

Machine learning-guided materials and system co-design for high-pressure hydrogen compression

Matthew D. Witman^{1,*}, Brendan Davis¹, Vitalie Stavila^{1,^}, and Terry Johnson^{1,†}

¹Sandia National Laboratories, Livermore, CA 94551, USA

^{*}*mwitman@sandia.gov*; [^]*vnstavi@sandia.gov*; [†]*tajohns@sandia.gov*

Abstract

Cost effective and reliable hydrogen compression remains a challenging barrier in the wide-spread adoption of hydrogen as an energy carrier. The prevailing technology of mechanical compression suffers from several drawbacks, some of which can be addressed by non-mechanical compression strategies (e.g., electrochemical or metal hydride-based thermal compression). Thermally driven metal hydride compression strategies typically rely on multi-stage metal hydride-based compressors; however, discovering or optimizing low-stability metal hydrides that can pressurize hydrogen upwards of 1000 bar is difficult, both with respect to computational predictions and experimental validation. Here we (1) demonstrate that simple machine learning-derived design rules can inform rational design of alloying strategies yielding low-stability hydrides, (2) validate their experimental pressure-composition-temperature (PCT) isotherms up to 875 bar, and (3) utilize a dynamic systems-level model of a metal hydride compressor design to evaluate their performance under realistic operating conditions. Importantly, this analysis yields predicted operational efficiencies of both 2-stage (90-875 bar) and 3-stage (20-875 bar) metal hydride compressors to enable further evaluation of this technology and its techno-economic outlook.

Introduction

Hydrogen is an energy carrier that presents distinctive opportunities to advance a variety of technologies, due to its notable gravimetric energy density of 120 MJ kg^{-1} and versatile end uses in combustion, in fuel cells, as a chemical reductant, etc.¹ Currently, most hydrogen is produced from Steam Methane Reforming (SMR), which involves reacting natural gas with steam.² Additionally, other fossil fuel-based methods, such as coal gasification, can also be employed to produce hydrogen, as the demand for hydrogen increases.³ Hydrogen also displays considerable potential for production through renewable energy sources⁴ or via the direct mining of natural hydrogen.⁵ Regardless of the production method employed, hydrogen is usually generated as a gas at relatively low pressures ranging from 1 to 50 bar, necessitating additional compression for effective storage and transportation.⁶ While mechanical compressors, including piston and diaphragm types, are commonly employed, they exhibit limitations such as high operating costs and limited reliability. In addition, the complex designs of most mechanical compressors suffer from hydrogen leakage, produce significant noise during operation, and lead to increased maintenance requirements.⁷

New technologies focused on metal hydrides for hydrogen compression are being developed, utilizing the re-

versible reaction between metal alloys and hydrogen at various pressures. These compressors offer several advantages, including high discharge pressures of up to 1000 bar H_2 , excellent scalability, and a modular design that allows the use of low-grade thermal energy to enhance energy efficiency.⁷ The absence of moving parts simplifies their design, increases reliability, and reduces noise, making them an attractive option for hydrogen compression. Metal alloys based on AB, AB₂, or AB₅ phases are typically considered as active materials for the development of single-stage and multi-stage hydride compressors.^{8–10} However, as noted by Lototsky et al.⁸ and Yartys et al.,⁹ only a limited number of interstitial-type metal hydrides possess the necessary reversibility and reaction kinetics to be effective in these applications. In addition, the pressure-composition-temperature (PCT) curves of many alloys proposed for H_2 compression exhibit sloped plateaus and significant absorption-desorption hysteresis, which limits the usable capacity and compression ratio.¹¹

Furthermore, *a priori* computational prediction and optimization of metal hydrides for high-pressure hydrogen compression is challenging for a variety of reasons. Compositional machine learning (ML) models for metal hydride thermodynamic properties provide an excellent preliminary screening tool,^{12–15} but highly accurate predictions for ultra-low stability hydrides remain a challenge. This primarily arises due to the imbalance of existing experimental data on which these models are trained, i.e.,

the extreme lack of experimental data for materials whose hydride plateaus require experimental PCT measurements greater than 100-200 bar. First-principles-based calculations or simulations enabling metal hydride phase equilibrium predictions^{16,17} (i.e., PCT curves), which can themselves be accelerated by various machine-learning approaches,^{18,19} have been proposed as well and can in principle overcome out-of-distribution accuracy issues, but these typically cannot account for sloping plateaus and hysteresis which also substantially impact a metal hydride's compression performance; therefore, experimental validation is required anyway, requiring highly specialized experimental equipment for accurate PCT measurements up to 1000 bar.

In this work we leverage simple metal hydride thermodynamic design rules, derived from explainability analysis of compositional ML models, to propose substitution strategies to fine-tune an existing and already promising materials' hydriding thermodynamics for high-pressure compression applications. Upon experimentally measuring PCT curves of the fine-tuned material up to 900 bar at several temperatures, we confirm the ML predicted design rule for desorption enthalpy decrease and demonstrate the material's ability to compress 1 wt% hydrogen from ~400 bar to ~875 bar under a temperature swing of 20 to 150 °C. Using a comprehensive system dynamic model for a prototype metal hydride compressor²⁰ and the experimentally measured PCTs for this new material up to 875 bar (rather than modeled PCTs extrapolated to higher temperatures and pressures), we quantitatively predict efficiency of various multi-stage, high-pressure metal hydride compressor designs. This paves the way to further improve projected efficiencies of materials-based hydrogen compression for various use cases as we continue to further optimize this material.

Results & Discussion

Optimal materials for hydrogen compression.

The principle of hydrogen compression using a metal hydride is based on the thermodynamic relationship described by the van't Hoff equation, which correlates the equilibrium pressure of hydrogen with temperature, enthalpy, and entropy changes associated with hydride formation.^{8,9} Figure 1 shows a schematic representation of hydrogen compression using a metal hydride, illustrating hydrogen absorption at a low temperature (T_L) and low pressure (P_L), followed by desorption at a higher temperature (T_H), generating an elevated pressure (P_H). At reduced temperature T_L , the metal hydride absorbs hydrogen gas at low pressure P_L to form a stable hydride phase, a process that is thermodynamically exothermic. Upon heating to temperature T_H , the chemical equilibrium shifts, favoring the endothermic desorption of hydrogen, thereby elevating the hydrogen delivery pressure P_H . The compression ratio is defined as the ratio of the high

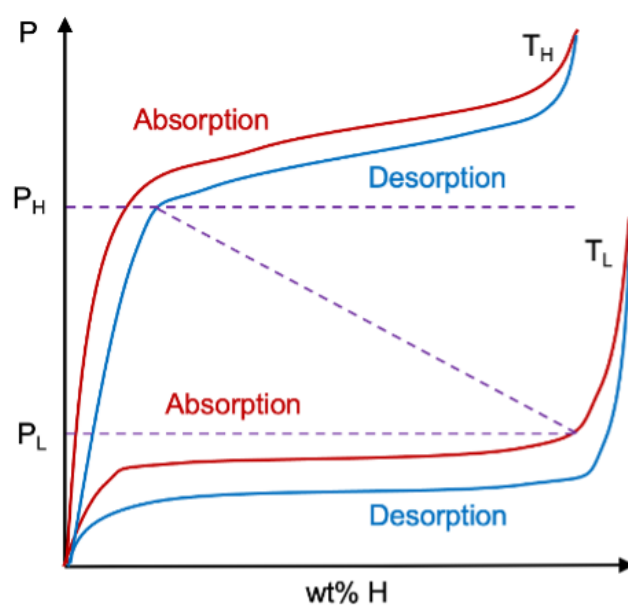


Figure 1: Schematic representation of hydrogen compression using a metal hydride. Hydrogen is absorbed at temperature T_L and H_2 pressure P_L , and desorbed upon heating at temperature T_H , generating pressure P_H .

pressure at desorption (P_H) to the low pressure at absorption (P_L).²¹ This behavior underlies the functionality of metal hydride-based hydrogen compressors, where cyclic cooling and heating enable hydrogen absorption at low pressures and subsequent desorption at high pressures. Effective thermal management, achieved through the incorporation of high-conductivity materials such as copper, aluminum, or various carbons, is imperative to facilitate rapid heat transfer and uniform temperature distribution within the hydride bed.^{10,11}

Explainable ML design rules for hydride destabilization.

Compositional ML models have been widely used to predict metal hydride thermodynamic properties and subsequently screen for new or modified (high entropy) alloys with targeted thermodynamic destabilization. See Supplementary Information (SI) for more details on the ML methodology. Briefly, descriptors derived purely from a given alloy composition are generated using Magpie.²² These features correspond to, for example, the alloy composition-weighted averages over an elemental property, $\bar{p} = \sum_i x_i p_i$ ($x_i \equiv$ atomic fraction of element i in the alloy, $p_i \equiv$ the elemental property) and serve as input to a gradient boosting regressor²³ machine learning model trained to predict thermodynamic properties of Equation (1), i.e., $\ln(P_{eq}^o/P_o)$, ΔH_d , and/or ΔS_d ,

$$\ln(P_{eq}^o/P_o) = -\Delta H_d/RT + \Delta S_d/R. \quad (1)$$

Here $\ln(P_{eq}^o/P_o)$ is the equilibrium plateau pressure at 25 °C with reference $P_o = 1$ bar, and ΔH_d and ΔS_d are the desorption enthalpy and entropy, respectively. How-

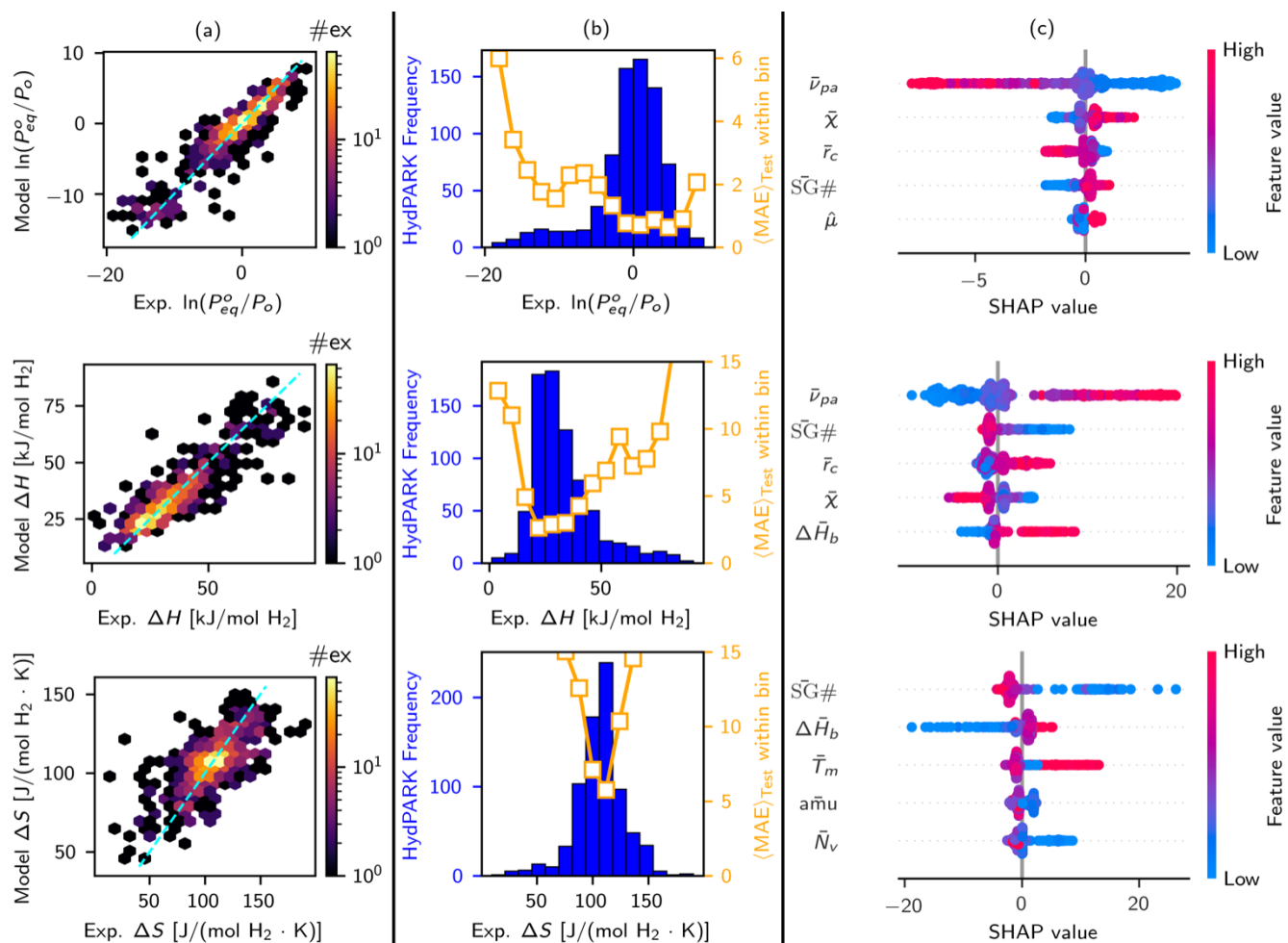


Figure 2: For target properties of $\ln(P_{eq}^o/P_o)$, ΔH , and ΔS in rows 1-3, respectively: (a) Parity plot of $K = 10$ -fold test set predictions vs. experiment from a gradient boosting tree (GBT) regressor trained on v0.0.6 of the ML-ready HydPARK dataset. (b) Histogram of the underlying target property (blue) and test prediction MAE in each individual bin. (c) SHAP values for the 5 most important features for the trained GBT model. These features correspond to the following properties $p = \{\nu_{pa} \equiv \text{volume per atom of the elemental solid}, \chi \equiv \text{Pauling electronegativity}, r_c \equiv \text{covalent radius}, SG\# \equiv \text{space group number of the elemental solid}, \Delta H_b \equiv \text{formation enthalpy of the binary elemental hydride}, T_m \equiv \text{melting temperature of the elemental solid}, N_V \equiv \text{number of valence electrons}\}$.

e' :	Cr	Mn	Fe	Ni
$\{\downarrow \nu_{pa}\}$	[Co, Fe, Cu, Ni, Mn]	[Co, Ni]	[Co, Mn, Ni]	[Co]
$\{\downarrow \nu_{pa}, \uparrow \chi\}$	[Co, Fe, Cu, Ni]	[Co, Ni]	[Co, Ni]	
$\{\downarrow \nu_{pa}, \uparrow \chi, \downarrow r_c, \}$	[Co, Fe, Cu, Ni]	[Co, Ni]	[Co, Ni]	
$\{\downarrow \nu_{pa}, \uparrow \chi, \downarrow r_c, \uparrow SG\#\}$		[Ni]		

Table 1: Possible substitution elements for select target elements, e' , that are likely to destabilize the hydride phase (e.g., increase $\ln(P_{eq}^o/P_o)$) for an increasing number of SHAP design rules from Figure 2c.

ever, a substantial challenge is the large imbalance of existing experimental data upon which such models are trained (e.g., the ML-ready HydPARK v0.0.6). Few van't Hoff-derived experimental data from Equation (1) exist for exceptionally low or high stability hydrides due to the difficulty of these PCT measurements, since a typical experimental apparatus is limited to pressure in the range of $10^{-3} \leftrightarrow 100$ bar and temperature in the range of $-75 \leftrightarrow 400$ °C. As shown in Figure 1b, lack of experimental data in the extrema for all target properties in turn leads to modeling predictions with increased mean absolute error (MAE) and, specifically, a systematic overestimation of stability in low stability hydrides, i.e., those with high $\ln(P_{eq}^o/P_o)$ arising from low ΔH and/or high ΔS . For materials with $\ln(P_{eq}^o/P_o) \gtrsim 6$, the test set error rapidly increases and is $\sim 3\times$ larger than for materials exhibiting $\ln(P_{eq}^o/P_o)$ near ambient.

To target low-stability hydrides for potential use as a high-pressure compression material, we adopt a simple heuristic strategy rather than relying on quantitative ML prediction accuracy in these data-limited regimes. We first identify a metal hydride from the literature that already shows promise for high-pressure H₂ compression based on an appropriately low hydride stability, low hysteresis, and relatively flat PCT plateau, but whose thermodynamic stability is still not optimal or does not have a validated PCT measurement up to 1000 bar. Then, we select a substitution strategy that is consistent with the SHAP analysis (Figure 1c) design rules²⁴ for tuning $\ln(P_{eq}^o/P_o)$ (or ΔH), i.e., the compositional features most likely to increase plateau pressure (by decreasing ΔH) are reducing $\bar{\nu}_{pa}$, increasing $\bar{\chi}$ decreasing \bar{r}_c , and increasing SG#. For the set of transition metal elements E , Table 1 shows the subset of elements e that can be substituted for some target e' , while satisfying an increasing number of SHAP design rules for increasing $\ln(P_{eq}^o/P_o)$. Based on these simple heuristics, Co and Ni-substitution for Mn, Cr, or Fe tend to simultaneously satisfy the most design criteria, while substitution by Co instead of Ni will yield the largest decrease in ν_{pa} .

Materials selection, synthesis, characterization, and high-pressure PCT measurements. AB₂ alloys, particularly in the composition space of Ti(Cr,Mn,Fe)₂, have been extensively studied as potential high-pressure compression materials since specific regions of this ternary B space can yield very large plateau pressures (e.g., $\ln(P_{eq}^o/P_o) \gtrsim 6$).^{21,25,26} For example, Ti_{1.02}Cr_{1.1}Mn_{0.3}Fe_{0.6} demonstrates a $\Delta H_d = 16.5$ kJ mol⁻¹ and $\Delta S_d = 103.8$ kJ mol⁻¹, maintains relatively flat plateau with small hysteresis, and exhibits faster hydrogen absorption kinetics upon small amounts of rare earth addition with minimal effect on hydride thermodynamics.²⁷ For example, a small modification to Ti_{1.02}Cr_{1.1}Mn_{0.3}Fe_{0.6}La_{0.03} (hereon referred to as "base TCMFL") induces a negligible change in the thermodynamics and exhibits $\Delta H_d = 16.6$ kJ mol⁻¹

and $\Delta S_d = 104.2$ kJ mol⁻¹, while substitution with Ce or Ho instead of La yields slightly larger ΔH_d . We therefore chose TCMFL as a base material to test our ML-destabilization design rules (Table 1), choosing a small amount of Co substitution for Mn: Ti_{1.02}Cr_{1.1}Mn_{0.2}Co_{0.1}Fe_{0.6}La_{0.03} (hereon referred to as "modified TCMCFL").

TCMCFL was prepared by arc-melting, followed by high-energy ball-milling to produce a fine powder. The resulting phase exhibits a C14 Laves structure (Figure 3a), with no other major crystalline phases detected. The as-milled sample was characterized using scanning electron microscopy with energy dispersive spectroscopy (SEM/EDS), revealing micron-sized particles that display a homogeneous distribution of all metallic elements present in the alloy (Figure 3b). This uniformity suggests effective mixing and milling processes during preparation. The EDS maps further indicate the presence of some oxygen in the sample, despite all precautions taken during the synthesis and handling of the alloy to minimize air exposure. A quantitative analysis of the EDS data estimates the oxygen content to be approximately 1.6 wt%. This oxygen incorporation may have occurred during the brief exposure to air when the sample was transferred into the SEM/EDS chamber for analysis, as no oxides were detected in the XRD pattern collected in a capillary under argon.

A MS-16 micro reactor was loaded with 48.3 grams of the TCMCFL sample, and the reactor was coupled with a custom high-pressure PCT system capable of measurements up to approximately 1000 bar (see Supplementary Information for additional details). The reactor was leak checked, calibrated, and then the sample was activated at 200 °C in vacuum. An initial set of isotherms were measured (Figure 4a), that demonstrate the promise of this material for high pressure compression applications. The material exhibits a reasonably flat plateau at 20 °C and minimal hysteresis, achieving an absorption capacity of 1.5 wt% at a pressure of 400 bar. Accessing the full plateau at 151 °C would exceed the pressure limitations of the PCT machine (>1000 bar), but we observe a capacity of 0.5 wt% at a desorption pressure of 875 bar. Therefore, as much as 1.0 wt% can be compressed from 400 to 875 bar via a $20 \leftrightarrow 151$ °C temperature swing. At a reduced desorption temperature of 103 °C, 0.7 wt% could be pressurized from 400 to 750 bar. This material also maintains a long shelf-life, with no major change observed in the PCT isotherm after 12 months maintained under inert atmosphere. Taken 12 months after Figure 4a, Figure 4b shows additional PCT measurements at additional temperatures for performing the van't Hoff analysis (using fugacity, f , instead of pressure^{28,29}) in Figure 4c. The observed value of $\Delta H_d = 14.1$ kJ mol⁻¹ indeed represents a 2.5 kJ mol⁻¹ decrease compared to the base TCMFL and is qualitatively consistent with the expected decrease according Table 1. However, the observed value $\Delta S_d = 95.8$ J mol⁻¹ K⁻¹ represents an accompanying decrease of

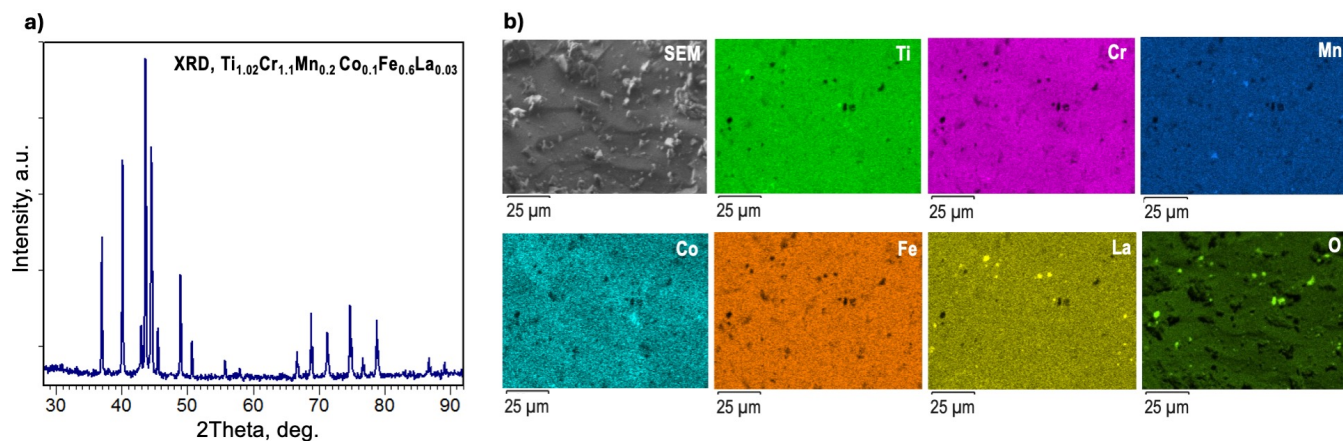


Figure 3: Experimental characterization of the TCMCFL material includes XRD of the as-milled C14 Laves phase (a), as well as SEM and EDS maps (b) confirming homogeneous elemental distributions.

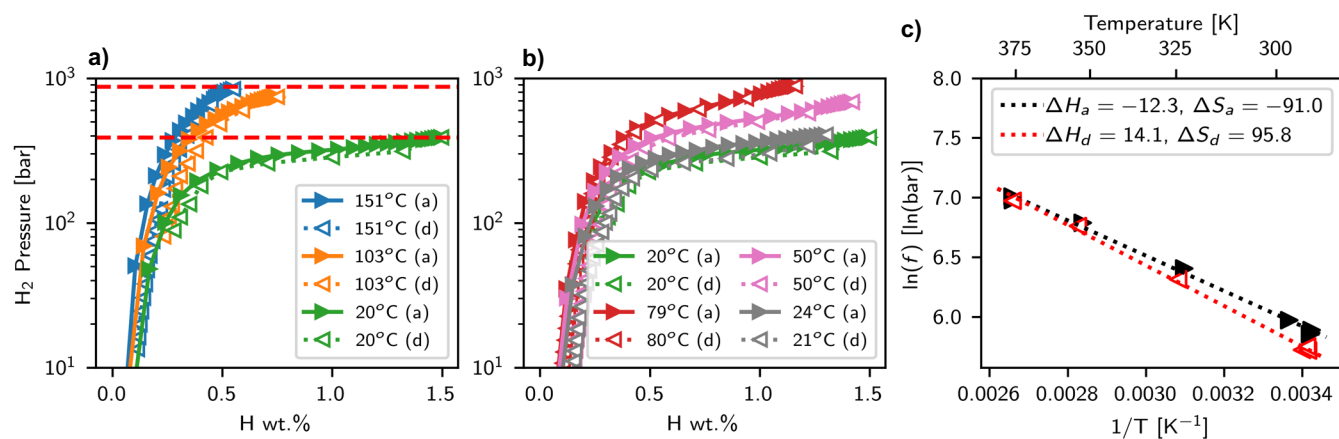


Figure 4: (a) High-pressure PCT absorption (closed markers) and desorption (open markers) for TCMCFL at three different temperatures. Dashed red lines indicate 400 bar and 875 bar. (b) Additional high-pressure PCT isotherms measurements taken 1 year after those in (a). (c) van't Hoff analysis to obtain the absorption and desorption enthalpy and entropy.

$8.4 \text{ J mol}^{-1} \text{ K}^{-1}$ in the desorption entropy compared to the base TCMFL. This enthalpy-entropy compensation is common in materials design, and often limits the extent to which one can freely tune the free energy of the hydriding reaction by modulating ΔH .^{18,30}

Systems-level modeling of high-pressure compressor operation. To show the potential for the TCMCFL material in a realistic compressor application, a system-level dynamic model of a metal hydride-based compressor was used to predict 2-stage and 3-stage compressor performance using the TCMCFL hydride for the high-pressure stage. Such modeling has been performed by others^{31–33} to take into account the complex process involving chemical kinetics and thermodynamics coupled with heat and mass transfer as multiple metal hydride beds undergo synchronized hydrogen absorption and desorption in a realistic system geometry. Here we use a model developed at Sandia using Matlab products Simulink and Simscape and described in detail elsewhere²⁰ and summarized in the SI.

To use the dynamic system model with measured data for candidate materials, parameterized thermodynamic models were fit using the Purdue Metal Hydride Toolbox³⁴ which was made available at <https://github.com/PurdueH2Lab/MetalHydrideToolbox>. The process and figures showing comparisons of each model to the measured data can be found in the SI. Four high performing materials investigated in previous work²⁰ were chosen for low pressure (LP) and medium pressure (MP) stages: A1= $\text{Ti}_{0.955}\text{Zr}_{0.045}\text{V}_{0.43}\text{Fe}_{0.12}\text{Al}_{0.03}\text{Mn}_{1.52}$ (known as Hydralloy C5), A2= $\text{Ti}_{0.95}\text{Zr}_{0.05}\text{Mn}_{1.55}\text{V}_{0.45}\text{Fe}_{0.09}$, A3= $\text{TiCrMn}_{0.7}\text{Fe}_{0.2}\text{V}_{0.1}$, A4= $\text{Ti}_{1.1}\text{CrMn}$). TCMCFL was chosen from the current work as the only viable high-pressure (HP) stage candidate. Several combinations of these metal hydrides, selected to best match the low- and high-temperature isotherms for the transitions between LP→MP stages and MP→HP stages, were used to predict the performance of 3-stage and 2-stage compressors as shown in Table 2. For each design, a limited parameter study (varying source pressure, heating and cooling temperatures, and cycle time) was carried out to determine the best performance in terms of compression ratio, hydrogen flow rate at 875 bar delivery pressure, and energy efficiency (calculated as heat required per mass of hydrogen delivered). The parameters that were varied included source pressure, heating and cooling temperatures, and cycle time. Source pressure was varied between 20 and 30 bar for the 3-stage systems and 90–120 bar for the 2-stage systems, hot and cold loop temperatures were constrained to ranges of 150 to 165 °C and 10 to 20 °C, respectively, and half cycle times between 12 to 18 minutes were considered.

As expected, Table 2 shows the effect and importance of the relationship between the operating parameters of a metal hydride compressor. Alloy combinations must be chosen to match thermodynamics at the desired operating

temperatures to maximize hydrogen flow rate and energy efficiency. For accurate predictions this must be done using realistic representations of the PCTs for each hydride that account for plateau slope and hysteresis at relevant pressures and temperatures. The study shows that trade-offs must be made between compression ratio and energy efficiency with a larger pressure range requiring higher energy use. At some compression ratio, the number of stages must increase assuming a fixed or limited temperature range. This not only reduces energy efficiency as more heat must be used for the additional stage (sensible heating and desorption enthalpy) but also increases the capital cost of the compressor which was not considered in this study. With these effects accounted for, the study shows that the TCMCFL material enables delivery of 875 bar hydrogen at the desired flow rate over reasonable operating temperature range in either a 2-stage or 3-stage compressor configuration, depending on the source pressure.

Conclusions

Compositional machine learning models of metal hydride thermodynamics provide an excellent tool to high-throughput screen hydride thermodynamic properties for new alloy compositions,^{13–15,35–38} but their performance degrades on highly out-of-distribution materials’ predictions. These include ultra-low stability hydrides (e.g., those useful for high pressure hydrogen compression), since the difficulty of PCT measurements in pressure regimes greater than 100–200 bar has led to a dearth of experimental characterization of these types of alloys. Therefore, we utilized SHAP explainability analyses of these models to obtain the composition-property design rules, and subsequent chemical substitution strategies, that are most likely to further destabilize an existing alloy’s hydride phase. We used one such substitution strategy to further destabilize an already promising metal hydride for hydrogen compression, then fully synthesized, characterized, and tested TCMCFL, including high-pressure PCT measurements to confirm its ability to compress hydrogen up to 875 bar.

Using experimental PCT data as the input upon which compression cycles were designed, a system dynamic model was used to predict operational performance and efficiencies of a multi-stage metal hydride compressor using TCMCFL in the final high-pressure stage. The performance study results indicate the lower overall energy efficiency of metal hydride compressors compared to other mechanical compression technologies. This is largely unavoidable given the heat required to overcome the metal hydride reaction enthalpy and the sensible heating required to cycle the metal hydride from low to high temperature. Furthermore, despite the compression performance of TCMCFL, we were unable to design a 2-stage metal hydride system that can produce a compression ra-

	LP	MP	HP	Half Cycle [min]	CR	P range [bar]	T range [°C]	H ₂ flow rate [kg _{H₂} hr ⁻¹]	Energy efficiency [kWh kg _{H₂} ⁻¹]
1	A1	A3	TCMCML	12	29.2:1	30-875	10-150	1.1	15.5
2	A2	A4	TCMCML	12	43.8:1	20-875	10-160	1.07	17.8
3	A3	–	TCMCML	12	7.3:1	120-875	10-160	1.1	10.9
4	A4	–	TCMCML	12	9.7:1	90-875	10-160	1.05	11.5

Table 2: System-level dynamic model predictions of operational performance and efficiency of four different TCMCFL-based high-pressure hydrogen compression designs including materials selection for LP, MP, and HP stages, half cycle time, T and P operating ranges, Compression Ratio (CR), H₂ flow rate, and Energy efficiency.

tio of 45:1 (e.g., ~20 to 875 bar) under the temperature swing conditions of interest (10 to 160 °C). However, the importance of energy efficiency is primarily in its relation to operating expense (OPEX). In many applications, the energy to operate the compressor must be paid for in the form of electricity and/or heat depending on the energy source. One advantage of a metal hydride compressor is that it is a heat-driven process requiring a proportionally small amount of electrical power.³⁹ In use-case scenarios where heat can be provided at a much lower cost than electricity, the energy usage of a metal hydride compressor may be higher than a comparable mechanical compressor but still be cost-competitive. For future work, there are clear opportunities to further optimize the TCMCFL alloy composition, since a further flattening of the PCT plateau could simultaneously improve its working capacity and compression ratio and lead to better operational efficiencies.^{7,40}

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