Supplemental material to "The effects of Al on the hydrogen storage properties of V from first-principles calculations"

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lattice coordinates. V Al Η VH_2 1.085 -0.542 - $V_{0.9}Al_{0.1}H_2$ 1.067 -0.603 2.323 $V_{0.8}Al_{0.2}H_2$ 1.044 2.307 -0.651 $V_{0.7}Al_{0.3}H_2 \\$ 1.034 2.245 -0.696 1.02 2.175 $V_{0.6}Al_{0.4}H_2$ -0.745

Supplementary Table S1. The average Bader charge (|e|) of constituent elements in *unrelaxed* FCC V_{1-x}Al_xH₂ (x= 0, 0.1, 0.2, 0.3, and 0.4), i.e. with atoms located at ideal lattice coordinates.

Systems	Desorption enthalpy
Y	206.13
Sc	191.17
Zr	165.06
Hf	139.75
Ti	136.05
TiVZrNbHf	116.52
TiZrNbHfTa	115.73
$Ti_{0.325}V_{0.275}Zr_{0.125}Nb_{0.275}$	103.91
TiZrHfMoNb	99.84
$Mg_{0.10}Ti_{0.30}V_{0.25}Zr_{0.10}Nb_{0.25}$	96.19
TiVNb	90.72
TiZrVMoNb	83.17
TiVNbTa	79.2
TiVCrNb	66.5
Nb	65.17
$Al_{0.10} Ti_{0.30} V_{0.25} Zr_{0.10} Nb_{0.25}$	62.7
V	54.87
V _{0.9} Al _{0.1}	29.89
Ta	23.5
V _{0.8} Al _{0.2}	8.84
V _{0.7} Al _{0.3}	-8.71
Мо	-19.22
Cr	-19.67
V _{0.6} Al _{0.4}	-25.26
W	-93.55

Supplementary Table S2. The calculated desorption enthalpies (KJ/mol H₂) of FCC metal dihydrides mentioned in figure 9.



Supplementary Figure S1. The average -ICOHP values of metal-hydrogen bonds in $V_{1-x}Al_xH_2$. The dotted line denotes the average -ICOHP value between V-H bond in VH_2 .



Supplementary Figure S2. The fraction of H-H bonds with net antibonding interactions (i.e., negative -ICOHP values) in $V_{1-x}Al_xH_2$ (x=0, 0.1, 0.2, 0.3, and 0.4).



Supplementary Figure S3. The average number of nearest neighboring H^* surrounding V and Al in V_{1-x}Al_xH₂ (x=0, 0.1, 0.2, 0.3, and 0.4). H^{*} is the hydrogen that forms net H-H antibonding state.



Supplementary Figure S4. The distribution of absolute atomic displacement Δd_i of the V and Al metal atoms for (a) V_{1-x}Al_x and (b) V_{1-x}Al_xH₂ (x=0.1, 0.2, 0.3, and 0.4) shown as box and whisker plots. The raw data and kernel distribution estimate are shown to the right of each box. The interquartile range (IQR) is the difference between the 75th percentile and the 25th percentile, and the maximum length of whisker plotted is restricted to 1.5IQR.