Supporting Information for

Unraveling CO adsorption behaviors and its poisoning effects on ZrCo surface

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Table S1 the only one imaginary vibrational frequency for the pathways of H_2 dissociative adsorption on clean and CO pre-adsorbed ZrCo(110) surface.

transition state	imaginary frequency (cm^{-1})	vibration mode
H_2 -ZrCo (path 1)	-1088	H-H bond stretching
H_2 -ZrCo (path 2)	-281	H_2 frustrated rotation
H_2-6CO/ZrCo	-880	H-H bond stretching

Table S2 The adsorption energies E_{ads} and bond length distances $d_{H\text{-}H}$ for H_2 adsorbed on different sites of ZrCo(110) surface, with the corresponding structures shown Fig. S5.

sites	T1	T2	B1	B2	H1
E_{ads} without ZPE (eV)	-1.19	-0.24	-0.06	-1.56	-0.03
E_{ads} (eV)	-1.17	-0.14	-0.02	-1.47	0.02
$d_{H\text{-}H}$ (Å)	3.07	0.79	0.75	2.39	0.75

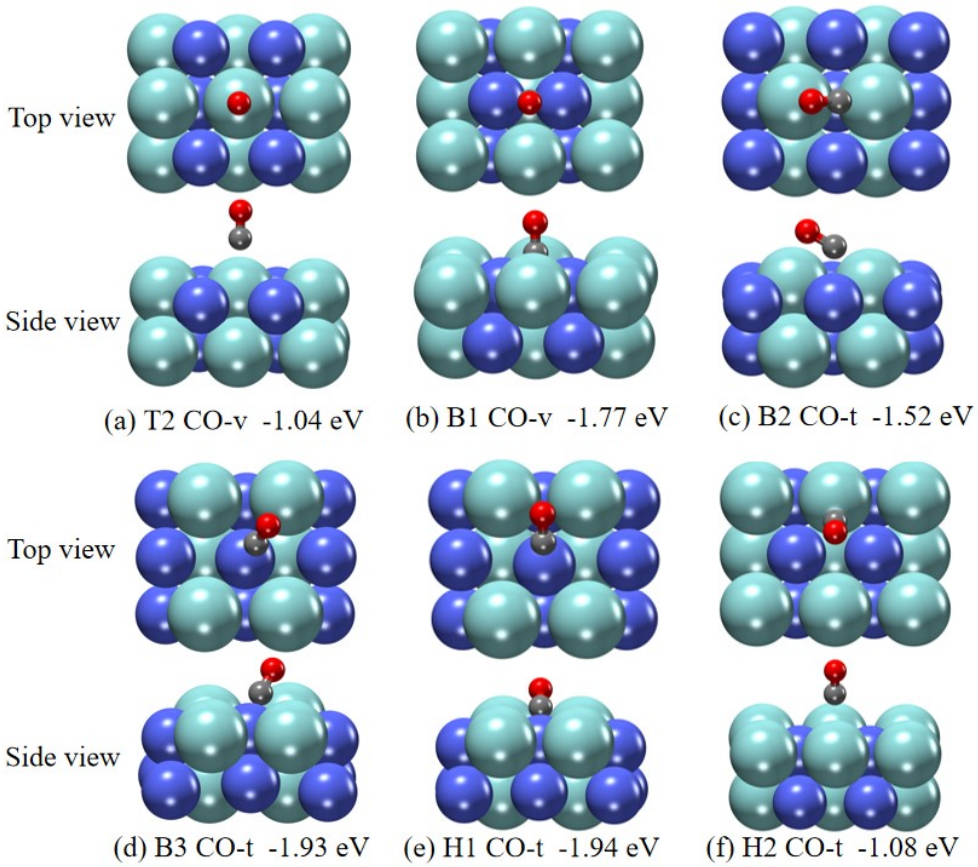

Fig. S1 The stable adsorption structures for CO-v and CO-t on different sites of ZrCo(110) surface. The Zr, Co, C, and O atoms are colored in cyan, blue, black, and red, respectively.

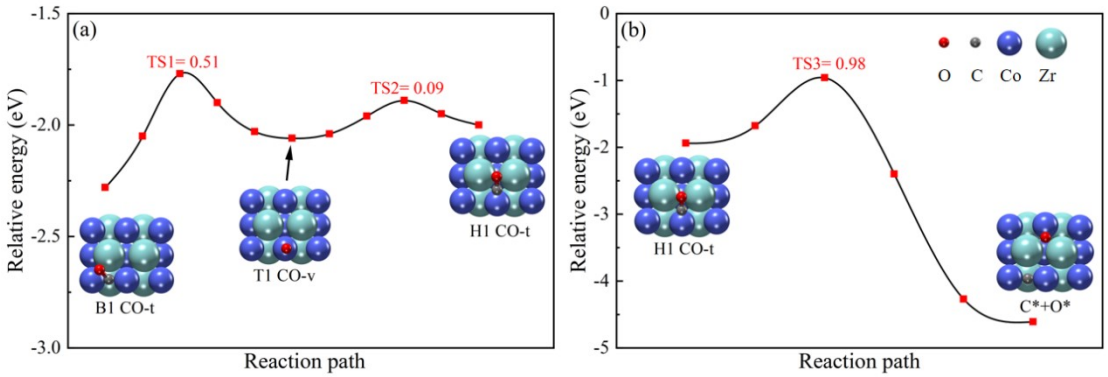

Fig. S2 (a)The diffusion of most stable CO-t on B1 site to CO-v on T1 site and then to CO-t on H1 sites. (b)The dissociation of CO-t at H1 site of the ZrCo(110) surface. The Zr, Co, C, and O atoms are colored in cyan, blue, black, and red, respectively.

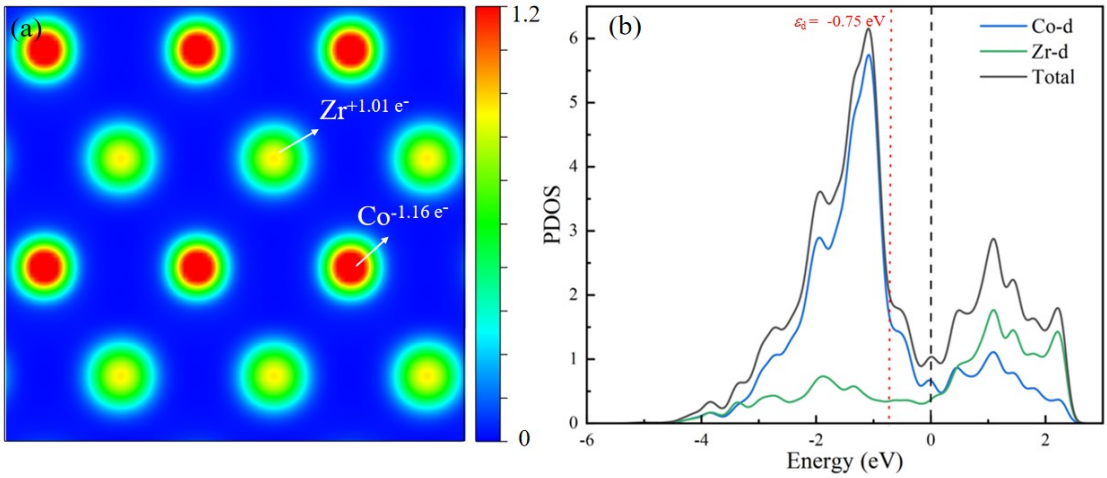

Fig. S3 The charge density and partial density of states (PDOS) of ZrCo(110) surface. The d-band center (ε_d) of surface is denoted with a red dotted line and Fermi energy is indicated by the vertical black dash line at 0 eV.

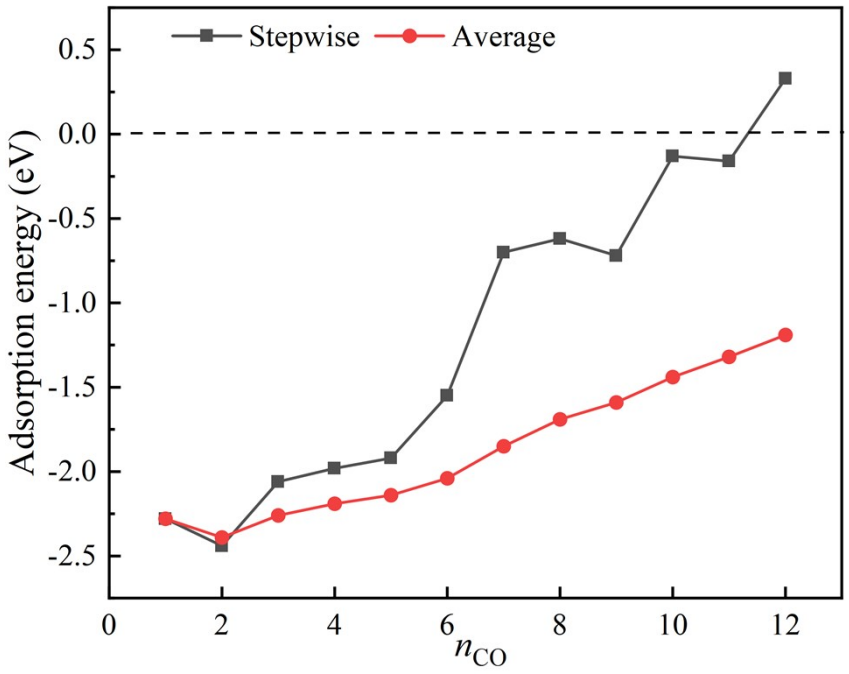

Fig. S4 The stepwise and average adsorption energies as a function of the number of CO molecules on ZrCo(110) surface.

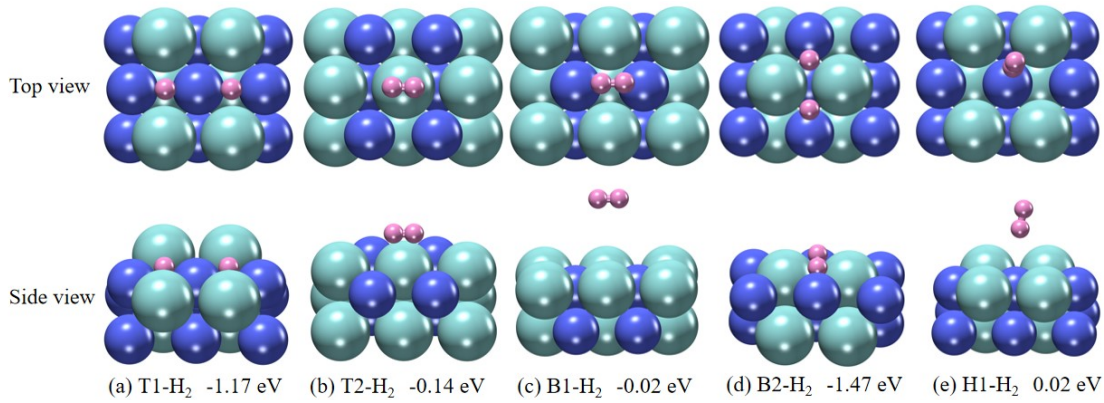

Fig. S5 The optimized structures of H_2 adsorbed on different sites of the clean ZrCo(110) surface. The Zr, Co, and H atoms are colored in cyan, blue, and pink, respectively.

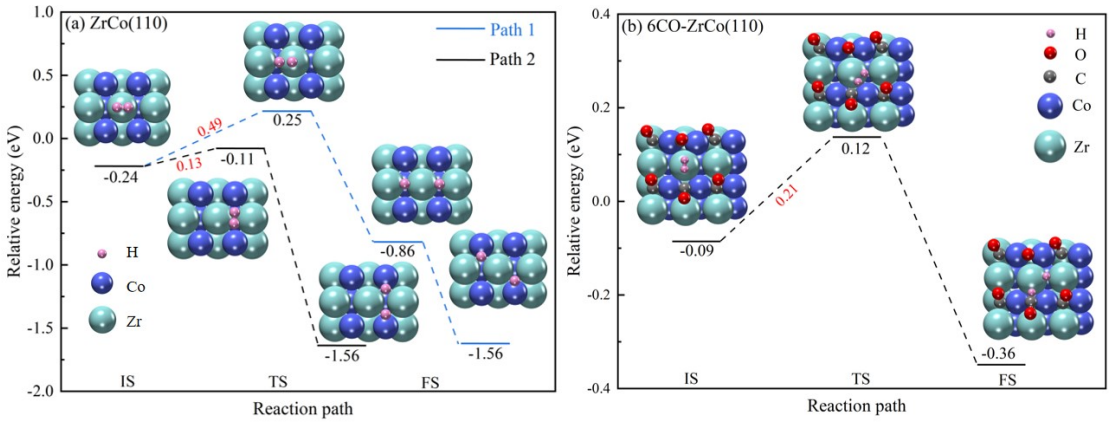

Fig. S6 The dissociation energy paths for H_2 adsorbed on T2 site of (a) the clean ZrCo(110) and (b) 6CO-ZrCo(110) surfaces. The energy values without ZPE are relative to the free gas H_2 and metal surface. The Zr, Co, C, O, and H atoms are colored in cyan, blue, black, red, and pink, respectively.

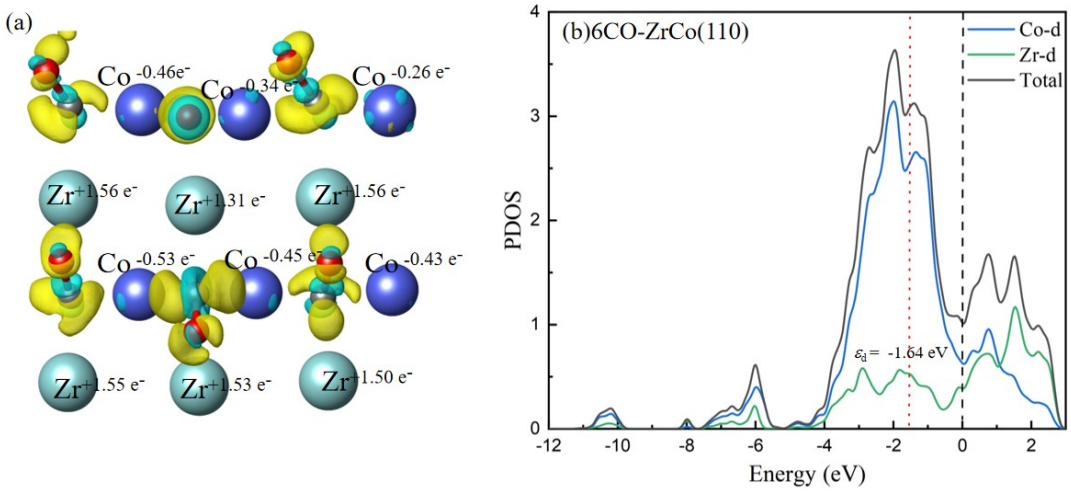

Fig. S7 (a) CDD and (b) PDOS for 6CO/ZrCo(110) surface. The iso-surface of charge density is 0.02 $e^-/bohr^3$. The yellow and light blue outlines represent electron accumulation and depletion regions, respectively. The Bader charges for each atom are also given. The d-band center of surface is denoted with a red dotted line and fermi energy is indicated by the vertical black dash line at 0 eV.

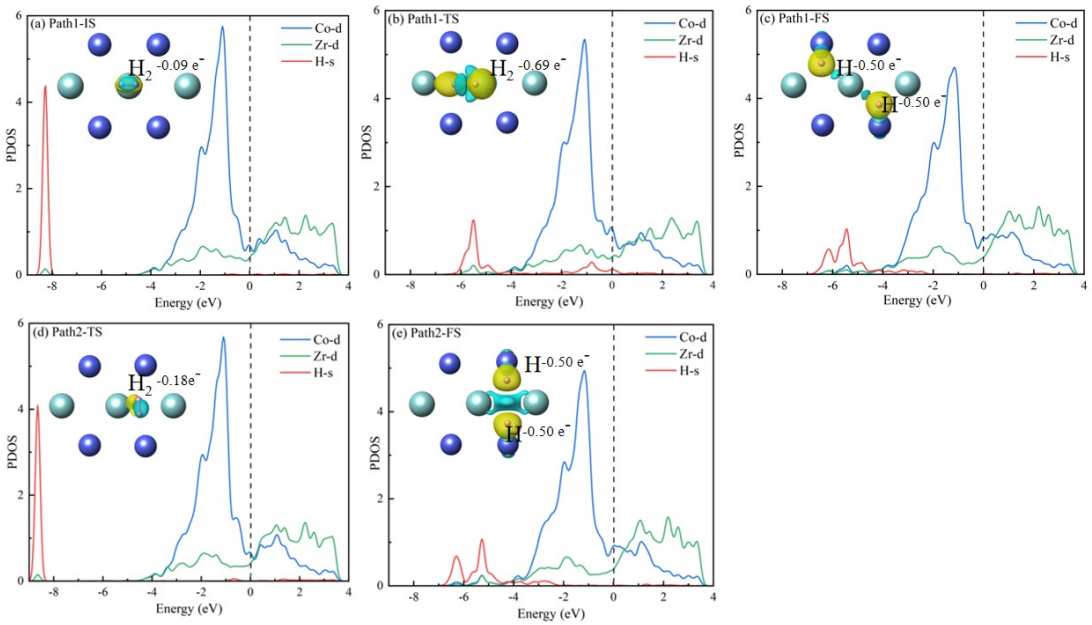

Fig. S8 PDOS for IS, TS, and FS points of two H_2 dissociation paths on the clean ZrCo(110) surface. The corresponding CDD are also plotted in inset, where the yellow and light blue outlines represent electron accumulation and depletion regions, respectively. The Zr, Co, C, and O atoms are colored in cyan, blue, black, and red, respectively.