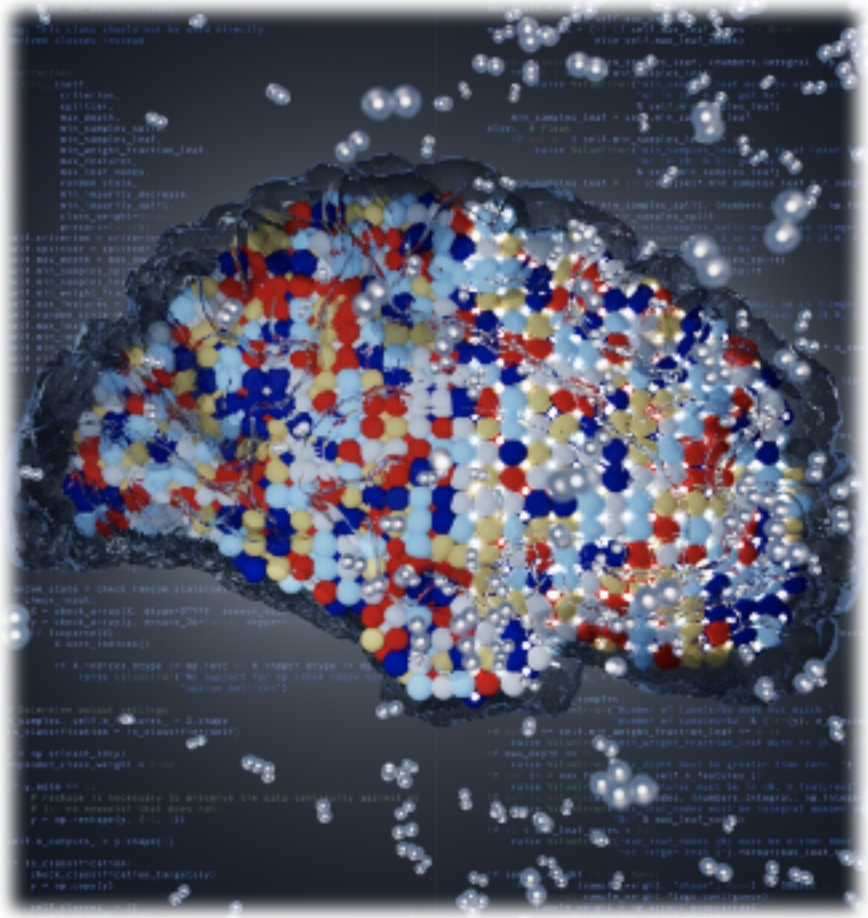
In preparation

(3) Identification of ~100 Pareto optimal materials for hydrogen compressors

Objectives / Quantity to maximize:

- P_{eq} @ T_L= 25 C -> P_L
- Compression ratio @ T_H= 150 C -> P_H/P_L
- High volumetric capacity -> H/M
- Raw material cost -> -cost





Conclusions:

- Given a set of ML models for various hydride properties, Pareto frontiers can be determined
- Random off-equimolar perturbations substantially advance the Pareto frontier, indicating potential for future improvement
- We have highlighted (with experimental validation pending) Pareto optimal compressor and stationary storage materials

Future Work:

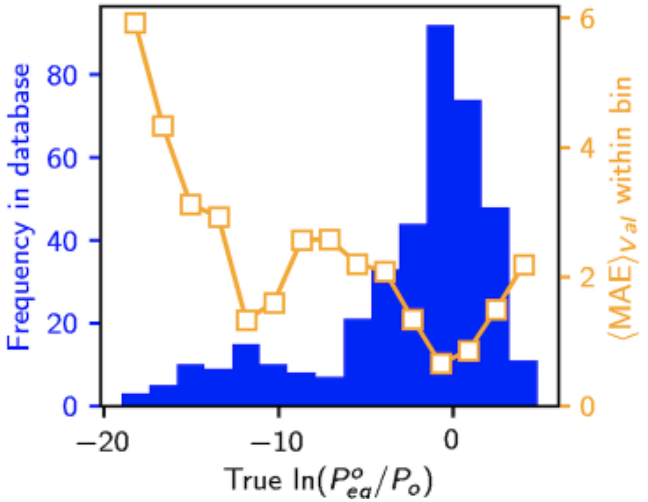
Overcoming barriers to ML-driven hydride discovery
(e.g. how to deal with limited data)

More/Better Experimental Data: Promoting standardization, reproducibility, and community-based "high-throughput" data acquisition

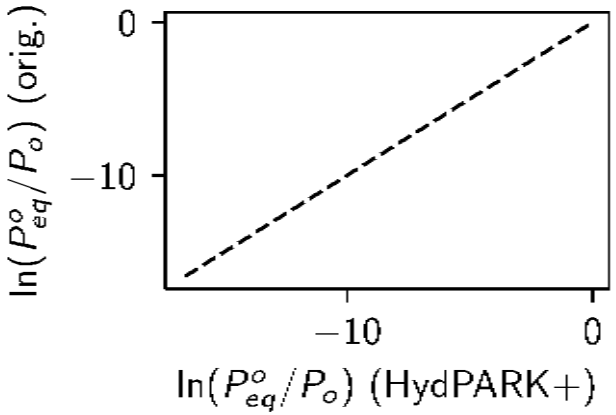
Challenge: Accuracy and accessibility of HydPARK data

Lack of updatability: data stored in "offline" CSV file
Lack of completeness: **15%** of HydPARK entries are usable
Lack of consistency: different values for identical materials

Very non-uniform distribution of target



Addition of new data systematically shifts predictions



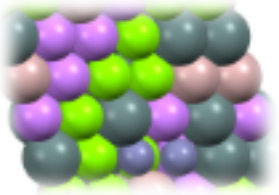
Solution: NREL DataHub app and backend for standardized PCT upload and storage

Much faster data acquisition than can be achieved by one group alone

More/Better Computational Data: If experimental measurements and physics-based simulation are too expensive, accelerate the latter with ML

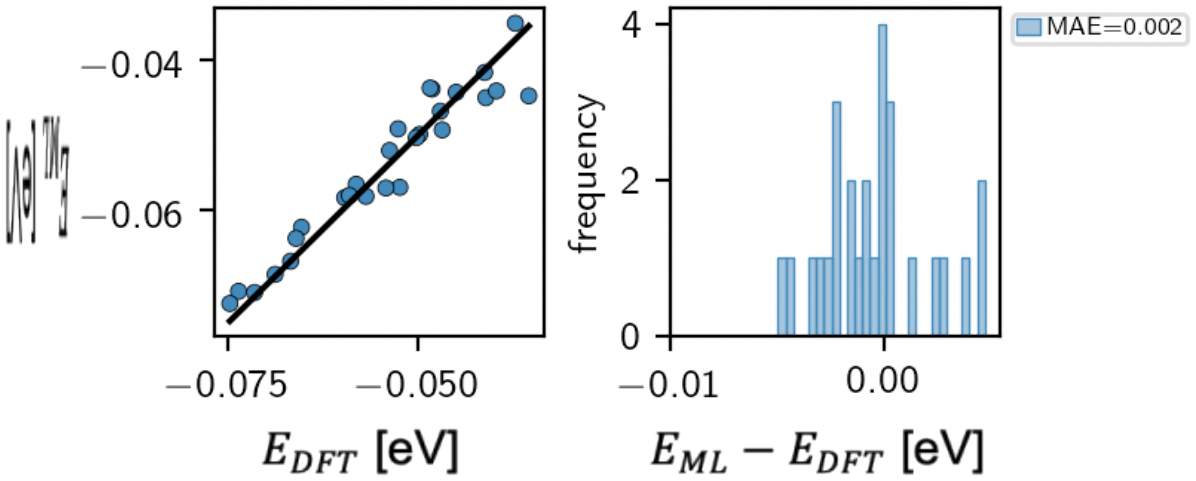
Challenge: Need an accurate, many-element potential energy surface (PES) for simulations (MD or MC)

PES choices: Force Field , ML PES, DFT



Not necessarily transferable/ generalizable

Too expensive for extensive sampling in complex systems



Solution: Direct atomistic simulation & screening of metal hydride thermodynamics?

Thank you for your attention.

Questions?

Contact: mwitman@sandia.gov