

Enhanced Cycle-Life Performance in Ti-V-Cr-Nb-Mo High-Entropy Alloys for Reversible Hydrogen Storage at Room Temperature

Andrei A. Agafonov,* Matthew D. Witman, Nayely Pineda-Romero, Mohamed El Hebri Rahmouni, Erik Elkaim, Vitalie Stavila, and Claudia Zlotea*



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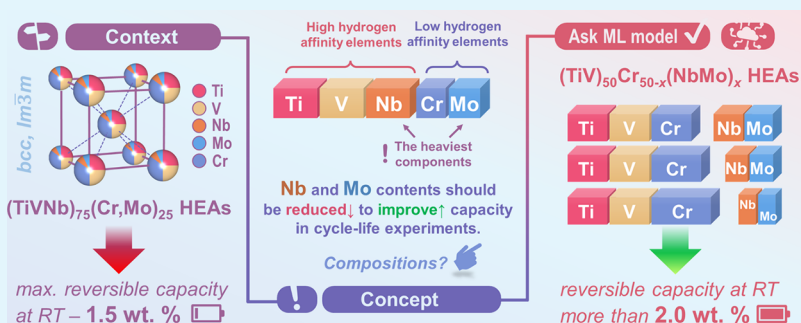
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ABSTRACT: This study focuses on the $(\text{TiV})_{50}\text{Cr}_{50-x}(\text{NbMo})_x$ ($x = 15, 20$, and 25) series of high-entropy alloys, which were predicted by machine learning to have high capacity and favorable thermodynamic properties. They are synthesized by arc melting and form single-phase *bcc* solid solutions. All three compositions rapidly absorb H_2 at room temperature and show strong tunability of the hydride phase formation thermodynamics, as well as of their total and reversible capacities. The most promising composition demonstrates a favorable enthalpy of hydride formation ($-35.5 \text{ kJ/mol} \cdot \text{H}_2$) and achieves a reversible capacity above 2.0 wt % upon 20 cycles of absorption-desorption at room temperature. These results are among the highest reported for this class of alloys.

KEYWORDS: high-entropy alloys, hydrogen storage, metal hydrides, synchrotron X-ray diffraction, high-capacity hydrides, room temperature cycle-life performance

Hydrogen energy is an emerging alternative to traditional fuels that serves as an energy carrier or reactant across diverse technological applications.^{1,2} However, one of the main challenges currently hindering the widespread adoption of hydrogen in energy applications is the difficulty of its storage and transportation.¹ Nowadays, most hydrogen is stored and transported in vessels under high pressure or in a liquefied state, which presents various challenges. Material-based hydrogen storage in the chemically bound form of metal hydrides has been proposed as an alternative that operates closer to ambient conditions and is potentially more cost-competitive.^{3–5}

High-entropy alloys (HEAs) have recently attracted attention for hydrogen storage due to their wide compositional variability, allowing for fine-tuning of their properties.^{6–8} However, many currently known potential candidates suffer from excessive stability of the resulting hydrides.^{2,9,10} Among many HEAs reported to date, the *bcc* Ti-V-Cr-Nb alloys forming *fcc* hydrides exhibited one of the best combinations of hydrogen storage properties: (1) high capacity (up to 2 H/M), (2) no significant secondary phase formation across variable alloy element ratios, and (3) promising cycle-life performance.^{11–14} Nevertheless, certain compositional limits are found. Thus, an excess of Cr content (more than 35 at %) leads to a sharp

capacity loss due to the secondary Laves phase formation, while a small concentration (less than 15 at %) results in insufficient destabilization of the hydride phase.¹³ In our recent study, we optimized the thermodynamic properties of Ti-V-Nb-Cr alloys by substituting Cr with Mo.¹⁵ However, Mo is a heavy component, which reduces the gravimetric capacity of the resulting material.¹⁶ The trade-off compositional limit between the HEA's thermodynamic properties and the total hydrogen storage capacity was found to be 10–15 Mo at %. On the other hand, the maximum reversible capacity in cycling experiments observed among the synthesized alloys is limited to 1.58 wt % of hydrogen (after 20 cycles). Room temperature (RT) “reversible” capacity refers to the amount of hydrogen that can be reversibly cycled at ambient or mild temperatures. Under these

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conditions, cycling occurs between the *fcc* dihydride and the *bcc* monohydride phases. While for some related compositions, the pressure-composition isotherm (PCI) experiments previously demonstrated the possibility to reach a reversible capacity slightly above 2.0 wt % at 50 °C; no cycling performance was reported.^{17,18} This suggests the need for further investigation to enhance the viability of these alloys for real-life applications. Additionally, Nb, as one of the heaviest components in this system along with Mo, also requires adjustment to improve the material gravimetric capacity. Particularly, considering all the above-mentioned limits for both Cr and Mo contents. Therefore, the next optimization step involves the simultaneous tuning of three elements (Cr, Nb, and Mo), which is a challenging task by the usual trial-and-error method. Modern materials design methods based on machine learning (ML), trained on experimental data or utilized in conjunction with density functional theory (DFT) calculations, have become a key tool for rationally navigating the composition space of alloys for hydrogen storage and compression applications. Thus, Witman et al.¹⁹ trained an ML model based on a large set of experimental data (HydPARK) to predict properties of new intermetallic and high-entropy hydrides.^{10,15,20,21} Similarly, Nefzi et al.²² in a recent study used a combined ML-DFT approach to identify promising *bcc* equimolar HEAs among over 8000 quinary systems.

In this study, we undertake a further attempt to optimize the Cr, Mo, and Nb contents in Ti-V-Cr-Nb-Mo *bcc* alloys to improve the reversible capacity at room temperature (RT) cycling. To this end, we first perform the ML predictions followed by experimental validation through laboratory techniques and synchrotron X-ray diffraction.

To identify the most promising direction in the compositional space of Ti-V-Cr-Nb-Mo alloys, considering the established solubility limits on the Cr, Nb, and Mo concentrations before secondary phase formation, our previously demonstrated¹⁵ ML (gradient boosting tree regressor) model is applied (see Supporting Information, Section 1 for more details). Simultaneously, the NbMo content was targeted for reduction (to increase the total gravimetric capacity), whereas the Cr content was intended to be increased (to destabilize the hydride phase additionally). According to the ML predictions, the $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{50-x}(\text{NbMo})_x$ series of alloys is expected to be promising and meet the stated goals in terms of hydride formation thermodynamics and capacity (Figure 1a and Table 1). The calculated model's Shapley Additive Prediction (SHAP) values, i.e., the impact on the model's output (*x*-axis) attributed to each feature for current prediction (scatter point), are presented in Figure 1b and indicate that the composition-

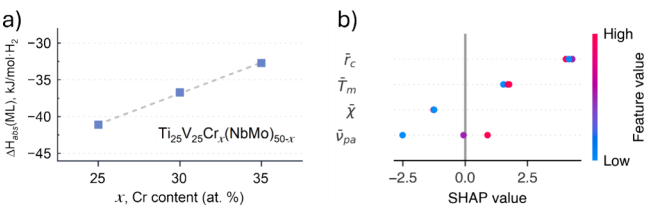


Figure 1. (a) ML-predicted absorption enthalpy for selected compositions and (b) SHAP values for the model's four most important features: the composition-weighted mean over elemental properties for covalent radius ($\bar{r}_c$), melting temperature ($\bar{T}_m$), Pauling electronegativity ($\bar{\chi}$), and volume per atom ($\bar{v}_{\text{pa}}$).

Table 1. Compositional Elemental A- and B-Type Metal Ratio, the Lattice Types and Parameters for All the As-Synthesized and Hydrogenated Alloys, Hydrogen Storage Capacities (Max. and Reversible after 20 Cycles of Absorption), Related ML Predicted and Experimental Hydrogenation Thermodynamics Parameters (ΔH_{abs} and ΔS_{abs})^a

composition	alloy	A/B ^a	phase	a, Å	max. capacity ^b		ΔH_{abs} (ML), kJ/mol H_2	ΔH_{abs} (exp.), kJ/mol H_2	ΔS_{abs} (exp.), J/(mol H_2 °K)	reversible capacity ^c	
					H/M	H wt %				H/M	H wt %
(TiV) ₅₀ Cr ₂₅ (NbMo) ₂₅	3.109(1)	1.67	<i>bcc</i>	4.341(1)	1.90	3.094	-41	-38.1(1.0)	-135.5(3.2)	1.14	1.862
(TiV) ₅₀ Cr ₃₀ (NbMo) ₂₀	3.097(1)	1.50	<i>bcc</i>	4.327(1)	1.88	3.179	-37	-39.6(2.0)	-142.0(0.7)	1.21	2.052
(TiV) ₅₀ Cr ₃₅ (NbMo) ₁₅	3.077(1)	1.35	<i>bcc</i>	4.308(1)	1.92	3.368	-33	-35.5(0.4)	-134.3(1.2)	1.18	2.078

^aA and B type elements: A—high and B—low affinity to hydrogen. ^b $T_{\text{abs}} = 25$ °C, $P_{\text{eq}} \sim 50$ bar. ^c $T_{\text{abs}} = T_{\text{des}} = 25$ °C, dynamic primary vacuum used for desorption between cycles; the reversible capacities are given after 20 cycles. ^dThe main phase in multiphase materials is provided in bold.

weighted mean volume per atom ($\bar{V}_{\text{pa}}$) is the primary feature by which the model differentiates its predictions of reaction enthalpy across the three alloys. To experimentally validate these findings, a series of $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{50-x}(\text{NbMo})_x$ ($x = 15, 20$, and 25) HEAs is prepared by electric arc-melting under an Ar atmosphere following the same procedure as reported previously.¹⁵ The crystalline structures (alloy ingots and powdered hydrides) are investigated by X-ray diffraction (XRD) using a D8 Advance Bruker diffractometer ($\text{Cu-K}\alpha$ radiation $\lambda = 1.5406 \text{ \AA}$, Bragg–Brentano Geometry). The Anton Paar TTK 450 environmental chamber is used for *in situ* XRD analysis of thermal desorption. All the XRD data are analyzed using the fundamental parameters approach implemented in TOPAS 7 Bruker software to evaluate the lattice parameters and phase composition. High-energy synchrotron X-ray powder diffraction (SR-XRD) experiments are performed at the CRISTAL beamline (SOLEIL synchrotron, France) for in-depth structural characterization of the powdered materials obtained after hydrogen absorption/desorption cycling experiments for $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{35}(\text{NbMo})_{15}$ ($\lambda = 0.46521 \text{ \AA}$). The results are refined by the Rietveld method using the FullProf software.²³ The microstructure of alloys is investigated by scanning electron microscopy (SEM) using a Zeiss Merlin instrument equipped with an EDS (energy-dispersive X-ray spectroscopy) detector,

as it was described earlier.¹⁵ The hydrogenation, PCI measurements, and cycling experiments are carried out for all the alloys using a home-made Sieverts apparatus following the same procedures as in our recent study.¹⁵ All materials are activated at 400°C under primary dynamic vacuum for 10 hrs prior to hydrogenation. For the acquisition of PCIs, temperature regulation is ensured by three different thermal devices: a refrigeration bath circulator (JULABO CORIO CD-1200F) for temperatures below 25°C , a thermostatic water bath for temperatures slightly above ambient conditions ($25\text{--}75^\circ\text{C}$), or a thermostatic oil bath for 100°C . The storage capacity in this study is estimated using GASPAC Version 3.32 (Horizon Technologies) and H_2 real gas density.

The XRD results (Figure 2a) confirm that all three samples are *bcc* single-phase solid solutions with a clear dependence (Figure 2b) of the lattice parameter (a_{bcc}) on the Cr content: as Cr has the smallest atomic radius within the composition ($r_{\text{Cr}} = 1.25 \text{ \AA}$),²⁴ the lattice parameter decreases as the Cr content increases. The microstructure of all HEAs is investigated by SEM-EDS. Typically for unannealed HEAs, the microstructure consists of dendritic (abundant in V, Nb, and Mo) and interdendritic (rich in Ti and Cr) regions (see Supporting Information, Section 2 for more details).²⁵ Despite this microstructural inhomogeneity, the overall composition agrees well

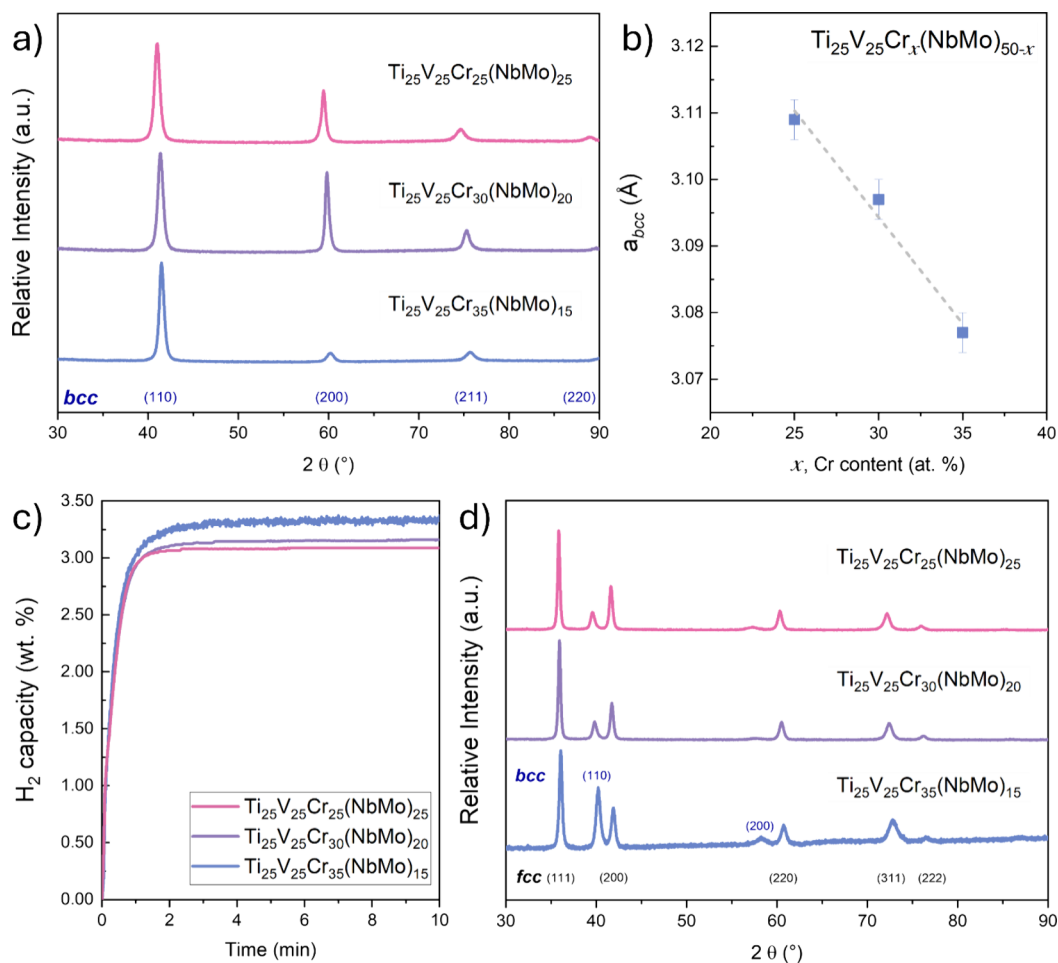


Figure 2. (a) XRD patterns ($\lambda = 1.5406 \text{ \AA}$) of the as-synthesized $(\text{TiV})_{50}\text{Cr}_{50-x}(\text{NbMo})_x$ ($x = 15, 20$, and 25); (b) a_{bcc} lattice parameters as the function of Cr content (the dashed line is a guide for the eyes); (c) first H_2 absorption kinetics, and (d) XRD patterns ($\lambda = 1.5406 \text{ \AA}$) of the hydrogenated materials.

with the nominal one. While the HEA concept presumes a probabilistic distribution of constituent metal atoms across crystallographic sites, XRD did not reveal any noticeable separation into multiple *bcc* phases. Thus, while explicitly considering compositional inhomogeneities at the level of dendritic and interdendritic regions, the materials exhibit effectively single-phase behavior in the following hydrogenation tests.

All alloys show fast H_2 absorption kinetics reaching capacities around 1.9 H/M (3.10–3.37 wt %) in 2 minutes at 25 °C under final H_2 pressure ~ 50 bar (Figure 2c, Table 1). The maximum gravimetric capacity exhibits a strong dependence on the average molar mass of the alloy, as expected: the highest capacity is achieved for $(TiV)_{50}Cr_{35}(NbMo)_{15}$, characterized by the lowest Nb and Mo content, whereas the lowest capacity is observed for $(TiV)_{50}Cr_{25}(NbMo)_{25}$. The materials obtained after hydrogenation consist of a mixture of *bcc* monohydrides and *fcc* dihydrides (Figure 2d, Table 1). As the Cr content increases, the *bcc* monohydride's phase fraction rises, which might indicate a strong dependence of hydride's destabilization on Cr concentration.

To evaluate the thermodynamics of the hydride formation, a set of PCIs is measured (Figure 3a). Even though all compositions exhibited sloped plateaus, the Van't Hoff approach was applied, considering the equilibrium pressure at the capacity of 1.20–1.25 H/M (in the middle of the second plateau). Due to compositional inhomogeneity in the alloys, the sloped PCI plateaus are typical characteristics of as-cast unannealed HEAs. However, a recent study by Enblom et al.²⁵ on two refractory HEAs with and without dendritic-interdendritic compositional fluctuations demonstrated that the derived thermodynamic parameters are indistinguishable within experimental uncertainty. Thus, in light of these considerations, the analysis proceeds based on experimentally estimated enthalpy and entropy of absorption/desorption and is confronted with ML predictions. The results obtained are in good agreement with the ML model's calculations within the error bar. The ratio between element concentration with high (A-type) and low (B-type) affinities to hydrogen plays an important role in thermodynamic destabilization. The reduction of the A/B-type elements content (A/B) ratio appears to destabilize the hydride phase (Table 1): the lowest A/B composition shows the best hydride destabilization ($\Delta H_{abs} = -35.5$ kJ/mol· H_2). Furthermore, the PCIs experiments allow the anticipation of promising cycle-life properties at RT for all three compositions. For instance, the $(TiV)_{50}Cr_{35}(NbMo)_{15}$ HEA, which is characterized by the most destabilized hydride phase, demonstrates an absorption plateau around 10 bar, while the desorption plateau exceeds 1 bar. Thus, it indicates potential in this composition for high-capacity retention during RT cycling. However, the compositions containing 20 and 25 at % of (NbMo), result in lower gravimetric capacities (Figure 2c) and a desorption plateau below 1 bar in the PCIs (Figure 3a). Therefore, slightly lower reversible capacities can be expected for both of them in cycling experiments.

The reversibility of the materials is investigated by *in situ* X-ray diffraction under 1 bar of inert Ar atmosphere during the stepwise heating from 25 to 400 °C (step is 25 °C; Figure 3b). The absorption mechanism of Ti-V-Cr-Nb-Mo *bcc* alloys involves the sequence phase transformation with increasing hydrogen content: *bcc* alloy $\rightarrow$ *bcc* monohydride $\rightarrow$ *fcc* dihydride.¹⁵ Meanwhile, the desorption is expected to occur reversibly in the same steps. The phase transition temperatures

from *fcc* dihydride to *bcc* monohydride phases for $(TiV)_{50}Cr_{25}(NbMo)_{25}$ and $(TiV)_{50}Cr_{30}(NbMo)_{20}$ are observed around 325 and 300 °C, respectively. For the lowest (NbMo)-content HEA, the phase transition starts at a lower temperature, around 250 °C. However, a significant presence of the *bcc* monohydride was already found in the initial $(TiV)_{50}Cr_{35}(NbMo)_{15}$ material (which is likely due to the high destabilization of the hydride and partial spontaneous desorption at ambient conditions). These results agree well with the thermodynamic properties evaluation of these alloys.

To study the cycle-life properties of the synthesized materials, several absorption/desorption cycling experiments were carried out. First, all HEAs are tested over 20 cycles under the same absorption/desorption conditions. H_2 absorption is performed at 25 ± 1 °C and the final pressure $P_{eq} \sim 40$ bar. Desorption between cycles is performed at 25 ± 1 °C and using a dynamic primary vacuum applied for ~ 60 minutes (until a base vacuum of $2\text{--}3 \times 10^{-3}$ mbar is reached). All three compositions show good reversible capacities ranging from 1.86 to 2.27 wt % (Figure 4a), which are around the highest values reported to date for *bcc* HEAs (e.g., 2.0 H wt % for Ti-V-Nb-Cr-Fe²⁶ or 1.85–2.04 H wt % for Ti-Cr-V-Nb-Fe/Mn²⁷ under almost similar conditions). The $(TiV)_{50}Cr_{35}(NbMo)_{15}$ alloy has the best performance with a reversible capacity at RT between 2.27 (*initial*) and 2.08 (*final*) wt % in 20 cycles. Despite a greater absolute value in the thermodynamics of hydride formation indicating a more stable hydride, the $(TiV)_{50}Cr_{30}(NbMo)_{20}$ also demonstrates good stability and reversible capacity (2.22 (*initial*) and 2.04 (*final*) wt %) under the same conditions. Therefore, the reversible capacity over 20 cycles exceeds 2.0 wt % for both mentioned alloys. Meanwhile, the $(TiV)_{50}Cr_{25}(NbMo)_{25}$ composition shows the lowest reversible capacity in this study (2.03 (*initial*) and 1.86 (*final*) wt %). However, all three HEAs demonstrate significantly higher reversible capacities over 20 cycles as compared to previous results within the same compositional space (1.58 wt % as a maximum).¹⁵ Figure 4b presents the XRD patterns of materials after 20 cycles, desorbed at 25 °C (*color*) and at 410 °C (*gray*). The difference in the *bcc* diffraction peak positions in the XRD pattern of the materials desorbed at high and room temperatures indicates the different states reached in the desorption process. High-temperature desorption ensures full recovery of the initial *bcc* alloy, while RT desorption results in the *bcc* monohydride phase. This is confirmed by the values of the *bcc* lattice parameters of desorbed materials at 25 and 410 °C (Table 2). The alloys desorbed at high temperature have lattice parameters close to the initial as-cast materials (Table 1), while the lattice parameters after RT desorption are higher because they are related to the *bcc* monohydride phase. For all compositions, the same capacity loss of around 8.1–8.4% in 20 cycles is observed.

The performance achieved for the $Ti_{25}V_{25}Cr_{35}(NbMo)_{15}$ marks a significant step forward for the Ti-V-Cr-Nb-Mo system, substantially outperforming the reversible capacity of $(TiV)_{75}Cr_{20}Mo_5$ (by $\approx 32\%$). Nevertheless, the reliance on vacuum during desorption in cycling experiments is impractical for real-life applications.²⁸ To assess the viability of the $Ti_{25}V_{25}Cr_{35}(NbMo)_{15}$ alloy under vacuum-free cycling conditions, two additional cycling tests were carried out (Figure 4c). The absorption conditions for both experiments remained constant ($T = 25 \pm 1$ °C and $P_{eq} \sim 60$ bar), while desorption is performed until an equilibrium hydrogen pressure

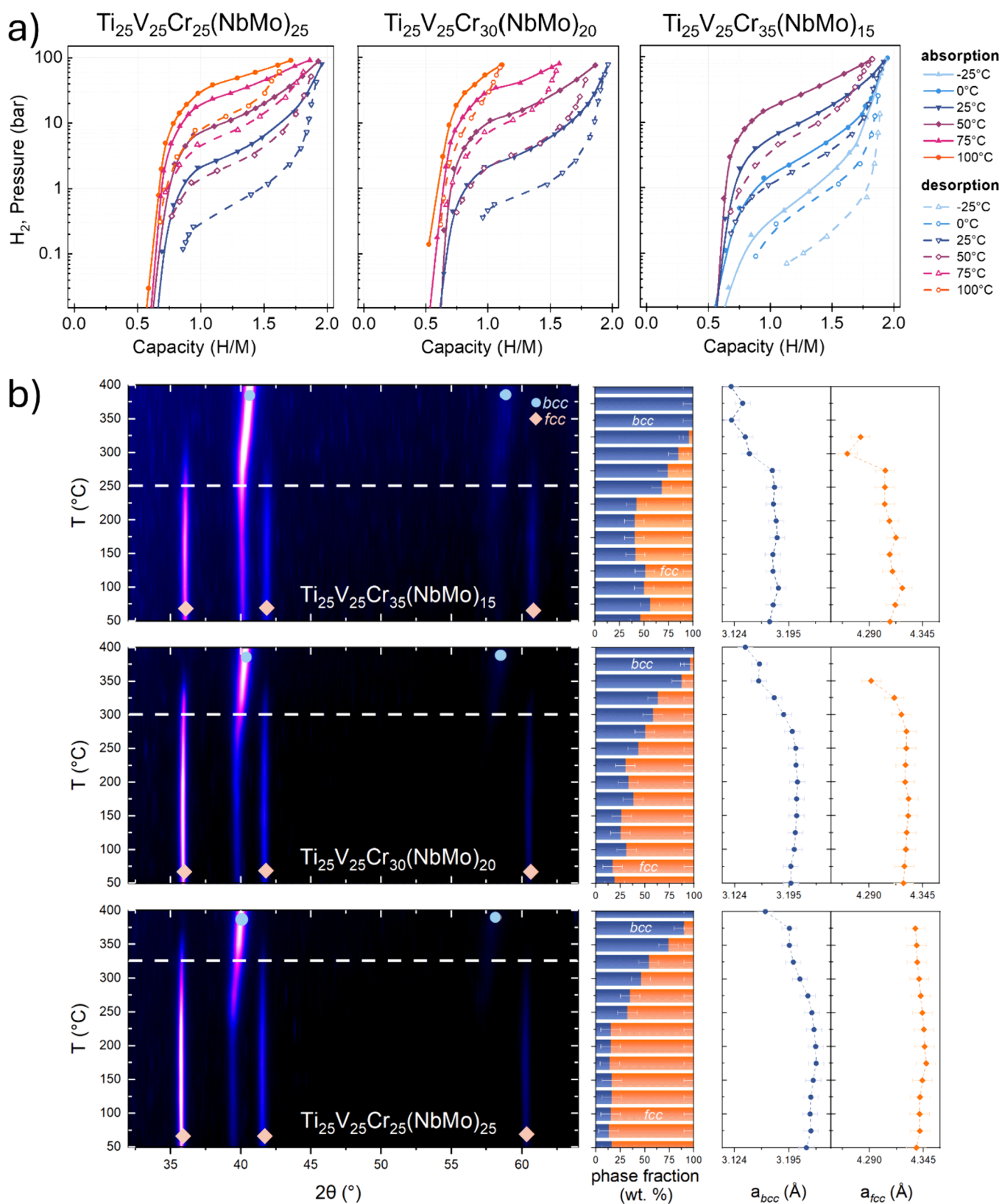


Figure 3. (a) Absorption and desorption PCIs performed at the $(\text{TiV})_{50}\text{Cr}_{50-x}(\text{NbMo})_x$ ($x = 15, 20, \text{ and } 25$) alloys; (b) *in situ* thermal desorption XRD patterns of hydrogenated $(\text{TiV})_{50}\text{Cr}_{50-x}(\text{NbMo})_x$ ($x = 15, 20, \text{ and } 25$) alloys (left), phase fractions (middle), and related lattice parameters (right) evolution during thermal desorption.

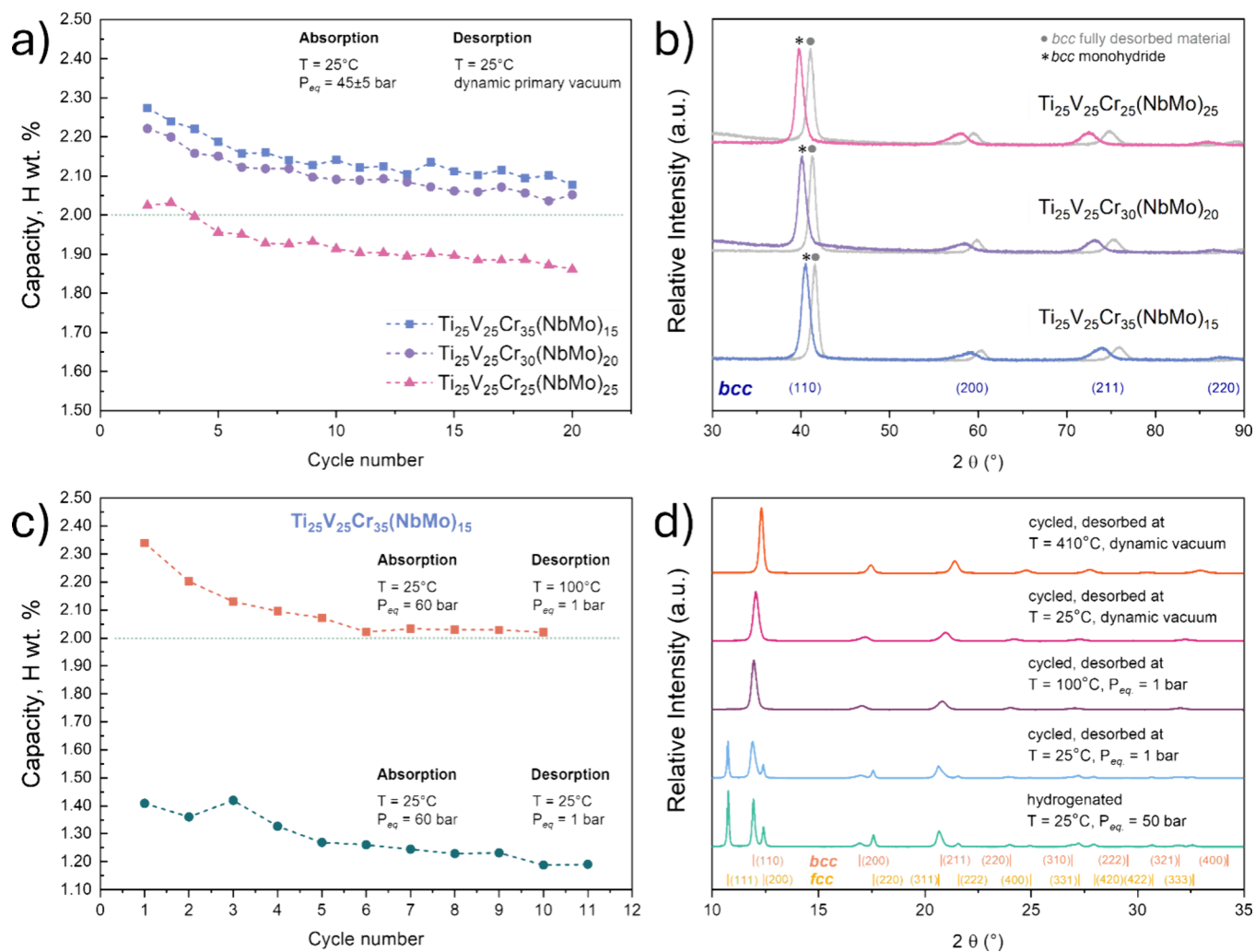


Figure 4. (a) Reversible capacity variation during 20 absorption/desorption cycles at 25 °C for all the $(\text{TiV})_{50}\text{Cr}_{50-x}(\text{NbMo})_x$ ($x = 15, 20$, and 25) alloys; (b) XRD patterns of desorbed materials after 20 cycles under vacuum at 25 °C (in colors) and after high temperature (410 °C) desorption are presented (in gray) for the sake of comparison; (c) the reversible capacity evolution under 10–11 cycles at constant absorption conditions ($T = 25$ °C and $P_{\text{eq}} = 60$ bar), but two different desorption regimes ($P_{\text{eq}} = 1$ bar) applied between cycles: $T = 25$ °C (in green) and $T = 100$ °C (in orange) for the $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{35}(\text{NbMo})_{15}$ alloy; (d) SR-XRD patterns ($\lambda = 0.46521$ Å) of the first hydrogenated and desorbed materials after cycling experiments on the $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{35}(\text{NbMo})_{15}$ alloy.

Table 2. *bcc* Lattice Parameters of the Materials after 20 Cycles, Desorbed at 25 and 410 °C

composition	a_{bcc} (Å), desorbed at 25 °C	a_{bcc} (Å), desorbed at 410 °C	Δa_{bcc} (Å)
$(\text{TiV})_{50}\text{Cr}_{25}(\text{NbMo})_{25}$	3.191(1)	3.106(1)	0.085
$(\text{TiV})_{50}\text{Cr}_{30}(\text{NbMo})_{20}$	3.165(1)	3.090(1)	0.075
$(\text{TiV})_{50}\text{Cr}_{35}(\text{NbMo})_{15}$	3.136(1)	3.068(1)	0.068

of 1 bar is reached at two different temperatures: 25 and 100 °C, respectively (to perform the desorption step, the material is heated up to 100 °C for each cycle). As shown in Figure 4c, even without high-temperature desorption, $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{35}(\text{NbMo})_{15}$ alloy enables the delivery of 1.2–1.4 wt % of hydrogen, which is comparable to the materials available on the market.²⁹ However, an increase in temperature during desorption conditions (100 °C) leads to a substantial improvement in the cycle-life performance, and it allows for a reversible capacity of 2 wt %. This highlights the potential of the $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{35}(\text{NbMo})_{15}$ alloy for applications in hydrogen storage systems capable of recycling dissipated energy from

industrial/urban processes to provide mild heating conditions during operation.

SR-XRD was employed to elucidate the structural mechanisms and phase evolution governing the alloy cycling behavior. The measurements were performed on the desorbed materials after cycling experiments as well as on the first hydrogenated nondesorbed material. As presented in Figure 4d (detailed in Table 3), the cycled material that is desorbed at $T = 25 \pm 1$ °C ($P_{\text{eq}} = 1$ bar) represents a combination of the *bcc* monohydride phase and the *fcc* dihydride phases, hinting at an incomplete dihydride decomposition between cycles. On the contrary, the cycled alloy with additional stimuli for the desorption step (vacuum or

Table 3. Phase Composition and Related Lattice Parameters According to the SR-XRD Results of the First Hydrogenated and Desorbed $\text{Ti}_{25}\text{V}_{25}\text{Cr}_{35}(\text{NbMo})_{15}$ Materials after Cycling Experiments^a

material	phase fraction	lattice parameter, a (Å)
hydrogenated at $T = 25\text{ }^{\circ}\text{C}$, $P_{\text{eq.}} = 50\text{ bar}$, nondesorbed	<i>fcc</i> – 51%	4.305(1)
	<i>bcc mh</i> – 49%	3.163(1)
cycled, desorbed at $T = 25\text{ }^{\circ}\text{C}$, $P_{\text{eq.}} = 1\text{ bar}$	<i>fcc</i> – 34%	4.309(1)
	<i>bcc mh</i> – 66%	3.165(1)
cycled, desorbed at $T = 100\text{ }^{\circ}\text{C}$, $P_{\text{eq.}} = 1\text{ bar}$	<i>bcc mh</i> – 100%	3.154(1)
cycled, desorbed at $T = 25\text{ }^{\circ}\text{C}$, dynamic vacuum	<i>bcc mh</i> – 100%	3.130(1)
cycled, desorbed at $T = 410\text{ }^{\circ}\text{C}$, dynamic vacuum	alloy <i>bcc</i> – 100%	3.068(1)

^a*bcc mh* = *bcc* monohydride.

mild heating up to 100 °C) shows only *bcc* phase formation, which confirms complete decomposition of the dihydride phase between cycles and drastically increases the reversible capacity to 2 wt %. This conclusion is proved by the shift in the *bcc* peak positions toward higher 2θ (a decrease in the crystalline lattice parameter due to the release of hydrogen, Table 3). The high-temperature desorption (410 °C) ensures the complete decomposition of all hydride phases (characterized by a subsequent decrease in a_{bcc}), resulting in the desorbed *bcc* alloy.

In conclusion, we demonstrate an integrated materials design framework that couples ML predictions with targeted synthesis, complemented by a detailed structural characterization to improve the thermodynamics, usable capacity, and cycle-life performance of Ti-V-Cr-Nb-Mo-based HEAs. The resulting compositions demonstrate good agreement between their thermodynamic properties and ML predictions. They also exhibit significantly improved cycling properties (the reversible capacity at RT is increased by up to 32% compared to previous materials in the same compositional space). A series of cycling experiments followed by SR-XRD analysis demonstrate that a reversible hydrogen capacity of 1.2–1.4 wt % can be achieved at 25 °C under an equilibrium pressure of 1 bar, demonstrating a potential to increase this reversible capacity beyond 2 wt % if the additional desorption stimulation between cycles (vacuum application or mild heating up to 100 °C under an equilibrium pressure of 1 bar) is applied. Ultimately, the coupling of data-driven screening and mechanistic studies provides a blueprint for the rapid discovery of transition-metal-based HEAs³⁰ designed for real-world hydrogen use cases.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsaem.6c01893>.

Machine learning details (Section 1); SEM-EDS results (Section 2) (PDF)

■ AUTHOR INFORMATION

Corresponding Authors

Andrei A. Agafonov – Univ. Paris-Est Créteil, CNRS, ICMPE, UMR 7182, 2 Rue Henri Dunant, Thiais 94320, France;

orcid.org/0009-0000-6758-9524;

Email: andrei.agafonov@cnrs.fr

Claudia Zlotea – Univ. Paris-Est Créteil, CNRS, ICMPE, UMR 7182, 2 Rue Henri Dunant, Thiais 94320, France;

orcid.org/0000-0002-4454-6991;

Email: claudia.zlotea@cnrs.fr

Authors

Matthew D. Witman – Sandia National Laboratories, Livermore, California 94551, United States; orcid.org/0000-0001-6263-5114

Nayely Pineda-Romero – Univ. Paris-Est Créteil, CNRS, ICMPE, UMR 7182, 2 Rue Henri Dunant, Thiais 94320, France

Mohamed El Hebri Rahmouni – Univ. Paris-Est Créteil, CNRS, ICMPE, UMR 7182, 2 Rue Henri Dunant, Thiais 94320, France

Erik Elkaim – Synchrotron SOLEIL, L'Orme des Merisiers Départementale 128, Saint-Aubin 91190, France

Vitalie Stavila – Sandia National Laboratories, Livermore, California 94551, United States; orcid.org/0000-0003-0981-0432

Complete contact information is available at:

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Author Contributions

A.A.: Conceptualization, investigation, validation, visualization, writing (original draft, review, and editing), and project administration; M.D.W.: conceptualization, investigation, validation, and writing (original draft, review, and editing); N.P.R.: writing (review and editing), validation; M.E.H.R. and E.E.: investigation and validation; V.S.: supervision and writing (review and editing); C.Z.: conceptualization, investigation, validation, writing (original draft, review, and editing), supervision, and funding acquisition.

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Notes

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