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Supplementary Information

Enhancement of single-atom catalytic activity by interlayer charge transfer in the electride-based heterostructures

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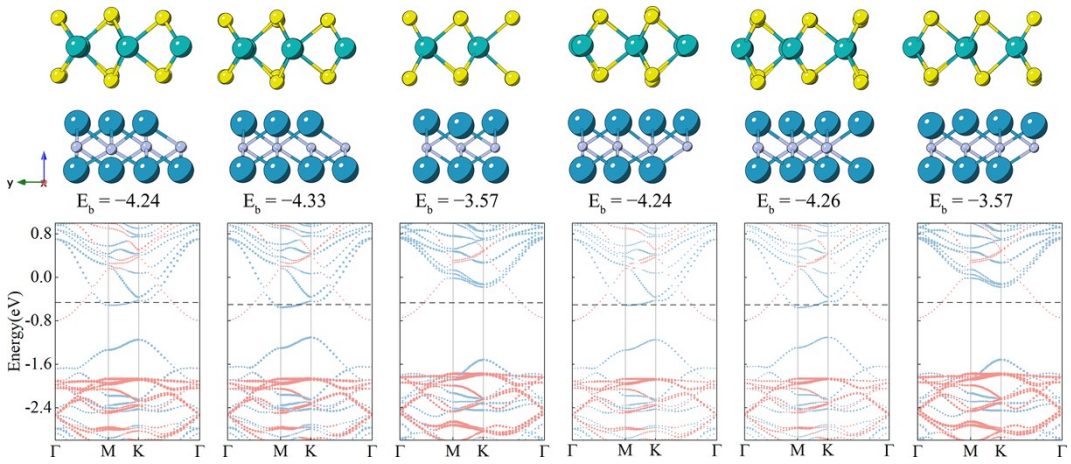

Fig. S1 The MoS_2/Ca_2N heterostructures and their band structures. The red and blue bubbles depict the projected band structure of MoS_2 and Ca_2N.

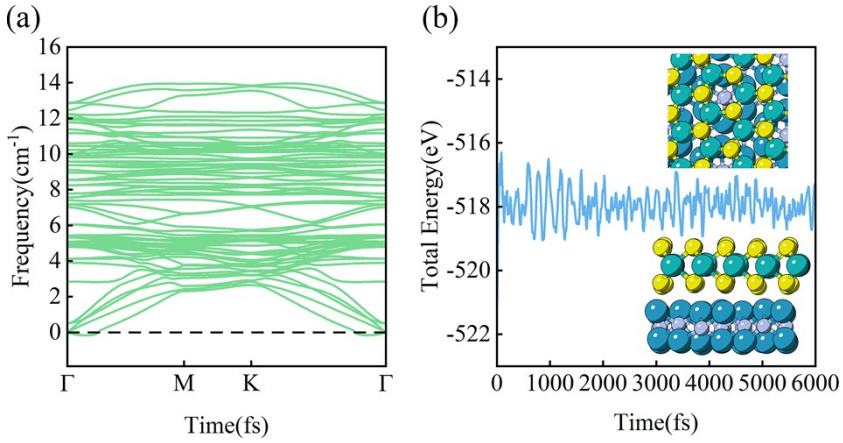

Fig. S2 (a) Phonopy dispersions of MoS_2/Ca_2N heterostructure and (b) the fluctuations of energy and the final configuration of MoS_2/Ca_2N heterostructure.

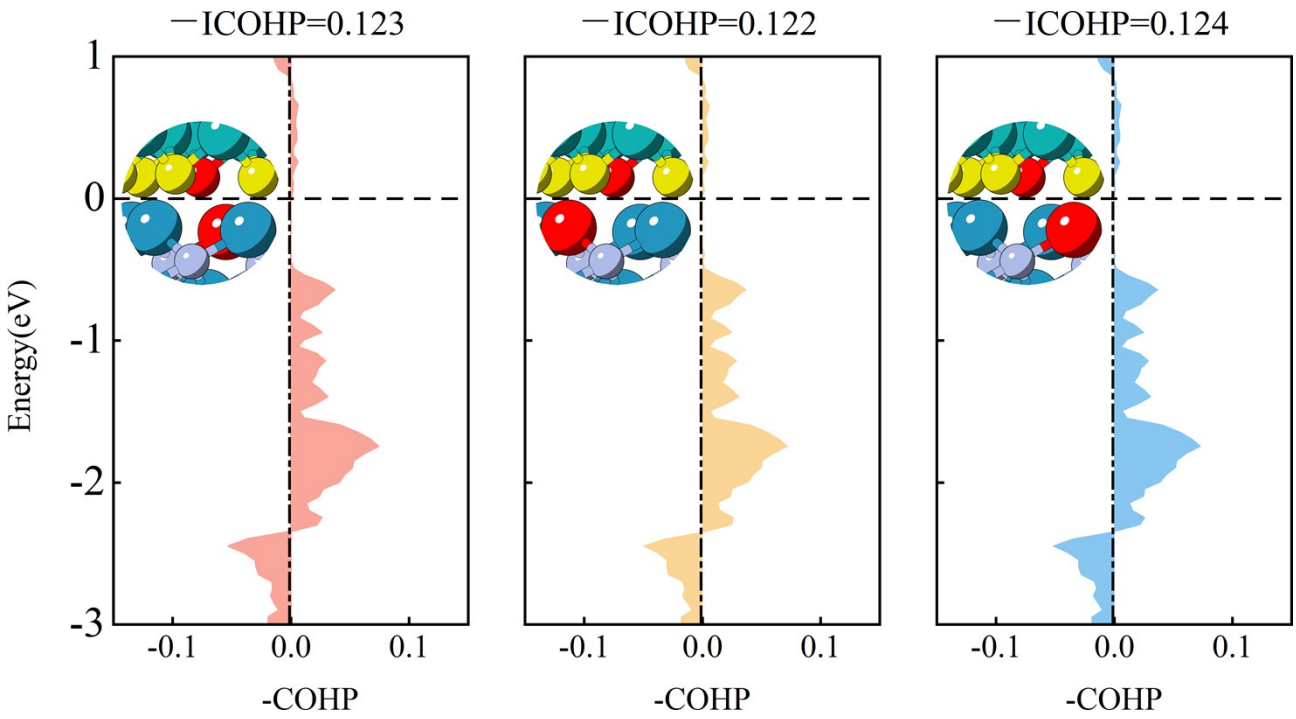

Fig. S3 The –COHP between S atom at the interface of MoS_2/Ca_2N heterojunction and the three closest Ca atoms is depicted in red.

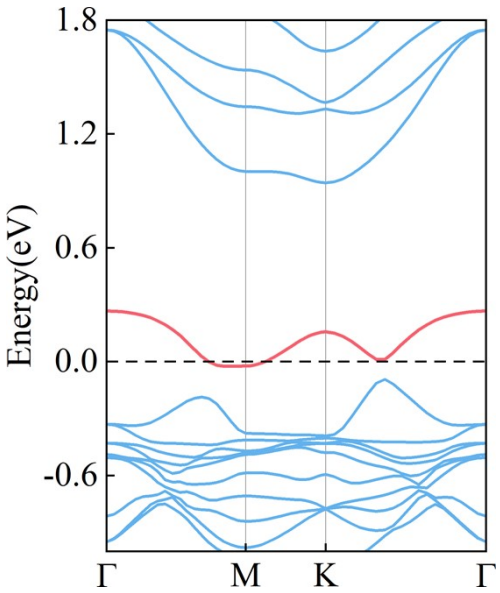

Fig. S4 The band structure of the MoS_2/Ca_2N heterostructure. The red line indicates electron layer related bands at the interface of MoS_2/Ca_2N heterostructure.

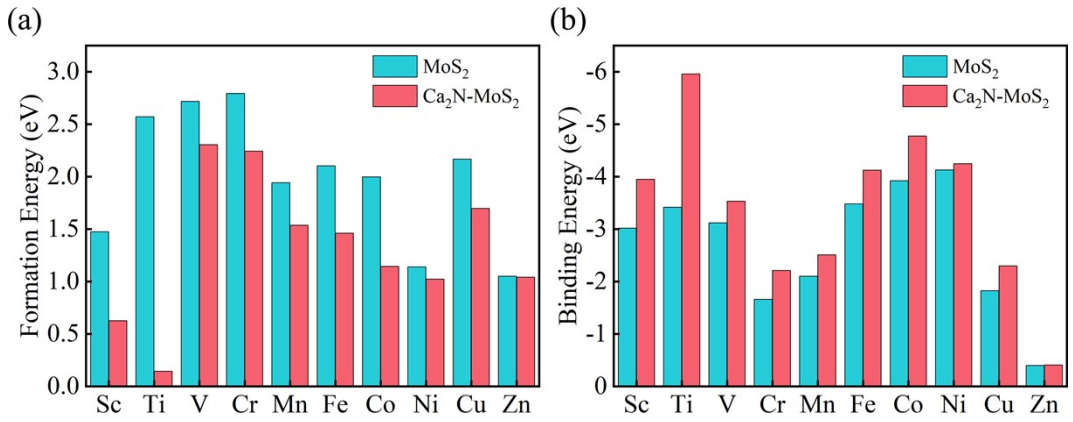

Fig. S5 (a) The formation energy and (b) binding energy of TM atoms in the catalyst.

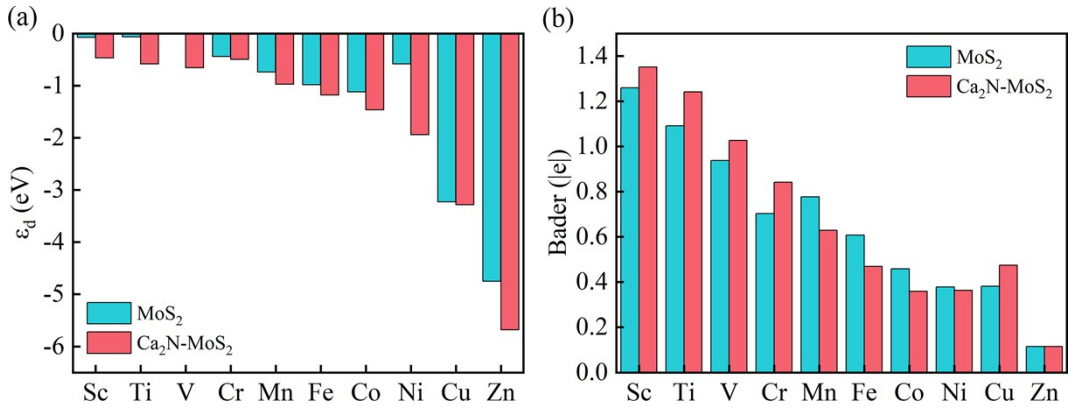

Fig. S6 (a) The *d*-band center and (b) charge transfer of TM atoms in the catalyst.

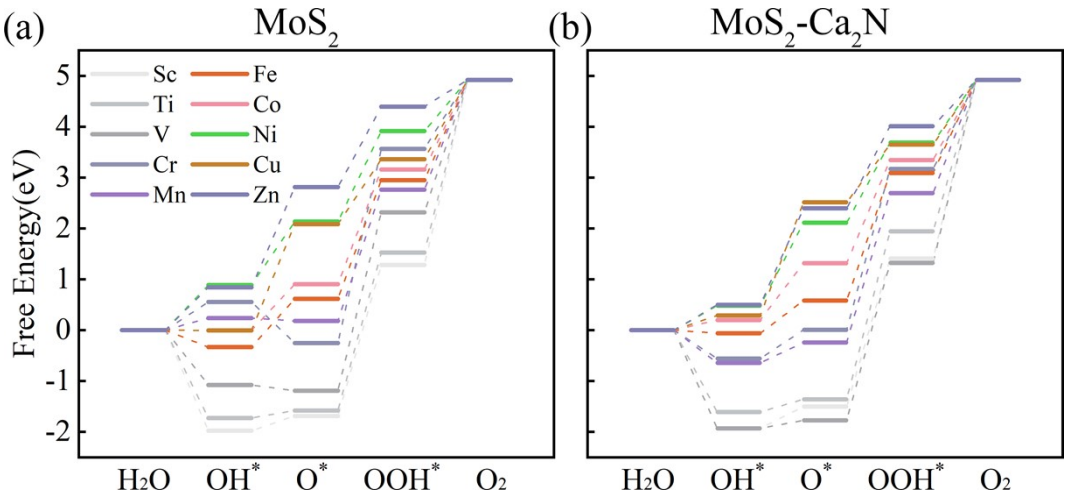

Fig. S7 Free energy paths of the OER for TM single atom on MoS2, and MoS_2/Ca_2N at U = 0 V

Table. S1 The energy consumptions of each reaction step and overpotential in MoS_2.

atom	ΔG_1	ΔG_2	ΔG_3	ΔG_4	Overpotential (eV)
Sc	-1.97411	0.288541	2.96982	2.96982	2.405749
Ti	-1.72438	0.146704	3.104847	3.392827	2.162827
V	-1.07642	-0.11327	3.504334	2.605357	2.274334
Cr	0.55552	-0.80949	3.820295	1.353673	2.590295
Mn	0.236223	-0.05476	2.57955	2.158989	1.34955
Fe	-0.33326	0.952117	2.328538	1.9726	1.098538
Co	-0.0042	0.906031	2.257347	1.760826	1.027347
Ni	0.886685	1.250345	1.77447	1.008499	0.54447
Cu	-0.00196	2.085197	1.27826	1.5585	0.855197
Zn	0.840583	1.974634	1.578548	0.526235	0.744634

Table. S2 The energy consumptions of each reaction step and overpotential in MoS_2/Ca_2N.

atom	ΔG_1	ΔG_2	ΔG_3	ΔG_4	Overpotential (eV)
Sc	-1.92824	0.429205	2.907719	3.511313	2.281313
Ti	-1.60638	0.244335	3.305766	2.976279	2.075766
V	-1.92773	0.156897	3.094221	3.596617	2.366617
Cr	-0.55611	0.560572	3.162978	1.752561	1.932978
Mn	-0.64414	0.400183	2.935482	2.228476	1.705482
Fe	-0.06092	0.642051	2.50625	1.832622	1.27625
Co	0.197897	1.119751	2.024946	1.577407	0.794946
Ni	0.483313	1.628964	1.572776	1.234946	0.398964
Cu	0.287181	2.22428	1.134103	1.274435	0.99428
Zn	0.495682	1.898775	1.615231	0.910312	0.668775