Harnessing structural complexity for hydrogen storage in multi-principal alloys

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Introduction

- Hydrogen storage is a central challenge for the development of efficient hydrogen technologies.
- Hydrogen stands out as a promising energy carrier thanks to its high energy density and environmental benefits. However, a major obstacle remains: identifying effective methods for hydrogen storage and release.
- Among various candidates, metal hydrides have attracted significant attention due to their unique properties - including high hydrogen storage capacity, favorable thermodynamics, and the potential for reversible uptake.
- This study explores advanced materials for hydrogen storage, focusing on the influence of composition and structure on hydrogen behavior.
- Advanced characterization techniques provide key insights into phase stability and kinetics, supporting the development of efficient materials for sustainable energy applications.

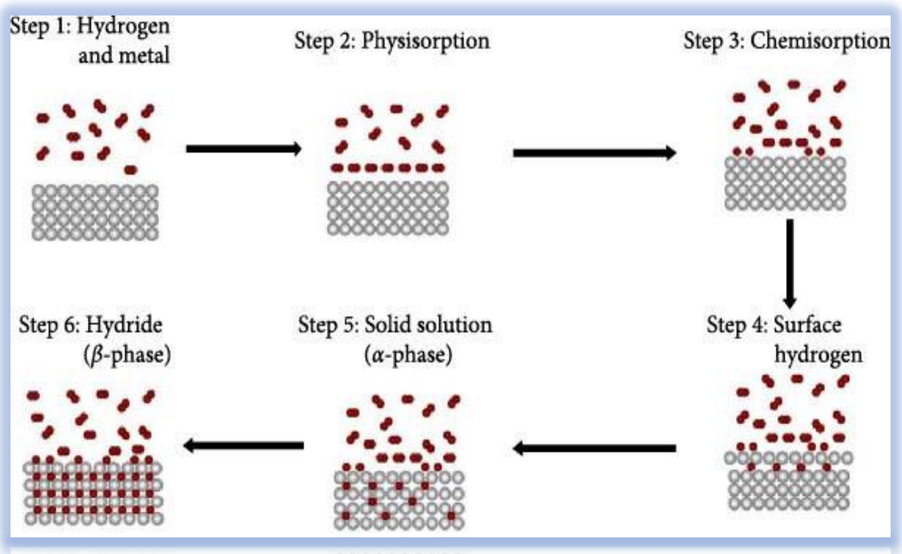

Figure 2. Steps involved in the formation of metal hydrides [2].

Methods

- To explore the role of compositional complexity and lattice distortions, a series of multi-principal alloys (MPAs) were synthesized and examined by diffraction and sorption techniques.
- Five equiatomic alloys with the compositions TiVZrNbTa, TiZrNbHfTa, TiZrNbMoTa, TiZrNbMoHf, and VNbMoHfTa were prepared by arc melting under an argon atmosphere.
- The resulting ingots were mechanically processed using planetary ball milling.
- The powder fraction with a particle size below 45 µm was collected by sieving and used for further analyses.

Hydrogen sorption kinetics

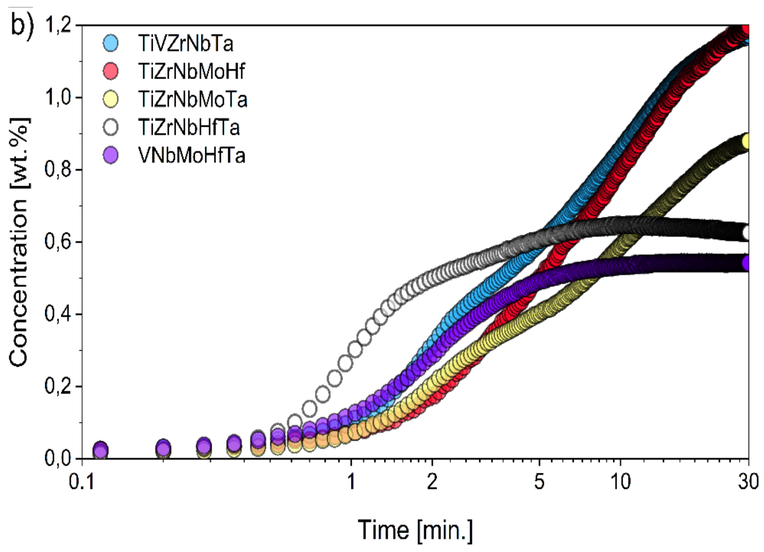

Figure 3. Hydrogen sorption kinetics of investigated samples (5 MPa, 250°C).

In-situ diffraction studies of hydrides desorption

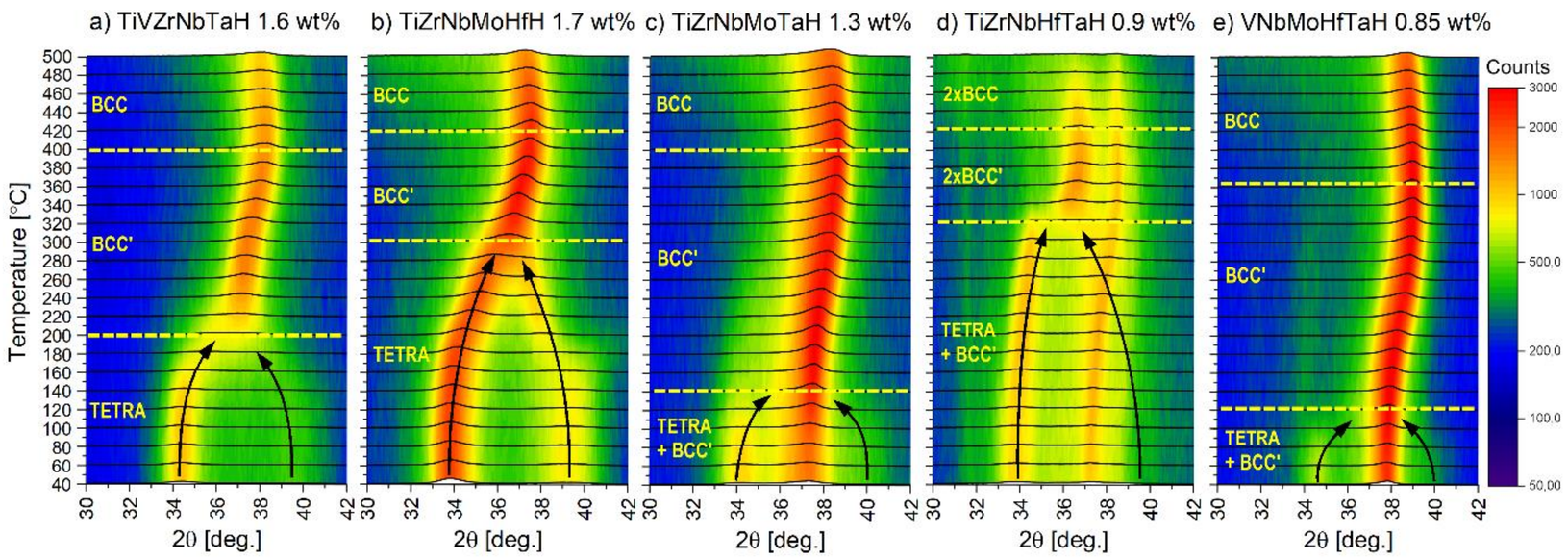

Figure 4. In-situ diffraction studies of hydrides desorption, the desorption was made in Ar flow at pressure of 0.02 MPa. The arrows guide decomposition of the tetragonal phases. Movement of the 101 and 110 reflections is marked for an eye with left and right arrow, respectively.

Theoretical prediction model

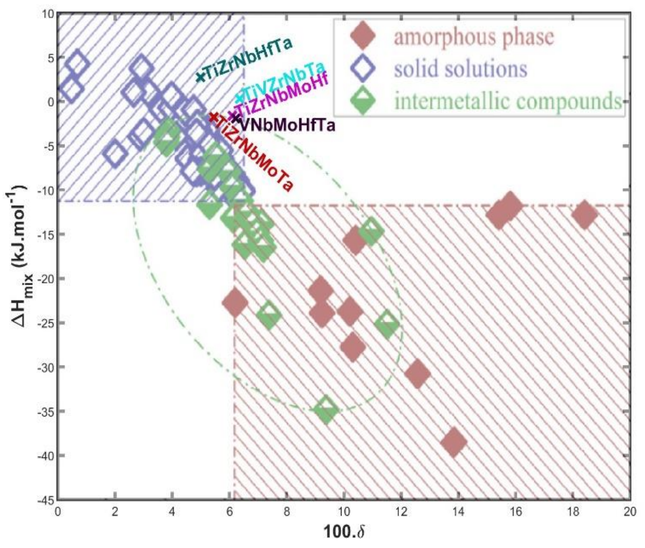

Figure 1. Prediction model of the formation of a solid solution given by the parameters 100.δ and ΔH_{mix}, taken from [1] and supplemented with the studied MPAs.

X-Ray diffraction

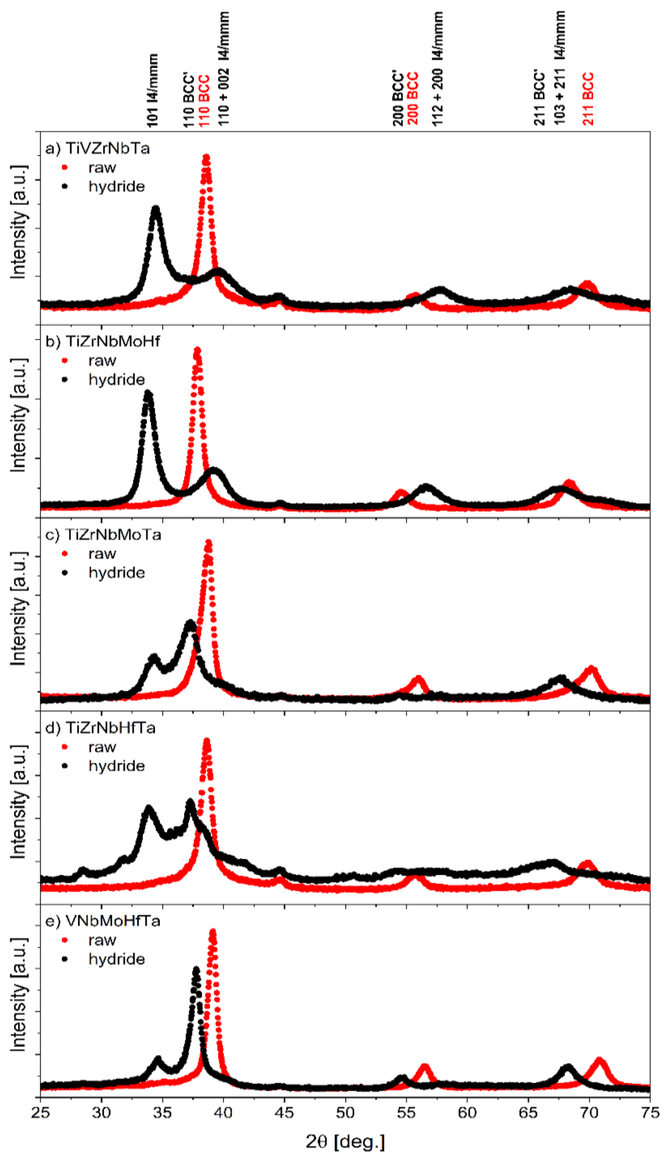

Figure 5. X-ray diffraction patterns for investigated samples. The comparison between raw (red points) and hydrided specimens (black points) is presented.

Conclusions

- Five-element MPAs, composed of Ti, V, Zr, Nb, Ta, Mo, and Hf, were successfully designed and synthesized.
- Hydrogen uptake induced distinct structural transitions, with BCC solid solutions evolving into hydrogen-rich tetragonal hydride phases.
- All as-cast alloys crystallized in a body-centered cubic structure.
- Upon hydrogenation:
- At high hydrogen concentrations (> 1.5 wt.%), the structure transformed completely into a tetragonal phase.
- At lower hydrogen concetrations, the tetragonal phase became dominant, while a residual BCC phase remained.
- The presence of the BCC is attributed to hydrogen-poor α-phases, whereas the tetragonal structure corresponds to hydrogen-rich β-phases.
- Among the studied alloys, TiVZrNbTa and TiZrNbMoHf exhibited significantly higher hydrogen uptake.
- The TiZrNbMoHf alloy demonstrated the highest hydrogen storage capacity, reaching 1.7 wt.% (corresponding to a D/M ratio of 1.7), while the lowest capacity was observed for VNbMoHfTa (0.85 wt.%).
- These results demonstrate that lattice strain and compositional tuning are decisive factors for tailoring hydrogen storage performance in MPAs.

References

[1] S. Guo, Q. Hu, C. Ng, and C. T. Liu, "More than entropy in high-entropy alloys: Forming solid solutions or amorphous phase" *Intermetallics*, vol. 41, pp. 96–103, 2013, doi: (10.1016/j.intermet.2013.05.002.).
[2] E. Nemukula, C.B. Mtshali, F. Nemangwele, "Metal Hydrides for Sustainable Hydrogen Storage: A Review" *International Journal of Energy Research*, Issue: 1, 2025, doi: (10.1155/er/6300225).

Acknowledgment

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