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Accelerating data-driven discovery of materials for hydrogen storage and generation

PRESENTED BY

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Collaborators & Acknowledgements

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HyMARC Hydrogen Materials Advanced Research Consortium

LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

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

Personal transportation (Toyota/Hyundai)

Air Travel (Airbus)

Home energy storage (Lavo)

Utility-scale generation (Shell)

Motivation: Do undiscovered materials exist that could improve upon conventional H_2 storage and generation technologies?

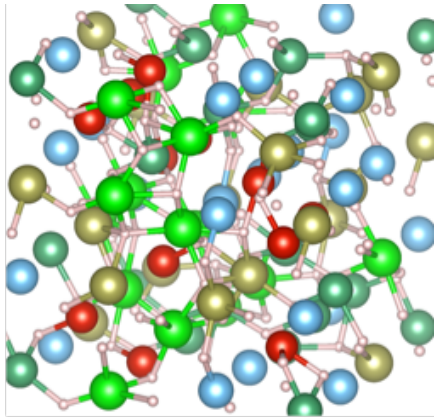
Storage objectives:

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

700 bar

"Conventional" (Compressed gas)

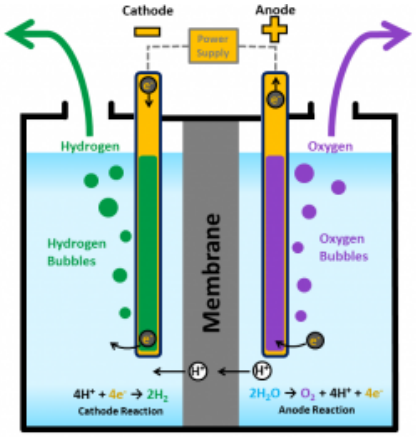
vs.



Material X ??

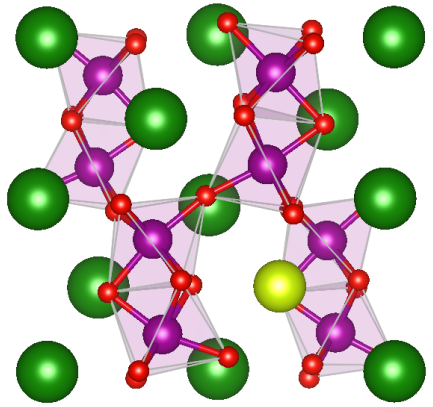
Generation objectives:

- Water-splitting using only renewable energy
- Lower overpotentials than electrolysis
- Practical/cost-effective



"Conventional"
1.2 V in theory
1.8 V in practice

vs.



Material Y ??

Approach: How are new materials discovered?

Experiments

Chemical intuition

↓

Synthesize material

↓

Characterize material

↓

Test Performance

Physics-based simulations

First-principles theory

↓

Run simulation &
predict performance

Machine/statistical learning (ML)

Training data labels/values:
y_i (from experiments or computation)

↓

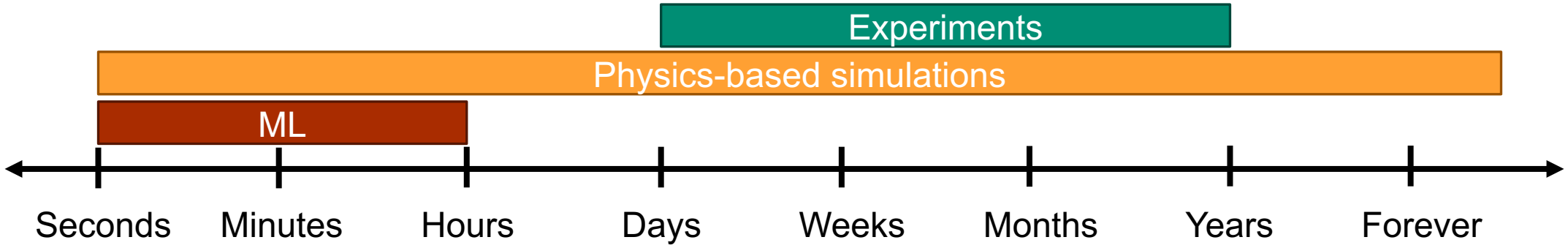
Featurize materials:
$LaNi_5 \rightarrow \boldsymbol{x} = \{\bar{v}_{pa}, \bar{r}_{cov}, \bar{\chi}, \ldots\}$

↓

Train a model:
$\hat{y}_i = f(\boldsymbol{x}_i; \theta)$ and $\mathcal{L} = \sum(\hat{y}_i - y_i)^2$

↓

Run model &
predict performance



Approach: Use of data science and machine learning techniques can…

1) Model material properties *and* provide interpretable predictions to improve fundamental understanding [1]

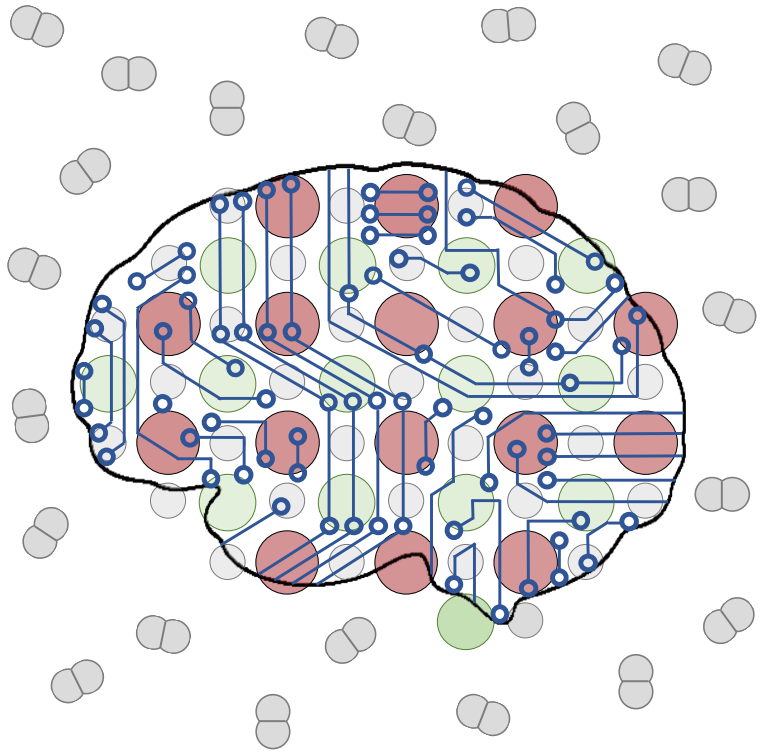
2) Elucidate the design rules (structure-property relationships) for optimal materials [1,2]

3) High-throughput screen large material space to identify top candidates orders of magnitude faster than brute-force experiments or physics-based simulations [3,4,5,6]

4) Accelerate physics-based simulations (critical when experimental training data is lacking) [7]

[1] Witman, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**
[2] Witman, Ling, Stavila, Wijeratne, Furukawa, Allendorf. *Mol. Sys. Des. & Eng.*, 5, **2020**
[3] Ek, Nygard, Pavan, Montero, Henry, Sorby, Witman, et al. *Inorg. Chem.*, 60 (2), **2021**
[4] Witman, Ek, Ling, Chames, Agarwal, Wong, Allendorf, Sahlberg, Stavila. *Chem. Mater.,* **2021***, Accepted*
[5] *In preparation*
[6] Ambrosini, Witman, McDaniel. *Provisional Patent*, **2020**.
[7] *In preparation*

H_2 **Storage**: A data driven roadmap to discover optimal hydrogen storage alloys



Milestone #1:
Develop an explainable ML framework to predict metal hydride thermodynamics and understand design rules [1]

Milestone #2:
Demonstrate our ability to identify and synthesize new hydrides exhibiting targeted thermodynamic properties [4]

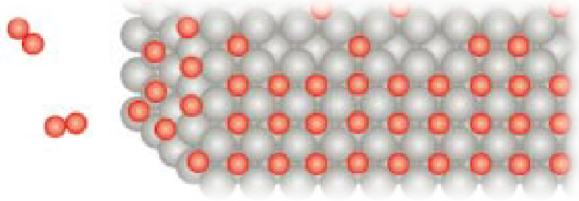
Milestone #3:
Discover the highest performing hydrides across multiple objectives (i.e., Pareto optimal) from an intractably large alloy space [5]

[1] **Witman**, Ling, Grant, Walker, Agarwal, Stavila, Allendorf. *J. Phys. Chem. Lett.*, 11 (1), **2020**
[4] **Witman**, Ek, Ling, Chames, Agarwal, Wong, Allendorf, Sahlberg, Stavila. *Chem. Mater.,* **2021***, Accepted*
[5] *In preparation*

Milestone #1: Explainable ML models predict metal hydride thermodynamics and elucidate simple, first-order design rules

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

H_2 @ P_{eq}, T Metal Hydride



*Intractable for rigorous, atomistic simulation in high-throughput

Training data

HydPARK metal-hydride database

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

400 out of 2500 are ML-ready…

Featurizing materials

Features derived **only from composition**

e.g. $Zr_{0.04}Ti_{0.96}Nb_{0.04}Fe_{0.95}$ →

$$\boldsymbol{x} = \{\bar{\nu}_{pa}, \bar{r}_{cov}, \bar{\chi}, \ldots\} \in \mathbb{R}^{145}$$

$$\bar{\boldsymbol{\nu}}_{\boldsymbol{pa}} = \sum_i f_i \nu_i$$

$f_i \equiv$ composition fraction

$\nu_i \equiv$ ground state vol. per atom

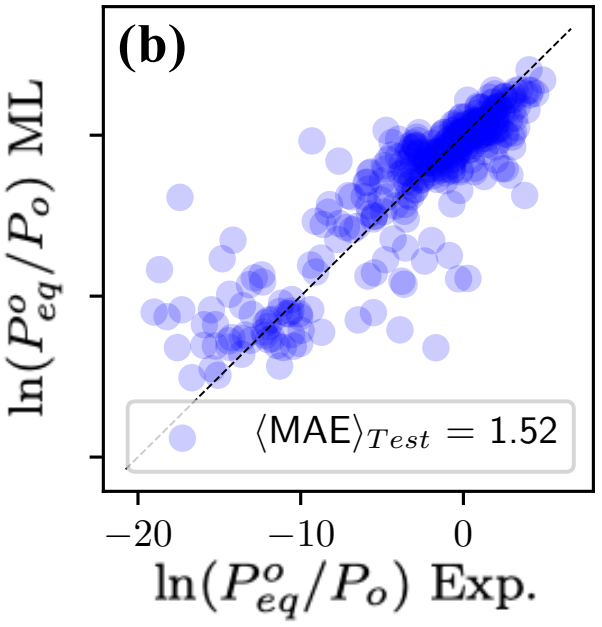
Milestone #1: Explainable ML models predict metal hydride thermodynamics and elucidate simple, first-order design rules

Gradient boosting trees performance

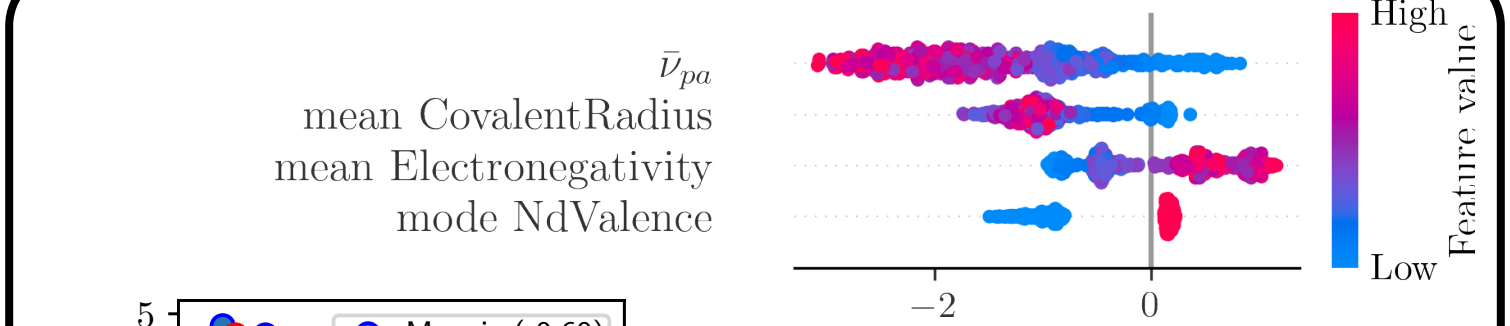
$y_i = \ln(P^o_{eq}/P_o)$ Exp.

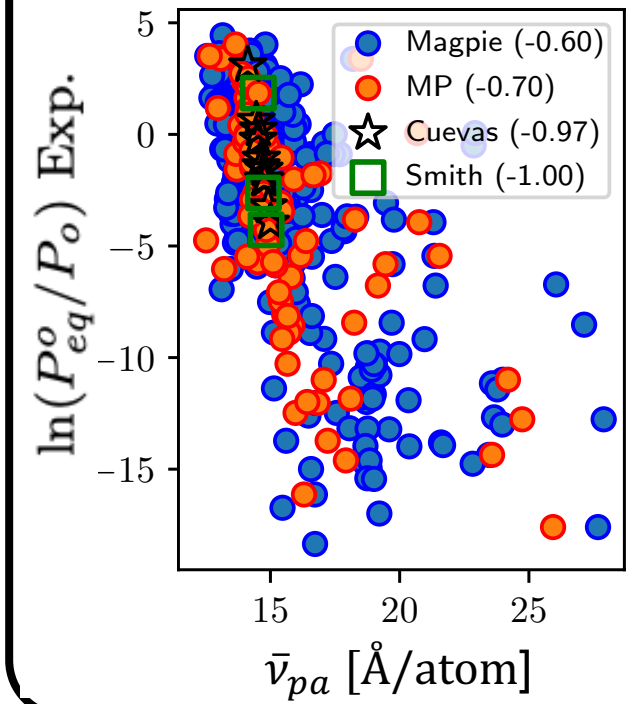
$\hat{y}_i = \sum_{k=1}^{K} f_k(\boldsymbol{x}_i)$ ML

$\mathcal{L} = \frac{1}{n}\sum_i (\hat{y}_i - y_i)^2$ Loss



Model explainability



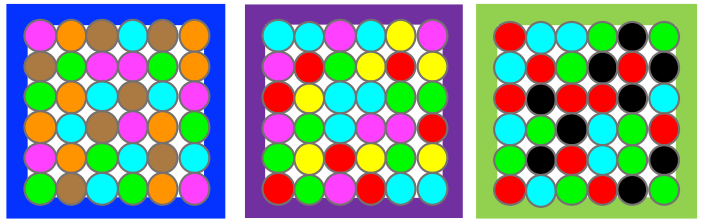


First-order design rule applicable across structural classes & diverse composition space!

$$\ln(P^o_{eq}/P_o) \approx C_1\,\bar{\nu}_{pa} + C_2$$

Milestone #2: ML model rapidly screens for high entropy alloy (HEA) hydrides with a desired thermodynamic stability

HEA overview:



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

Enumerating refractory HEA (rHEA) space:

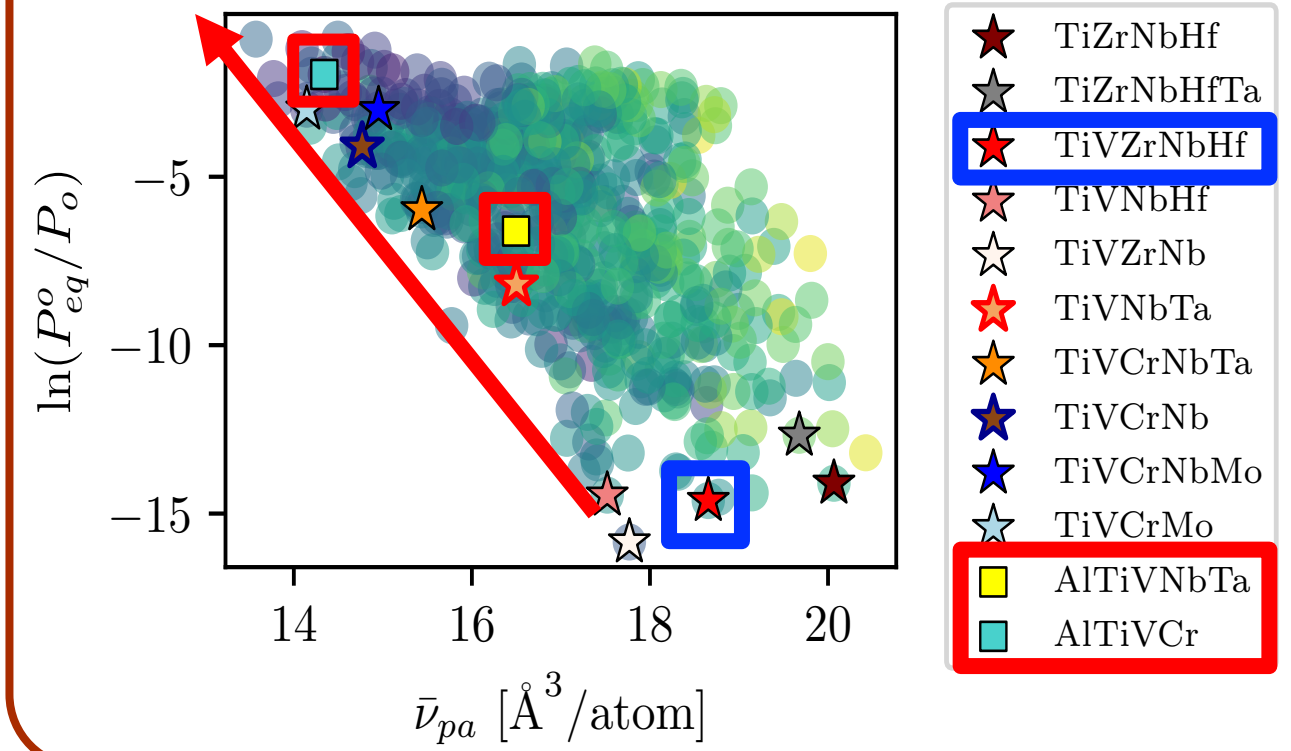
E = {Al, Ti, V, Cr, Zr, Nb, Mo, Pd, Hf, Ta}

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

Far too many for experiments…

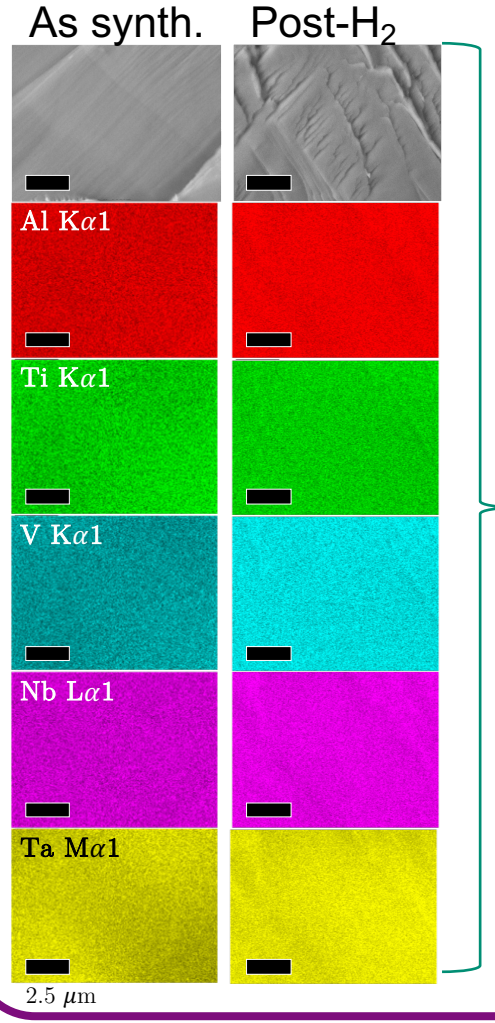
Screening rHEA space:

Novel, destabilized HEA hydrides



Milestone #2: ML model rapidly screens for high entropy alloy (HEA) hydrides with a desired thermodynamic stability

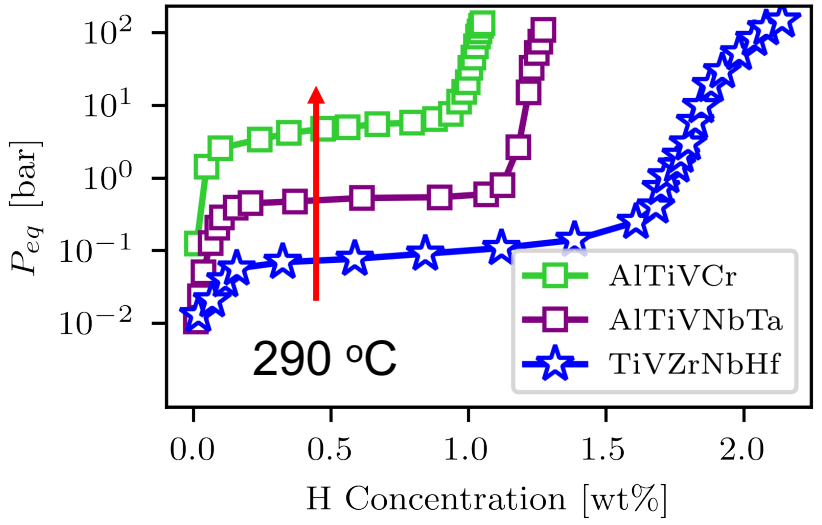
AlTiVNbTa & AlTiVCr synthesis

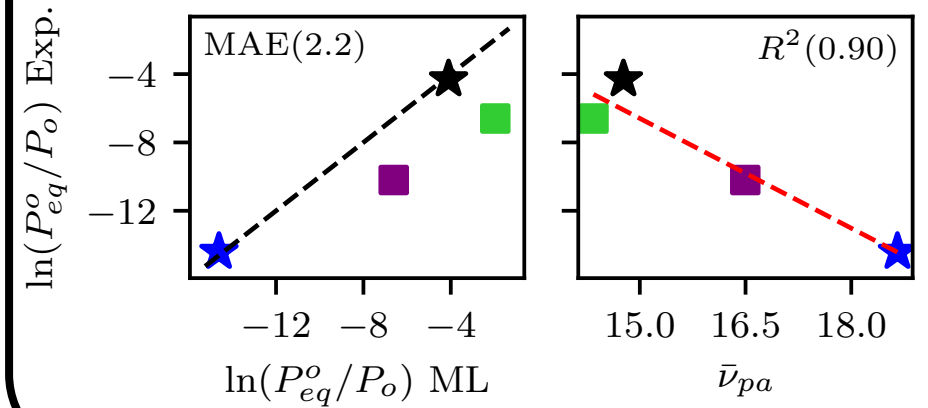


No elemental segregation

ML model & design rule confirmed by experiments

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





Validated ML model & design rule

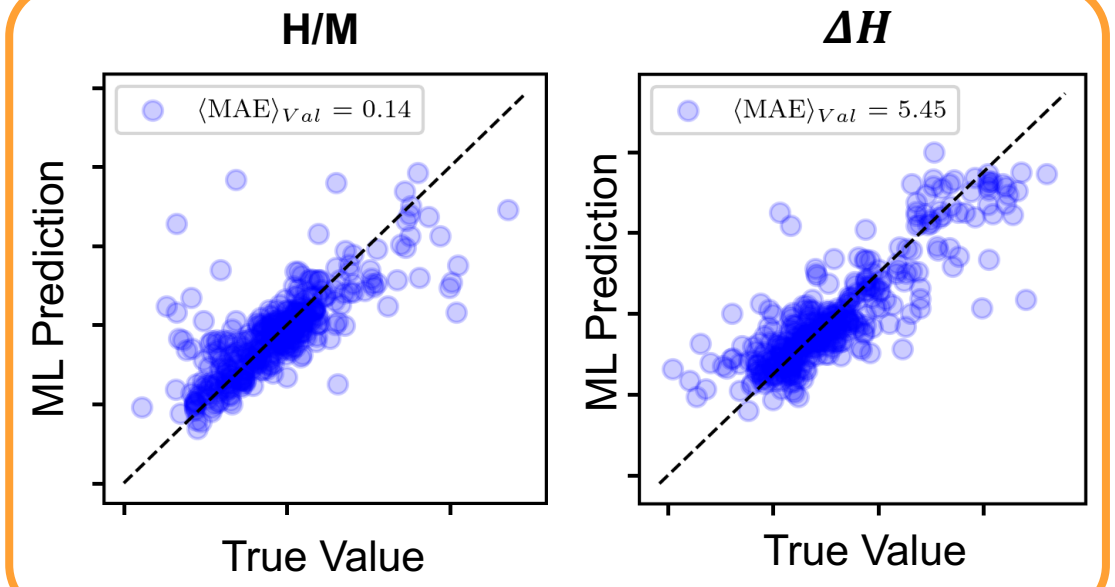
Milestone #3: Identify the Pareto optimal hydrides from an expansive space of ~21,000 candidate HEAs [3]

Screening a novel, expansive HEA space

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

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

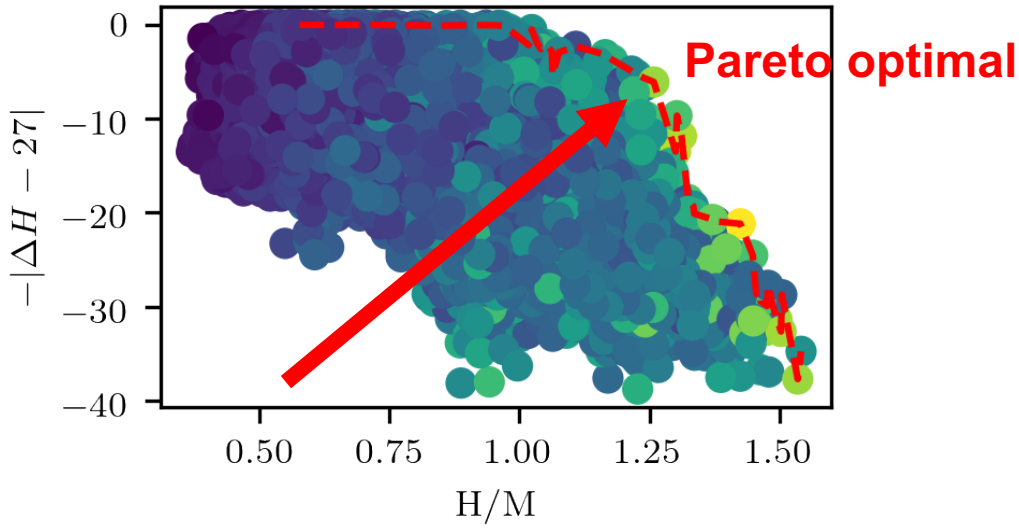
Multiple ML property predictions



Identification of ~100 Pareto optimal materials

Objectives / Quantity to maximize:

- Optimal thermodynamics -> $-|\Delta H - 27|$
- High volumetric capacity -> H/M
- High gravimetric capacity -> Hwt%
- Low Cost -> -cost [USD/kg]



ML: ***Seconds*** to identify all Pareto optimal candidates
Experiment: ***Years*** to identify one by trial-and-error

Ongoing Milestones: Address the lack of robust, standardized, and centralized hydrogen storage data

NREL Data Hub app for standardized experimental data reporting

- Better experimental reproducibility
- More standardized data reporting
- More efficient data collection
- Better ML models!

Experimental parameters/metadata (string fields):
1. Composition = TiVNbCr
2. Mechanical preparation = Arc Melting
3. Branch = Desorption

...TBD

Specialty parameters/metadata (string fields):
1. If nanoscale hydride: N/A
2. If nanoscale hydride: N/A
3. If nanoscale hydride: N/A

...TBD

Thermodynamic data (variable row data table):

dH	dS	van't Hoff Pts	T	P	H/M	Step
47	122	(des1_348K.csv, 16)	348 K	0.202	1.42	1
		(des2_373K.csv, 9)	373 K	0.600	1.39	
		(des3_388K.csv,10)	383 K	1.08	1.43	

$$\Delta H = -m * \frac{8.3145}{1000} * \left(\frac{1}{T}\right)$$
$$\Delta S = b * 8.3145$$

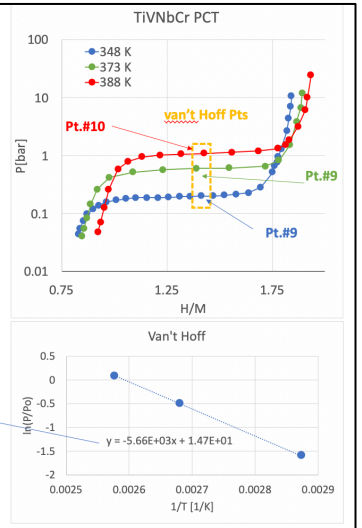
Capacity data:
1. Total capacity [H/M] = 1.9
2. Total reversible capacity [H/M] = 0.9

Step	H/M
1	0.9

# of steps/rows must equal max of Step col above

Kinetic data:

...TBD

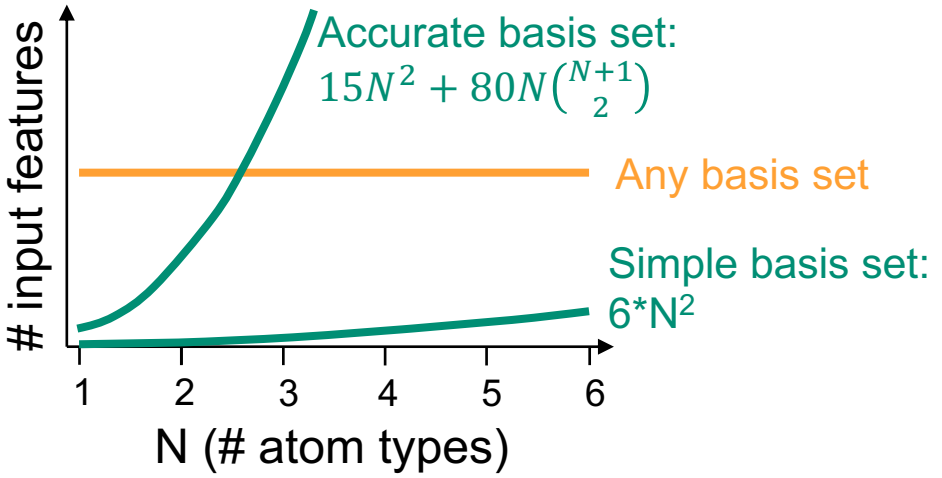


Data that is actually plotted on PCT:
(Each csv has col1=H/M, col2 = P)
des2_373K.csv
des1_348K.csv
pct3_388K.csv

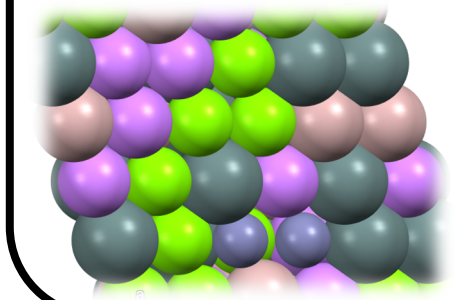
Raw PCT instrumentation file for reproducibility:
(User responsible for creating *.csv above from this file)
des2_373K.dat
des1_348K.dat
des3_388K.dat

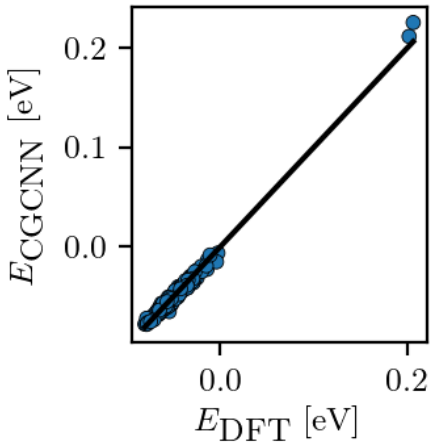
Surrogate models for DFT in many element systems

Conventional vs. *Graph Convolutional* NN



5 element system…
w/accurate potential:





Research Aspirations (in 5 years can we confidently say that we have…)

Storage

- Performed truly comprehensive HEA screening for ***Pareto optimal hydrides?***

Generation

- An ML framework to identify ***optimal metal oxides*** for thermochemical water-splitting?

Data

- Created ***standardized data management tools*** for hydrogen storage experiments?

Simulations

- ML driven simulations that can predict physical/chemical properties of ***many-element hydrogen/material systems***?

More data & better ML methods leads to

- Improved models
- More accurate materials predictions
- Faster experimental success
- *Quicker hydrogen technology adoption*

Thank you for your attention.

Questions?

Contact: mwitman@sandia.gov