SAND2021-4579 PE

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

2021 HFTO Postdoctoral Recognitional Award

PRESENTED BY

Matthew Witman

Sandia National Laboratories, Livermore, CA USA

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Collaborators & Acknowledgements

Experimental chemistry/materials science

Mark Allendorf
Vitalie Stavila
Anthony McDaniel
Andrea Ambrosini
Jeffery Chames
David Grant (Nottingham University)
Gavin Walker (Nottingham University)
Gustav Ek (Uppsala University)
Martin Sahlberg (Uppsala University)
Claudia Zlotea (Paris Est)

ML Theory

Sapan Agarwal
Justin Wong

Computational Chemistry

Sanliang Ling (Nottingham University)
Nathan Mahynski (NIST)
Harold Hatch (NIST)
Nick Wunder (NREL)
Nalinrat Guba (NREL)

The authors gratefully acknowledge research support from the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Hydrogen and Fuel Cell Technologies Office through the Hydrogen Storage Materials Advanced Research Consortium (HyMARC). This work was supported by the Laboratory Directed Research and Development (LDRD) program at Sandia National Laboratories.

LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT

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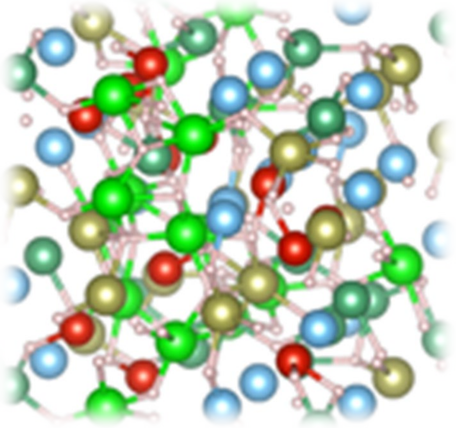
Motivation: Are there undiscovered materials that could improve upon conventional H_2 storage and generation technologies?

H_2 Storage objectives:

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

“Conventional”
(Compressed gas)

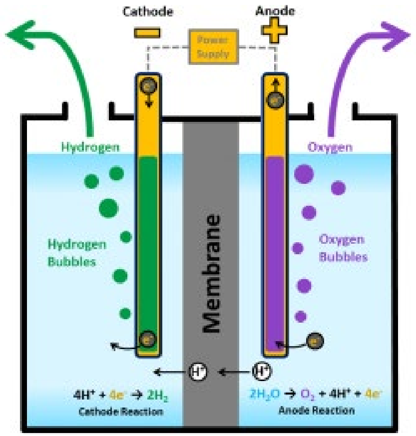
vs.



Material X ??

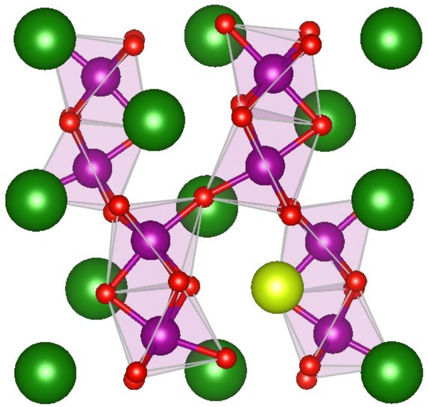
H_2 Generation objectives:

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



“Conventional”
1.2 V in theory
1.8 V in practice

vs.



Material Y ??

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Approach: How are new materials discovered?

Experiments

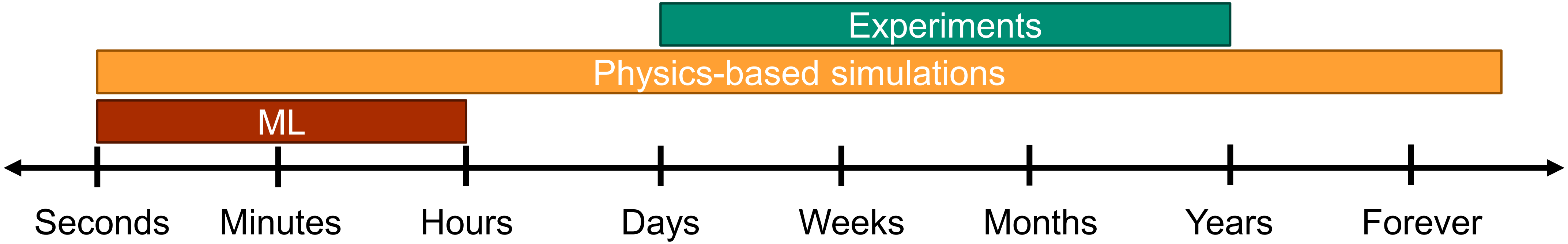
Chemical intuition
↓
Synthesize material
↓
Characterize material
↓
Test Performance

Physics-based simulations

First-principles theory
↓
Run *simulation* &
predict performance

Machine learning (ML)

Material property/training data
↓
Convert materials to features
↓
Train a model:
↓
Run *model* &
predict performance



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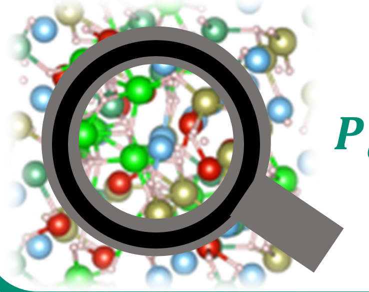
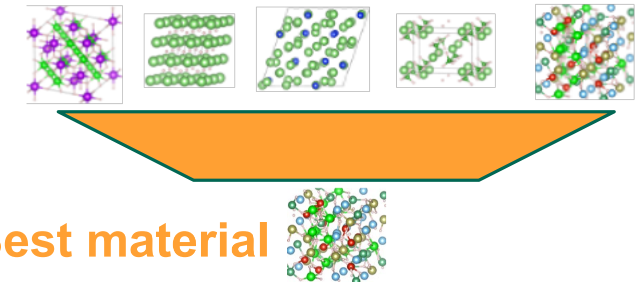
Approach: Data science and machine learning techniques can…

Predict properties and elucidate design rules for optimal materials[1,2]



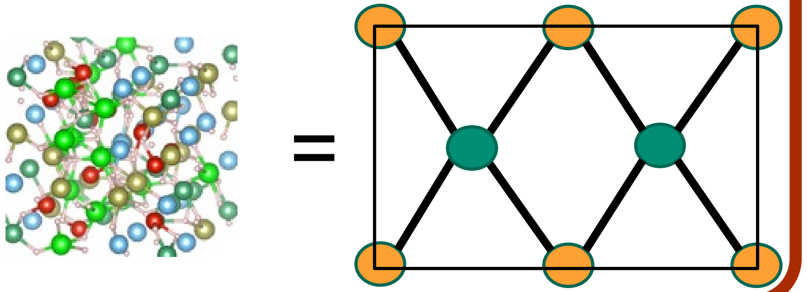
$$P_{eq} = mx + b$$

High-throughput screen materials orders of magnitude faster than experiments or simulations[3,4,5,6]



Best material

Accelerate physics-based simulations when lacking experimental training data[7]



Tools provide a roadmap for…

H2 storage: Data-driven discovery of optimal hydrogen storage alloys

H2 generation: Data-driven discovery of liquid metals for water splitting

[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**

[5] *In preparation*

[6] Ambrosini, Witman, McDaniel. *Provisional Patent*, **2020**.

[7] *In preparation*

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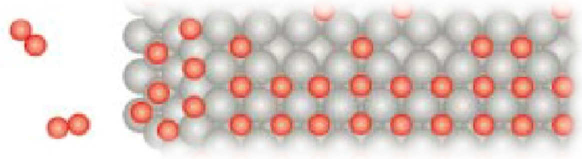
H_2 Storage Milestone #1: Explainable ML models predict metal hydride thermodynamics

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

H$_2$ Metal Hydride



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

From HydPARK database

(2) Compositional ML model

Features from composition:

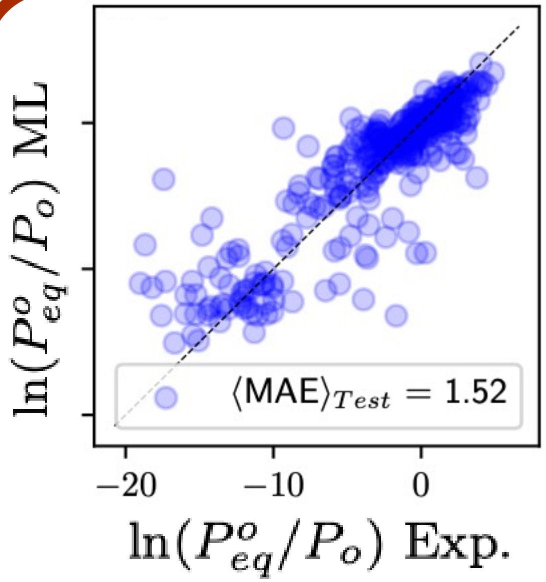
$TiFe_{0.96}Nb_{0.04} \rightarrow$

$\boldsymbol{x} = \{\bar{\nu}_{pa}, \bar{r}_{cov}, \bar{\chi}, \ldots\}$

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

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

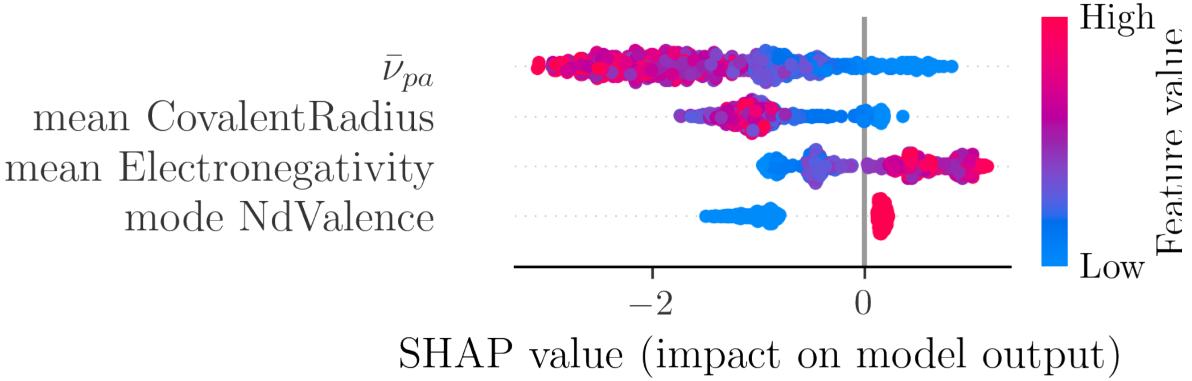
(3) Model validation and explainability



*ML model (gradient boosting trees) **can predict** $\ln(P^o_{eq}/P_o)$ with MAE = 1.5

*<u>**Linear correlation**</u> with $\bar{\nu}_{pa}$:

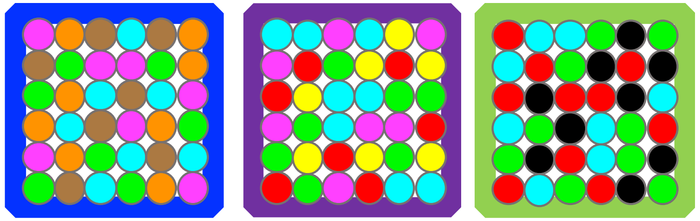
$$\ln\left(\frac{P^o_{eq}}{P_o}\right) \approx -m\,\bar{\nu}_{pa} + b$$



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

H_2 storage 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

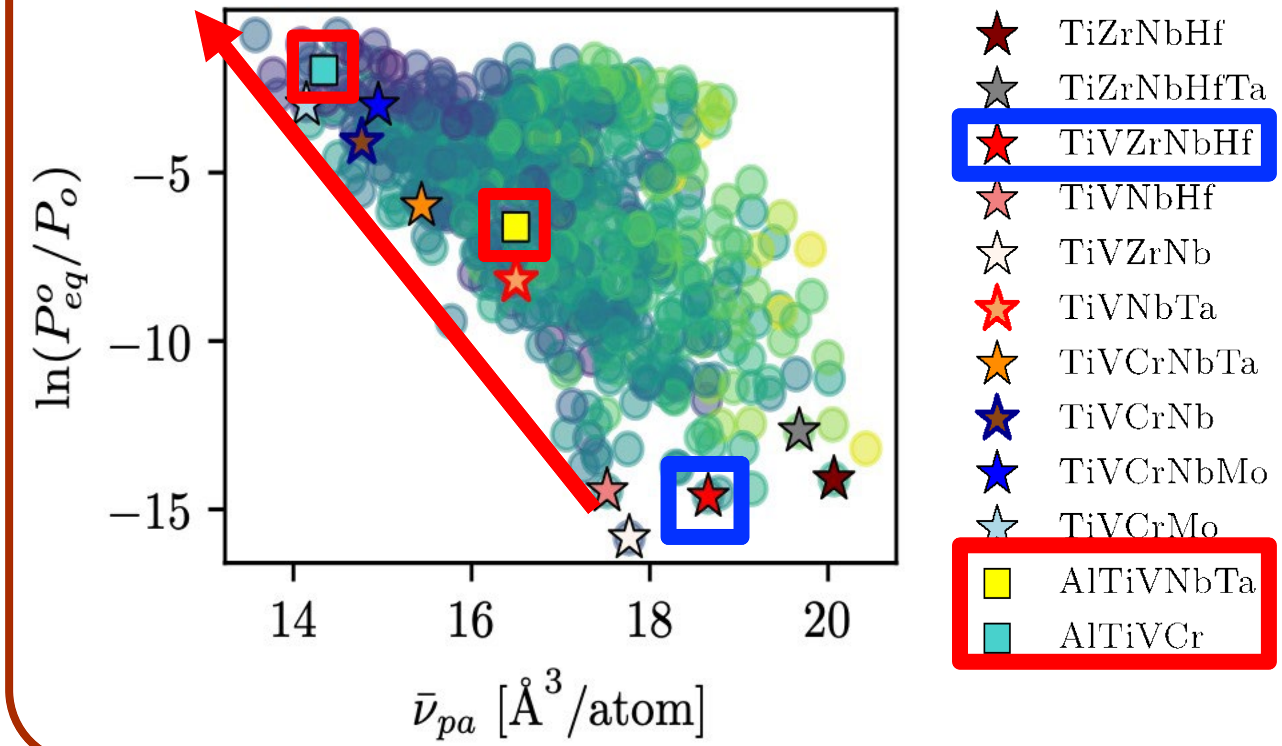
E = {Al, Ti, V, Cr, Zr, Nb, Mo, Pd, Hf, Ta}

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

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.*, **2021**

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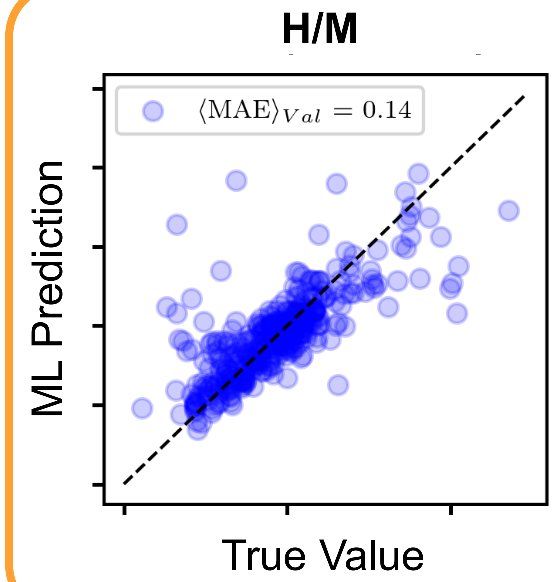
H_2 Storage Milestone #3:
Identify Pareto optimal HEA hydrides

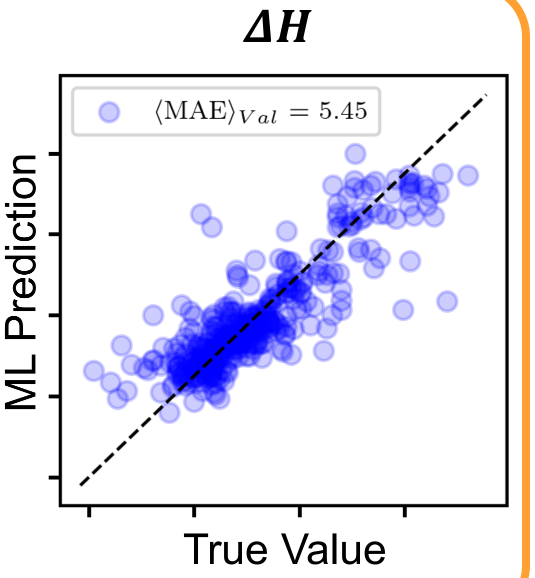
(1) Screening an expansive HEA space

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

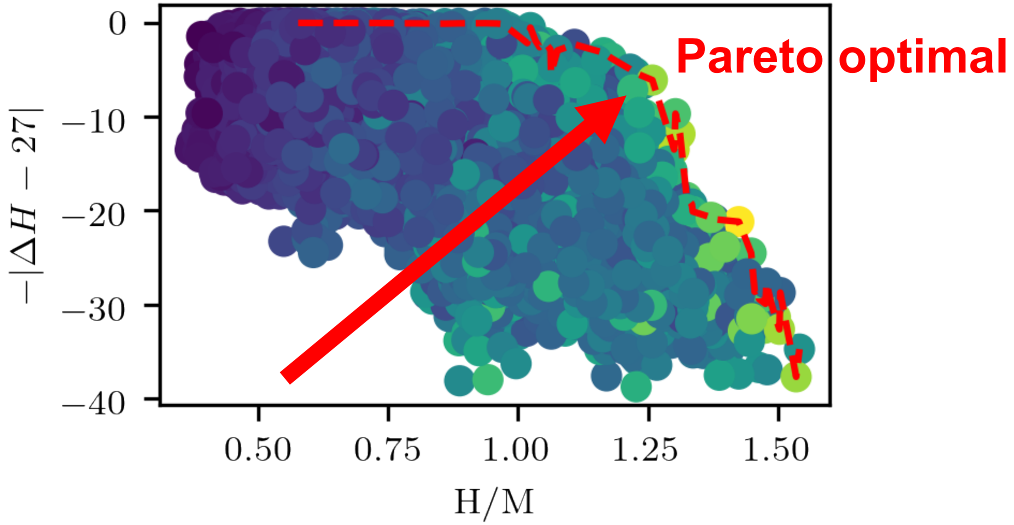




(3) Identification of ~100 Pareto optimal materials

Objectives / Quantity to maximize:

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



ML: ***Seconds*** to identify 100 Pareto materials

Experiment: ***Months*** to synthesize, characterize, and test 1 material

In preparation

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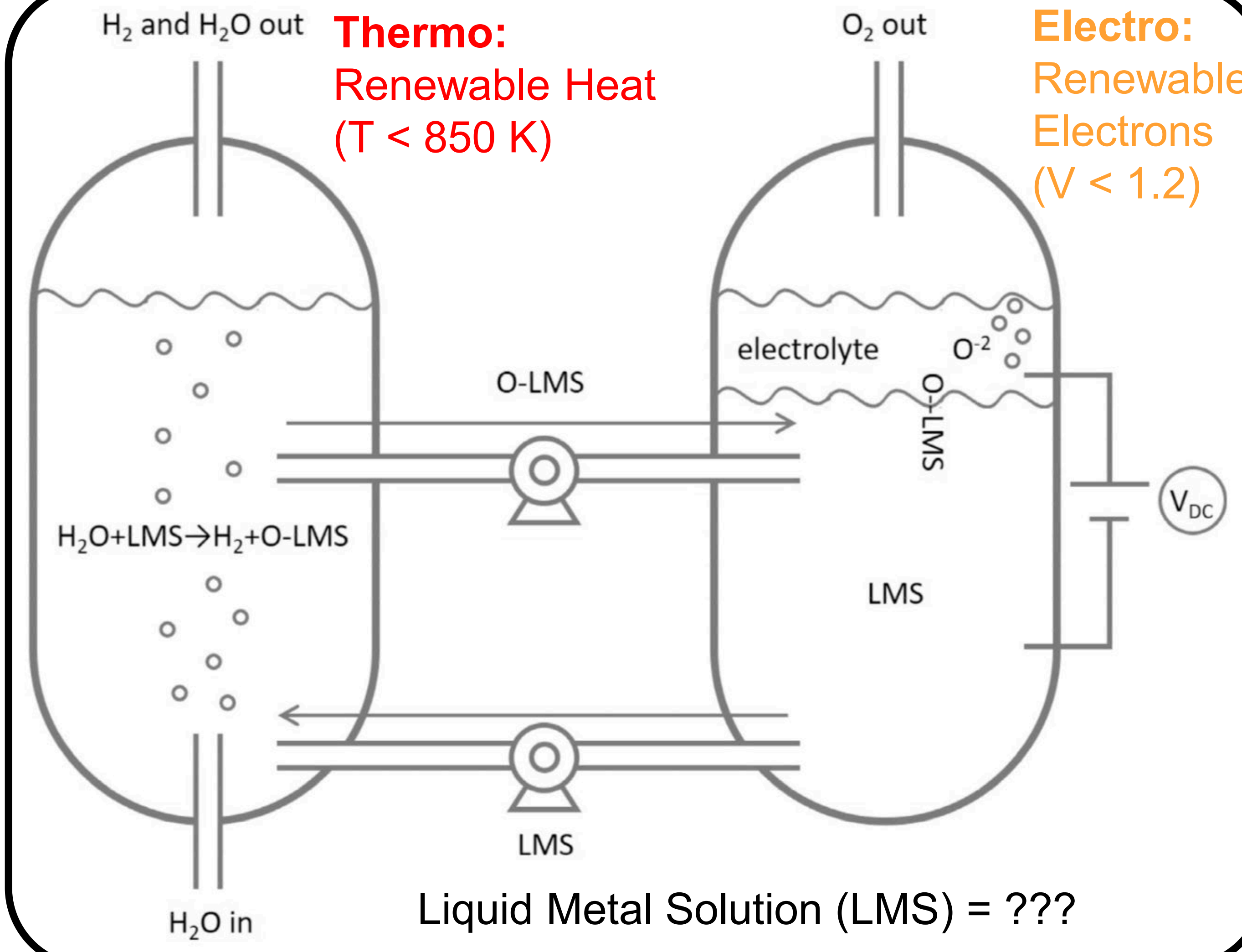
H_2 Generation Milestone #1:
Data-driven down selection of liquid metals for water splitting

Hybrid thermo/electrochemical concept

Thermo:
Renewable Heat
(T < 850 K)

Electro:
Renewable
Electrons
(V < 1.2)



Liquid Metal Solution (LMS) = ???

Down selection of candidate LMS

2415 possible binaries

ML modeling of liquidous curves

~100 w/melting below 850 K

Mining Pourbaix diagrams from Materials Project

~15 w/desired O-LMS electrochemical stability

Cost/practicality

~3 in laboratory testing

Ambrosini, Witman, McDaniel. *Provisional Patent*, **2020**.

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Planned H_2 research directions (0-5 year aspirations) include:

H_2 Storage Applications

- Identify Pareto optimal hydrides across ***all possible HEA composition space***
- Can they play a practical role in the stationary (or even transportation) space?

H_2 Generation Applications

- Improve hybrid concept by going beyond binary LMS (ternary, quaternary, …)
- Graph neural networks to model defects in STCH materials across all composition space

Methods/Data

- Develop ***ML surrogate models*** to accelerate previously intractable DFT simulations
- Improve quality of experimental training data via ***standardized data management tools***

More data + Improved ML models

- More accurate and faster materials predictions
  - More efficient use of experimental resources
    - ***Faster hydrogen technology adoption***

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Thank you for your attention.

Questions?

Contact: mwitman@sandia.gov