Electronic Supplementary Material for:

FeS_2-anchored transition metal single atoms for high-efficient overall water splitting: a DFT computational screening study

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Content includes: one Table and six Figures.

Table S1 The binding energy (E_b), cohesive energy (E_c), Mulliken charge (Q), magnetic moment (M), average bond length ($d_{M\text{-}S}$) and d-band center (ε_d) of TM atom adsorption on FeS_2(100) surface.

Fig. S1 Schematic illustration of the structure model of FeS_2 surface. (a) Top view. (b) Side view. a, b, c, and d denote hollow, S_4, S_3, and Fe sites, respectively. S_3 and S_4 denote 3-fold and 4-fold coordinated sulfur atoms, respectively.

Fig. S2 The most stable structure of TM single atoms adsorption on the FeS_2(100) surface.

Fig. S3 Band-edge positions and bandgaps of M@FeS_2 catalyst.

Fig. S4 Partial density of states (PDOS) of M@FeS_2 catalysts: (a) Sc@FeS_2, (b) Ti@FeS_2, (c)

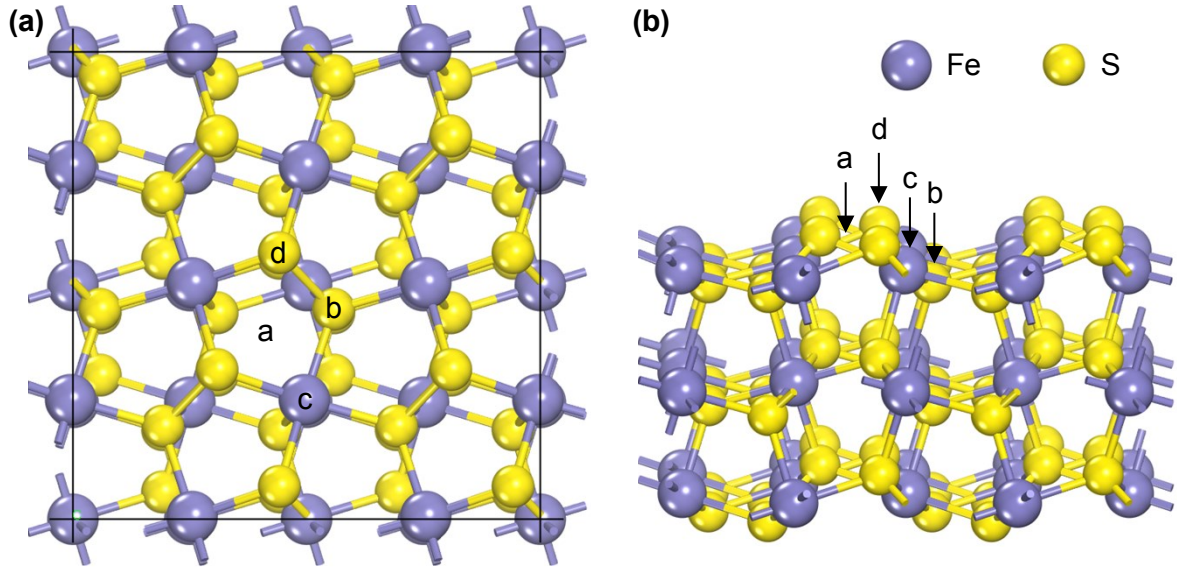
V@FeS_2, (d) Cr@FeS_2, (e) Mn@FeS_2, (f) Co@FeS_2, (g) Ni@FeS_2, (h) Cu@FeS_2, and (i) Zn@FeS_2. The dashed lines denote the Fermi level.

Fig. S5 Potential energy evolution of M@FeS_2 catalysts at 500 K in the water-phase environment. (a) Sc@FeS_2, (b) Ti@FeS_2, (c) V@FeS_2, (d) Cr@FeS_2, (e) Mn@FeS_2, (f) Fe@FeS_2, (g) Co@FeS_2, (h) Ni@FeS_2, (i) Cu@FeS_2, and (j) Zn@FeS_2.

Fig. S6 Gibbs free energy diagram of the HER over S atom of M@FeS_2 catalysts.

Table S1 The binding energy (E_b), cohesive energy (E_c), Mulliken charge (Q), magnetic moment (M), average bond length ($d_{M\text{-}S}$) and d-band center (ε_d) of TM atom adsorption on $FeS_2(100)$ surface.

TM atoms	E_b (eV)	E_c (eV)	Q (e)	M (μ_B)	$d_{M\text{-}S}$ (Å)	ε_d (eV)
Sc	−4.93	−4.16	0.71	−0.016	2.504	−2.13
Ti	−5.54	−5.91	0.35	0.000	2.275	−2.13
V	−5.79	−7.11	0.09	0.000	2.165	−1.77
Cr	−5.57	−7.44	−0.11	2.976	2.107	−1.47
Mn	−5.15	−6.77	−0.22	0.000	2.085	−1.64
Fe	−4.61	−6.30	0.32	1.701	2.072	−1.59
Co	−4.22	−5.68	−0.32	0.000	2.084	−1.88
Ni	−3.34	−4.68	−0.03	−0.026	2.141	−1.72
Cu	−1.96	−3.53	0.10	0.002	2.327	−2.36
Zn	−0.78	−0.46	0.31	0.001	2.505	−6.21



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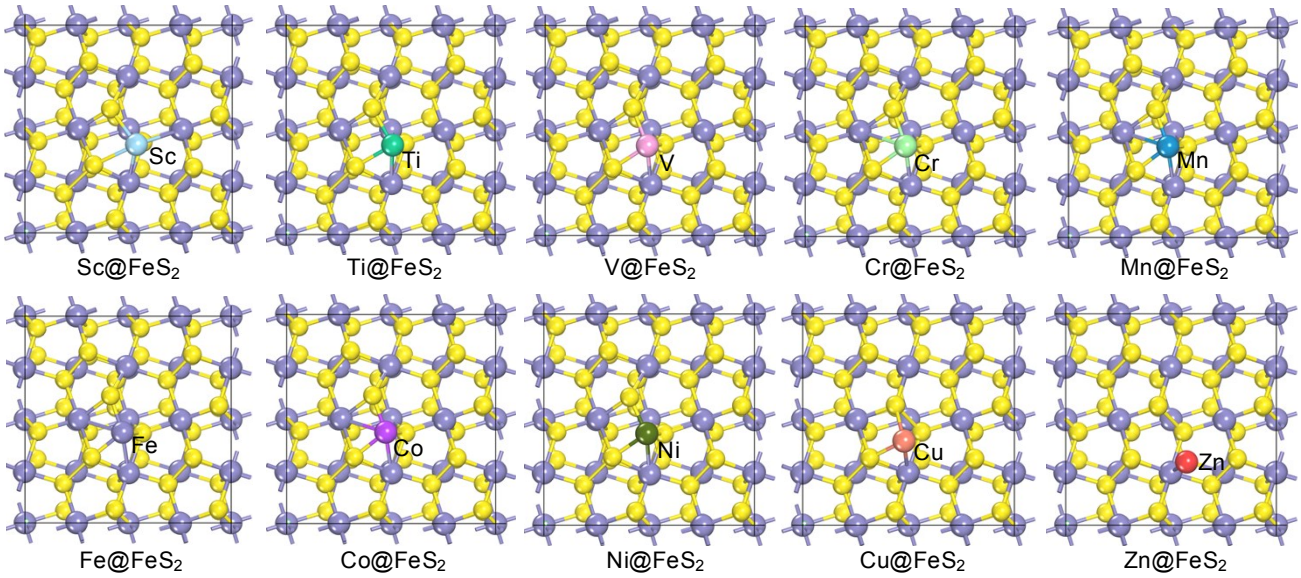
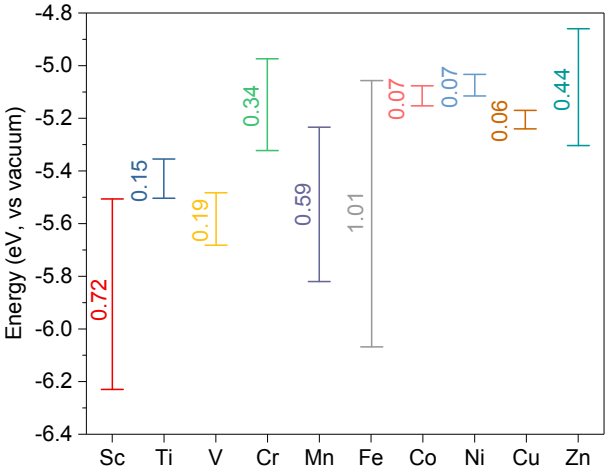


Fig. S2 The most stable structure of TM single atoms adsorption on the FeS_2(100) surface.



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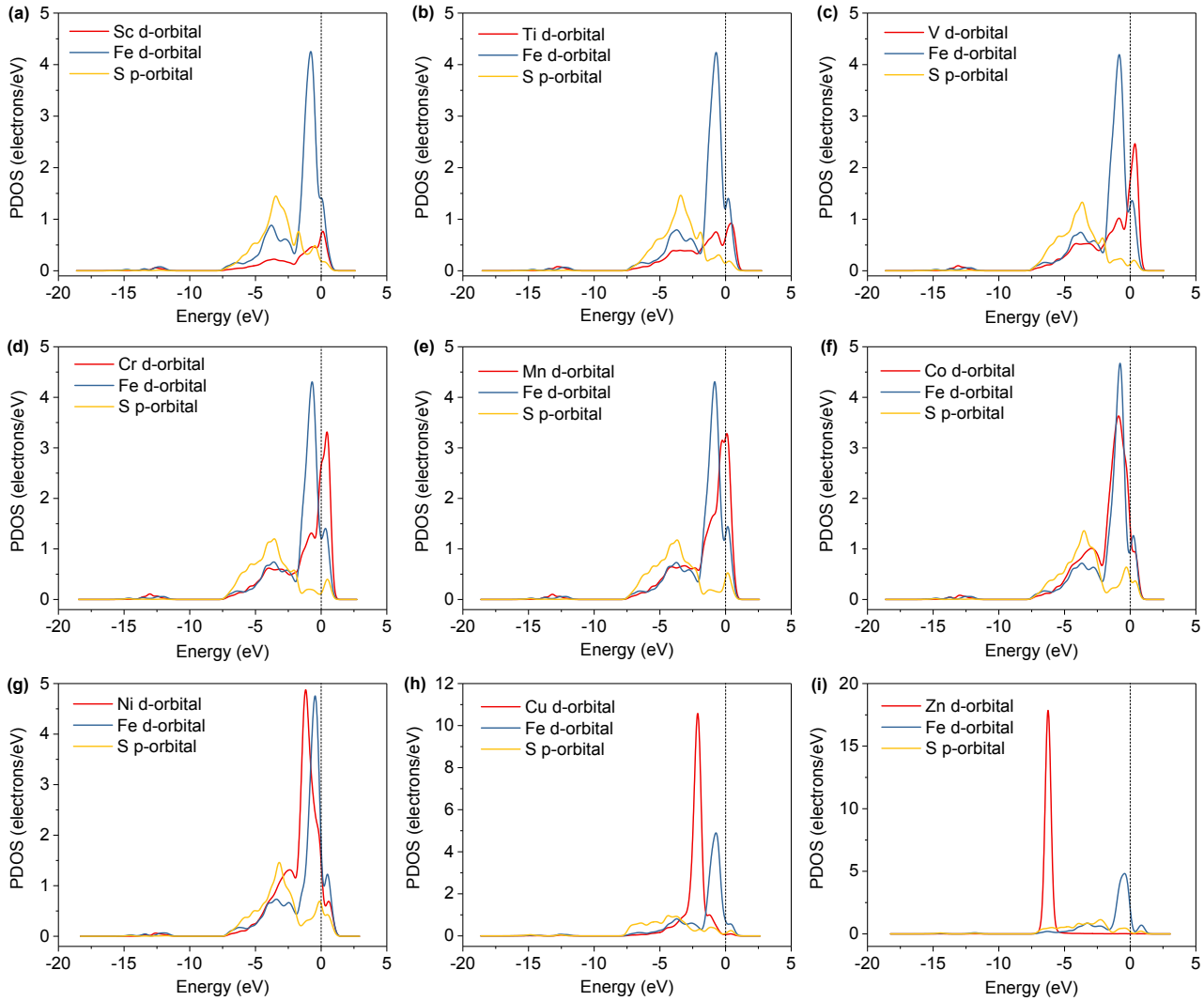

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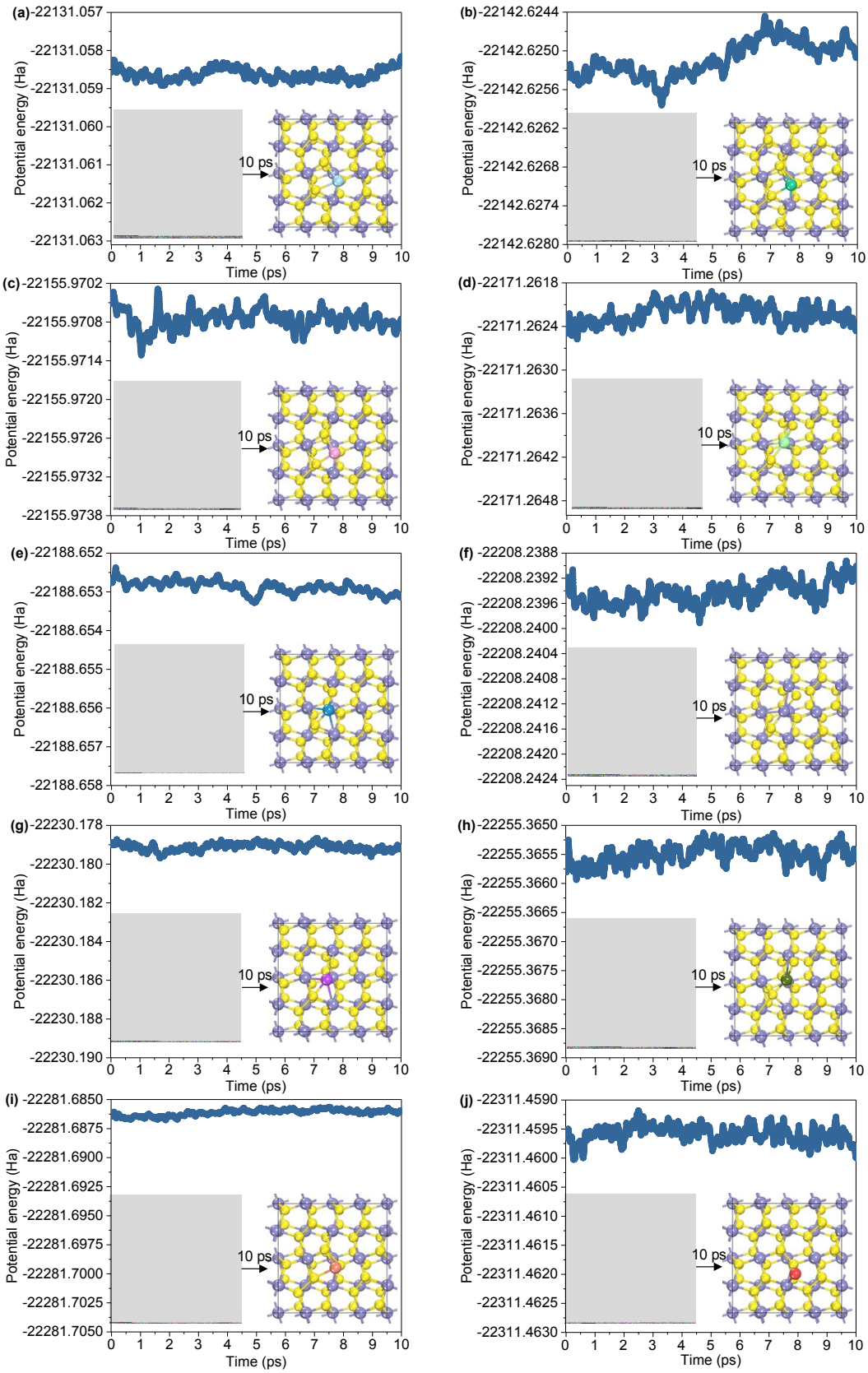

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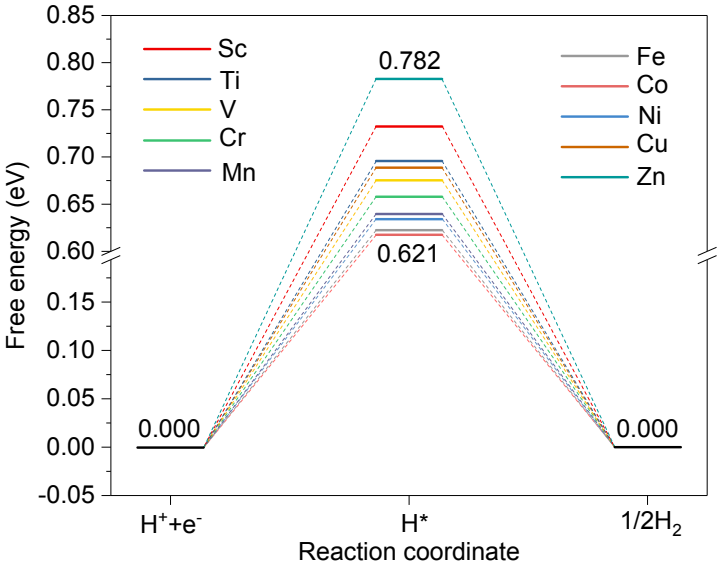

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