Extracting an Empirical Intermetallic Hydride Design Principle from Limited Data via Interpretable Machine Learning

PRESENTED BY

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Collaboration Group:

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HyMARC Hydrogen Materials Advanced Research Consortium

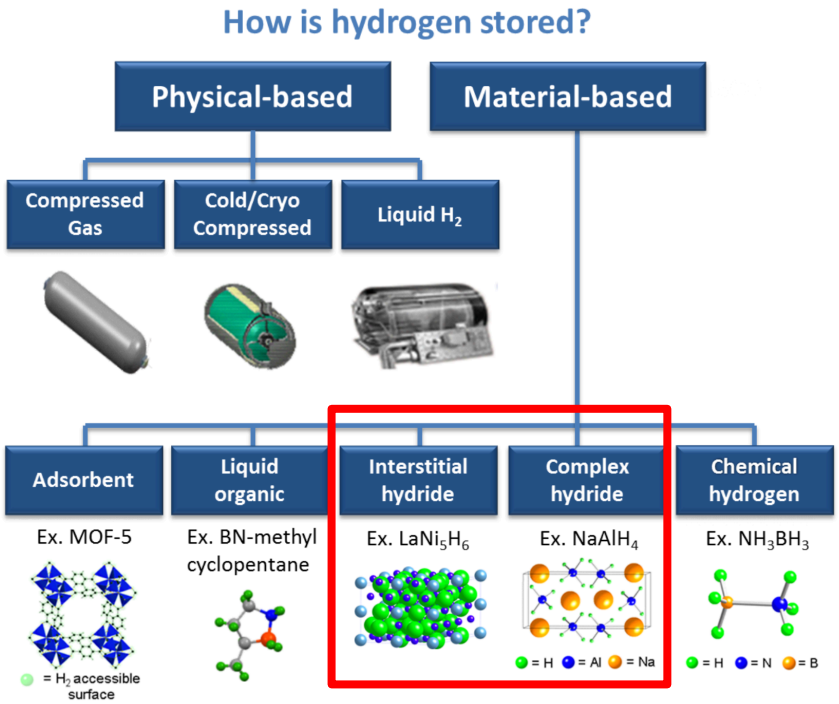
LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Economical storage of hydrogen is critical for enabling a variety of zero emission technologies

E.g., the Toyota Mirai is a commercially available fuel cell vehicle whose hydrogen is compressed and stored onboard

https://www.businessinsider.com/this-toyota-fuel-cell-car-can-power-your-house-2014-11

A material that meets all DOE technical targets for on board hydrogen storage could send the technology mainstream:



https://www.energy.gov/eere/fuelcells/hydrogen-storage

(1) Thermodynamics of hydriding, (2) hydrogen capacity, and (3) absorption kinetics decide applicability of the hydride

Table 2 Intermetallic compounds and their hydrogen-storage properties

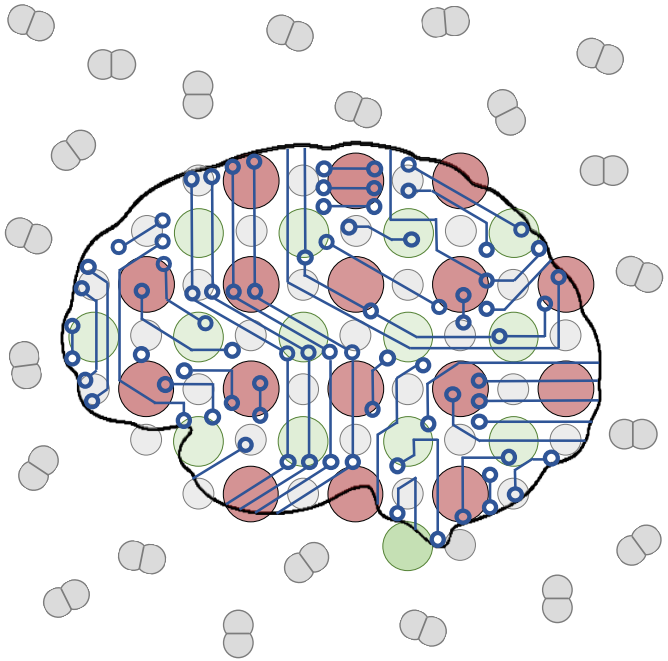
Type	Metal	Hydride	Structure	mass%	p_{eq}, T
Elemental	Pd	$PdH_{0.6}$	*Fm3m*	0.56	0.020 bar, 298 K
AB_5	$LaNi_5$	$LaNi_5H_6$	*P6/mmm*	1.37	2 bar, 298 K
AB_2	ZrV_2	$ZrV_2H_{5.5}$	*Fd3m*	3.01	10^{-8} bar, 323 K
AB	FeTi	$FeTiH_2$	*Pm3m*	1.89	5 bar, 303 K
A_2B	Mg_2Ni	Mg_2NiH_4	*P6222*	3.59	1 bar, 555 K
Body-centred cubic	TiV_2	TiV_2H_4	b.c.c.	2.6	10 bar, 313 K

Schlapbach, L and Züttel, A. Hydrogen-storage materials for mobile applications. *Nature*, **2001**, 414, 6861.

Near atmospheric equilibrium pressure of H_2 occurs at room temperature

Near atmospheric equilibrium pressure of H_2 occurs at > 280 C

Research Question: Can machine learning (ML) yield physics-based insight to facilitate the design of novel metal hydrides exhibiting targeted thermodynamic properties ?

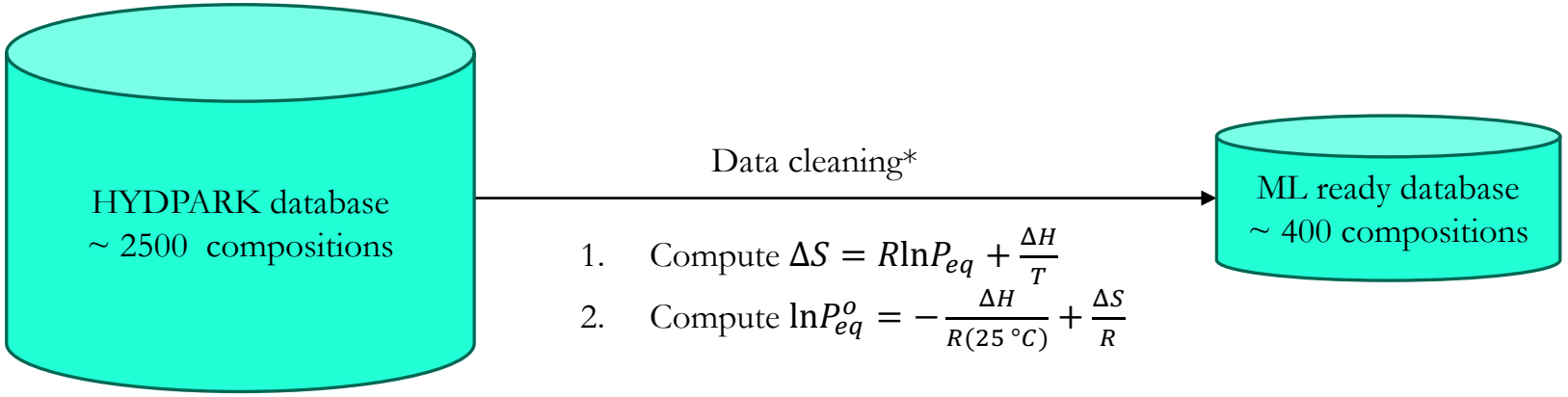


1. Train an ML model to predict the equilibrium plateau pressure, P_{eq}, of a metal hydride from *only the alloy composition*

2. Utilize the ML model's *interpretability* to understand the underlying structure-property relationships from which P_{eq} can be predicted

3. Use these structure-property relationships to *a priori* identify known intermetallic compositions whose hydrides have not been reported *and* are predicted to exhibit a desired P_{eq}

DOE's experimental HYDPARK database contains alloy compositions and their hydriding thermodynamics



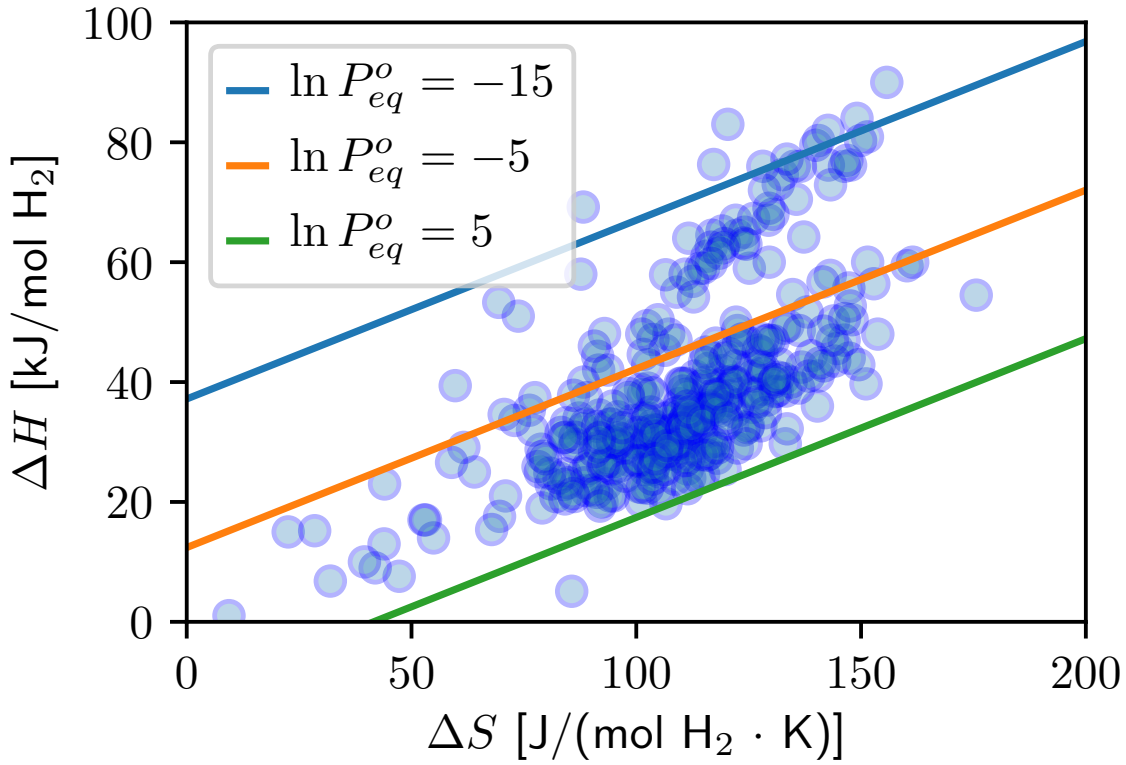
Comp.	ΔH	P_{eq}	T	…
$LaNi_5$				
…				
Er_6Fe_{23}				
…				
…				

Comp.	ΔH	P_{eq}	T	…	$\ln P^{o}_{eq}$
$LaNi_5$					
…					
Er_6Fe_{23}					

* Must remove incomplete and handle duplicate compositions
* Complex hydrides excluded b/c only ~10 entries are complete

Aside: Why train an ML model to predict $\ln P_{eq}^{o}$ and not ΔH?

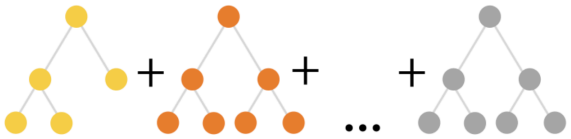
1. $\ln P_{eq}^{o}$ accounts for both the enthalpic and entropic contributions to the free energy of hydriding
2. Indicates the practical applicability of a given hydride for a given application (vehicular storage, high-pressure storage, H_2 getters, etc.)
3. A clear enthalpy-entropy trade-off exists, i.e. it is a "stretch" to argue an optimal ΔH exists for targeting a given $\ln P_{eq}^{o}$



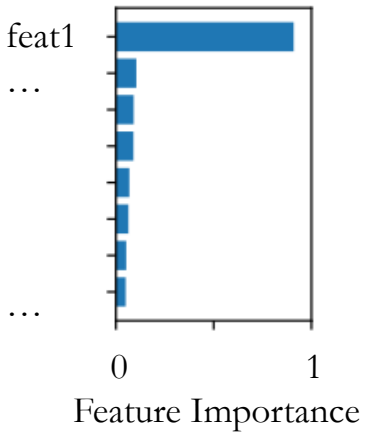
Structurally agnostic featurization is required and gradient boosting trees yield insights from these features

Magpie:
each of the ~ n = 400 compositions is mapped to a 145 dimensional vector computed from elemental properties

Gradient Boosting Trees:
train a model, F, by minimizing the mean squared error of its $\ln P_{eq}^{o}$ predictions



Feature importance:
Several ways to calculate, e.g. average number of times a feature is used to split data across all trees



$$\boldsymbol{X} \in \mathbb{R}^{n\times 145}$$

$$x_{LaNi_5} = \{v_{pa}^{Magpie}, \ldots, 145\}$$

$$\hat{\boldsymbol{y}} = F(\boldsymbol{X}) \in \mathbb{R}^{n\times 1}$$

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

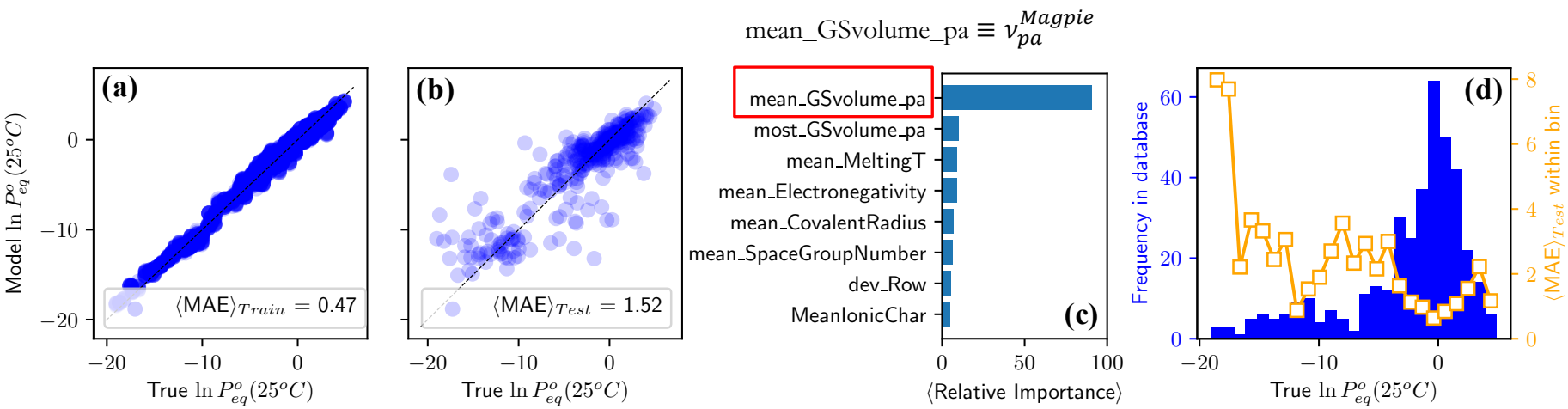
An example Magpie descriptor:

$$v_{pa}^{Magpie} = \sum_i f_i v_i$$

$f_i \equiv$ composition fraction of element i

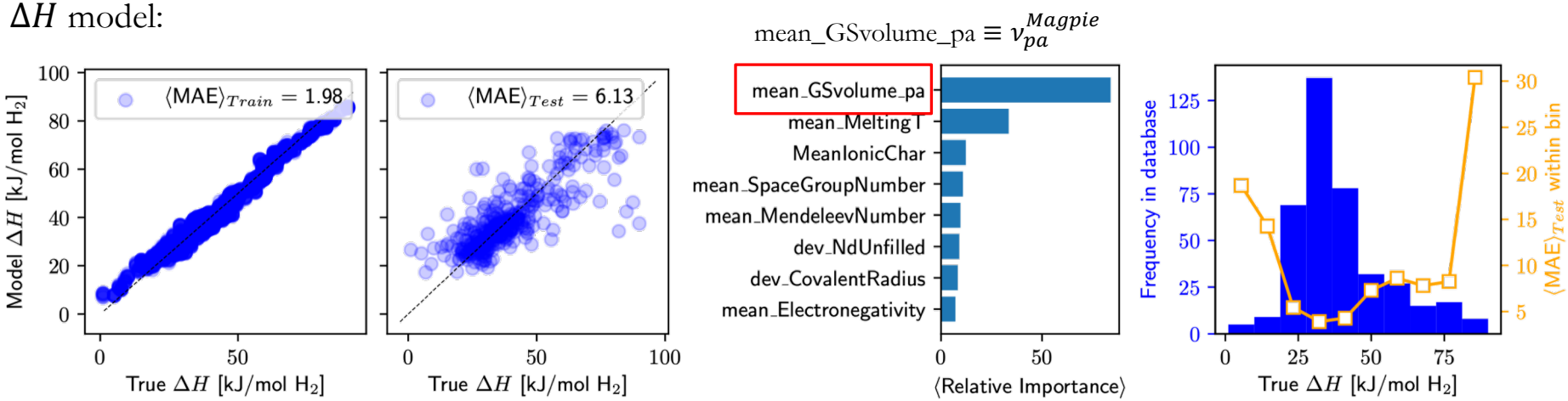
$v_i \equiv$ ground state volume per atom of elemental solid i

ML model can predict $\ln P_{eq}^{o}$ with decent accuracy using input features derived only from the intermetallic composition

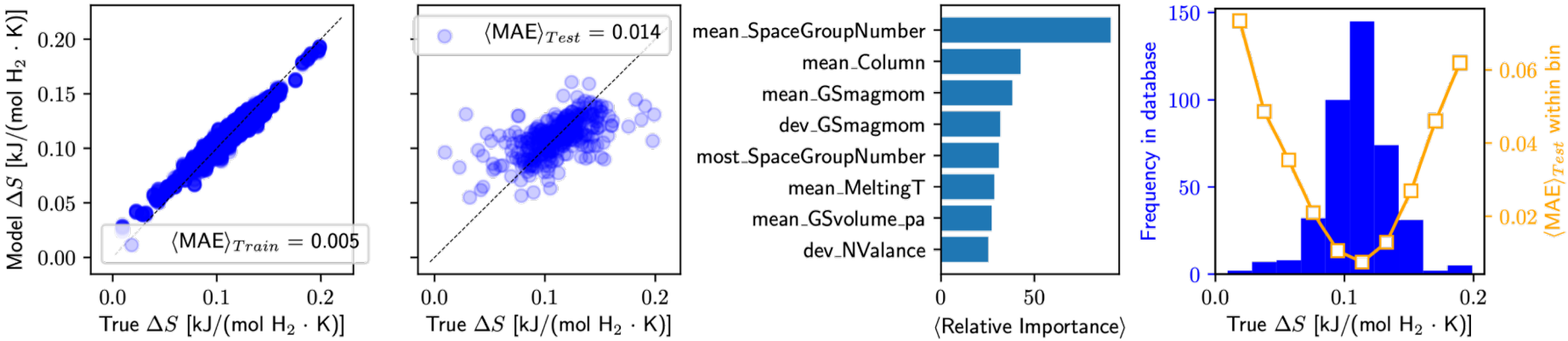


ML model can predict $\ln P_{eq}^{o}$ with decent accuracy using input features derived only from the intermetallic composition

ΔH model:



ΔS model:

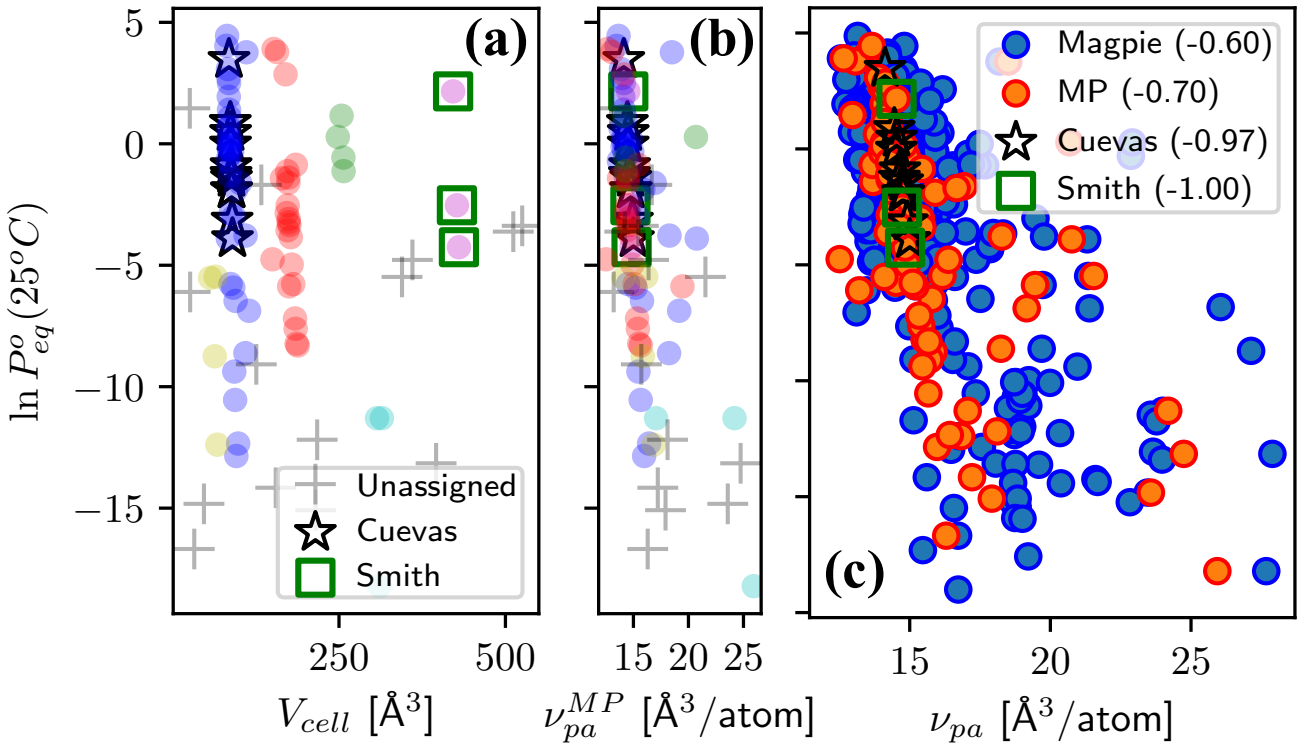


The ν_{pa}: $\ln P^o_{eq}$ structure-property relationship extends over a wide range of metal substitutions and intermetallic classes

1. Compute the structurally specific volume per atom for ~ 70 available structures in the Materials Project (MP) via:

$$V_{cell} \equiv \text{Volume of the intermetallic lattice computed in MP}$$
$$\nu_{pa}^{MP} = V_{cell}/n_{atoms}$$

2. Investigate equilibrium pressure as a function of ν_{pa}^{MP} and ν_{pa}^{MP}:

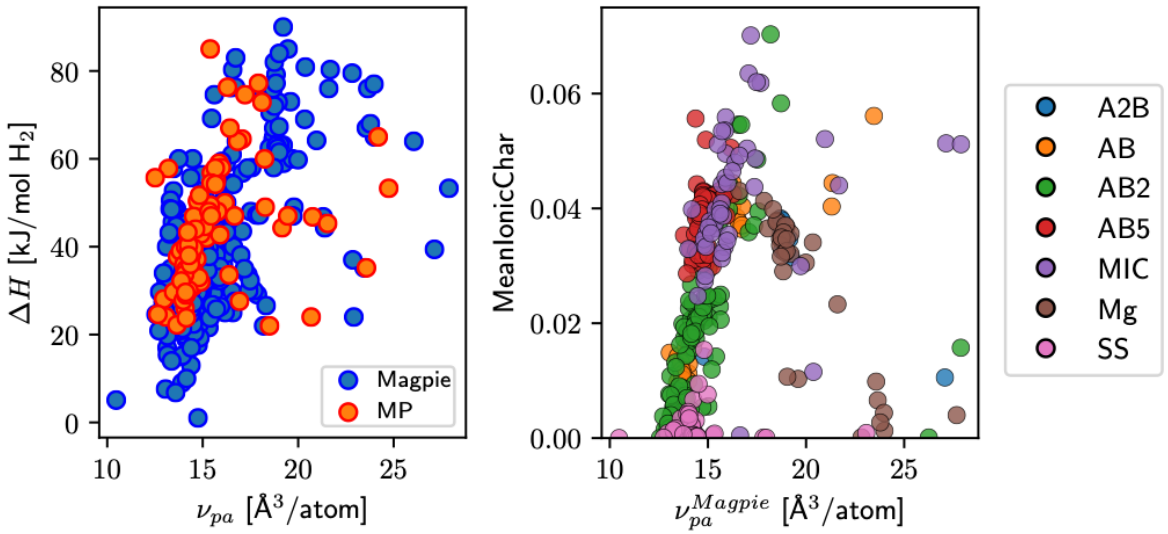


Cuevas et al. noted the dependence of $\ln P^o_{eq}$ on V_{cell} in $LaNi_5$ substitutions

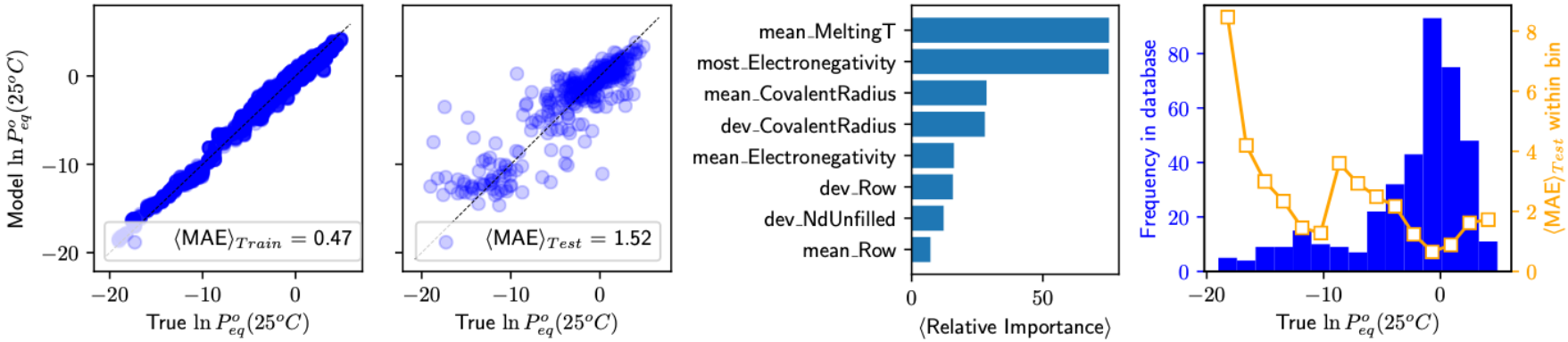
Smith et al. noted the same trend for R_6Fe_{23} [R=Ho,Er,Lu] substitutions

On the importance of ν_{pa}

ν_{pa} encodes information about other features:



An equally accurate ML model can still be trained after removing ν_{pa} from the feature list:



Novel hydride phase of a known intermetallic for high-pressure H_2 storage predicted based on ν_{pa} (and validated with DFT)

DFT computed properties for $AB_5 + 3.5H_2 \rightarrow AB_5H_7$:

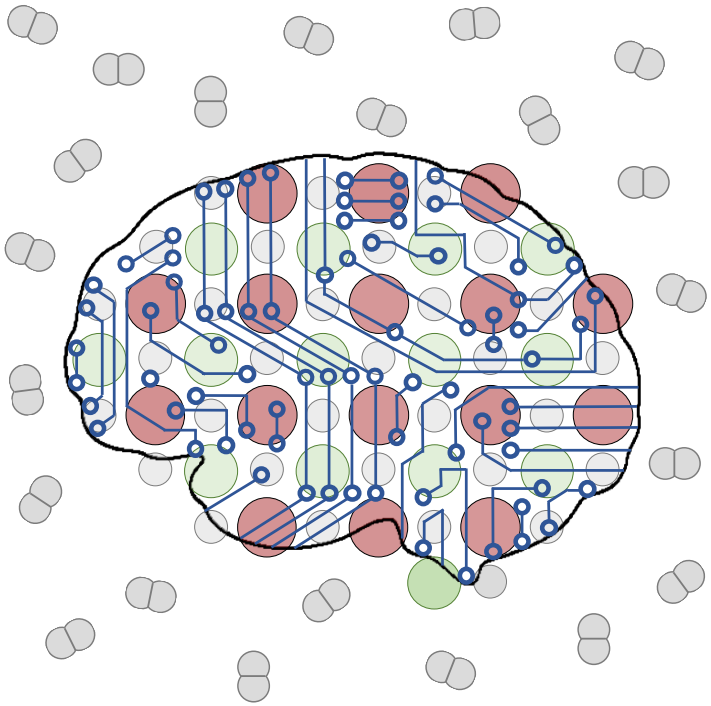
1. ΔH [kJ/molH_2] ≡ hydriding enthalpy
2. ΔE_{def} [kJ/molH_2] ≡ energy penalty to deform lattice to accommodate H absorption
3. ΔE_H [kJ/molH_2] ≡ binding energy of H

Δ ≡ forward hydriding reaction

	ν_{pa}	ΔH	E_f	ΔE_{def}	ΔE_H	V/V_0
UNi_5	13.17	-0.60	-285	65.2	-65.8	1.278
$CeNi_5$	13.76	-20.5	-353	49.3	-69.8	1.266
$LaNi_5$	14.38	-36.1	-224	44.3	-80.5	1.256

*Several U containing compounds in HydPARK, but no UNi_5 (even though it exists in ICSD)

Key Takeaways



1. ML models with experimental data provide a powerful tool to explore phenomena too expensive to simulate directly with computational approaches

2. Equilibrium pressure in intermetallic hydrides can be predicted just from the alloy composition, despite noisy/incomplete data

3. Explainable insights from the ML model permit the rational design of novel materials with targeted thermodynamic properties

Thank you for your attention.

Questions?

Backup: DFT computed properties

Notation: $E(X)_{\{Y\}}$ denotes the energy of chemical system X in the geometry of system Y

Enthalpy of hydriding:

$$\Delta H = \left[E(AB_5H_7)_{\{AB_5H_7\}} - E(AB_5)_{\{AB_5\}} - 3.5 \times E(H_2)_{\{H_2\}}\right] / 3.5 \; [=] \; [\mathrm{kJ/molH_2}]$$

Lattice deformation energy of hydriding:

$$\Delta E_{def} = \left[E(AB_5)_{\{AB_5H_7\}} - E(AB_5)_{\{AB_5\}}\right] / 3.5$$

Binding energy of hydriding:

$$\Delta E_H = \left[E(AB_5H_7)_{\{AB_5H_7\}} - E(AB_5)_{\{AB_5H_7\}} - 3.5 \times E(H_2)_{\{H_2\}}\right] / 3.5$$