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

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Introduction

An open question in the metal hydride community is whether there are simple, physics-based design rules that dictate the thermodynamic properties of these materials across the variety of structures and chemistry they can exhibit. While black box machine learning (ML)-based algorithms can predict these properties with some success, they do not directly provide the basis on which these predictions are made, therefore complicating the a priori design of novel materials exhibiting a desired property value. In this work we demonstrate how feature importance, as identified by a gradient boosting tree regressor, uncovers the strong dependence of the metal hydride equilibrium H_2 pressure on a volume-based descriptor that can be computed from just the elemental composition of the intermetallic alloy. Elucidation of this simple structure–property relationship is valid across a range of compositions, metal substitutions, and structural classes exhibited by intermetallic hydrides. This permits rational targeting of novel intermetallics for high-pressure hydrogen storage (low-stability hydrides) by their descriptor values, and we predict a known intermetallic to form a low-stability hydride (as confirmed by density functional theory calculations) that has not yet been experimentally investigated [1].

Data cleaning

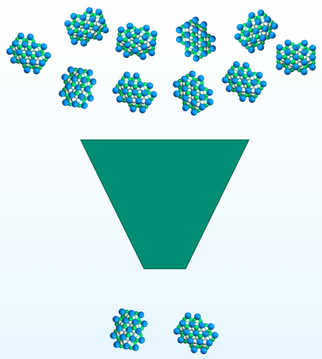
HydPARK is Sandia's historical dataset on metal hydride properties:

Class	Composition	H wt %	ΔH (kJ/molH_2)	T (K)	P_{eq} (atm)
A_2B	$Ce_{0.5}Th_{1.5}Al_1$	0.4	133	650	0.0003
AB_5	$La_{0.75}Ce_{0.25}Ti_{0.1}Cu_{0.9}Ni_4$	1.4	38.4	22	1.1

Target property: Compute equilibrium pressure at room temperature,

$$\ln P^o_{eq} = -\Delta H/RT^o + \Delta S/R$$

to gauge a material's thermodynamic utility in a given storage application



Initial: ~ 2500 HydPARK compositions

1. Remove duplicate compositions
2. Remove entries with missing data, i.e. $\ln P^o_{eq}$ can't be computed

Final: ~ 400 "ML ready" compositions

ML Model Development

1. Magpie presents a useful way to featurize inorganic materials in terms of elemental properties when exact crystal structures are not known [2]

$$\mathbf{x}_{\mathrm{LaNi5}} = [\,\bar{v}^{Magpie}_{pa}, \bar{T}_{melt}, \dots\,] \in \mathbb{R}^{145}$$

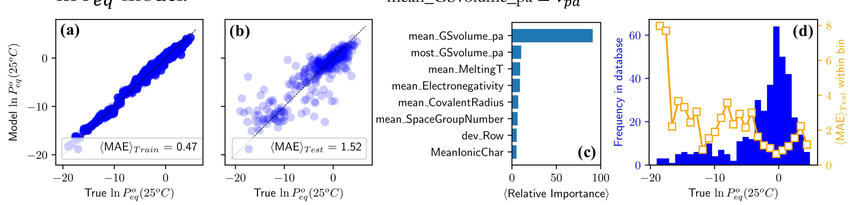
$$\bar{v}^{Magpie}_{pa} = \sum f_i v_i \quad , \quad f_i \equiv \text{composition fraction of element } i$$
$$v_i \equiv \text{ground state volume per atom of solid element } i$$

2. Materials Project (MP) provides electronic structure data if crystal structure known (only ~70 HydPARK compositions found in MP)

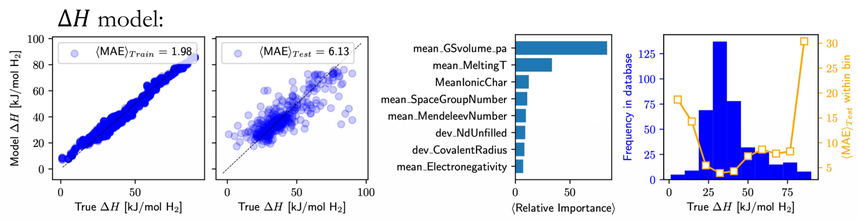
$$v^{MP}_{pa} = V_{cell}/n_{atoms} \qquad V_{cell} \equiv \text{DFT optimized cell volume}$$

3. Gradient Boosting Regressors are "interpretable" : they reveal the relative importance of each feature in the model's prediction [3]

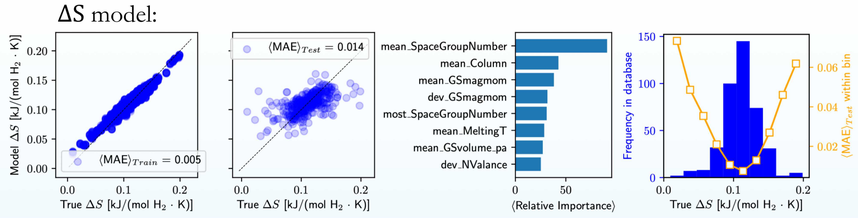
$\ln P^o_{eq}$ model:



ΔH model:

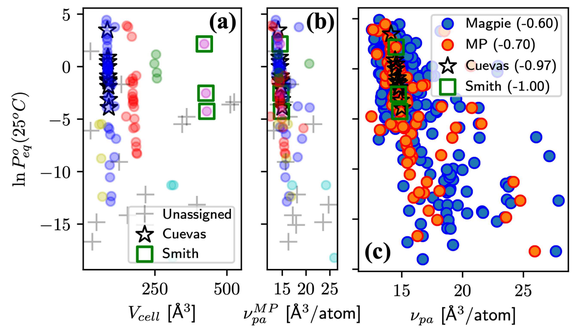


ΔS model:



5-fold Train Performance | 5-fold Test Performance | 5-fold Averaged Feature Importance | Distributed Error

Exploiting Explainable Insights



1. Reveals log-linear relationship with v_{pa}
2. Unifies previous experimental results with a single structure-property relationship
3. Provides an empirical design rule to target low, moderate, or high stability hydrides

Novel Hydride with Targeted Thermodynamics

Utilize DFT to (1) gain insight into the v_{pa} structure-property relationship and (2) validate the hypothesis that a known intermetallic (UNi_5 whose hydriding properties have not yet been tested) would form an ultra low-stability (high-pressure) hydride:

For $ANi_5 \rightarrow ANi_5H_7$ (A=U, Ce, La)

$E_f \equiv$ formation energy of ANi_5H_7

$\Delta E_{def} \equiv$ deformation energy to accommodate H absorption

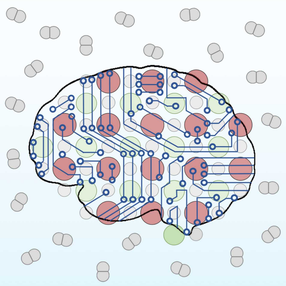
$\Delta E_H \equiv$ binding energy of H

$V/V_0 \equiv$ volume expansion upon H absorption

	v_{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

Conclusions

1. ML models and experimental data provide a powerful tool to explore phenomena too expensive for 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



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