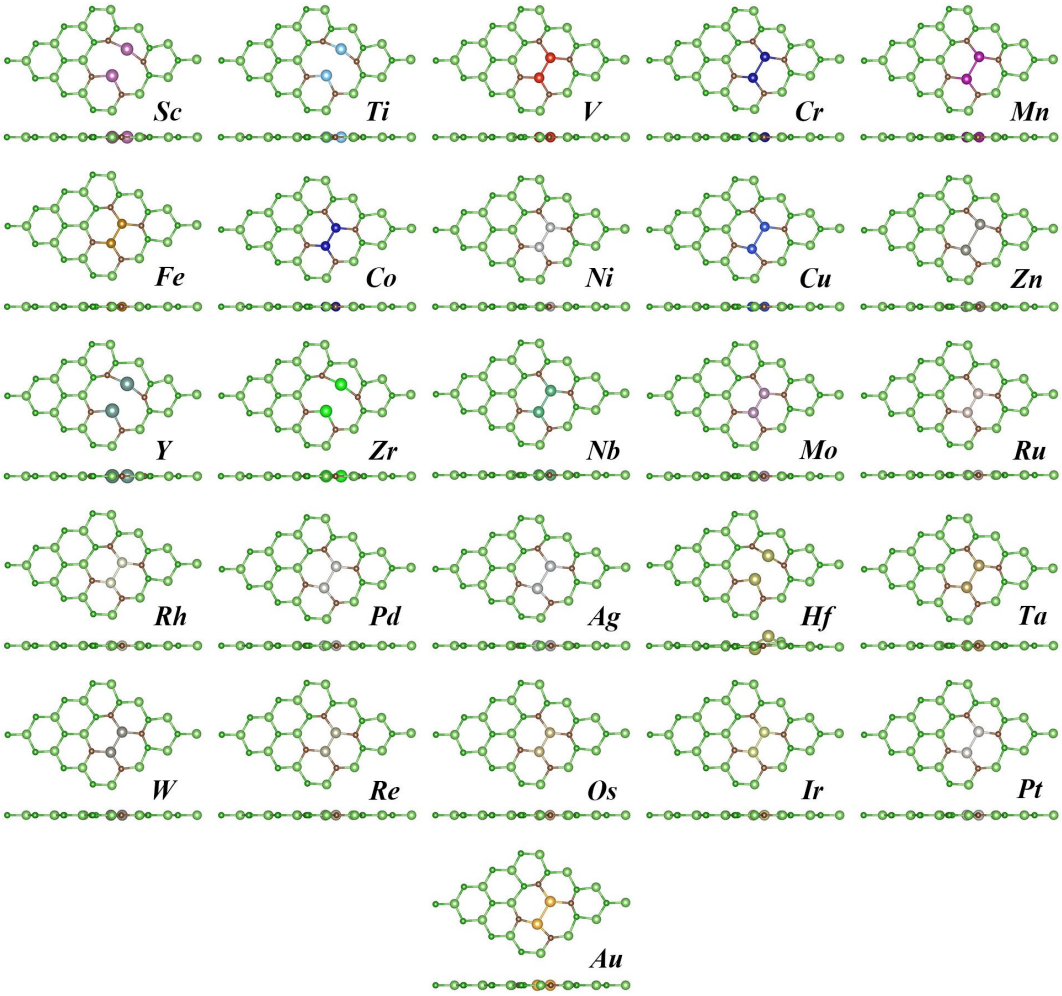

Fig. S1 Top and side views of the optimized structures of TM_2C_4@BAs surface.

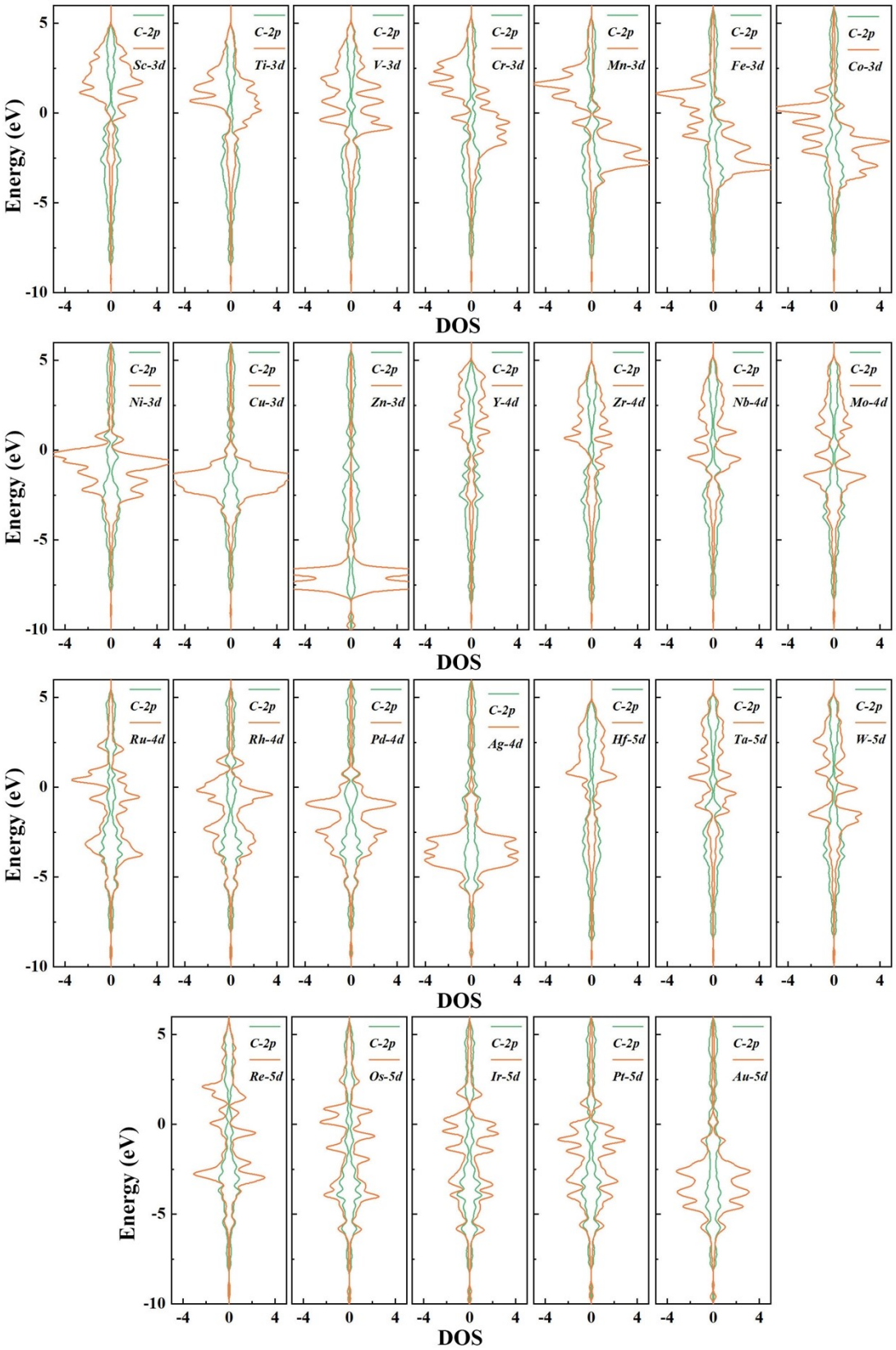

Fig. S2 Projected densities of states (PDOS) of TM_2C_4@BAs surface.

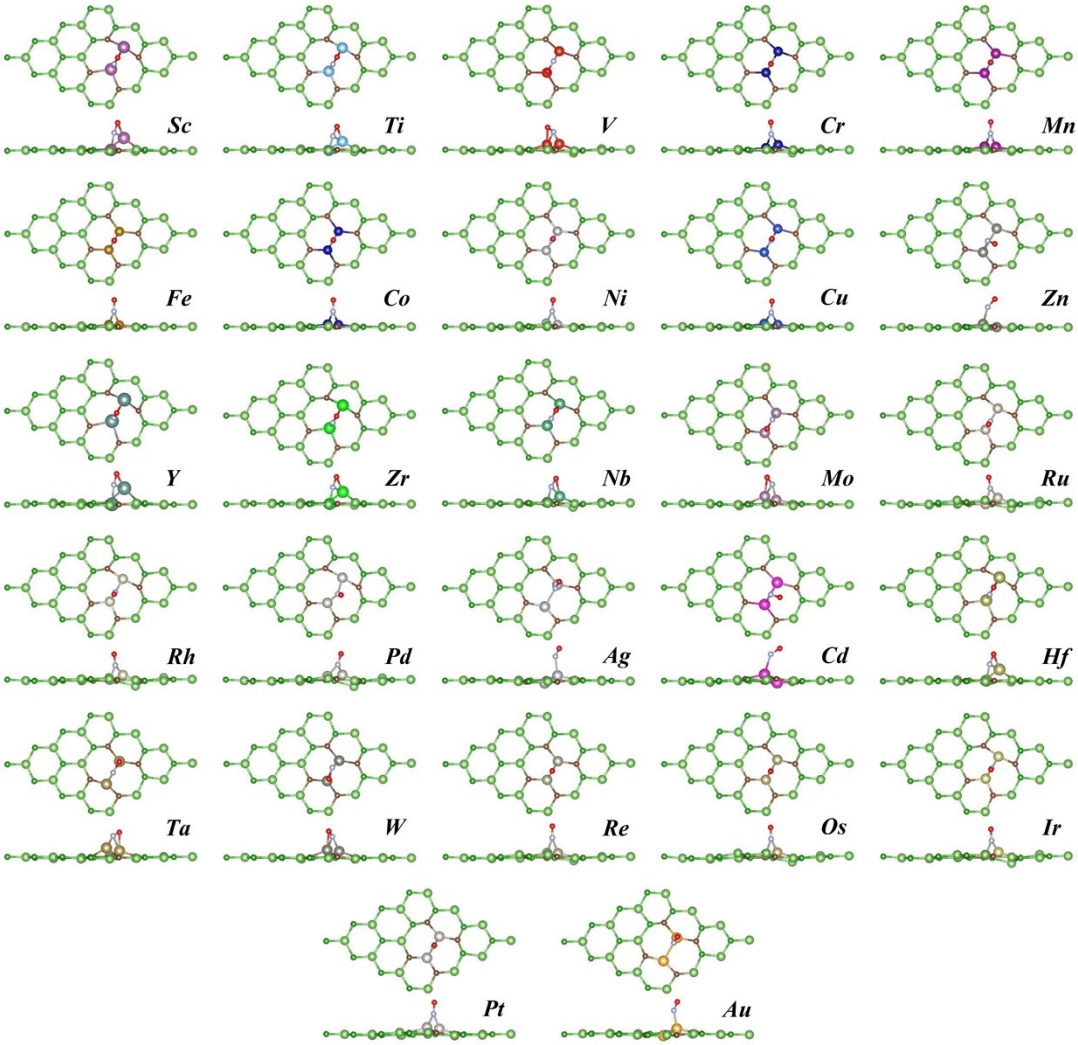

Fig. S3 Top and side views of the optimized structures of NO adsorbed on TM_2C_4@BAs surface.

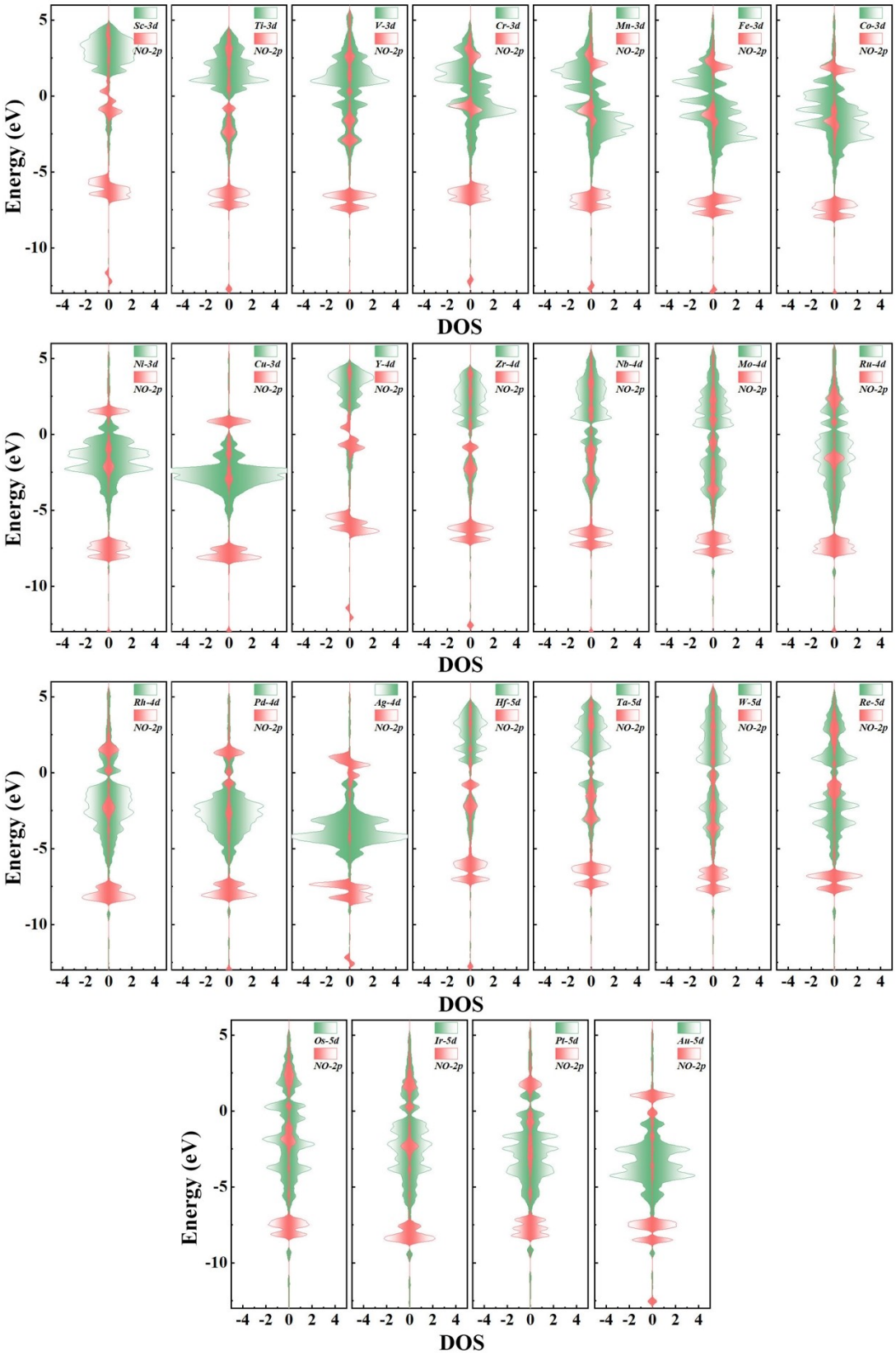

Fig. S4 Projected densities of states (PDOS) of NO adsorbed on TM_2C_4@BAs surface.

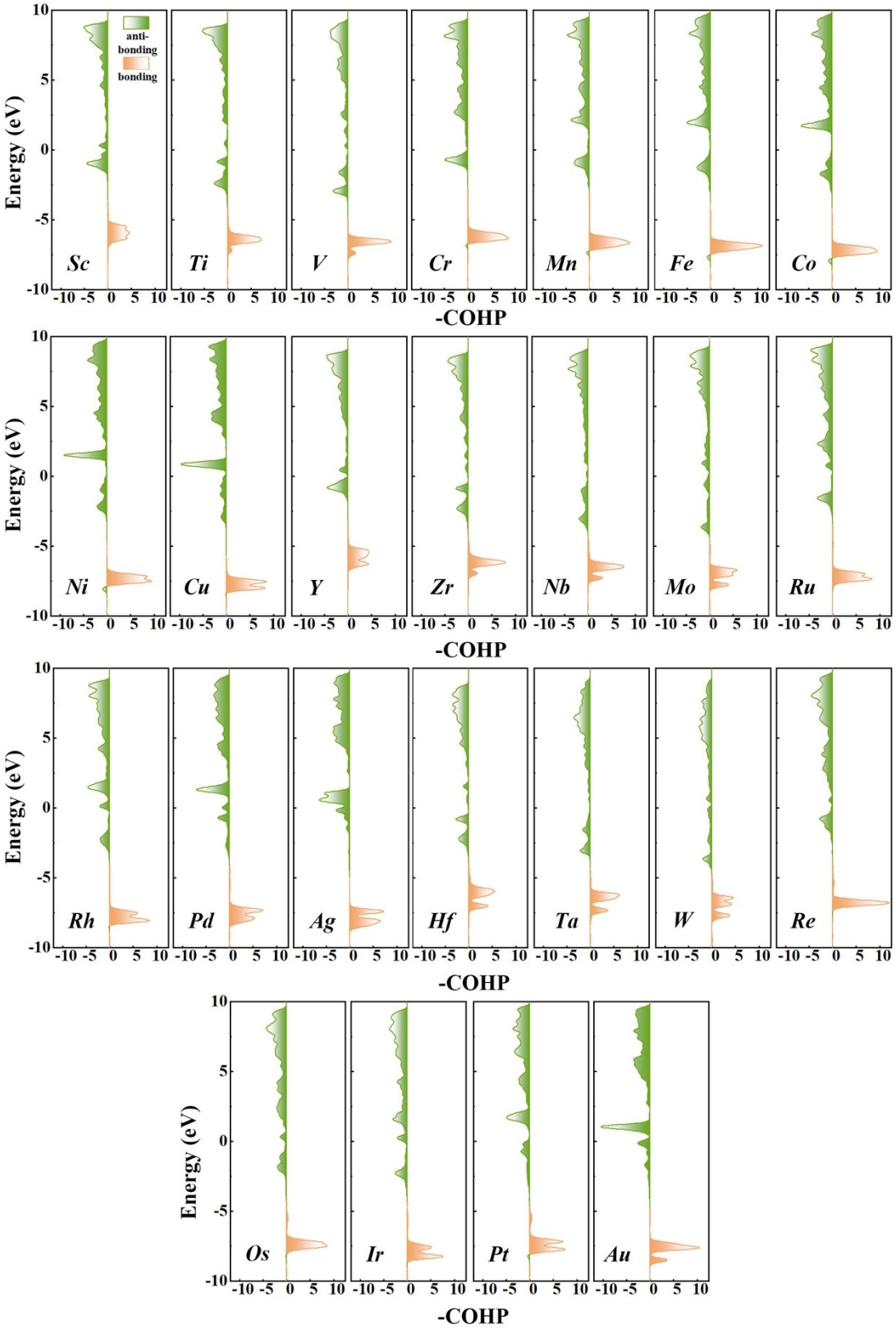

Fig. S5 Projected crystal orbital Hamilton population (-pCOHP) for N-O interaction of NO adsorbed on TM_2C_4@BAs surface.

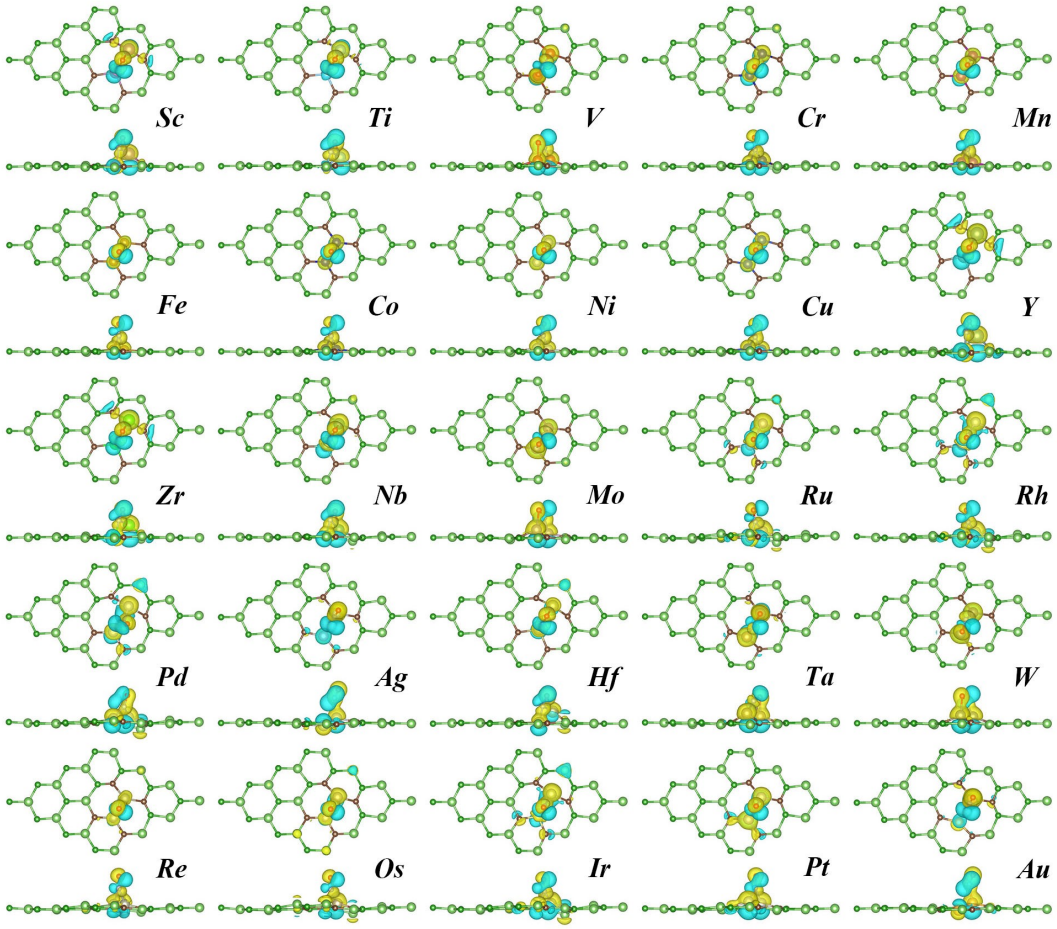

Fig. S6 Top and side views of the difference charge density of NO adsorbed on TM_2C_4@BAs surface. The charge accumulation region and depletion region are depicted by the yellow and green, respectively. The isosurface value is set to be 0.05 e/Å^3

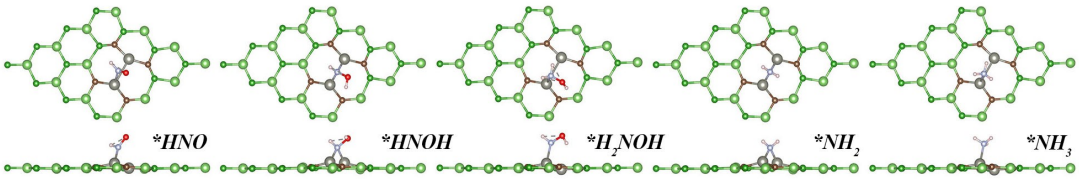

Fig. S7 Top and side views of the intermediate optimization structure in the favorable NORR pathway on the Zn_2C_4@BAs surface.

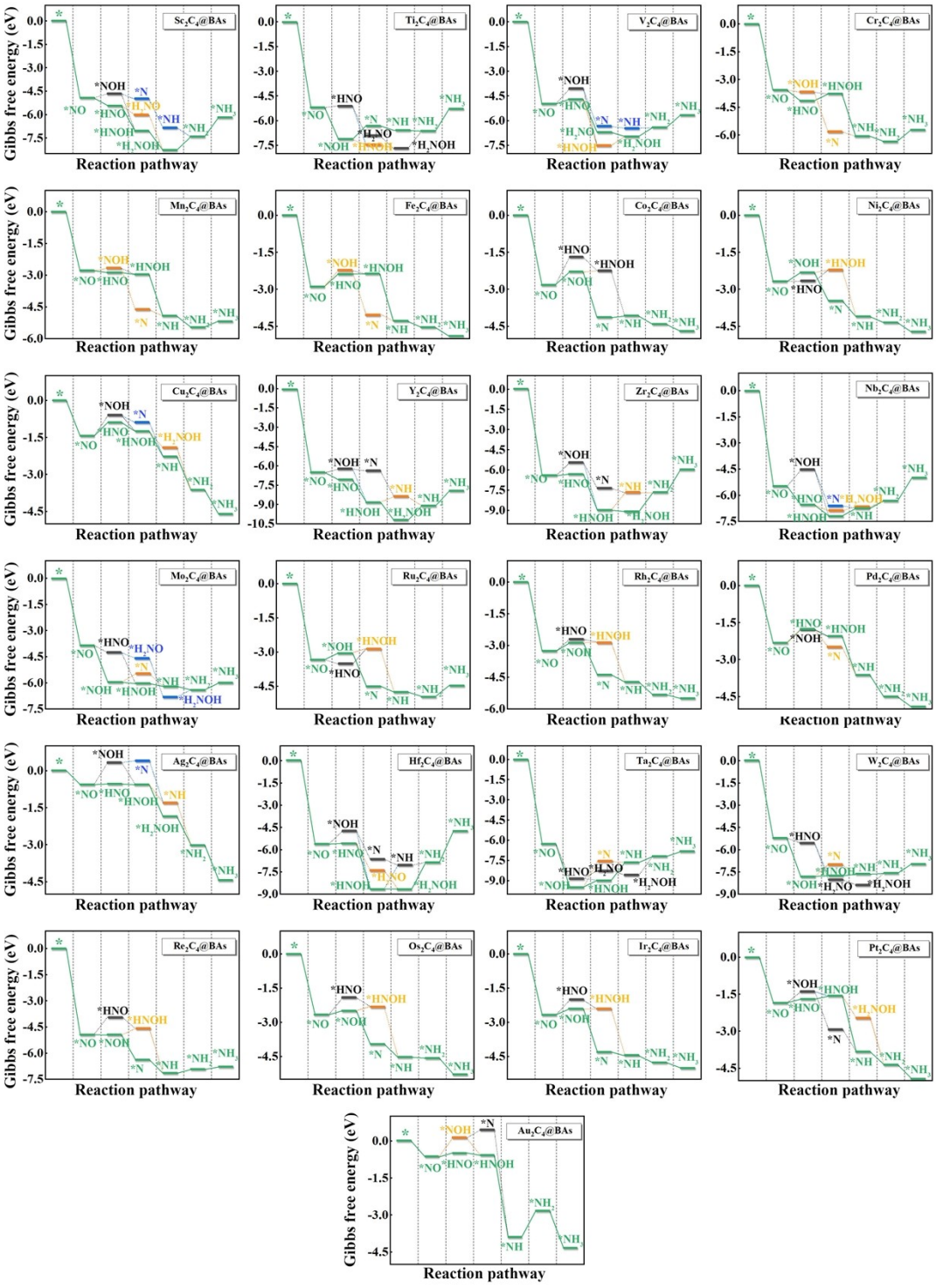

Fig. S8 NORR free energy diagrams of the considered pathways on TM_2C_4@BAs surface.

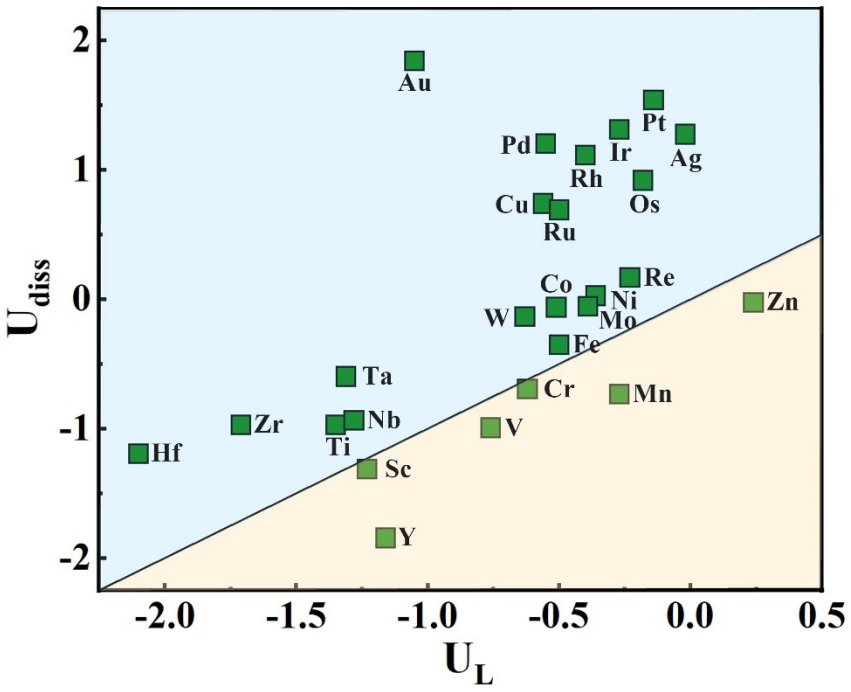

Fig. S9 The U_L(NORR) and U_{diss} of the TM_2C_4@BAs.

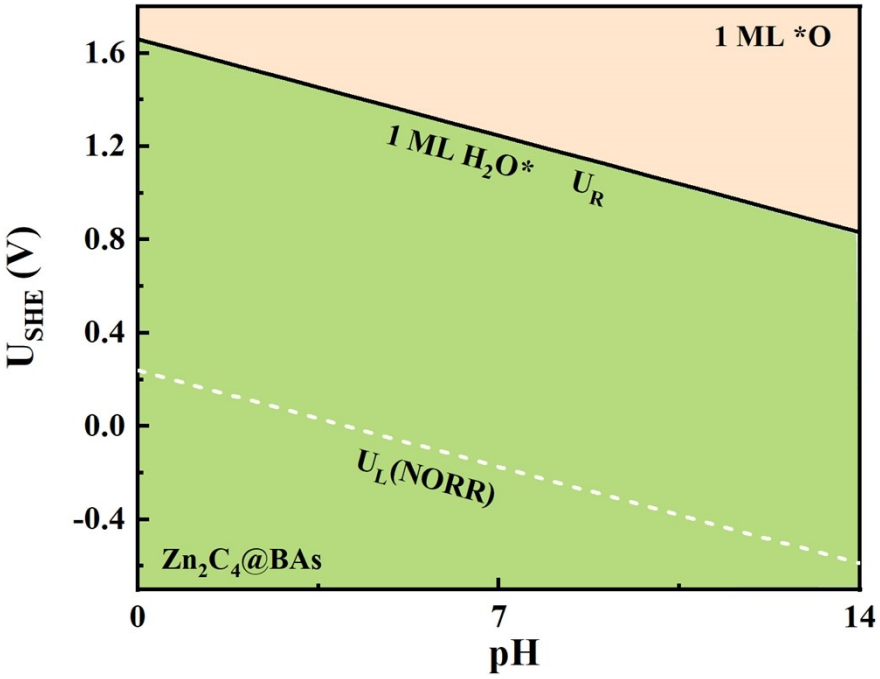

Fig. S10 Surface Pourbaix diagrams of Zn_2C_4@BAs.

Table S1 The formation energy of surface structures in all different coordination environments.

	E_form/atom (eV)	
C_4@BAs	-4.41	/
N_4@BAs	-6.07	geometric structure collapse
P_4@BAs	-4.56	/
O_4@BAs	-3.95	/
S_4@BAs	-4.18	/

Table S2 The adsorption free energy (ΔG_{ads}) of NO, NH_3 and H, the NO charge accumulation ΔQ(NO), the N-O bond length (d_{N-O}) on TM_2C_4@BAs, respectively.

TM_2C_4	ΔG_{ads}(NO/eV)	d_{N-O} (Å)	ΔQ(NO/e)	ΔG_{ads}(NH_3/eV)	ΔG_{ads}(H/eV)
Sc	-4.93	1.30	1.0747	-1.59	-2.05
Ti	-5.19	1.33	1.1455	-0.67	-1.12
V	-5.00	1.35	1.0549	-1.06	-1.26
Cr	-3.56	1.24	0.7773	-1.10	-1.44
Mn	-2.79	1.22	0.6485	-0.57	-0.73
Fe	-2.88	1.21	0.4796	-0.29	0.04
Co	-2.80	1.20	0.4372	-0.09	-0.08
Ni	-2.68	1.20	0.3863	-0.12	-0.34
Cu	-1.45	1.19	0.2830	0.01	-0.04
Zn	-0.43	1.19	0.2628	0.07	0.58
Y	-6.52	1.31	1.1292	-3.31	-3.87
Zr	-6.43	1.35	1.2774	-1.37	/
Nb	-5.47	1.35	1.1241	-0.41	-1.55
Mo	-3.84	1.34	0.9700	-1.40	-1.29
Ru	-3.33	1.21	0.4593	0.15	-0.78
Rh	-3.27	1.20	0.3409	-0.89	-1.10
Pd	-2.32	1.20	0.3192	-0.30	-0.74
Ag	-0.56	1.18	0.0538	0.19	0.44
Hf	-5.62	1.37	1.3129	-0.14	-1.32
Ta	-6.27	1.38	1.1914	-2.24	-1.74

W	-5.20	1.39	1.1129	-2.36	-2.19
Re	-4.94	1.22	0.6165	-2.18	-2.36
Os	-2.67	1.22	0.5049	-0.67	-0.74
Ir	-2.67	1.21	0.3874	-0.39	-0.76
Pt	-1.85	1.2	0.3539	-0.32	-0.51
Au	-0.64	1.18	0.0580	0.28	0.22