

Supplementary Information

How 10 at% Al addition in the Ti-V-Zr-Nb high entropy alloy changes the hydrogen sorption properties?

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Figure SI1. XRD pattern comparison between the quaternary $\text{Ti}_{0.325}\text{V}_{0.275}\text{Zr}_{0.125}\text{Nb}_{0.275}$ (top) and the quinary $\text{Al}_{0.10}\text{Ti}_{0.30}\text{V}_{0.25}\text{Zr}_{0.10}\text{Nb}_{0.25}$ (bottom) alloys as-cast by arc melting technique.

Figure SI2. Pressure composition isotherm of $\text{Al}_{0.10}\text{Ti}_{0.30}\text{V}_{0.25}\text{Zr}_{0.10}\text{Nb}_{0.25}\text{H}_{1.6}$ measured at 100 °C. Sample activated at 340 °C under dynamic vacuum for 2 hours.

Figure SI3. *In-situ* neutron diffraction of $\text{Ti}_{0.325}\text{V}_{0.275}\text{Zr}_{0.125}\text{Nb}_{0.275}\text{D}_{1.8}$ under a heating ramp of 1 °C/min while under dynamic vacuum, along with the corresponding desorption profile on the right.

Figure SI4. Reversible hydrogen absorption capacity at 25 °C of $\text{Ti}_{0.325}\text{V}_{0.275}\text{Zr}_{0.125}\text{Nb}_{0.275}$ upon 20 cycles. The cycling evaluation consisted in measuring the hydrogen capacity at room temperature followed by complete hydrogen desorption. The latter step was done by heating at 400 °C while evacuating under secondary vacuum (10^{-5} mbar) for 10 hours.

Figure SI5. Chemical mapping (SEM-EDX) of the hydride $\text{Al}_{0.10}\text{Ti}_{0.30}\text{V}_{0.25}\text{Zr}_{0.10}\text{Nb}_{0.25}\text{H}_{1.5}$ after 20 hydrogen absorption/desorption cycles. Similar Al-Zr-rich and Al-Zr-poor regions can be observed as the initial alloy.

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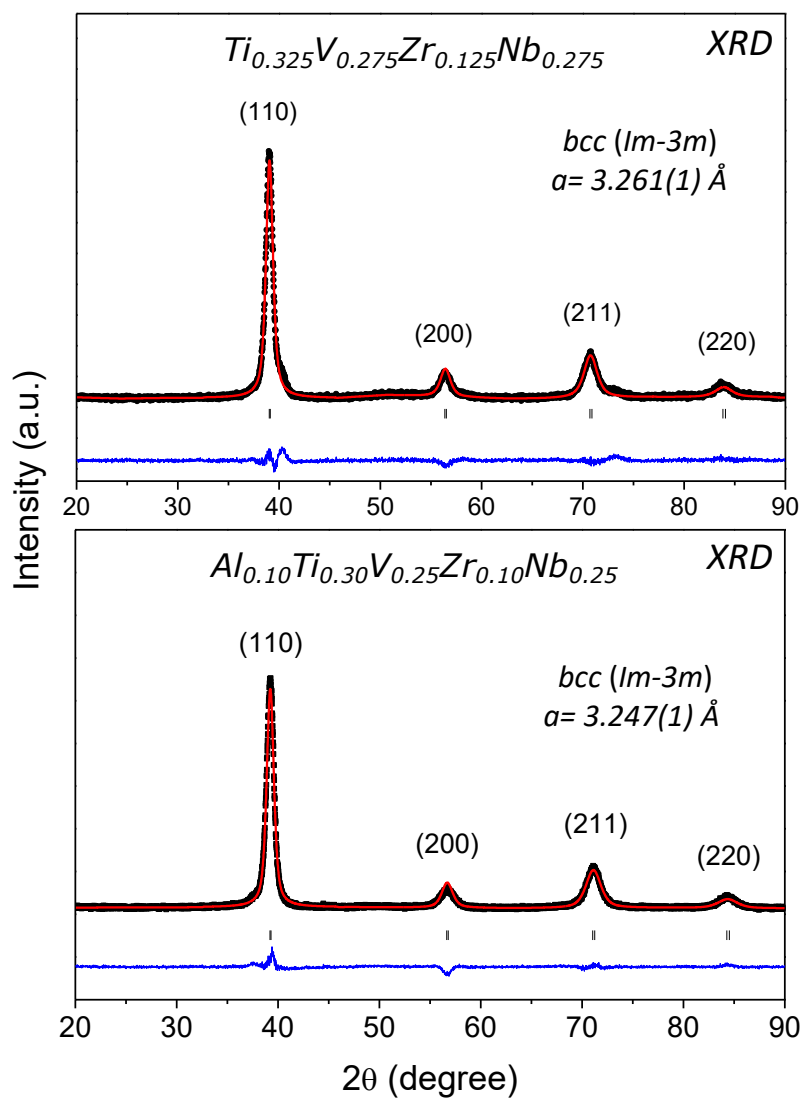


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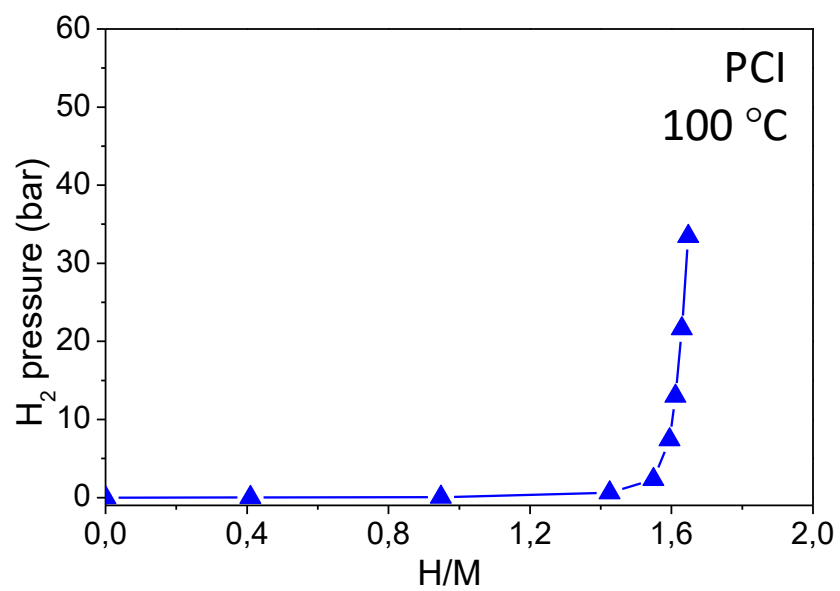


Figure S13. *In-situ* neutron diffraction of $\text{Ti}_{0.325}\text{V}_{0.275}\text{Zr}_{0.125}\text{Nb}_{0.275}\text{D}_{1.8}$ under a heating ramp of 1 °C/min while under dynamic vacuum, along with the corresponding desorption profile on the right.

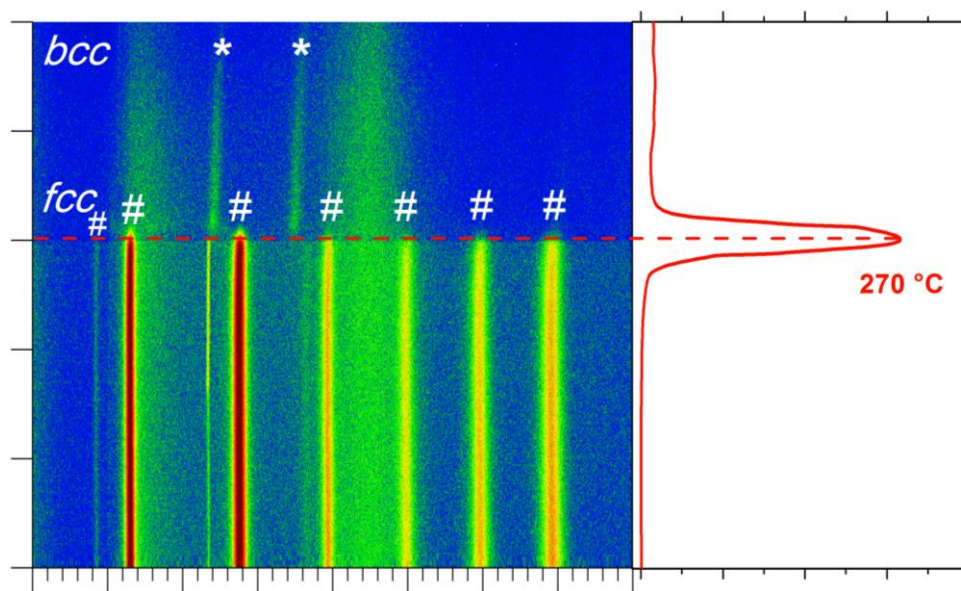


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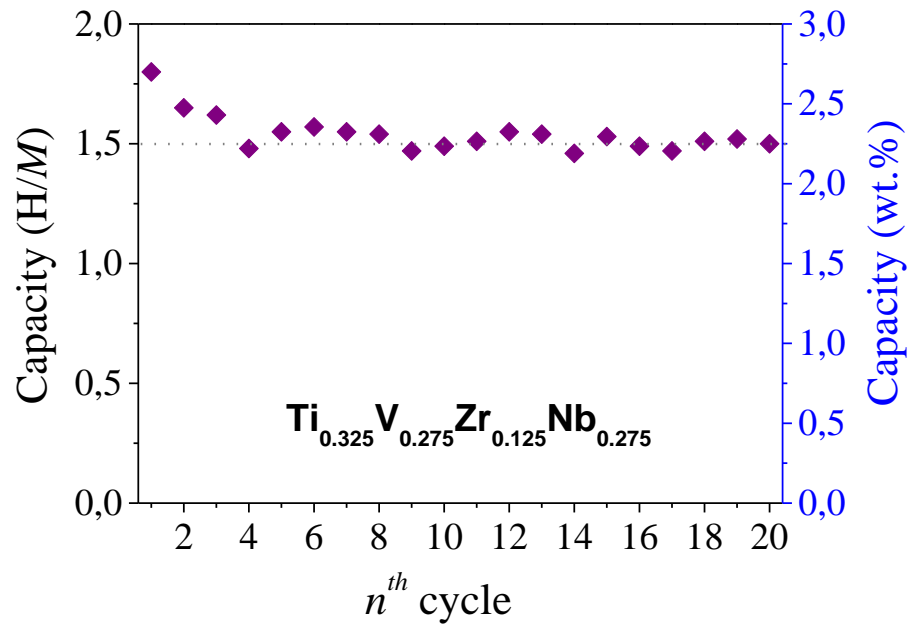


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