

Supplementary materials

1T-MoS₂ Coordinated Bimetal Atoms as Active Centers to Facilitate Hydrogen Generation

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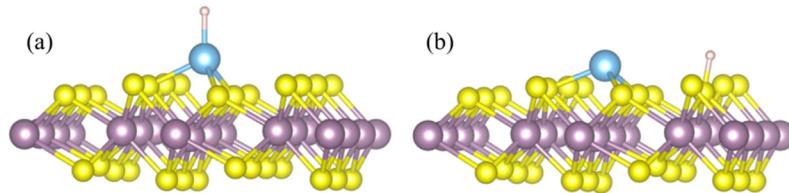


Figure S1. Two different adsorption sites of H atom on the Ti@1T-MoS₂ surface. (a) H adsorption on the Ti atom; (b) H adsorption on the S atom.

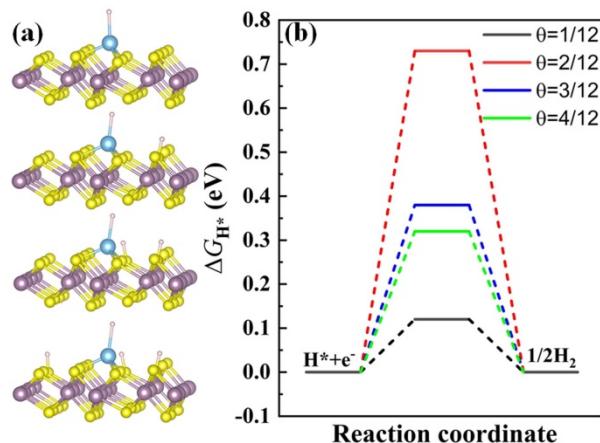


Figure S2. (a) Side views of the lowest energy structure and (b) corresponding Gibbs free energy of H* adsorption (ΔG_{H^*}) at different H coverage on the Ti@1T-MoS₂ surface.

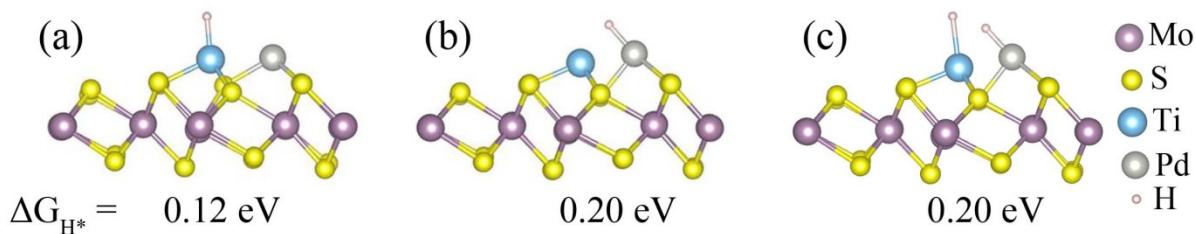


Figure S3. One H atom adsorption on (a) the Ti and (b) Pd site of PdTi@1T-MoS₂ catalyst, where the H atom is inclined to be directly bonded to the Ti atom with lower H* adsorption free energy ΔG_{H^*} . (c) The second H atom adsorption on the Pd site.

Table S1. The elastic constants C_{11} , C_{22} , C_{12} , C_{66} (N/m) used to evaluate whether the TM@1T-MoS₂ systems meet the mechanical stability criteria, namely, $C_{11} > 0$, $C_{66} > 0$ and $C_{11} \times C_{22} > C_{12} \times C_{12}$.

System	C_{11}	C_{22}	C_{12}	C_{66}	Meeting Criteria
graphene	357.0	357.0	63.0	147.0	yes
Graphene [1]	352.0	352.0	62.6	144.7	yes
2H-MoS ₂	135.9	135.9	33.3	51.3	yes
2H-MoS ₂ [2]	128.4	128.4	32.6	47.9	yes
1T-MoS ₂	96.5	96.5	-7.5	52.0	yes
Ti@1T-MoS ₂	86.7	63.3	46.8	9.1	yes
V@1T-MoS ₂	78.2	94.0	21.8	1.4	yes
Co@1T-MoS ₂	114.5	102.5	14.4	9.6	yes
Ni@1T-MoS ₂	116.1	108.1	13.7	10.2	yes
Cu@1T-MoS ₂	119.0	127.8	16.1	10.4	yes
Zr@1T-MoS ₂	108.3	-80.7	112.3	9.1	no
Nb@1T-MoS ₂	77.7	96.1	25.1	-3.4	no
Rh@1T-MoS ₂	114.8	106.2	14.9	9.3	yes
Pd@1T-MoS ₂	111.6	117.8	16.5	10.0	yes
Ag@1T-MoS ₂	116.7	126.4	17.2	10.2	yes
Hf@1T-MoS ₂	95.2	88.3	32.5	8.1	yes

Table S2. The total energy E_{tot} and free energy ΔG_{H^*} of H* adsorption on site (a) and (b) shown in Figure S1, as well as the overpotential η_{HER} (V). The bold font marks the more negative E_{tot} , corresponding to the energetically most favorable configurations of H* adsorption.

System	E_{tot} (eV)		ΔG_{H^*} (eV)		η_{HER} (V)
	site-a	site-b	site-a	site-b	
Ti@1T-MoS ₂	-273.28	-272.10	0.12	1.31	0.12
V@1T-MoS ₂	-273.53	-272.45	0.13	1.21	0.13
Co@1T-MoS ₂	-270.74	-270.76	0.44	0.41	0.41
Ni@1T-MoS ₂	-269.45	-269.31	0.58	0.72	0.58
Cu@1T-MoS ₂	-267.51	-267.55	0.81	0.77	0.77
Rh@1T-MoS ₂	-270.86	-270.55	0.42	0.73	0.42
Pd@1T-MoS ₂	-269.06	-269.23	0.53	0.36	0.36
Pd@1T-MoS ₂ [3]	-	-	0.49	0.35	0.35
Ag@1T-MoS ₂	-266.36	-266.66	0.92	0.62	0.62
Hf@1T-MoS ₂	-276.15	-274.26	-0.46	1.42	0.46

Table S3. Total energy E_{tot} and free energy ΔG_{H^*} of H* adsorbed on different sites shown in Figure 5b for bimetal atoms immobilized on the 1T-MoS₂ surface. The bold font marks the energetically most favorable configurations of H* adsorption.

System	E_{tot} (eV)				ΔG_{H^*} (eV)	
	I	II	III	IV	I	II
TiTi@1T-MoS ₂	-281.09	-281.06	-279.42	-279.38	-0.62	-0.59
VTi@1T-MoS ₂	-280.99	-280.46	-280.19	-279.79	0.00	0.53
NiTi@1T-MoS ₂	-277.80	-277.57	-277.41	-276.98	0.19	0.43
CuTi@1T-MoS ₂	-276.36	-275.73	-275.36	-275.13	0.01	0.64
PdTi@1T-MoS ₂	-277.33	-277.26	-277.05	-276.54	0.12	0.20

Table S4. H adsorption Gibbs free energy ΔG_{H^*} of $\text{TM}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$ ($\text{TM} = \text{Ti}, \text{V}, \text{Ni}, \text{Cu}, \text{Pd}$ and Pt) at H coverage $\theta = 1/12$ (one H adsorption) and $2/12$ (two H adsorption), where Ti atom was coordinated with three S atoms (site-S) and the other TM was deposited into the S vacancy (site-Sv).

System	$\Delta G_{\text{H}^*} (\theta = 1/12)$		$\Delta G_{\text{H}^*} (\theta = 2/12)$
	site-S	site-Sv	
$\text{Ti}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$	0.08	0.45	0.63
$\text{V}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$	0.18	0.15	0.34
$\text{Ni}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$	0.64	-0.36	0.40
$\text{Cu}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$	0.24	-0.07	0.36
$\text{Pd}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$	0.35	0.09	-0.05
$\text{Pt}_{\text{Sv}}\text{Ti}@1\text{T-MoS}_2$	0.37	0.28	0.05

References

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