Supporting Information

Elucidating the effects of composition on hydrogen sorption in TiVZrNbHf-based High Entropy Alloys

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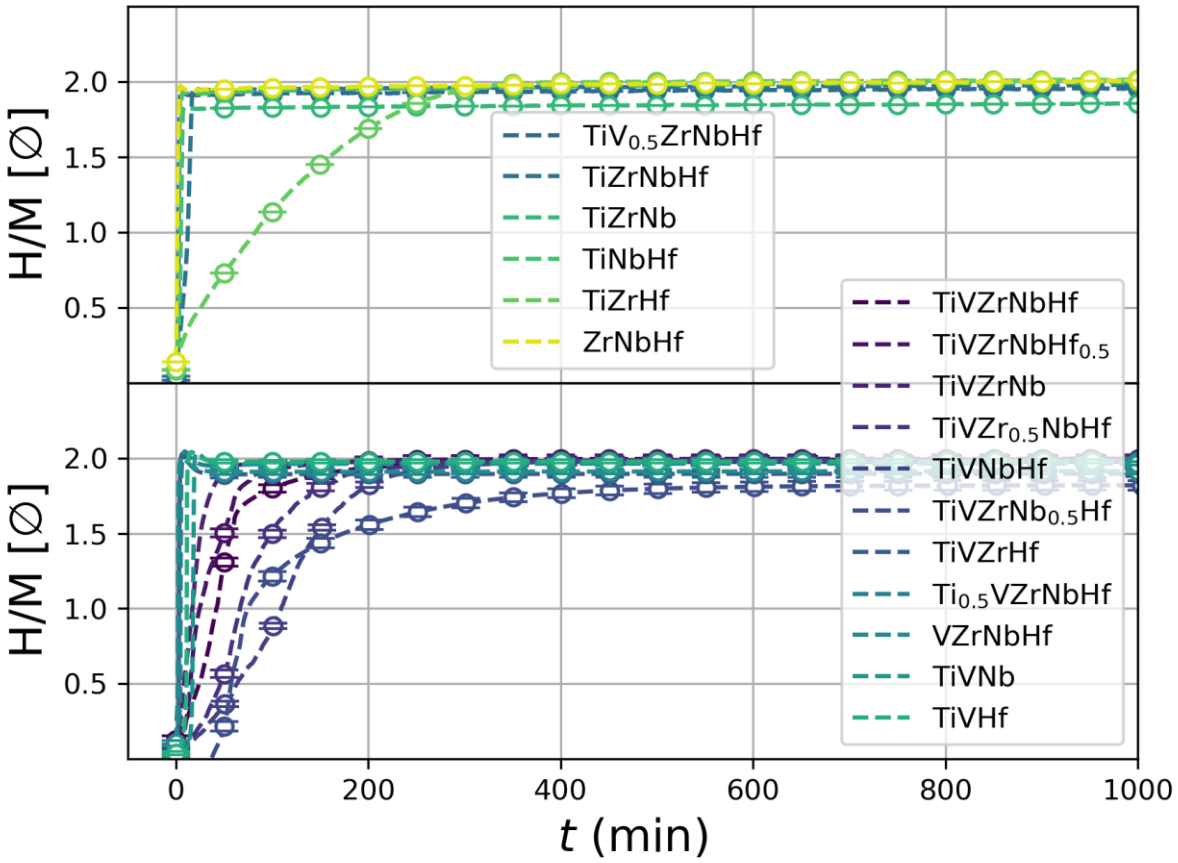

Figure S1: Hydrogen absorption kinetics after exposure to 40 bar H2 at RT (bottom) and 300°C (top).

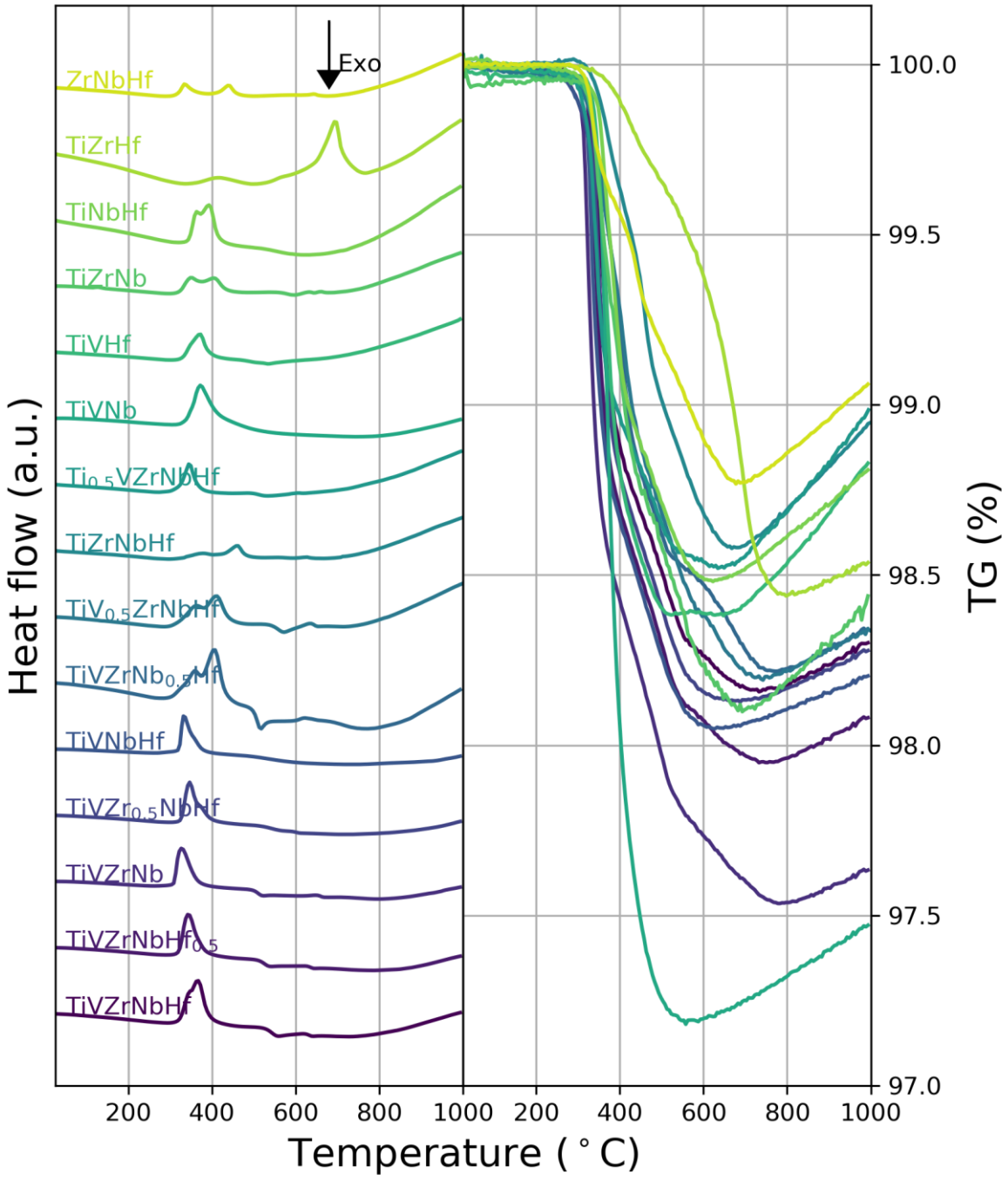

Figure S2: TG (right) and DSC (left) traces during hydrogen desorption at 10 °C/min.

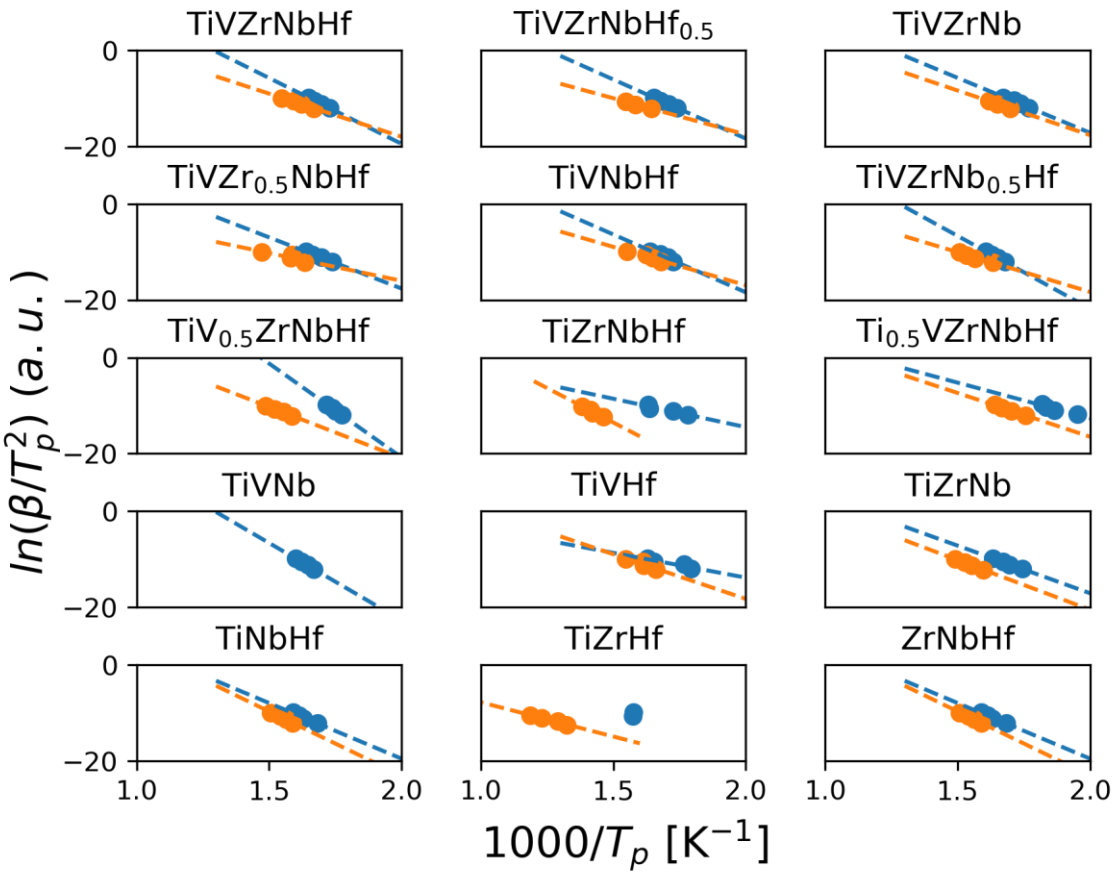

Figure S3: Kissinger analysis for the hydrogen desorption for the first (blue) and second (orange) desorption events.

Table S1: Calculated coherent neutron scattering cross sections for selected compositions using neutrons with wavelength 1.5 Å.

Composition	**Neutron scattering cross section (1/cm)**
$TiVNbD_6$	0.402
TiVNb	0.009
$TiVZrNbD_8$	0.458
TiVZrNb	0.047
$TiVZrNbHfD_{10}$	0.505
TiVZrNbHf	0.087

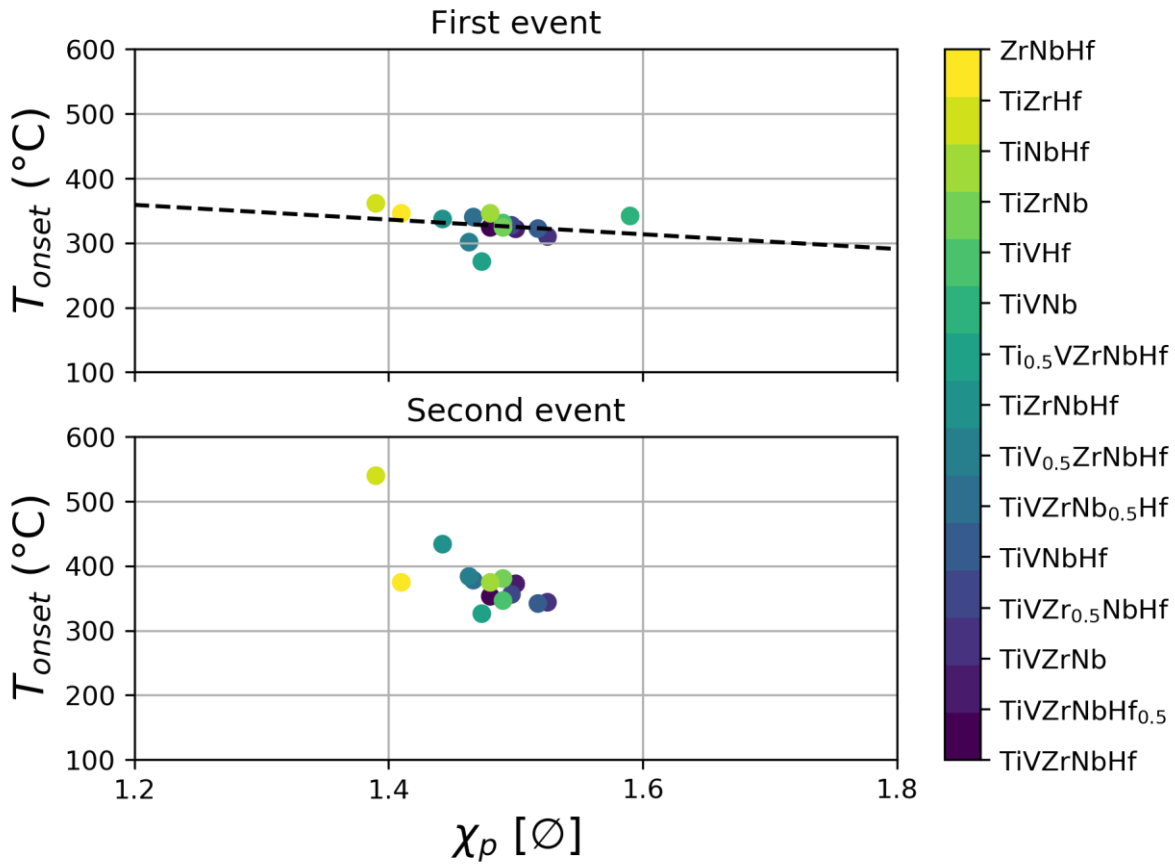

Figure S4: The onset temperature of the first (top) and second (bottom) desorption events as a function of the mean Pauling electronegativity.

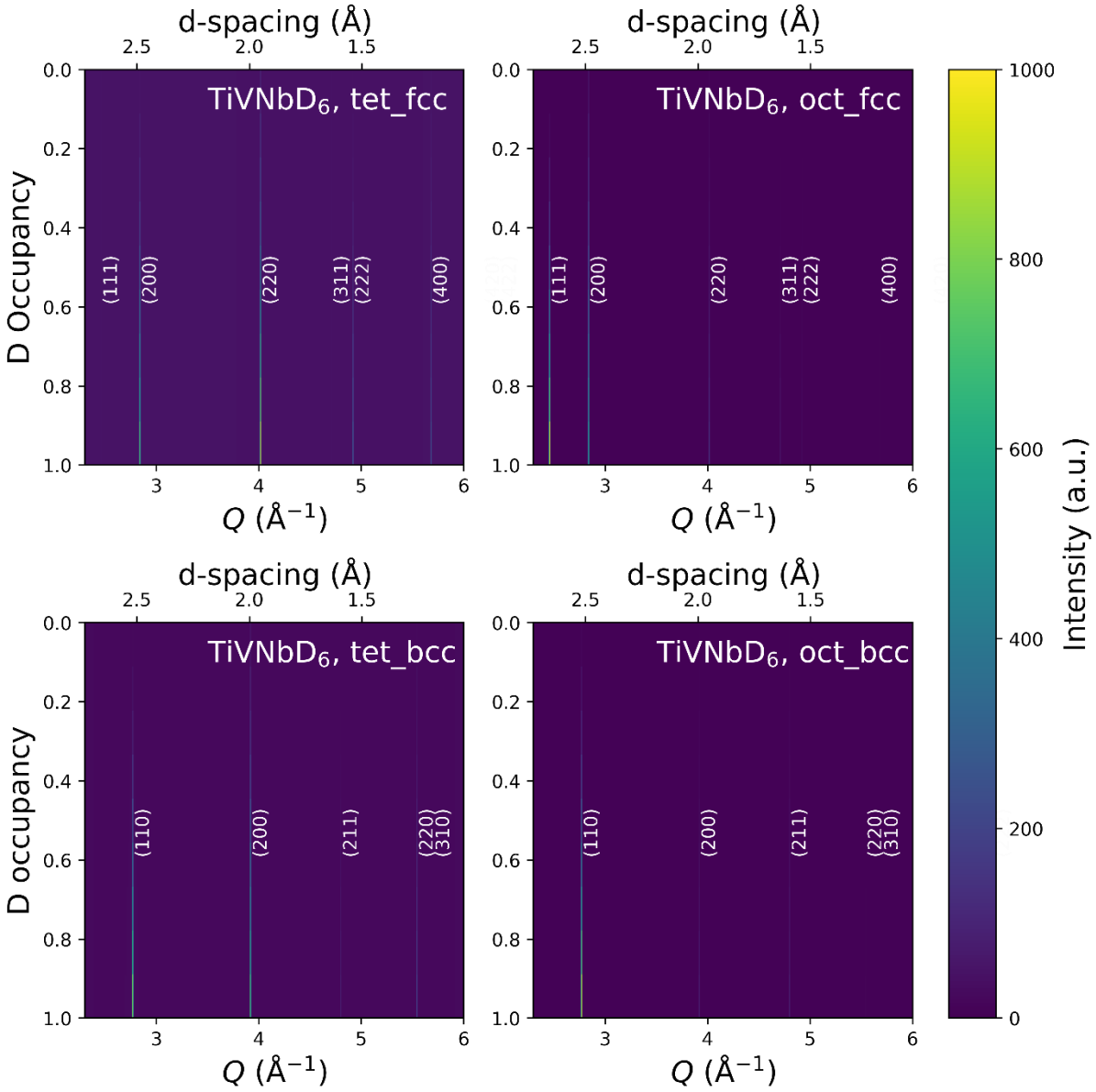

Figure S5: Simulated neutron powder diffraction patterns for $TiVNbD_6$ with different deuterium occupancy factors and constant unit cell parameter.

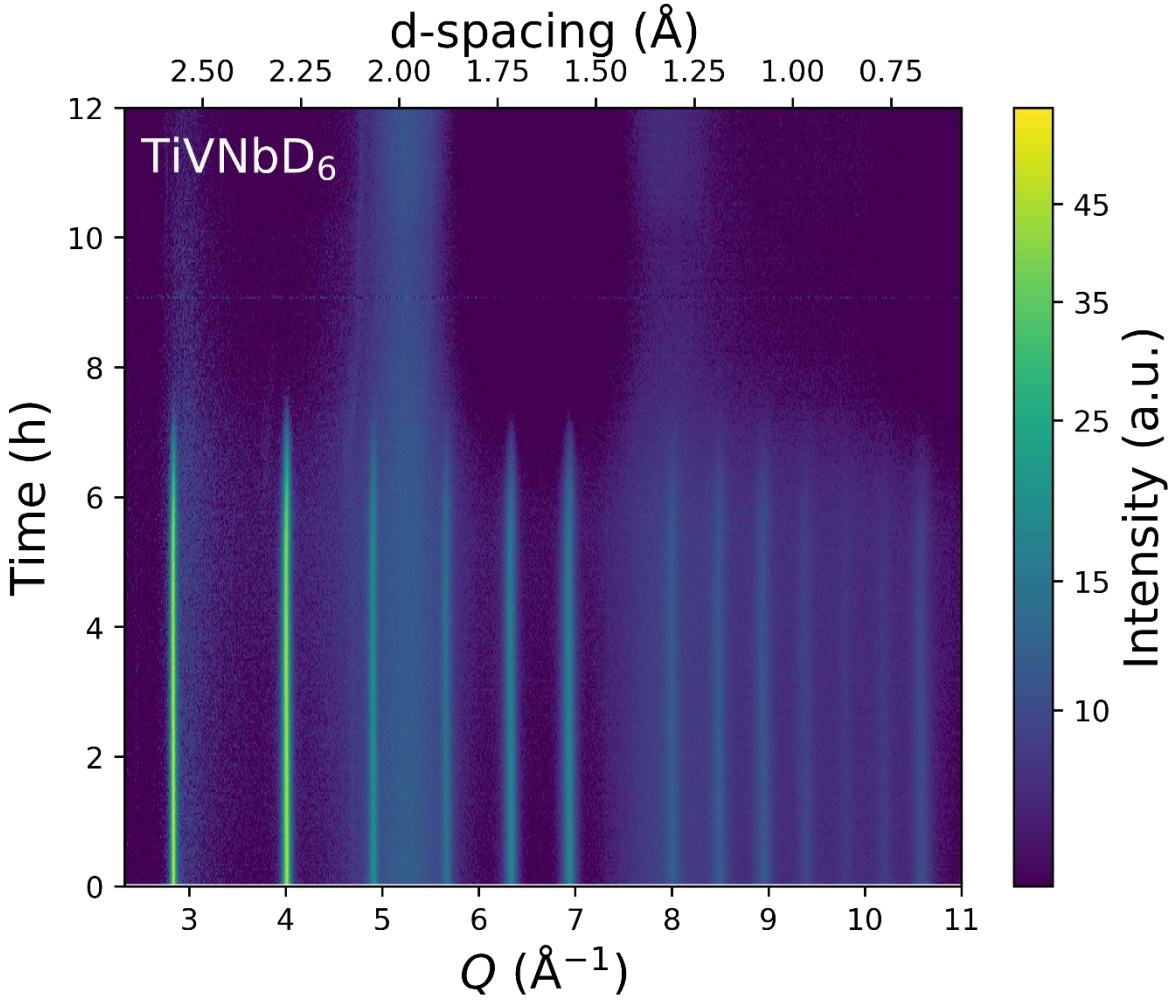

Figure S6: In-situ neutron powder diffraction from the backscattering bank of Polaris during desorption of $TiVNbD_6$.

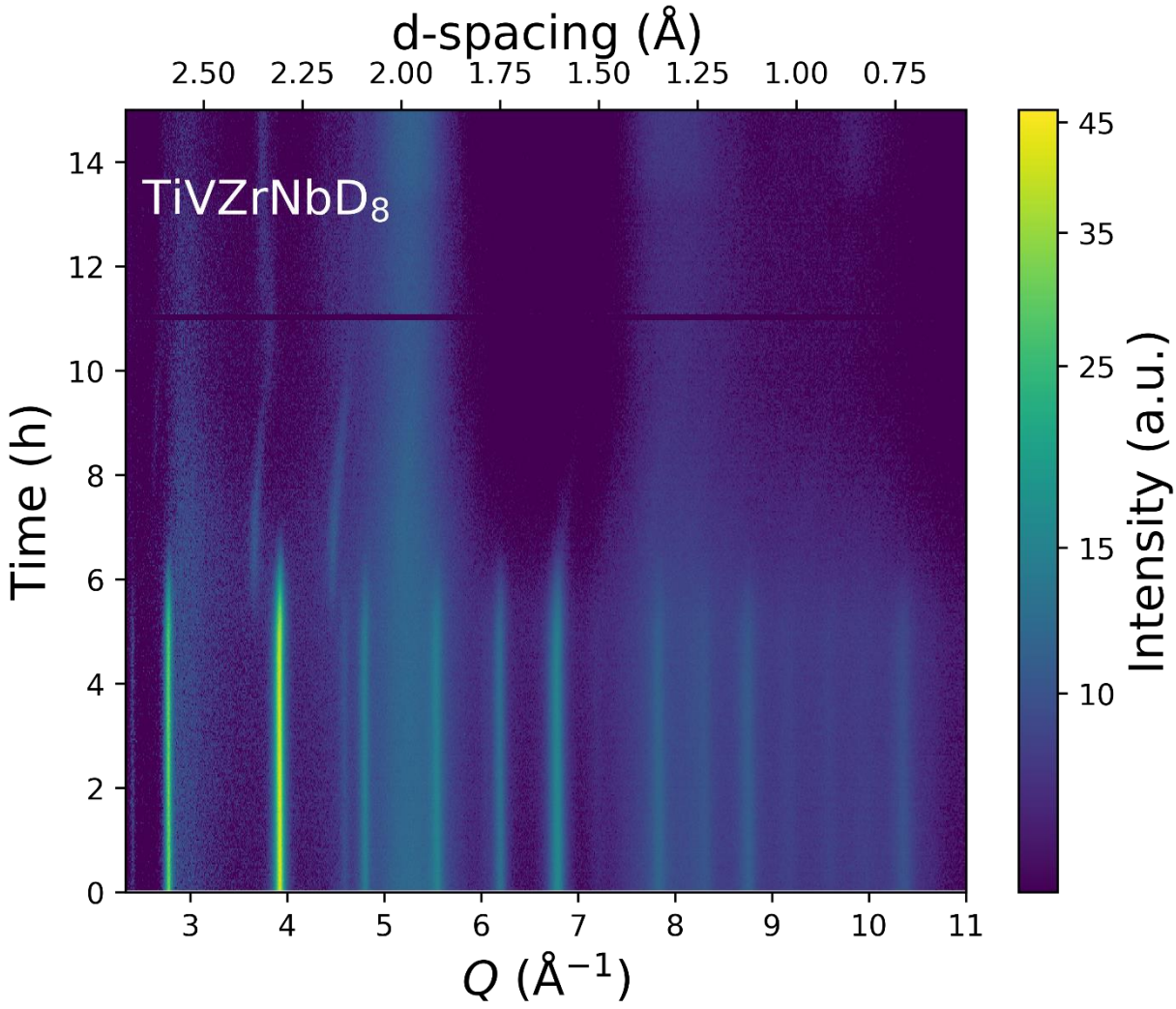

Figure S7: In-situ neutron powder diffraction from the backscattering bank of Polaris during desorption of $TiVZrNbD_8$.

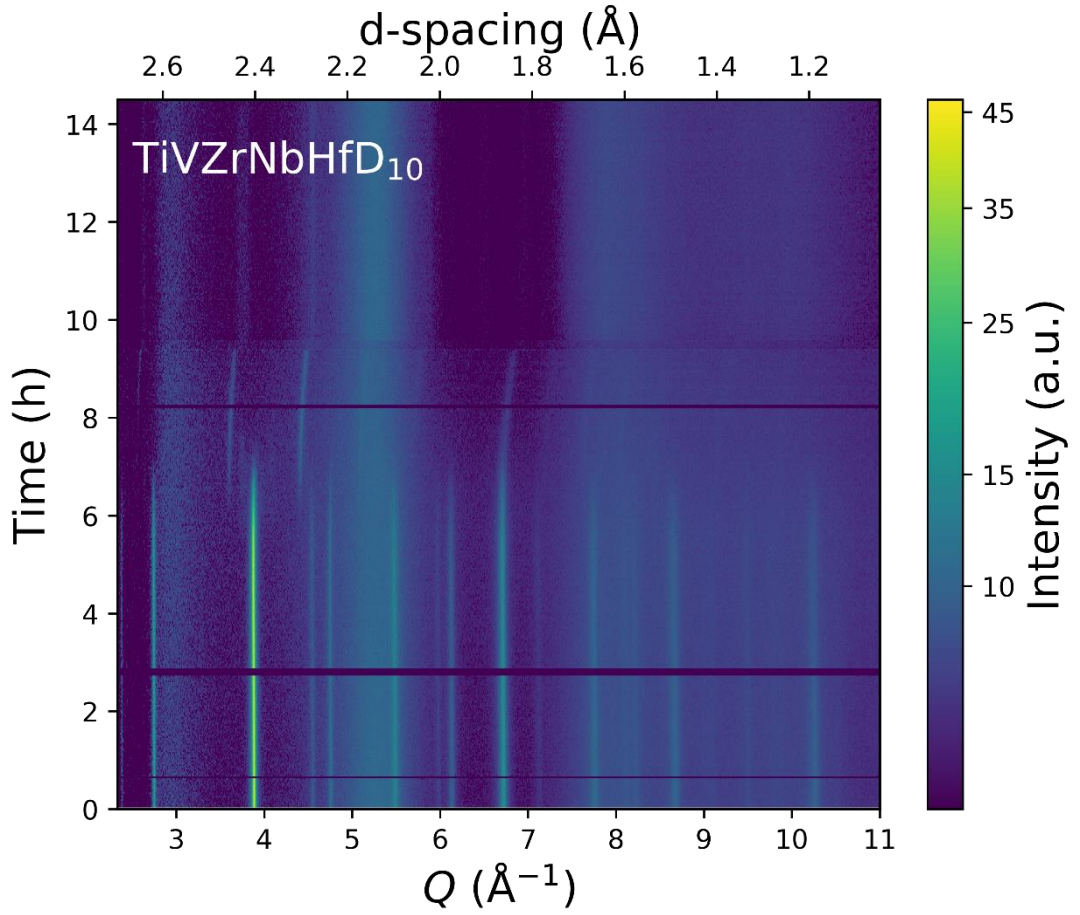

Figure S8*:* In-situ neutron powder diffraction from the backscattering bank of Polaris during desorption of $TiVZrNbHfD_{10}$.

Table S2: Crystallographic information and refinement strategies for the in-situ neutron diffraction data.

	Site	**Wyckoff**	**x**	**y**	**z**	**occupancy**	**flag**	**limits**
Fm-3m								
	Metal	4a	0	0	0	nominal	fixed	none
	D_tet	8c	0.25	0.25	0.25	0 < x < 1	refined	Total occupancy constrained by TG information
	D_oct	4b	0.5	0	0	0 < x < 1	refined	
Im-3m								
	Metal	2a	0	0	0	nominal	fixed	none
	D_tet	6b	0	0.5	0.5	0 < x < 1/3	refined	Total occupancy constrained by TG information
	D_oct	12d	0	0.5	0.25	0 < x < 1/6	refined	

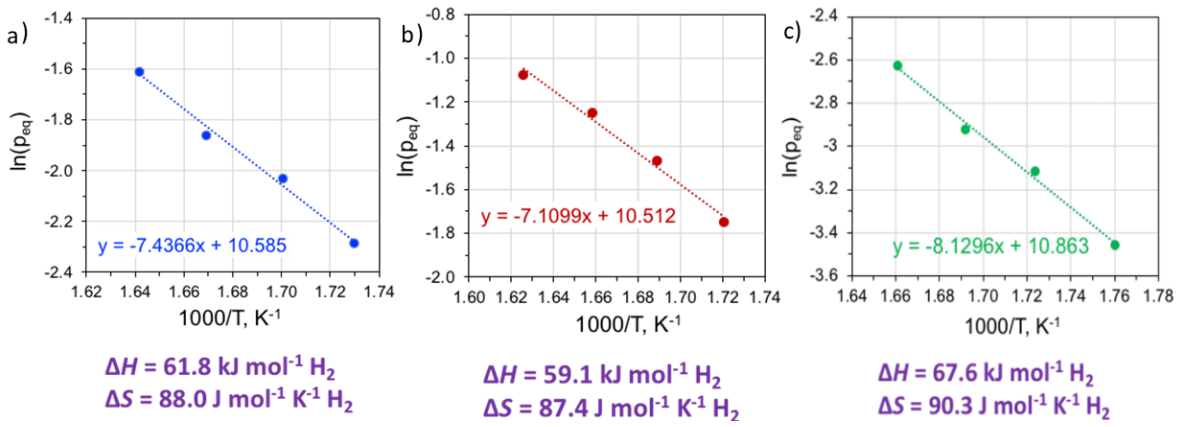

Figure S9: Van 't Hoff plot for a) TiVZrNbHf, b) $TiV_{0.5}ZrNbHf$ and c) TiVZrNb