

Supporting Information for

Why the abnormal phenomena of D-band center theory exist? A new BASED theory for surface catalysis and chemistry

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1. DFT calculation details.

The Density Functional Theory (DFT) calculations were performed through the projector augmented wave (PAW) method by using the Vienna ab initio simulation package (VASP).[1] The Generalized gradient approximation (GGA) method with the Perdew-Burke-Ernzerhof (PBE) was adopted as the exchange-correlation functional. The k-mesh in Brillouin zones was determined based on Monkhorst–Pack kpoint grids. All the calculations use gamma-centered k-points $3\times3\times1$, zero damping DFT-D3 method of Grimme, 500 eV cutoff energy and the spin-polarized calculations (collinear). The convergence tolerance for the residual force and energy on each atom during structure relaxation were set to $0.015\text{ eV}\text{\AA}^{-1}$ and $1\times10^{-5}\text{ eV}$. VASP-sol package[2] is used to simulate the solution environment for all calculations, where the dielectric constant (ϵ_r) is set to 78.5. For semiconductor systems like SACs, the Hubbard U correction (only for 3d TM) was also employed within the DFT+U approach, U-J: 2.11 eV for Sc, 2.58 eV for Ti, 2.72 eV for V, 2.79 eV for Cr, 3.06 eV for Mn, 3.29 eV for Fe, 3.42 eV for Co, 3.4 eV for Ni, 3.87 eV for Cu, 4.12 eV for Zn[3]. In order to accurately obtain the DOS information, we use the most precise tetrahedron method with Blöchl corrections, scatter 2000 points and completely turns off the symmetry for all electronic structure calculations to pursue sufficient accuracy. The z-direction is set to $20\text{ \AA}$ to make the vacuum distance is more than $10\text{ \AA}$ for all the models, which is to avoid steric hindrance and interaction due to periodicity. The optimization of the unit cell parameters is carried out by the method of fixing the lattice vector, and the cell parameters are obtained as $14.903\times12.735\times20.000$, $\alpha = \beta = \gamma = 90^\circ$ for related calculations of SACs. And the cell parameters of the Bulks model

can be obtained through separate optimization calculations: $9.370 \times 10.796 \times 20.000$, $\alpha = \beta = \gamma = 90^\circ$ for Pt bulk, $8.429 \times 9.724 \times 20.000$, $\alpha = \beta = \gamma = 90^\circ$ for Ni bulk, $9.943 \times 11.399 \times 20.000$, $\alpha = \beta = \gamma = 90^\circ$ for Ag bulk, $8.682 \times 9.976 \times 20.000$, $\alpha = \beta = \gamma = 90^\circ$ for Cu bulk. The Lobster software[4, 5] performed the COHP analysis to obtain the bonding and anti-bonding information in the d orbital.

Calculate the adsorption energy using the following equation:

$$\Delta E_{\text{ado}*} = E_{\text{slab-ado}*} - E_{\text{slab}} - E_{\text{ado}*} \quad \#(1)$$

Where the $\Delta E_{\text{ado}*}$ represents the adsorption energy of the active center atom on the adsorbent, $E_{\text{slab-ado}*}$ represents the total energy of slab and adsorbent, E_{slab} represents the energy of slab and $E_{\text{ado}*}$ represents the energy of adsorbent. Notably, The structure of E_{slab} needs to directly remove the adsorbent from the structure of $E_{\text{slab-ado}*}$, and then obtain its static energy to ensure that the energy of E_{slab} can represent the spin state of the active site during adsorption, which we also use in previous work[3, 6]. The adsorbents include * H, * O, * OH, and * OOH, and their corresponding energies are E_{ado} shown in **Table S30**.

When using the BASED theory to calculate Q, for sites with multiple active centers (e.g. Pt/Ag/Cu/Ni Bulk), multiple bonds should be formed, so each Q needs to be stacked and summed, as follows:

$$Q_{\text{SUM}} = \sqrt{Q_1^2 + Q_2^2 + Q_3^2 + \dots} \quad \#(2)$$

The detailed Q_{SUM} data obtained from this formula is shown in **Table S2-S13**.

2. Supplementary Figures.

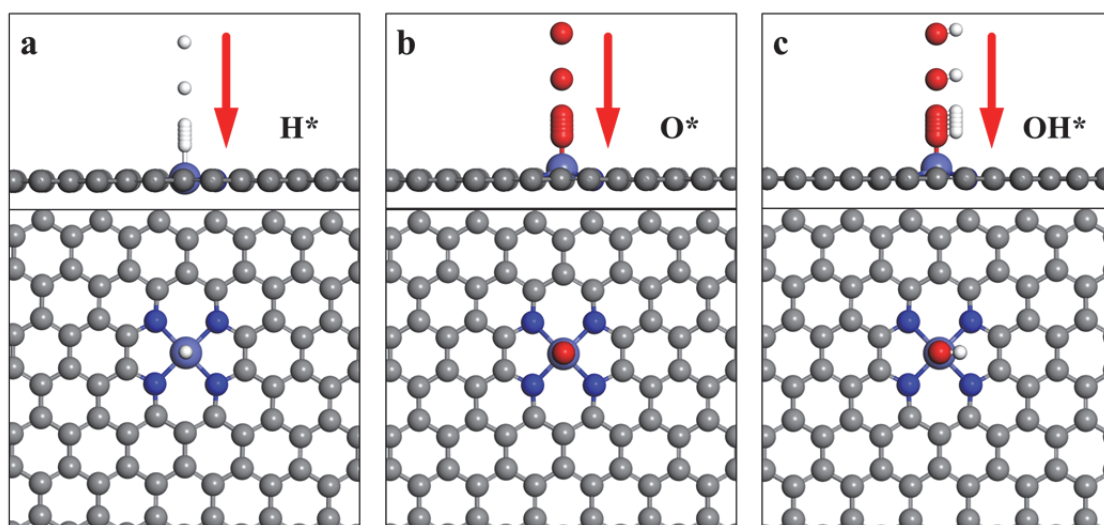


Fig. S1 Illustrations of SACs adsorption model. The H* (a), O* (b), O*H (c) adsorption models of SAC with different distance, including all kinds of 3d transition metal SACs.

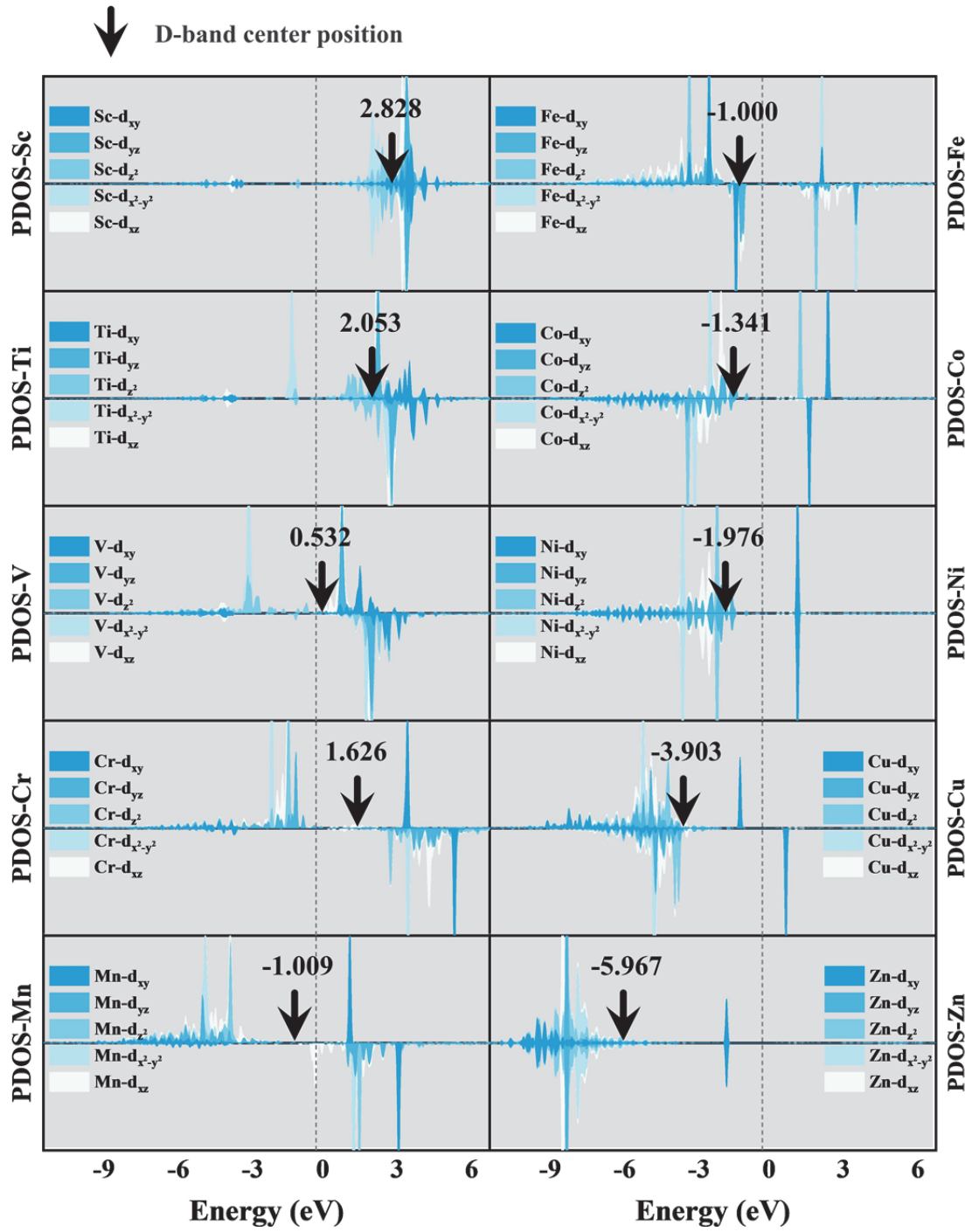


Fig. S2 Schematic diagram of pDOS. Projected DOS (PDOS) diagram of five types d orbit of all 3D metal SACs slab. The black arrow represents the center position of the D band, and the black number represents the value of ϵ_d .

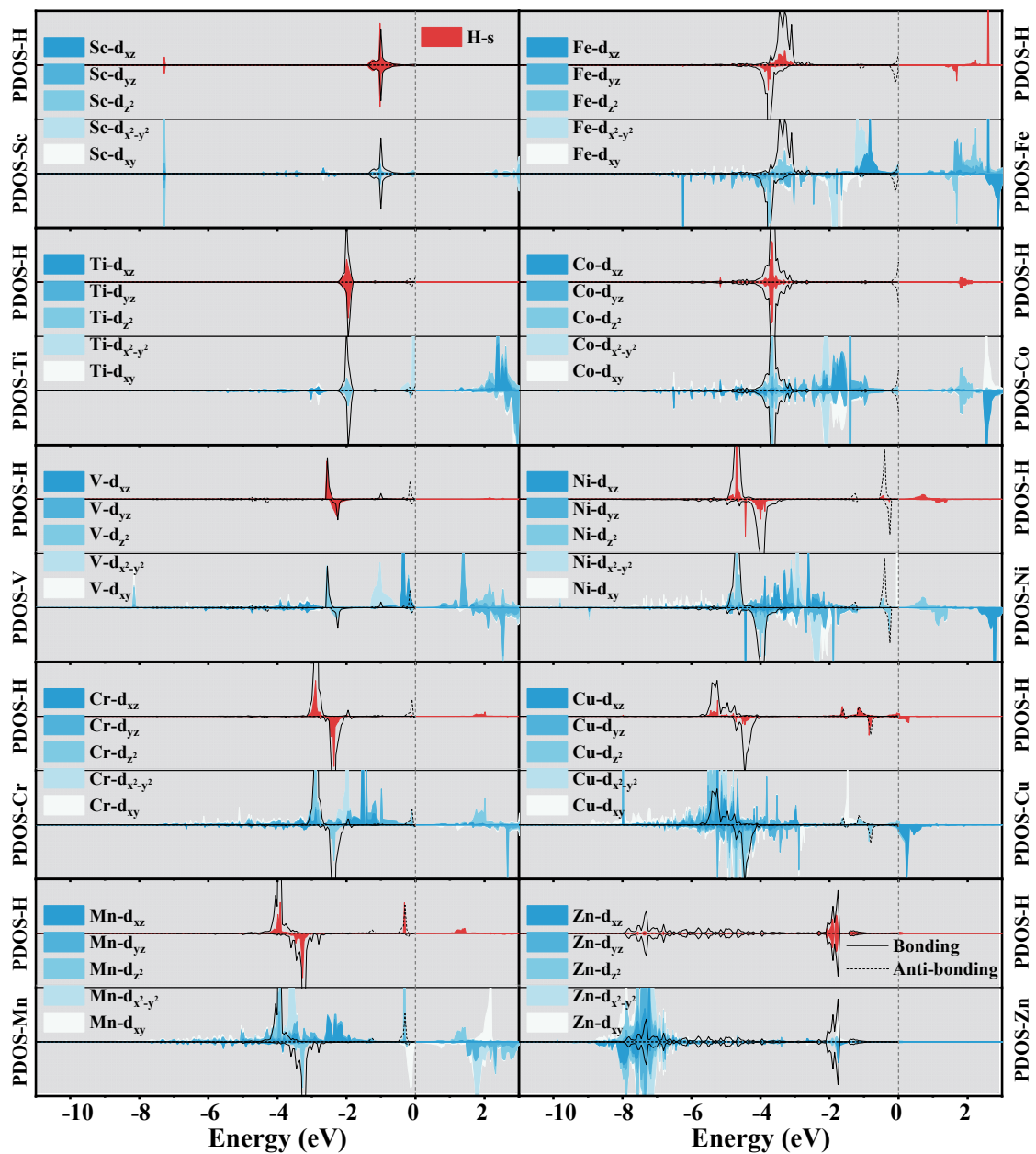


Fig. S3 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of SACs and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements.

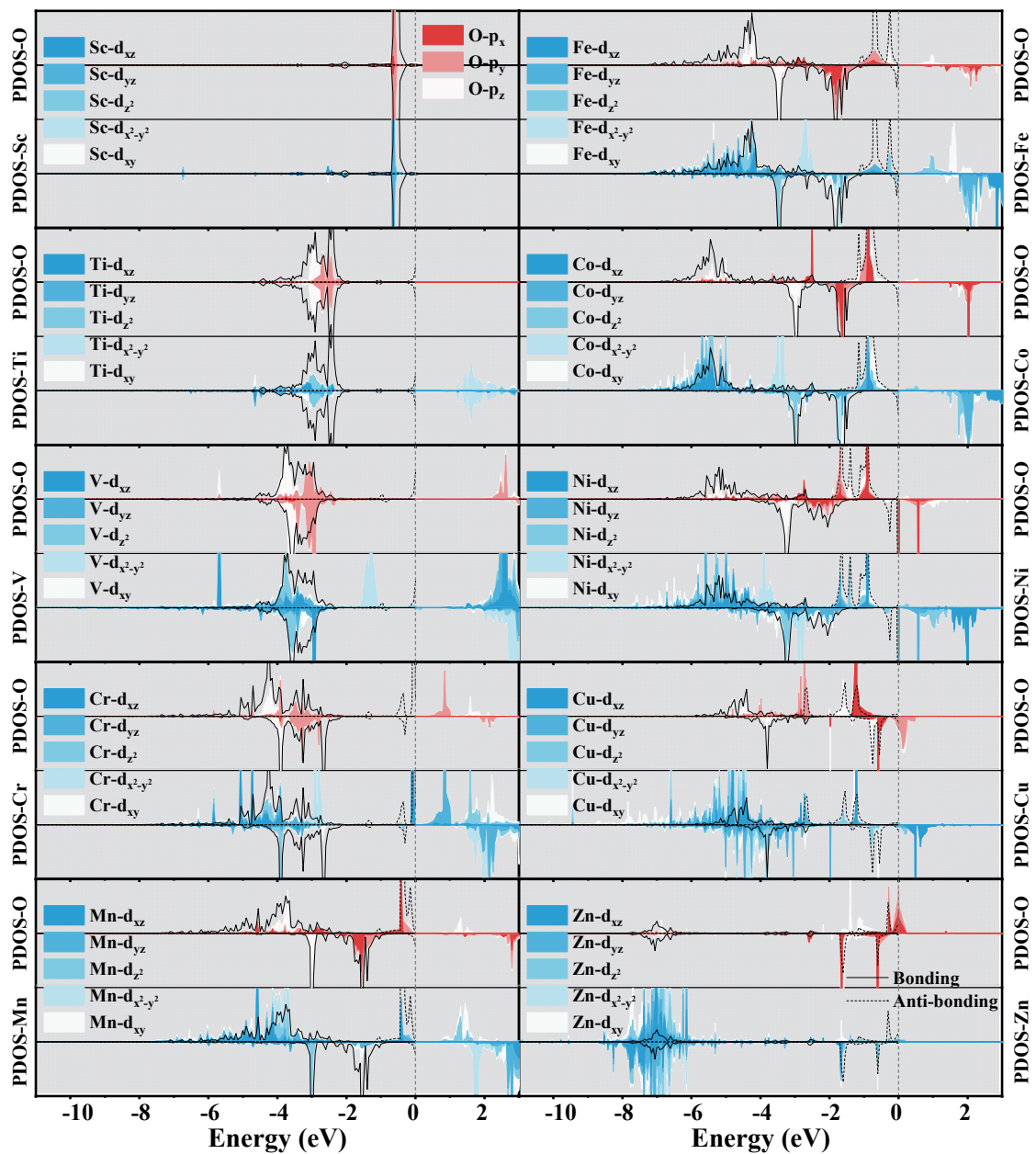


Fig. S4 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of SACs and O* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements.

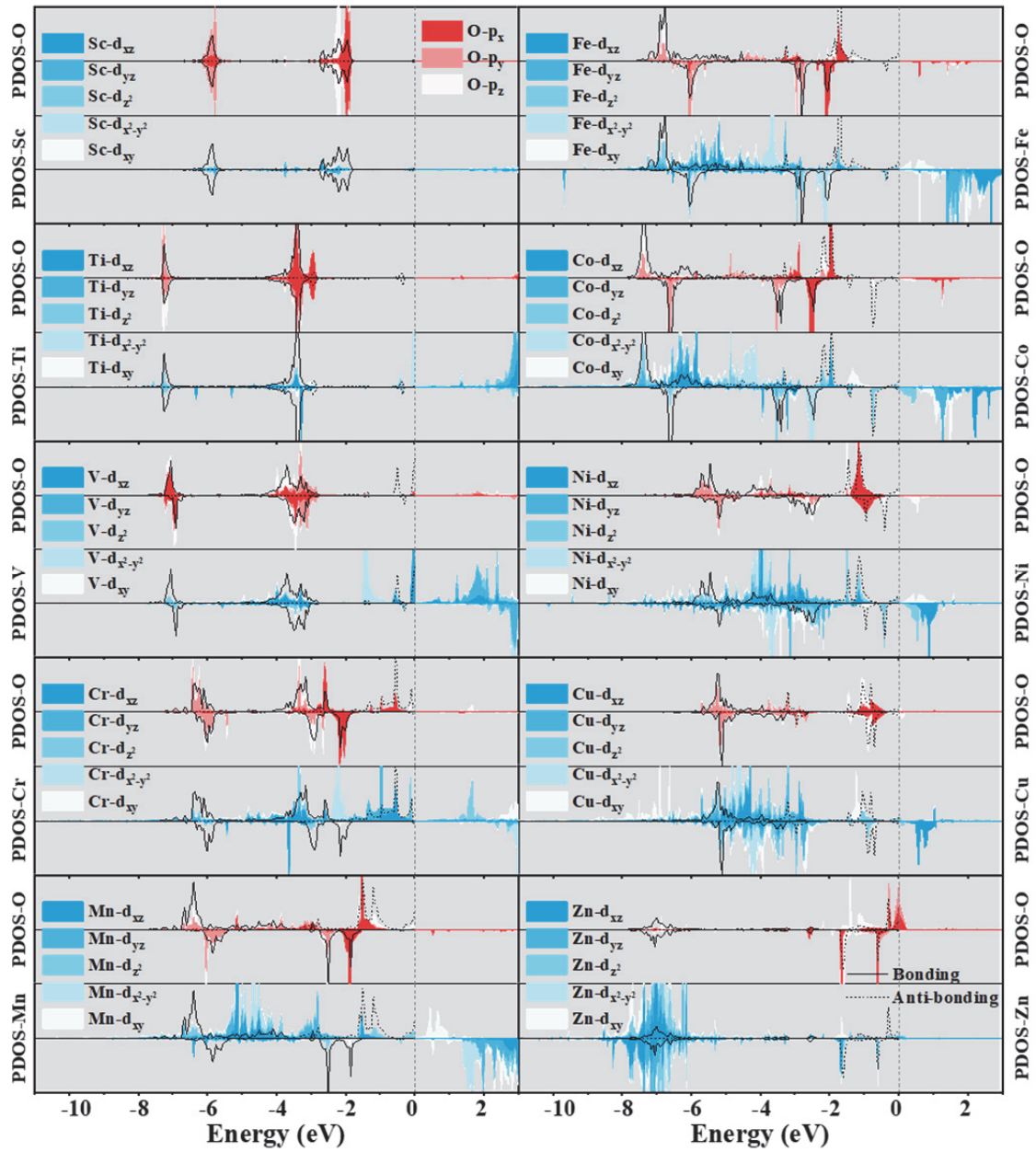


Fig. S5 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of SACs and O* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements.

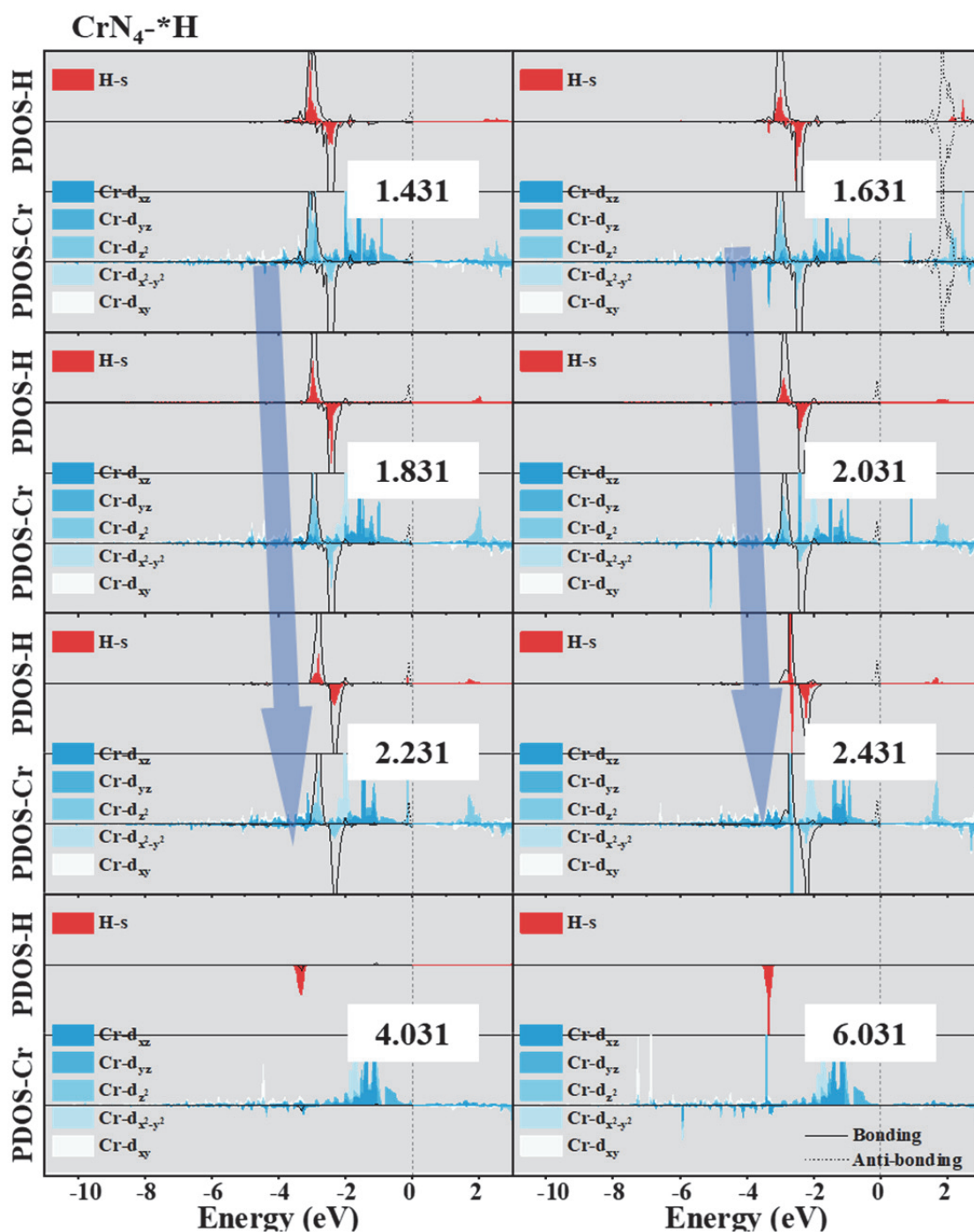


Fig. S6 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CrN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

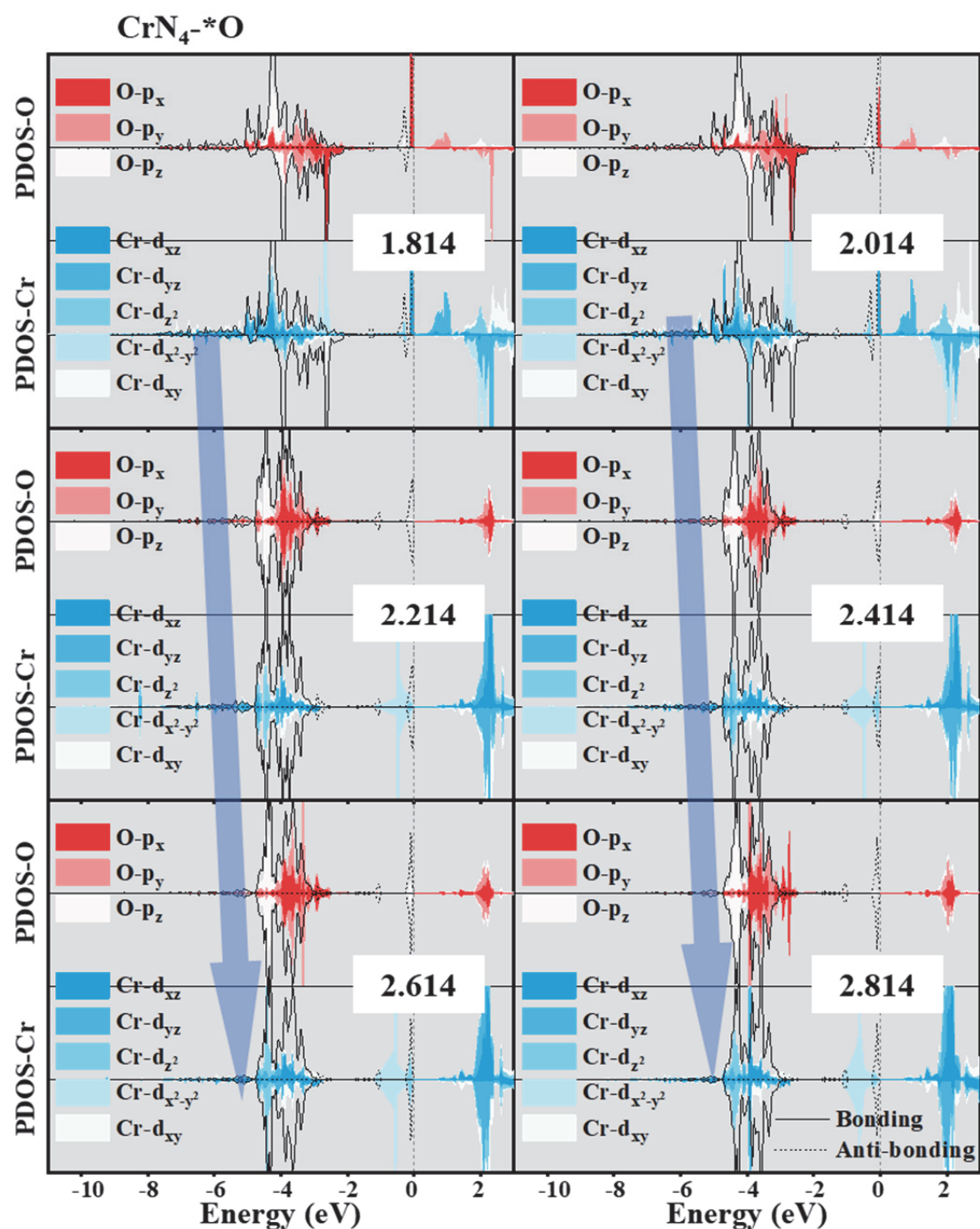


Fig. S7 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CrN_4 and O^* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

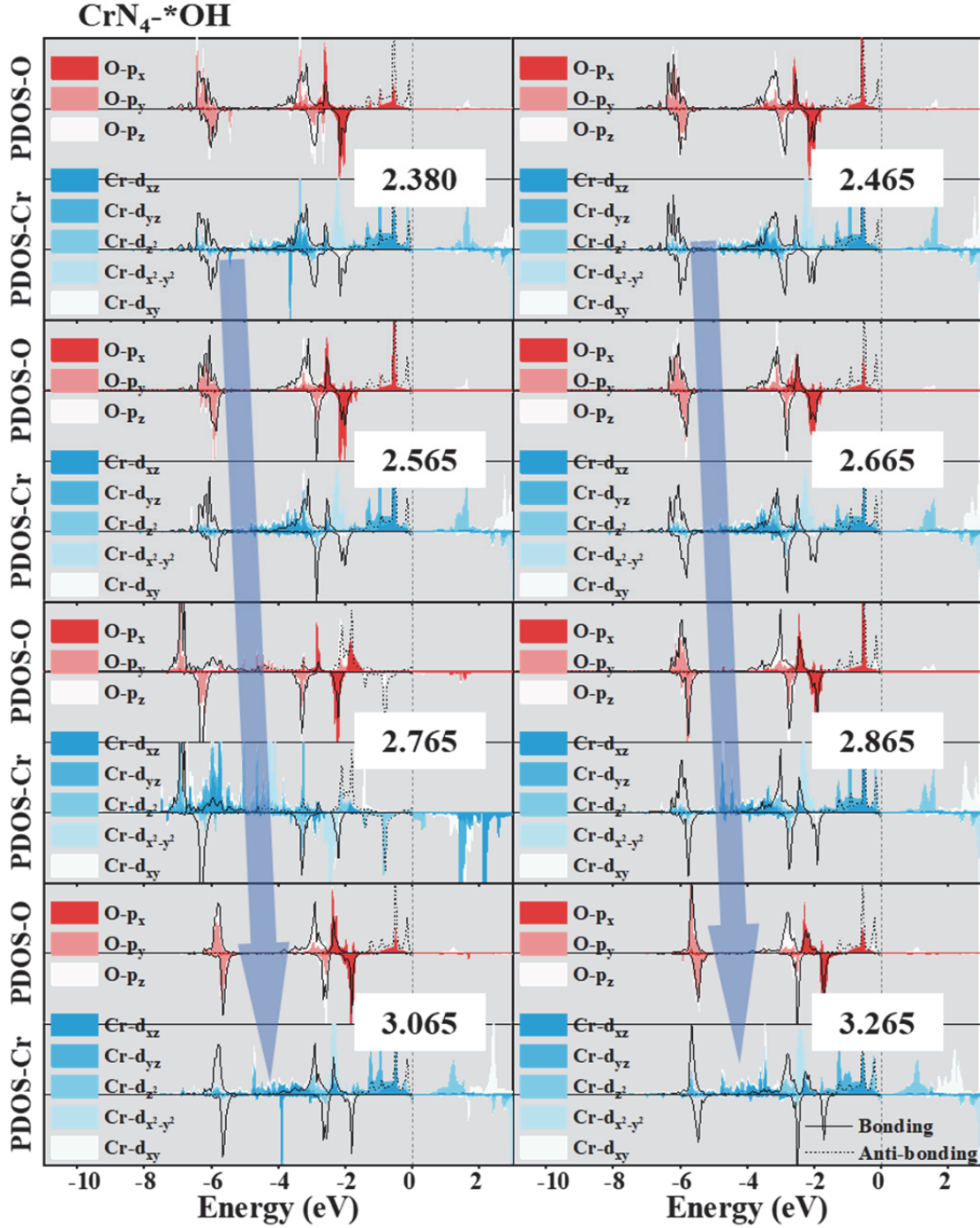


Fig. S8 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CrN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

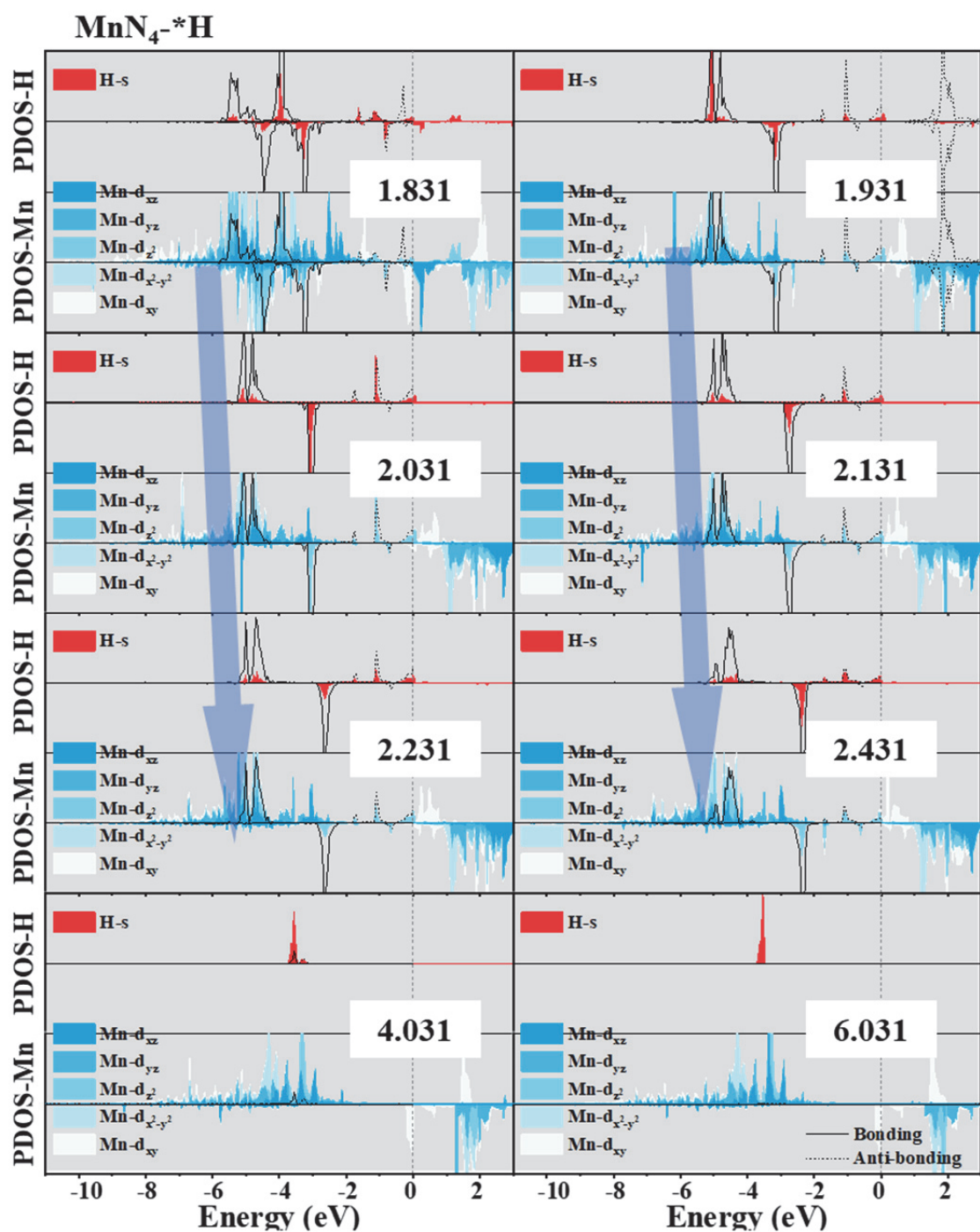


Fig. S9 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of MnN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

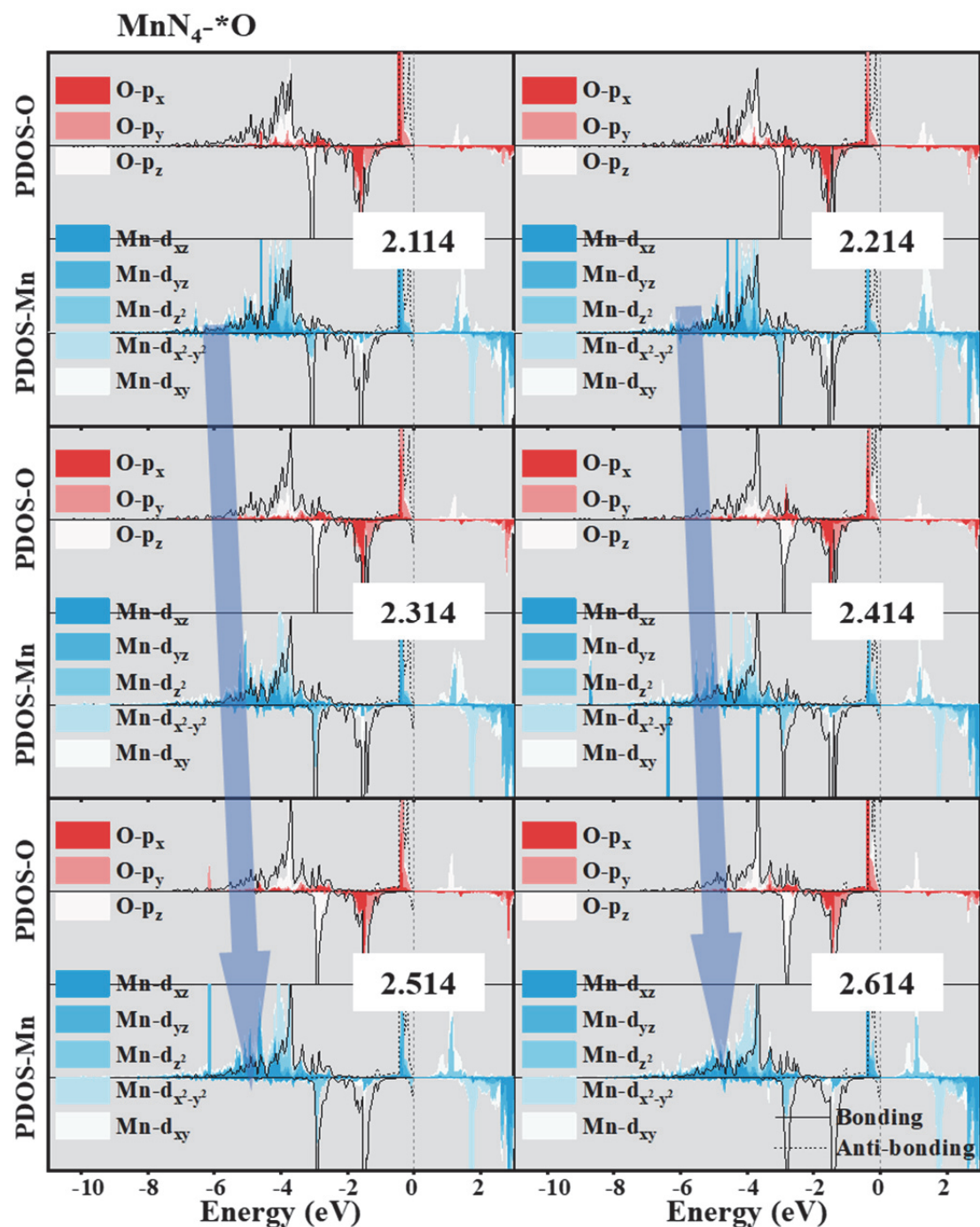


Fig. S10 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of MnN₄ and O* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

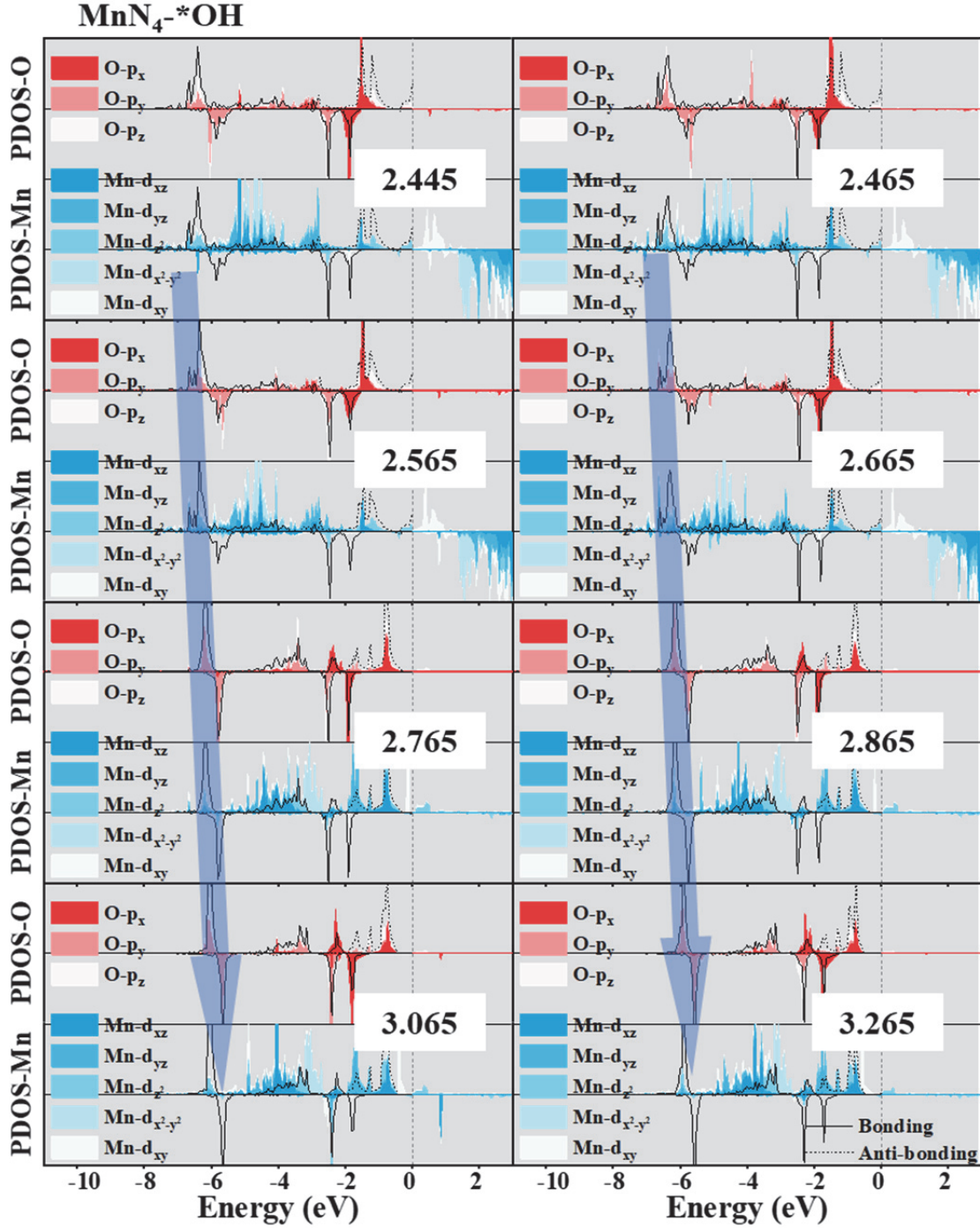


Fig. S11 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of MnN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

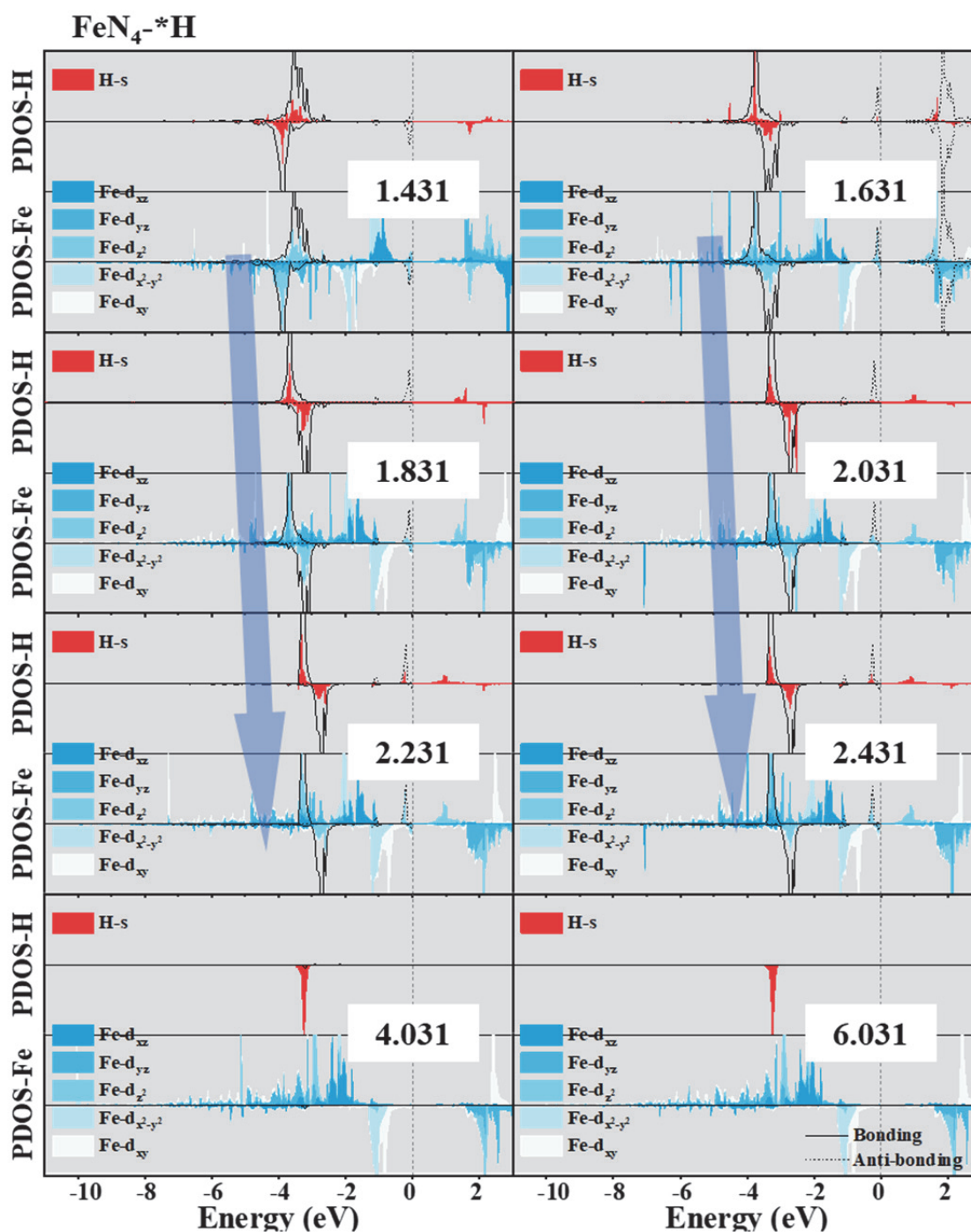


Fig. S12 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of FeN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

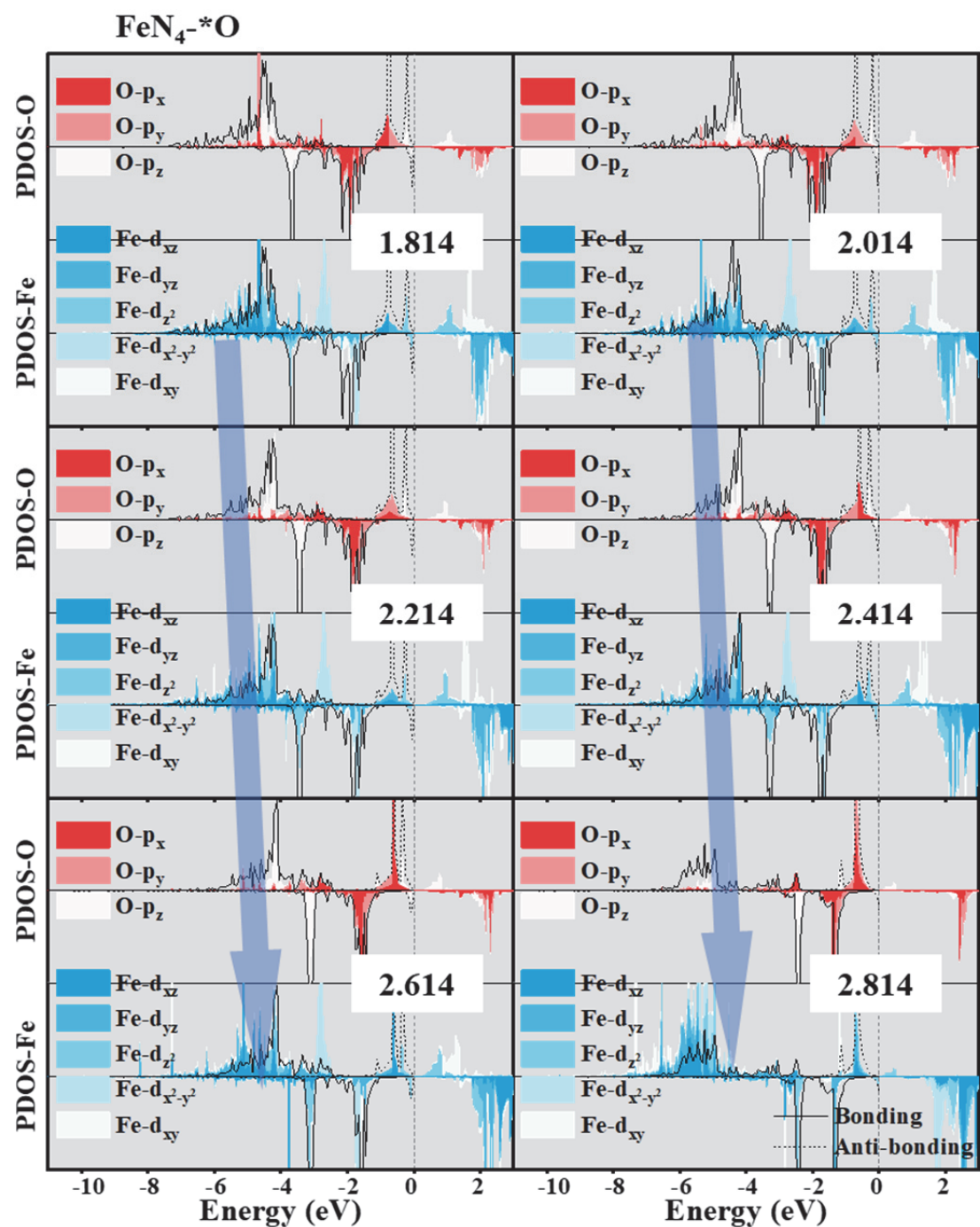


Fig. S13 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of FeN₄ and O* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

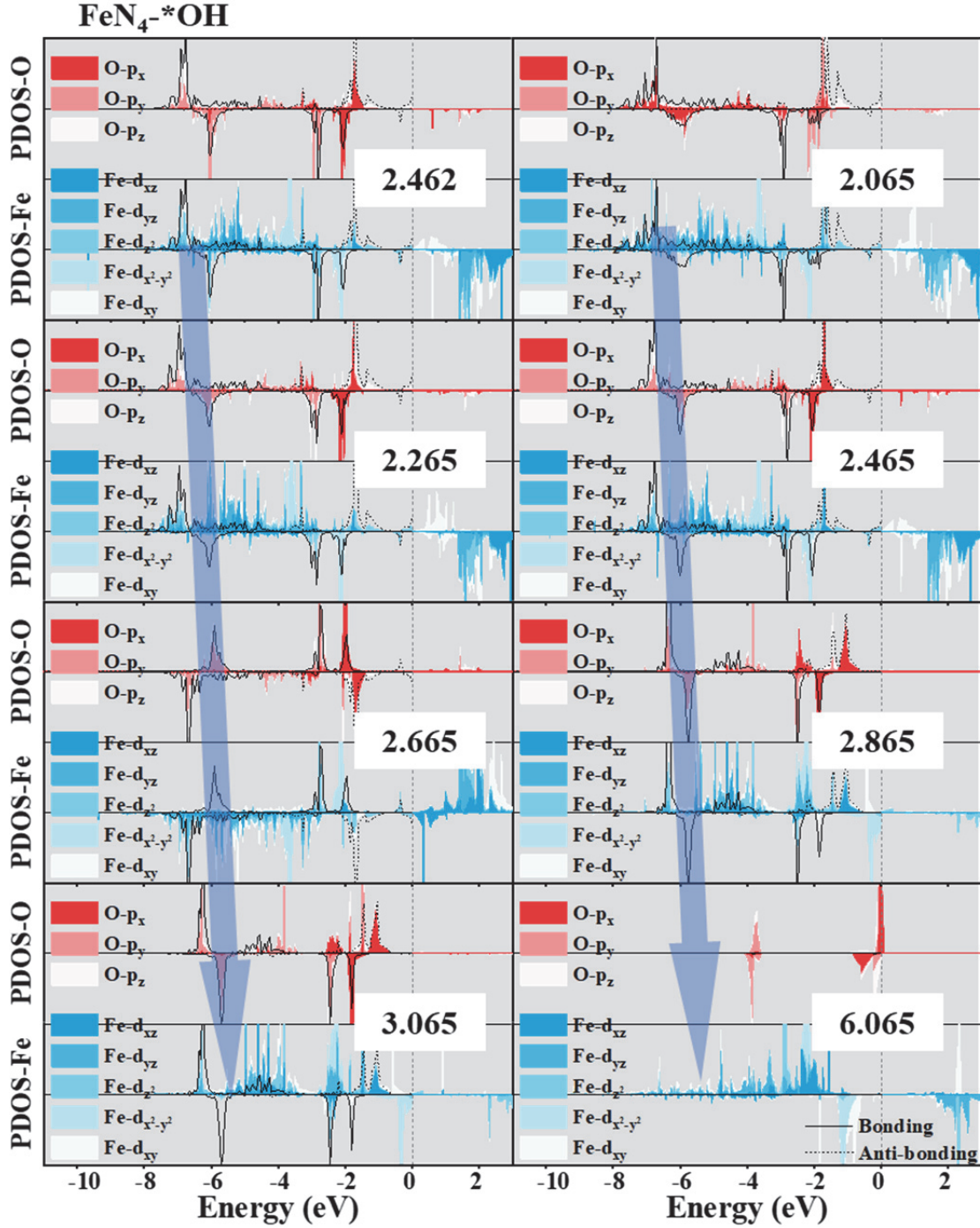


Fig. S14 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of FeN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

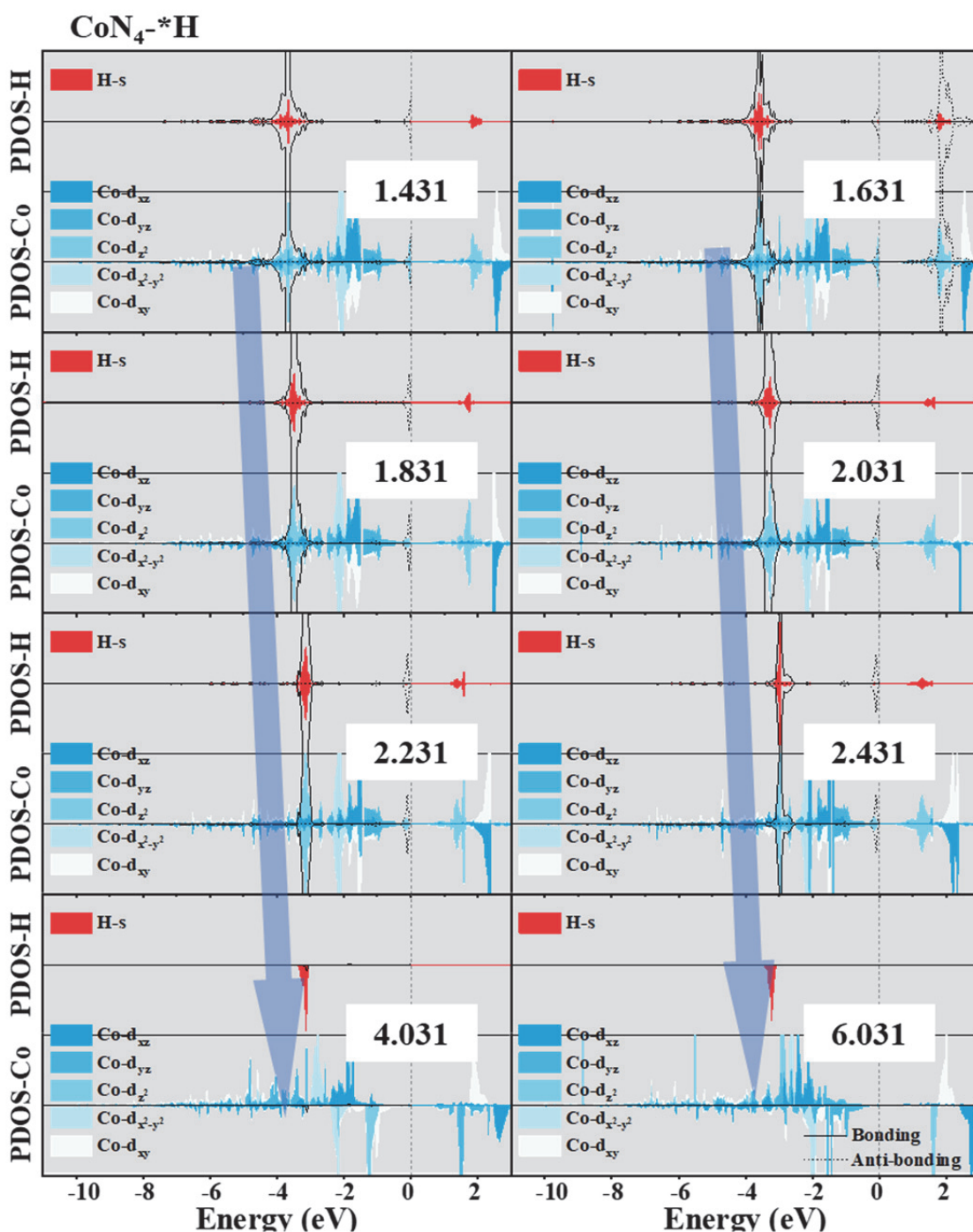


Fig. S15 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CoN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

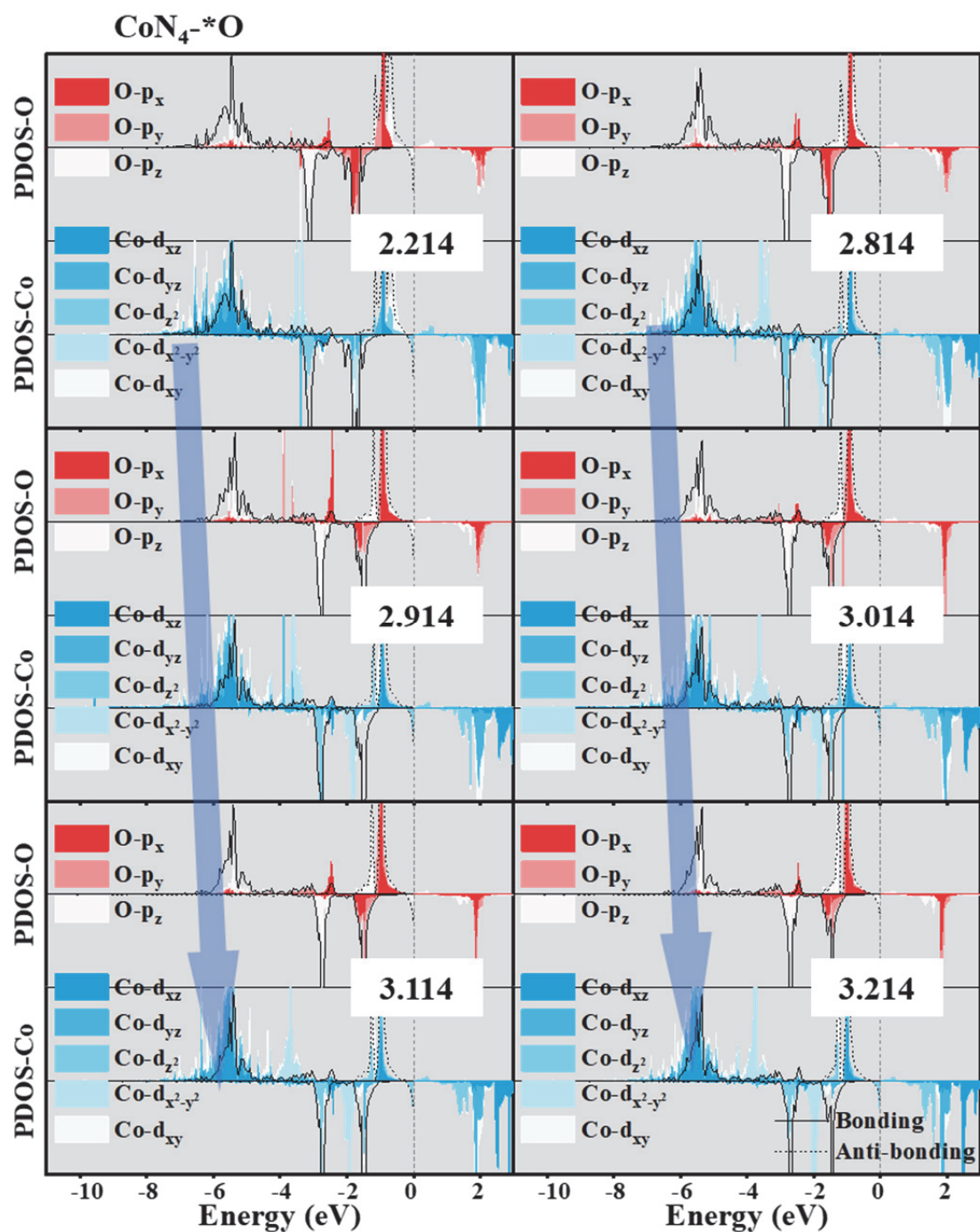


Fig. S16 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CoN₄ and O* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

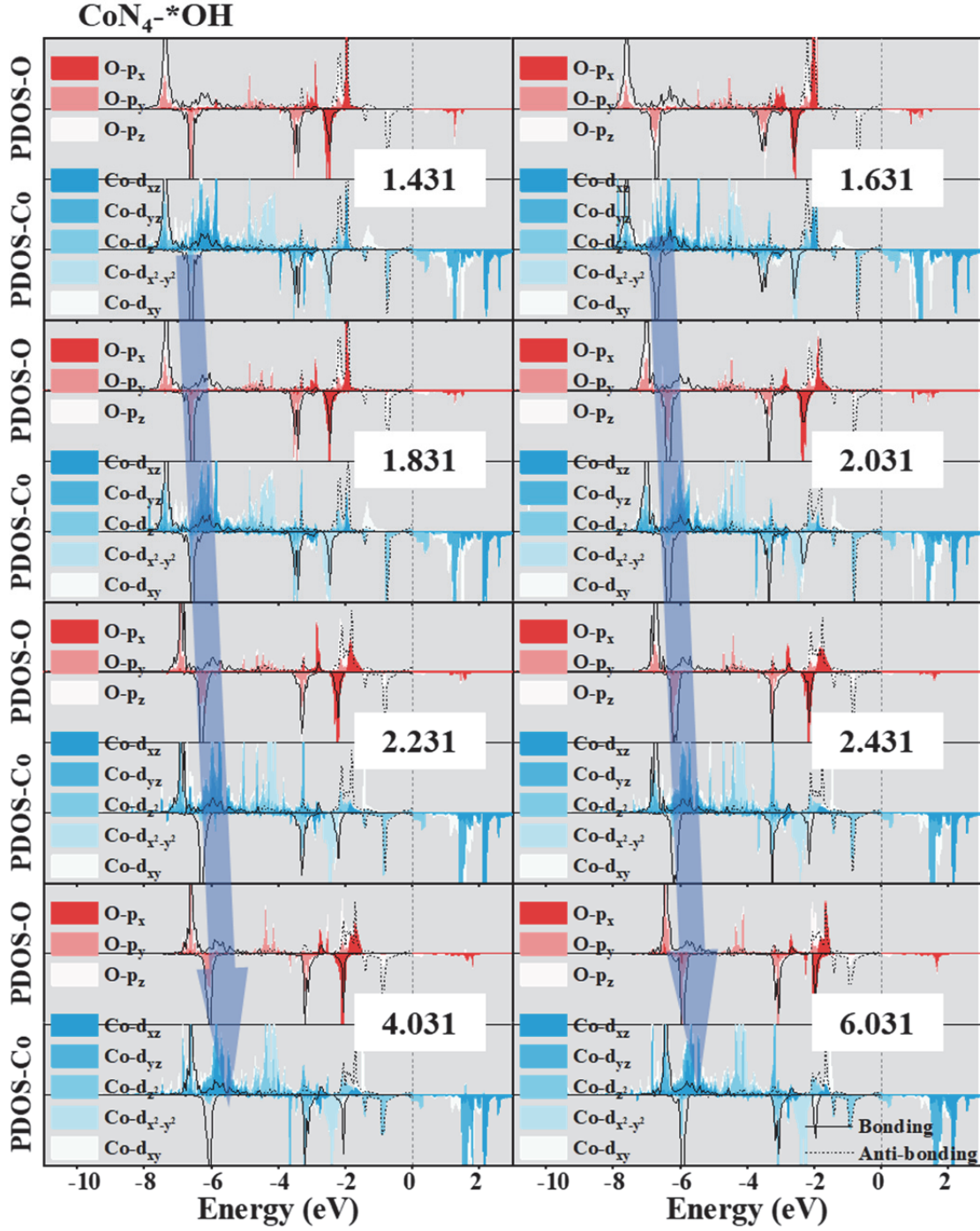


Fig. S17 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CoN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

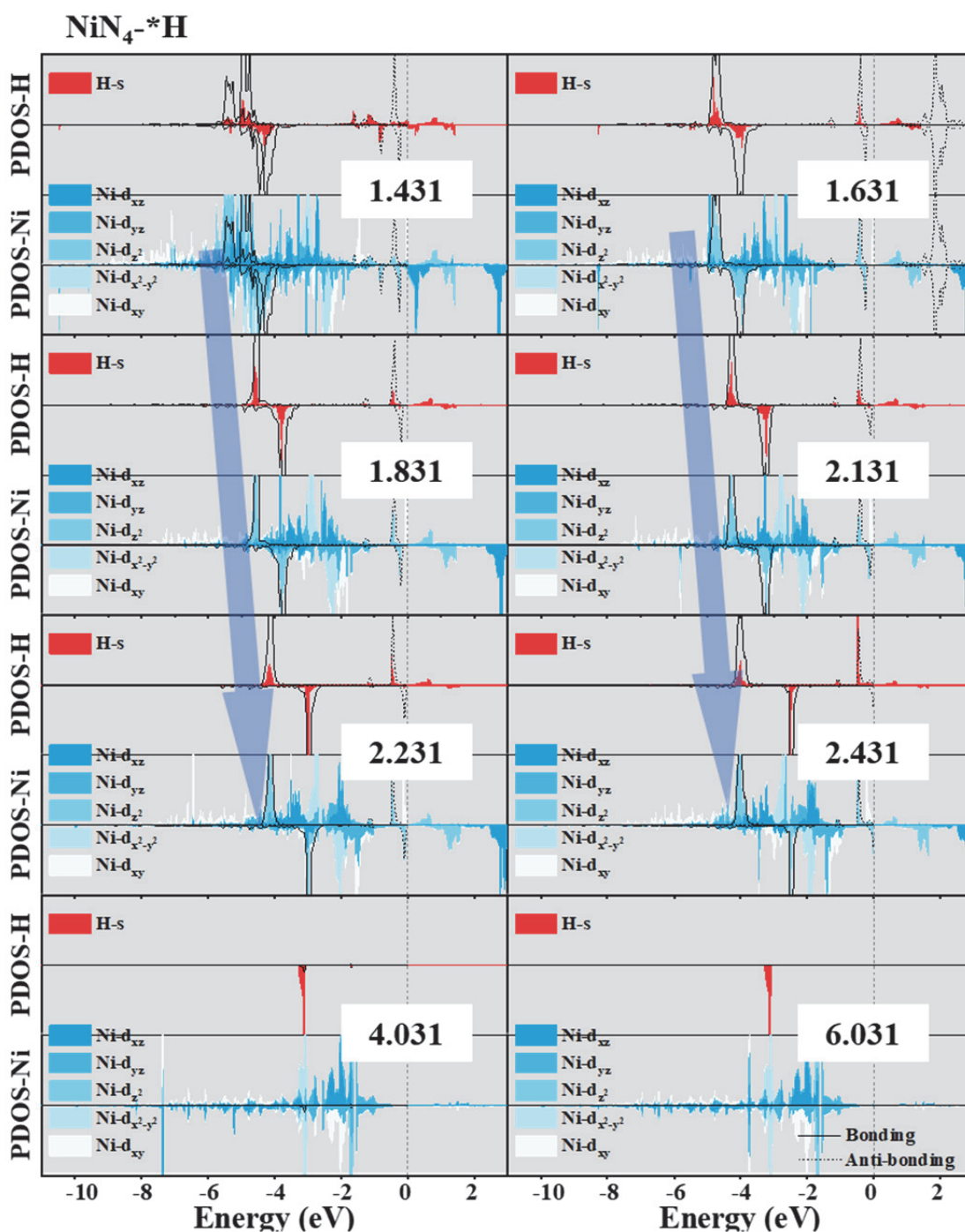


Fig. S18 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of NiN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

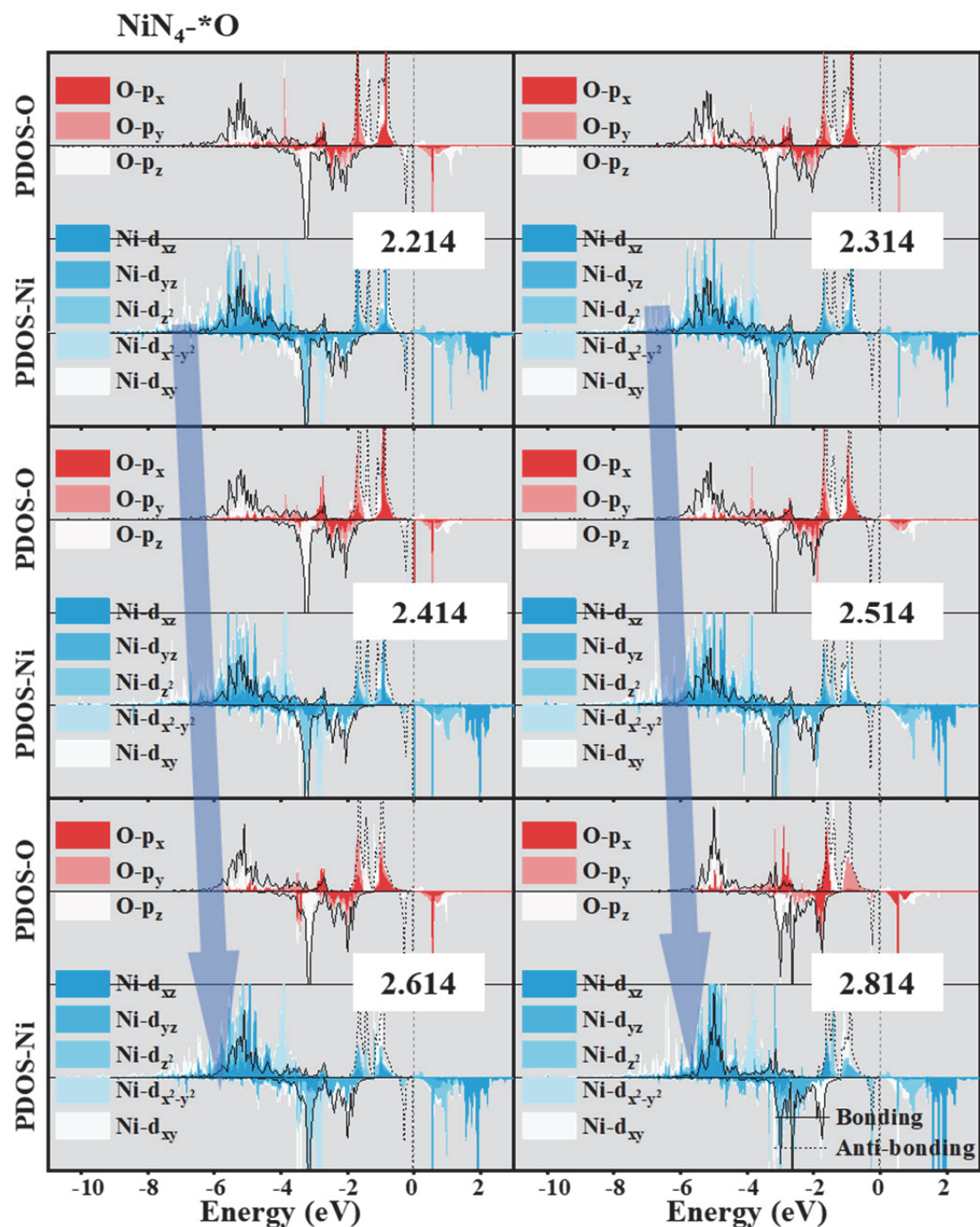


Fig. S19 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of NiN_4 and O^* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

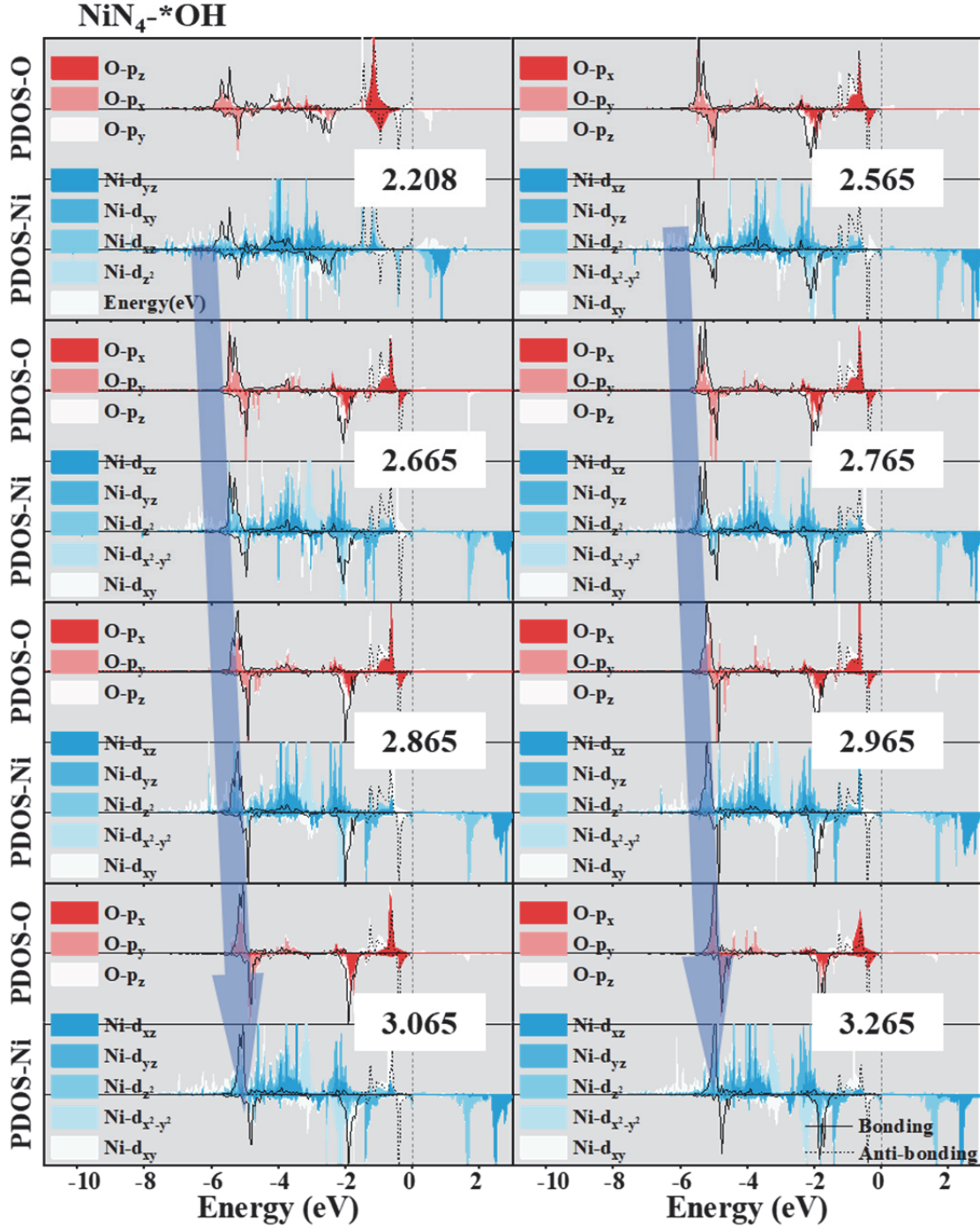


Fig. S20 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of NiN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

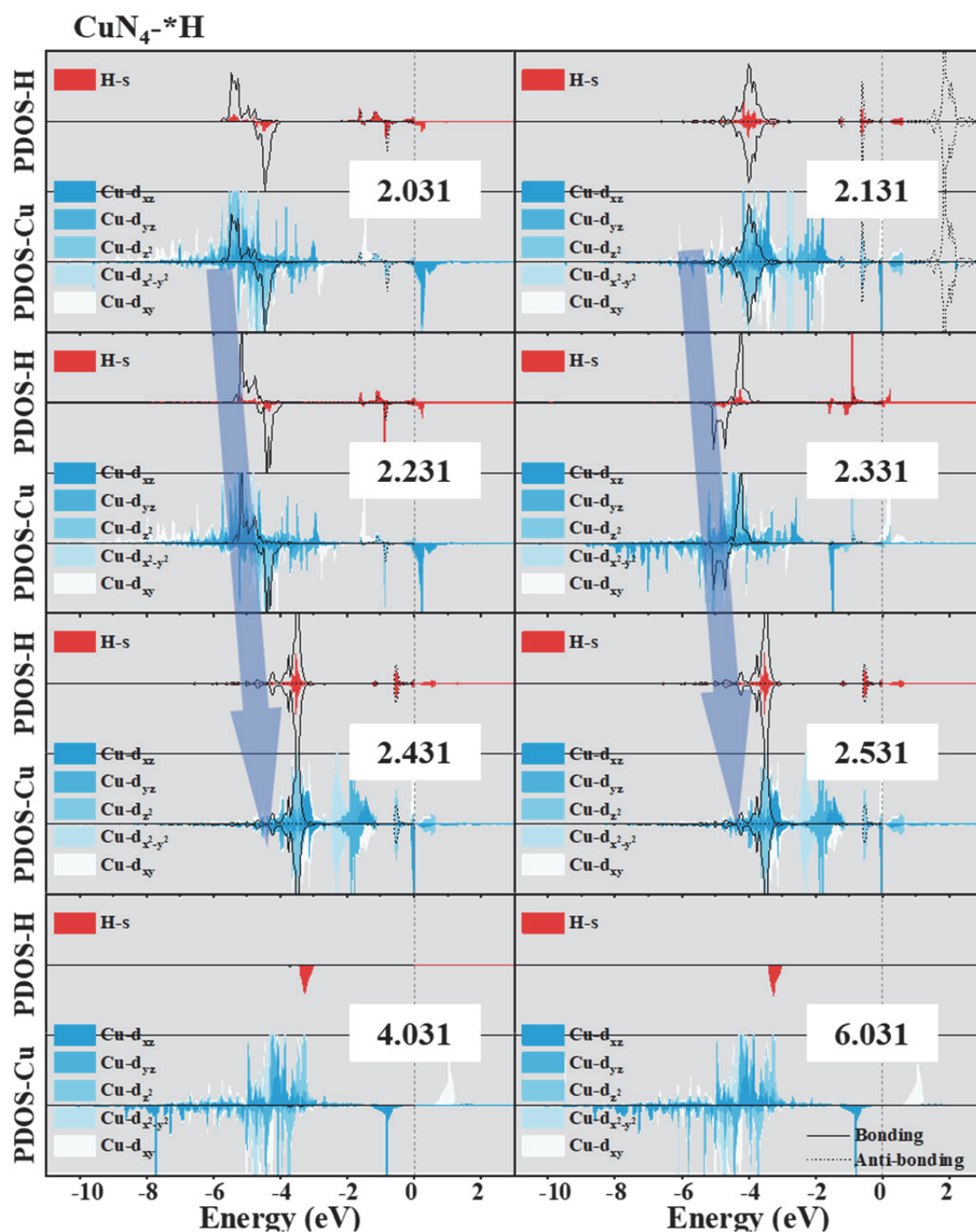


Fig. S21 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CuN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

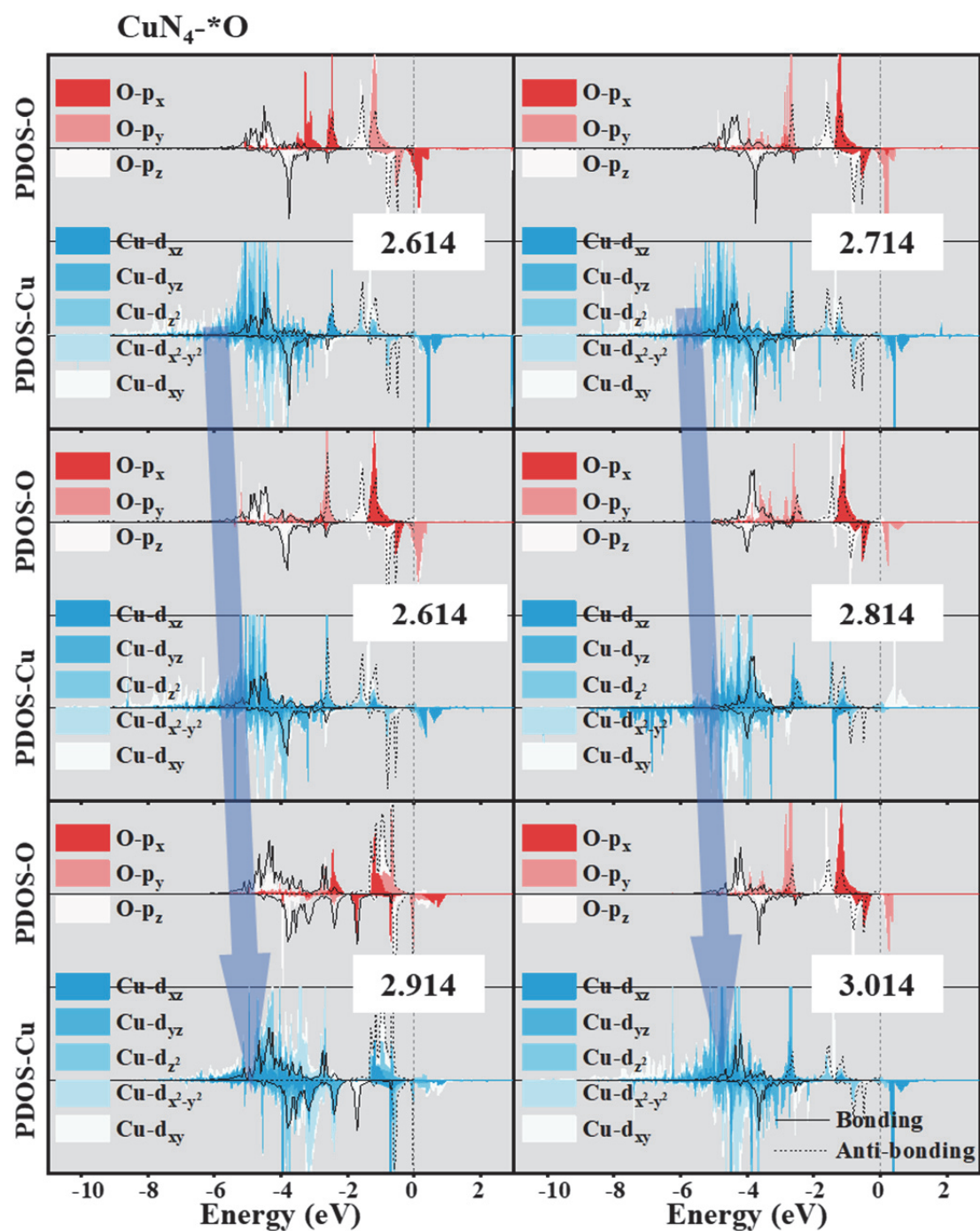


Fig. S22 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CuN_4 and O^* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

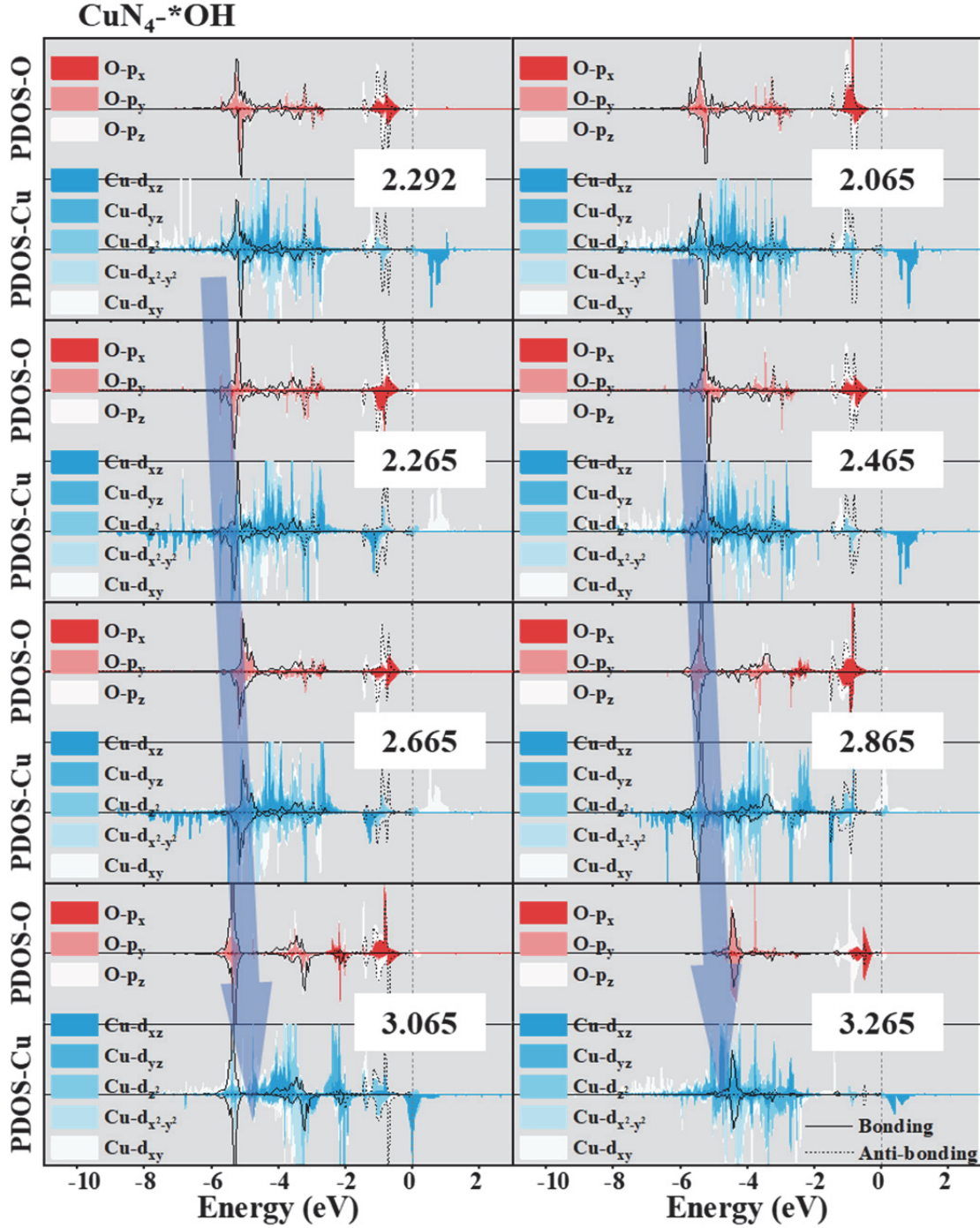


Fig. S23 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of CuN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

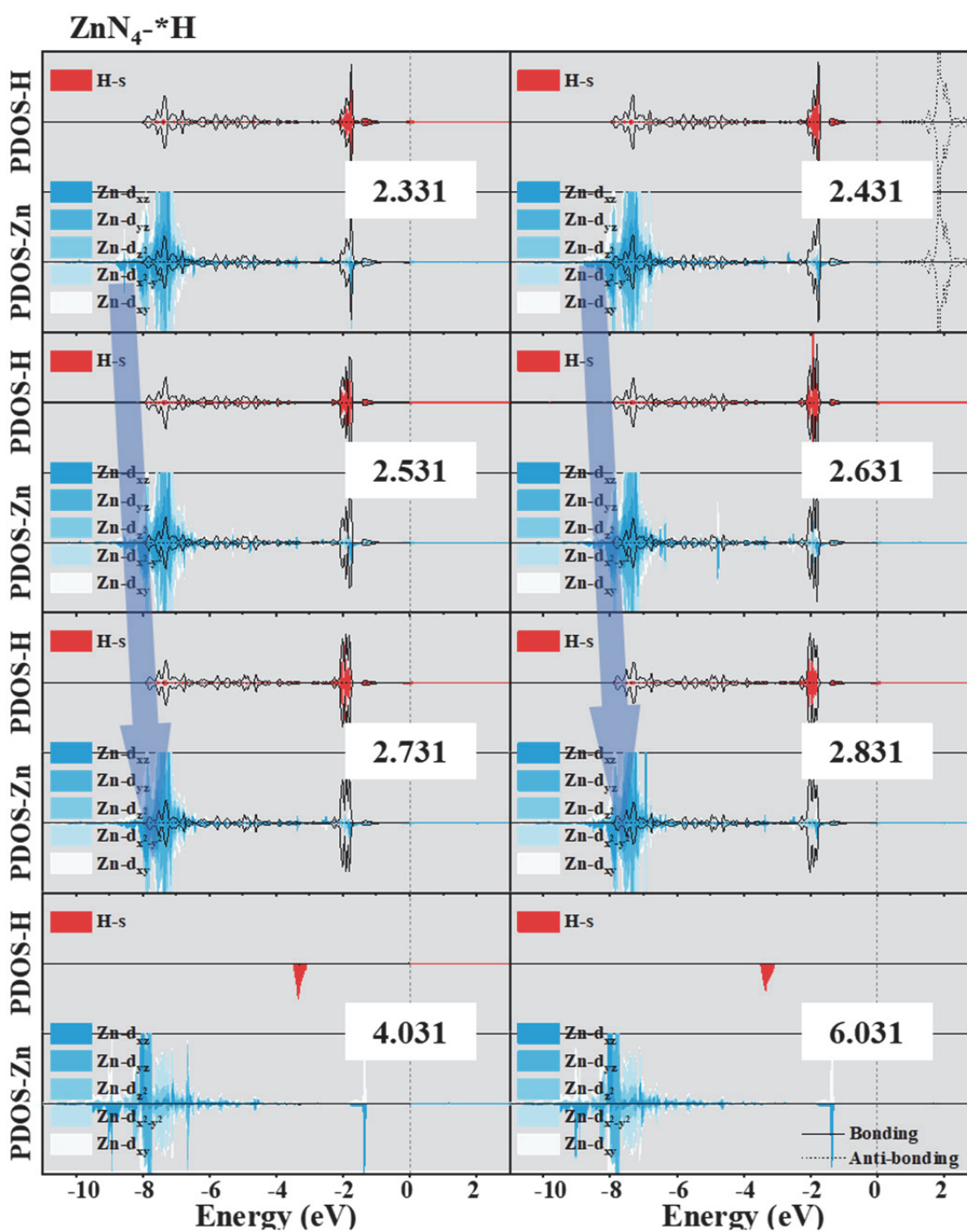


Fig. S24 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of ZnN₄ and H* species, blue-white are metal d orbitals, red-white are H s orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and H, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

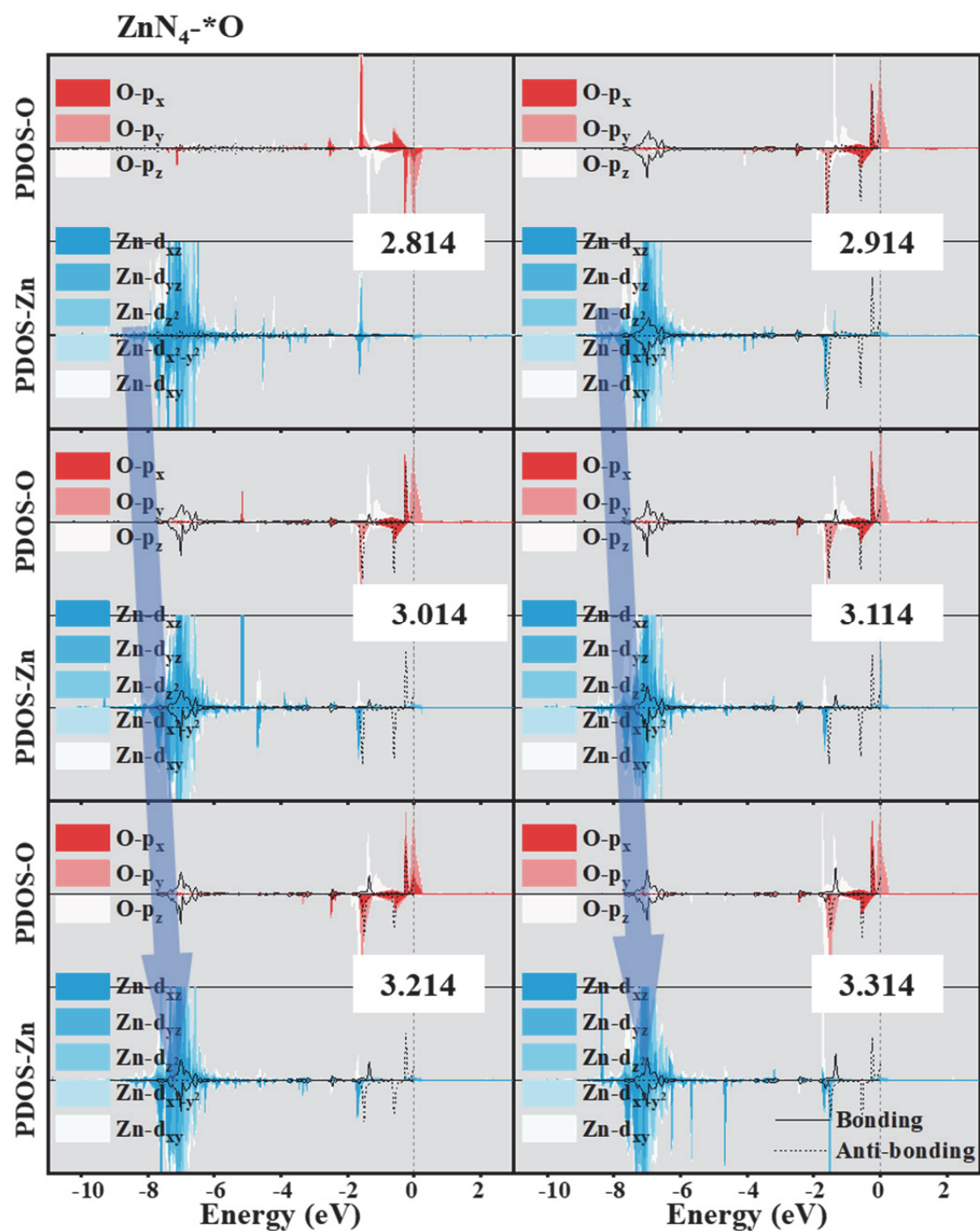


Fig. S25 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of ZnN₄ and O* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

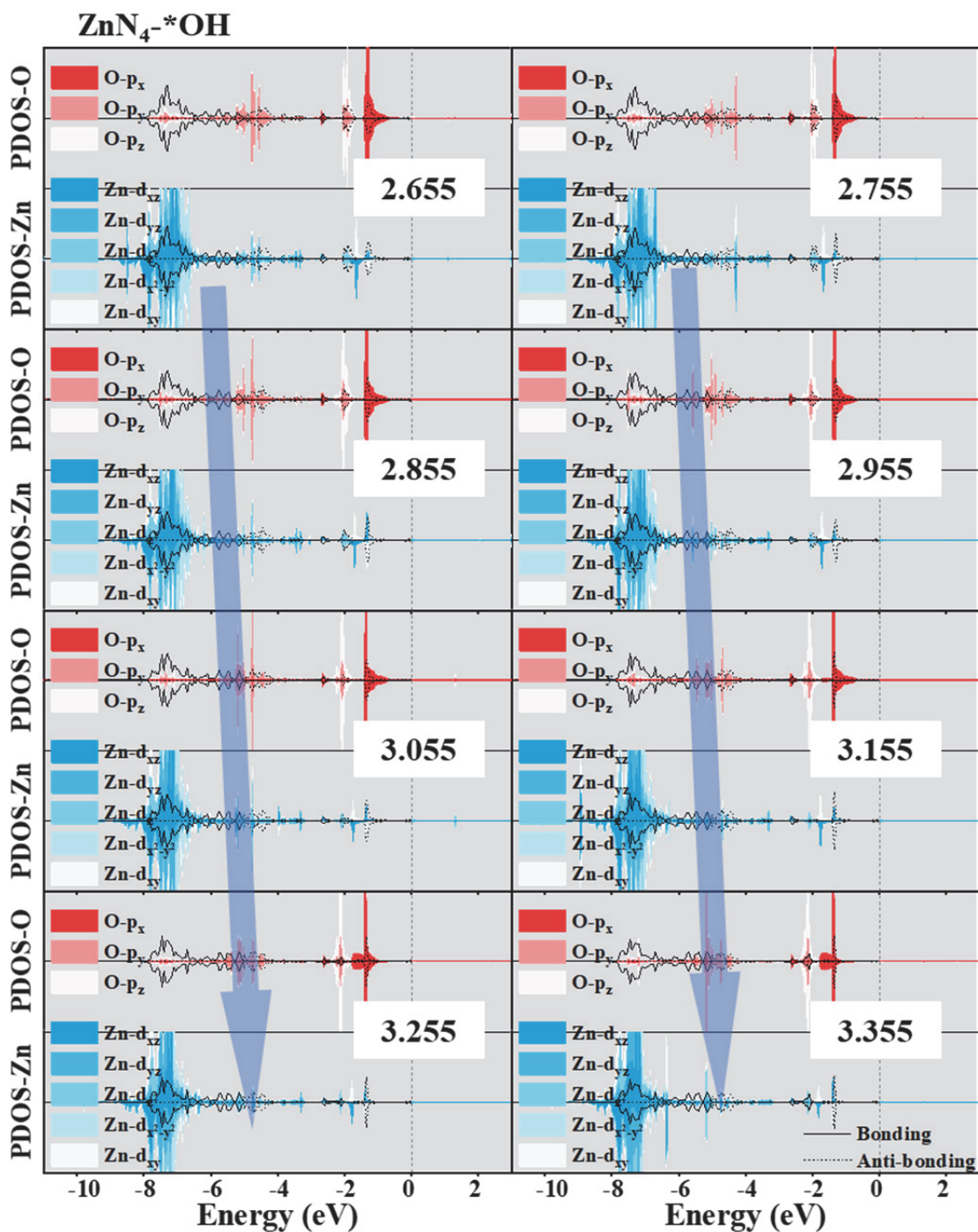


Fig. S26 Bonding information between SACs and adsorbed species. COHP-DOS diagrams of ZnN₄ and OH* species, blue-white are metal d orbitals, red-white are O p orbitals, solid lines represent bonding orbitals, and dashed lines represent antibonding orbitals. The overlapping part of DOS and COHP is the bonding method between metal and O, and the non-overlapping part is the bonding information of metal and other elements. The black letters on a white background represent the height between the adsorbent and the slab.

IIIB	IVB	VB	VIB	VIIB	VIII			IB	IIB		
Sc 0.3	Ti 0.3	V 0.3	Cr 0.1	Mn 0.1	Fe 0.1	Co 0.1	Ni 0.1	Cu 0.1	Zn 0.1	H 0	C 0.6
Y 0.4	Zr 0.4	Nb 0.4	Mo 0.2	Tc 0.2	Ru 0.2	Rh 0.2	Pd 0.2	Ag 0.2	Cd 0.2	O 0.6	N 0.6
La 0.5	Hf 0.5	Ta 0.5	W 0.3	Re 0.3	Os 0.3	Ir 0.3	Pt 0.3	Au 0.3	Hg 0.3		

Sc 0.3	Element α
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Fig. S27 Atomic radius correction parameter. Due to the differences in atomic radii, we need to make corrections to the atomic radius when calculating the relationship between bond length and BASED. The red font in the figure represents the correction parameter α . When calculating the bond length, the corresponding parameter needs to be subtracted. For example, when calculating Fe active site adsorb O*, the bond length needs to be subtracted by 0.1 Å of Fe and 0.6 Å of O (= bond length – 0.1 – 0.6). The black font represents the α of elements that have been calculated and validated, while the gray font represents the parameters that have been calculated but not yet validated.

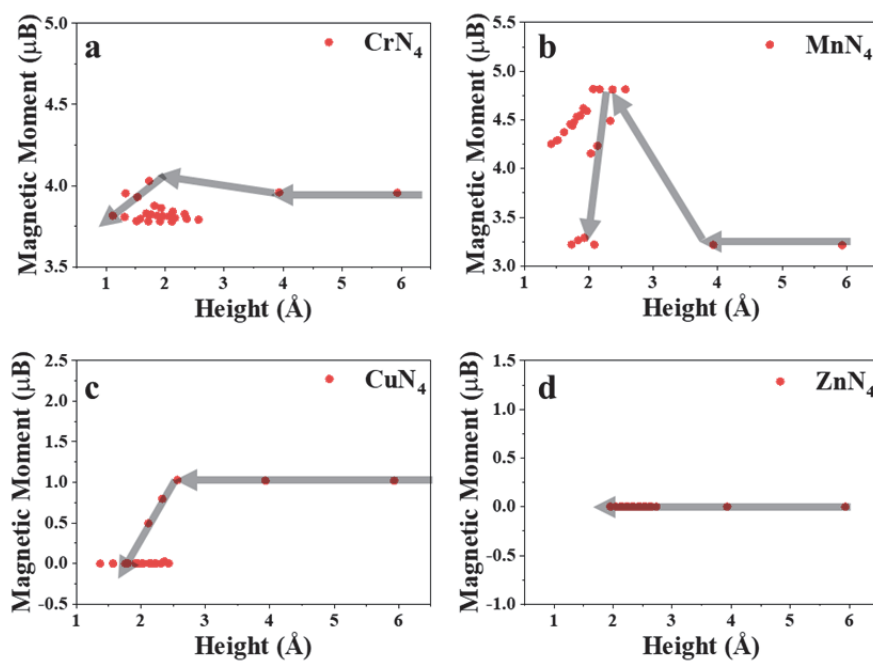


Fig. S28 Illustrations of STS phenomenon. (a-d). Schematic diagram of the relationship between magnetic moment of active center atoms and the height of the adsorbent (H^* , O^* and O^*H) distance slab, based on CrN_4 (a), MnN_4 (b), CuN_4 (c), ZnN_4 (d).

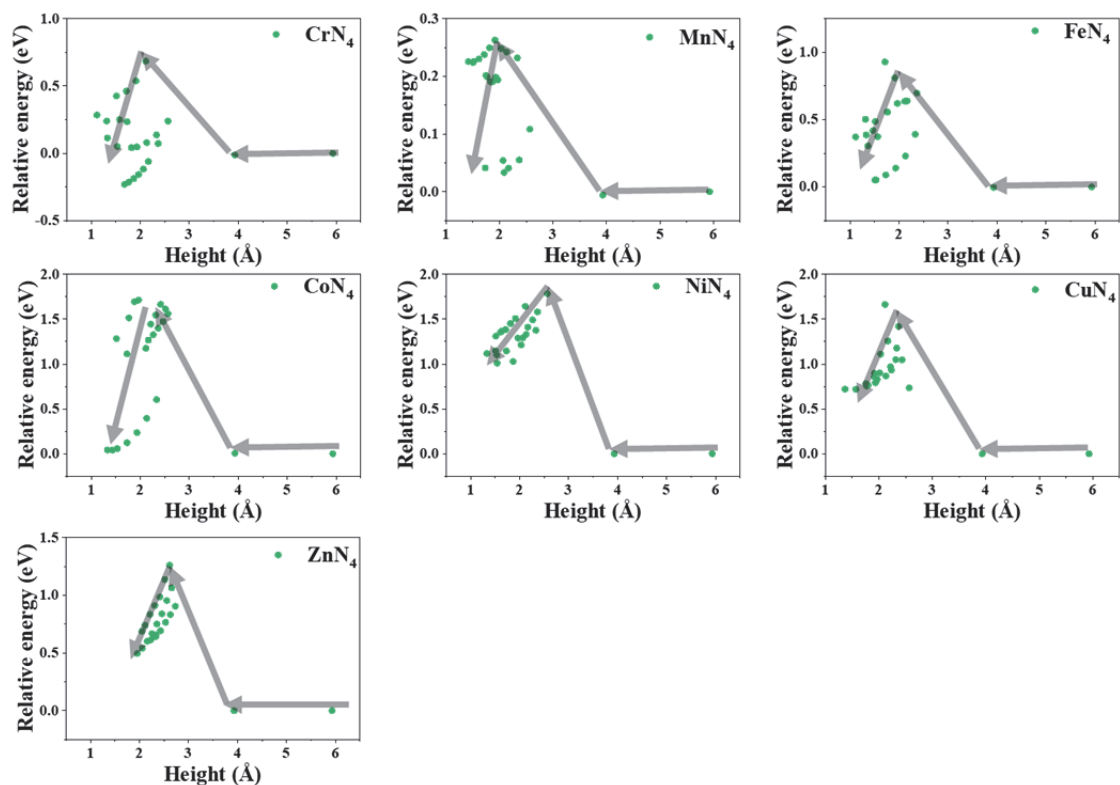


Fig. S29 Illustrations of STS phenomenon. Schematic diagram of the relationship between relative energy of slab and height of the adsorbent (H*, O* and O*H) distance slab, based on CrN₄ ~ ZnN₄. The gray arrow represents the trend of the relative energy of slab with respect to height, high relative energy represents instability.

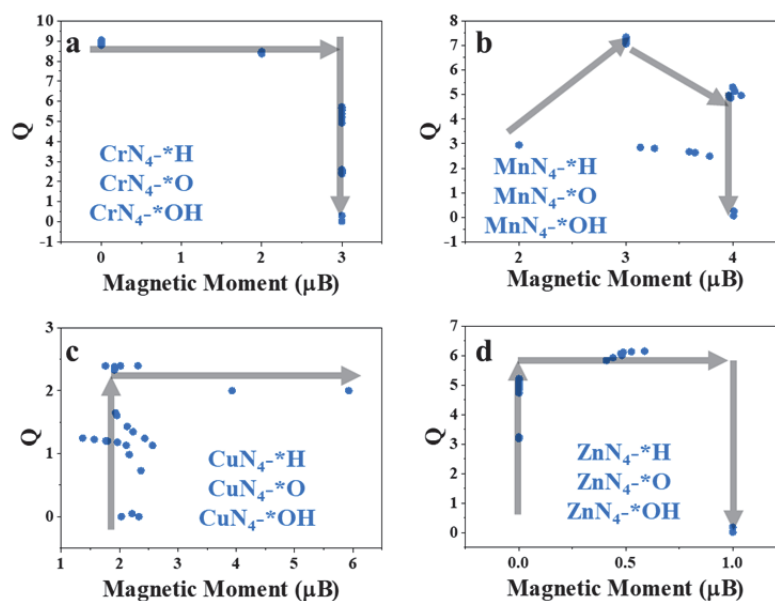


Fig. S30 Illustrations of STS phenomenon. Schematic diagram of the relationship between magnetic moment of active center atoms and Q , based on CrN₄ (a), MnN₄ (b), CuN₄ (c), ZnN₄ (d). High value of Q represents strong adsorption ability of the active center atom. The gray arrow represents the trend of the adsorption ability indicator Q with respect to magnetic moment.

3. Supplementary Table.

Table. S1 D-band center and adsorption energy. The D-band center position ϵ_d and the adsorption energy of H* (ΔE_{*H}) and O*H (ΔE_{*OH}).

Species		ϵ_d	ΔE_{*H}	ΔE_{*O}	ΔE_{*OH}
SACs	CoN ₄	-1.341	-1.955	-4.881	-3.766
	CrN ₄	1.626	-1.967	-5.232	-3.286
	CuN ₄	-3.903	-1.499	-2.865	-2.512
	FeN ₄	-1.000	-1.582	-4.580	-3.167
	MnN ₄	-1.009	-1.536	-4.738	-3.219
	NiN ₄	-1.976	-1.867	-3.697	-2.617
	ScN ₄	2.828	-1.168	-4.342	-3.134
	TiN ₄	2.053	-1.771	-6.349	-3.477
	VN ₄	0.532	-2.292	-6.625	-4.104
	ZnN ₄	-5.967	-2.202	-3.660	-3.192
Bulk	Pt	-2.330	-3.036	-4.227	-2.734
	Ni	-1.424	-2.692	-4.770	-3.017
	Ag	-3.300	-1.743	-2.987	-2.186
	Cu	-2.293	-2.127	-5.209	-2.560

Table. S2 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of CrN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	1.431	1.224	0.053	1.175	0.000	-3.098	-0.201	-2.548	-7.565	2.602	2.346
	1.631	1.220	0.069	1.151	0.002	-3.079	-0.373	-2.571	-5.233	2.584	2.300
	1.831	1.161	0.087	1.112	0.009	-3.021	-0.578	-2.545	-1.502	2.505	2.177
	2.031	1.187	0.096	1.100	0.014	-2.954	-0.474	-2.498	-1.070	2.488	2.177
	2.231	1.177	0.106	1.084	0.016	-2.914	-0.509	-2.464	-0.752	2.457	2.139
	2.431	1.141	0.112	1.066	0.017	-2.832	-0.495	-2.391	-0.599	2.390	2.078
	4.031	0.006	0.001	0.030	0.000	-1.361	-8.578	-3.401	-18.727	0.308	0.034
	6.031	0.000	0.001	0.001	0.000	-19.800	-7.428	-3.332	-16.626	0.028	0.000
	1.919	1.186	0.091	1.102	0.011	-2.970	-0.482	-2.503	-1.218	2.494	2.185
O*	1.814	3.920	0.999	3.731	0.126	-9.617	-0.327	-9.224	-2.105	8.457	6.526
	2.014	3.883	0.990	3.736	0.132	-9.642	-0.345	-9.375	-2.296	8.475	6.497
	2.214	4.182	0.278	4.182	0.278	-9.884	-1.593	-9.884	-1.593	9.043	7.808
	2.414	4.109	0.274	4.109	0.274	-9.867	-1.554	-9.867	-1.552	8.958	7.671
	2.614	4.039	0.268	4.040	0.268	-9.874	-1.536	-9.875	-1.534	8.885	7.542
	2.814	3.954	0.263	3.955	0.263	-9.871	-1.502	-9.871	-1.502	8.791	7.382
	2.278	3.812	0.971	3.634	0.135	-9.659	-0.384	-9.306	-2.178	8.365	6.339
OH*	2.465	2.079	0.807	1.728	0.042	-8.550	-0.840	-8.783	-3.274	5.669	2.959
	2.565	2.042	0.794	1.695	0.043	-8.489	-0.821	-8.744	-3.212	5.601	2.900
	2.665	2.016	0.783	1.673	0.044	-8.452	-0.797	-8.718	-3.205	5.555	2.862
	2.765	1.981	0.770	1.642	0.044	-8.378	-0.775	-8.654	-3.179	5.483	2.808
	2.865	1.931	0.749	1.598	0.044	-8.299	-0.763	-8.629	-3.250	5.394	2.736
	3.065	1.840	0.712	1.509	0.043	-8.079	-0.763	-8.496	-3.410	5.196	2.594
	3.265	1.720	0.663	1.391	0.041	-7.781	-0.816	-8.320	-3.751	4.925	2.407
	2.380	2.113	0.816	1.755	0.041	-8.592	-0.867	-8.810	-3.368	5.724	3.011

Table. S3 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of MnN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	1.831	1.169	0.177	1.162	0.026	-4.064	-0.711	-3.493	-0.286	2.946	2.129
	1.931	0.978	0.468	1.104	0.037	-5.044	-1.028	-3.358	-0.927	2.850	1.577
	2.031	0.960	0.463	1.100	0.037	-5.024	-1.000	-3.245	-0.948	2.810	1.560
	2.131	0.896	0.396	1.077	0.033	-4.895	-0.960	-2.919	-0.928	2.668	1.544
	2.231	0.879	0.374	1.072	0.031	-4.853	-0.926	-2.842	-0.911	2.633	1.545
	2.431	0.802	0.228	1.051	0.026	-4.675	-0.913	-2.574	-0.902	2.494	1.599
	4.031	0.050	0.035	0.000	0.001	-3.570	-3.349	-20.669	-10.730	0.249	0.015
	6.031	0.001	0.000	0.000	0.000	-3.532	-3.518	-20.234	-9.920	0.066	0.000
	2.183	1.166	0.169	1.165	0.024	-4.066	-0.673	-3.499	-0.247	2.949	2.138
O*	2.114	3.170	1.495	3.298	0.111	-9.360	-0.537	-7.698	-3.671	7.338	4.862
	2.214	3.131	1.475	3.259	0.112	-9.336	-0.535	-7.672	-3.717	7.282	4.802
	2.314	3.085	1.453	3.212	0.114	-9.326	-0.535	-7.669	-3.788	7.225	4.731
	2.414	3.047	1.433	3.176	0.115	-9.271	-0.496	-7.638	-3.849	7.166	4.675
	2.514	3.013	1.417	3.142	0.114	-9.301	-0.537	-7.666	-3.938	7.134	4.624
	2.614	2.967	1.394	3.100	0.114	-9.264	-0.494	-7.624	-4.010	7.069	4.559
	2.201	3.135	1.477	3.263	0.112	-9.327	-0.525	-7.666	-3.698	7.285	4.809
OH*	2.465	1.564	1.171	1.473	0.039	-9.413	-1.445	-8.001	-4.925	4.962	1.826
	2.565	1.525	1.109	1.447	0.040	-9.378	-1.453	-7.940	-4.874	4.898	1.823
	2.665	1.501	1.071	1.433	0.040	-9.367	-1.457	-7.911	-4.826	4.863	1.824
	2.765	1.980	1.092	1.514	0.058	-8.354	-1.033	-8.568	-4.860	5.301	2.344
	2.865	1.971	1.072	1.505	0.060	-8.303	-1.040	-8.537	-4.870	5.273	2.344
	3.065	1.902	0.998	1.450	0.060	-8.144	-1.069	-8.412	-4.900	5.131	2.294
	3.265	1.806	0.912	1.385	0.057	-8.016	-1.109	-8.271	-4.818	4.965	2.223
	2.445	1.570	1.191	1.476	0.038	-9.418	-1.442	-8.006	-4.958	4.969	1.817

Table. S4 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of FeN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	1.431	1.334	0.050	1.289	0.136	-3.705	-0.125	-4.055	-0.297	3.181	2.436
	1.631	1.255	0.151	1.294	0.054	-3.903	-0.321	-3.553	-0.159	3.072	2.345
	1.831	1.221	0.164	1.255	0.055	-3.802	-0.353	-3.443	-0.210	2.982	2.257
	2.031	1.014	0.191	1.044	0.029	-3.399	-0.443	-2.994	-0.529	2.544	1.839
	2.231	1.037	0.192	1.067	0.034	-3.403	-0.475	-2.984	-0.534	2.569	1.878
	2.431	1.038	0.192	1.074	0.037	-3.412	-0.540	-2.965	-0.602	2.569	1.882
	4.031	0.004	0.001	0.020	0.001	-3.456	-10.505	-3.317	-3.328	0.263	0.023
	6.031	0.000	0.001	0.001	0.000	-17.417	-8.494	-3.215	-5.210	0.065	0.000
	1.608	1.299	0.053	1.260	0.149	-3.562	-0.143	-3.907	-0.305	3.082	2.357
O*	1.814	3.466	1.639	3.740	0.138	-10.065	-0.798	-8.286	-2.336	8.015	5.429
	2.014	3.420	1.620	3.693	0.142	-10.013	-0.764	-8.225	-2.518	7.939	5.351
	2.214	3.343	1.590	3.618	0.145	-9.942	-0.739	-8.160	-2.743	7.822	5.225
	2.414	3.304	1.596	3.590	0.148	-9.921	-0.700	-8.093	-3.001	7.764	5.150
	2.614	3.143	1.525	3.444	0.146	-9.818	-0.704	-7.952	-3.236	7.530	4.917
	2.814	2.740	1.444	3.449	0.124	-11.064	-0.802	-7.586	-3.925	7.405	4.621
	4.014	0.053	0.002	0.082	0.001	-5.816	-4.624	-3.791	-10.130	0.775	0.132
	2.168	3.361	1.600	3.636	0.146	-9.955	-0.757	-8.175	-2.668	7.848	5.252
OH*	2.065	1.702	1.215	1.777	0.071	-10.309	-1.855	-8.212	-2.776	5.448	2.193
	2.265	1.605	1.057	1.706	0.074	-10.263	-1.910	-8.110	-2.739	5.299	2.179
	2.465	1.640	0.072	1.524	0.969	-7.980	-2.697	-10.158	-1.887	5.152	2.123
	2.665	1.855	1.037	2.027	0.172	-11.195	-2.852	-8.762	-2.046	5.934	2.673
	2.865	1.821	0.900	1.700	0.063	-9.462	-1.365	-8.328	-4.652	5.465	2.558
	3.065	1.722	0.800	1.647	0.063	-9.486	-1.386	-8.218	-4.720	5.335	2.506
	6.065	0.003	0.000	0.005	0.000	-1.235	-9.729	-1.332	-14.947	0.095	0.008
	2.462	1.617	1.071	1.716	0.074	-10.273	-1.906	-8.127	-2.738	5.322	2.189

Table. S5 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of CoN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	1.431	1.335	0.111	1.335	0.111	-3.948	-0.205	-3.948	-0.205	3.240	2.448
	1.631	1.304	0.117	1.305	0.117	-3.834	-0.223	-3.833	-0.223	3.154	2.375
	1.831	1.271	0.122	1.271	0.122	-3.710	-0.260	-3.710	-0.260	3.060	2.297
	2.031	1.224	0.126	1.224	0.125	-3.541	-0.269	-3.541	-0.269	2.932	2.196
	2.231	1.183	0.128	1.183	0.128	-3.383	-0.297	-3.383	-0.297	2.816	2.109
	2.431	1.139	0.129	1.138	0.129	-3.204	-0.302	-3.204	-0.302	2.686	2.019
	4.031	0.005	0.001	0.024	0.008	-2.749	-10.750	-3.179	-1.644	0.264	0.020
	6.031	0.000	0.001	0.001	0.000	-16.009	-8.714	-3.212	-20.417	0.057	0.000
	1.527	1.323	0.114	1.323	0.114	-3.898	-0.214	-3.898	-0.214	3.204	2.418
O*	2.214	3.070	1.837	3.719	0.181	-11.218	-0.954	-8.014	-2.718	7.874	4.772
	2.814	2.837	1.611	3.530	0.173	-11.168	-1.010	-7.828	-2.983	7.561	4.583
	2.914	2.781	1.556	3.485	0.170	-11.184	-1.036	-7.794	-2.975	7.493	4.541
	3.014	2.735	1.515	3.450	0.167	-11.205	-1.069	-7.780	-2.971	7.441	4.503
	3.114	2.684	1.466	3.409	0.164	-11.243	-1.114	-7.772	-2.951	7.386	4.464
	3.214	2.624	1.407	3.363	0.161	-11.275	-1.145	-7.742	-2.894	7.317	4.419
	4.014	0.051	0.018	0.030	0.001	-6.316	-2.126	-8.766	-9.877	0.730	0.062
	2.428	2.984	1.749	3.647	0.180	-11.156	-0.963	-7.933	-2.859	7.747	4.702
OH*	2.465	1.949	1.175	2.113	0.421	-10.987	-2.562	-8.891	-1.271	6.054	2.466
	2.665	1.879	1.070	2.047	0.436	-10.721	-2.471	-8.729	-1.322	5.898	2.420
	2.865	1.774	0.935	1.943	0.463	-10.348	-2.347	-8.487	-1.341	5.659	2.318
	2.965	1.732	0.885	1.898	0.470	-10.189	-2.323	-8.379	-1.399	5.554	2.276
	3.065	1.667	0.810	1.841	0.462	-10.027	-2.288	-8.251	-1.415	5.422	2.236
	3.165	1.601	0.739	1.779	0.450	-9.841	-2.219	-8.119	-1.432	5.283	2.191
	3.265	1.507	0.643	1.696	0.423	-9.689	-2.163	-7.970	-1.438	5.111	2.138
	2.580	1.899	1.104	2.055	0.427	-10.785	-2.488	-8.788	-1.317	5.935	2.422

Table. S6 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of NiN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	1.431	1.237	0.300	1.323	0.184	-5.095	-0.658	-4.599	-0.461	3.480	2.077
	1.631	1.208	0.318	1.301	0.183	-4.918	-0.665	-4.364	-0.452	3.365	2.007
	1.831	1.174	0.327	1.279	0.176	-4.722	-0.656	-4.093	-0.411	3.240	1.951
	2.131	1.101	0.312	1.241	0.149	-4.423	-0.607	-3.613	-0.306	3.019	1.880
	2.231	1.049	0.288	1.217	0.129	-4.298	-0.619	-3.339	-0.271	2.891	1.849
	2.431	0.958	0.245	1.185	0.091	-4.150	-0.534	-2.873	-0.166	2.690	1.807
	4.031	0.005	0.001	0.022	0.008	-2.629	-11.057	-3.173	-1.961	0.245	0.019
	6.031	0.000	0.000	0.000	0.000	-15.133	-9.686	-3.118	-2.472	0.056	0.000
	1.641	1.201	0.320	1.295	0.182	-4.867	-0.675	-4.306	-0.455	3.335	1.993
O*	2.214	2.619	1.658	3.060	0.693	-10.767	-1.320	-8.341	-0.712	7.144	3.329
	2.314	2.598	1.623	3.050	0.689	-10.799	-1.347	-8.341	-0.724	7.128	3.336
	2.414	2.590	1.609	3.049	0.687	-10.826	-1.358	-8.347	-0.730	7.128	3.344
	2.514	2.552	1.559	3.020	0.680	-10.812	-1.382	-8.320	-0.750	7.075	3.334
	2.614	2.526	1.531	2.999	0.676	-10.804	-1.396	-8.295	-0.760	7.037	3.318
	2.814	2.311	1.336	2.783	0.624	-10.624	-1.391	-8.062	-0.772	6.682	3.134
	2.335	2.604	1.622	3.060	0.690	-10.818	-1.358	-8.353	-0.726	7.143	3.352
OH*	2.565	1.294	0.756	1.519	0.468	-8.893	-1.163	-6.988	-0.974	4.560	1.589
	2.665	1.256	0.703	1.498	0.454	-8.901	-1.153	-6.933	-0.959	4.508	1.597
	2.765	1.227	0.656	1.478	0.440	-8.871	-1.160	-6.867	-0.979	4.454	1.609
	2.865	1.178	0.595	1.440	0.417	-8.836	-1.163	-6.774	-1.021	4.364	1.606
	2.965	1.147	0.543	1.418	0.399	-8.784	-1.168	-6.696	-1.046	4.303	1.623
	3.065	1.092	0.475	1.373	0.371	-8.712	-1.166	-6.566	-1.080	4.192	1.619
	3.265	0.990	0.346	1.294	0.315	-8.578	-1.204	-6.321	-1.178	3.986	1.623
	2.208	1.174	0.865	1.244	0.387	-8.837	-1.426	-7.587	-1.164	4.258	1.166

Table. S7 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of CuN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	2.031	0.740	0.172	0.770	0.146	-5.412	-1.338	-4.643	-0.923	2.686	1.192
	2.131	1.068	0.190	1.068	0.190	-4.186	-0.702	-4.189	-0.703	2.946	1.756
	2.231	0.687	0.064	0.736	0.091	-5.171	-1.291	-4.572	-0.869	2.600	1.269
	2.331	0.709	0.033	0.669	0.000	-4.467	-0.839	-4.923	-7.072	2.536	1.345
	2.431	1.116	0.127	1.116	0.127	-3.816	-0.551	-3.816	-0.551	2.894	1.977
	2.531	0.734	0.000	0.736	0.000	-4.144	-8.265	-4.266	-15.297	2.485	1.469
	4.031	0.002	0.001	0.020	0.001	-4.655	-9.690	-3.626	-5.863	0.254	0.020
	6.031	0.000	0.001	0.001	0.000	-17.492	-7.546	-3.268	-11.042	0.052	0.000
	2.051	0.723	0.144	0.759	0.134	-5.369	-1.333	-4.643	-0.921	2.663	1.204
O*	2.614	1.373	0.731	1.381	0.523	-11.446	-1.840	-10.546	-1.204	5.321	1.501
	2.714	1.326	0.693	1.318	0.482	-11.304	-1.925	-10.536	-1.260	5.190	1.469
	2.614	1.530	0.852	1.494	0.578	-11.172	-1.897	-10.525	-1.173	5.525	1.595
	2.814	2.439	1.267	2.597	0.743	-10.483	-1.087	-9.287	-0.677	6.914	3.026
	2.914	1.242	0.626	1.058	0.313	-10.549	-1.763	-11.301	-1.419	4.848	1.360
	3.014	1.135	0.513	1.132	0.343	-11.300	-1.969	-10.592	-1.370	4.831	1.411
	2.457	1.434	0.821	1.402	0.562	-11.256	-1.907	-10.536	-1.189	5.355	1.453
OH*	2.065	1.119	0.722	1.152	0.711	-9.384	-1.668	-8.976	-1.336	4.323	0.838
	2.265	1.132	0.675	1.100	0.680	-8.923	-1.322	-9.307	-1.654	4.280	0.878
	2.465	1.045	0.600	1.081	0.607	-9.265	-1.625	-8.857	-1.305	4.182	0.919
	2.665	1.012	0.529	0.968	0.505	-8.722	-1.299	-9.170	-1.601	4.026	0.946
	2.865	1.398	0.615	1.347	0.590	-8.841	-1.155	-9.196	-1.381	4.819	1.540
	3.065	1.382	0.560	1.501	0.662	-8.807	-1.175	-8.215	-1.130	4.806	1.662
	3.265	0.597	0.120	0.597	0.116	-8.299	-2.495	-8.162	-1.450	3.059	0.958
	2.494	1.031	0.586	1.068	0.596	-9.271	-1.619	-8.859	-1.297	4.159	0.917

Table. S8 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of ZnN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	2.331	1.103	0.000	1.103	0.000	-4.757	-16.012	-4.757	-16.012	3.238	2.206
	2.431	1.127	0.000	1.127	0.000	-4.652	-15.885	-4.652	-15.884	3.237	2.254
	2.531	1.152	0.000	1.153	0.000	-4.531	-15.870	-4.530	-15.869	3.231	2.305
	2.631	1.175	0.000	1.175	0.000	-4.406	-13.220	-4.406	-13.220	3.217	2.349
	2.731	1.186	0.001	1.186	0.001	-4.308	-3.747	-4.308	-3.747	3.196	2.371
	2.831	1.195	0.001	1.195	0.001	-4.243	-1.884	-4.243	-1.875	3.184	2.388
	4.031	0.002	0.001	0.010	0.000	-1.423	-10.907	-3.802	-6.925	0.174	0.011
	6.031	0.000	0.001	0.000	0.000	-15.025	-8.214	-3.323	-12.831	0.010	0.000
	2.439	1.136	0.000	1.136	0.000	-4.609	-15.872	-4.610	-15.871	3.235	2.271
O*	2.814	1.454	0.392	1.421	0.261	-13.446	-1.579	-13.313	-1.038	6.130	2.223
	2.914	1.421	0.236	1.440	0.351	-13.213	-1.000	-13.430	-1.528	6.111	2.274
	3.014	1.408	0.219	1.414	0.321	-13.121	-0.994	-13.436	-1.515	6.063	2.282
	3.114	1.394	0.202	1.387	0.289	-13.004	-0.989	-13.422	-1.503	6.009	2.290
	3.214	1.373	0.183	1.354	0.254	-12.838	-0.992	-13.358	-1.481	5.930	2.290
	3.314	1.356	0.169	1.325	0.223	-12.581	-0.997	-13.170	-1.455	5.833	2.290
	2.747	1.465	0.410	1.431	0.277	-13.449	-1.582	-13.320	-1.047	6.149	2.208
OH*	2.755	1.395	0.502	1.395	0.502	-10.771	-3.098	-10.771	-3.098	5.190	1.785
	2.855	1.365	0.457	1.365	0.458	-10.739	-3.170	-10.738	-3.170	5.140	1.815
	2.955	1.336	0.421	1.336	0.420	-10.693	-3.242	-10.693	-3.242	5.083	1.830
	3.055	1.297	0.376	1.297	0.376	-10.665	-3.328	-10.665	-3.328	5.016	1.842
	3.155	1.254	0.332	1.254	0.332	-10.620	-3.429	-10.620	-3.429	4.935	1.844
	3.255	1.224	0.301	1.224	0.301	-10.465	-3.506	-10.465	-3.506	4.848	1.845
	3.355	1.185	0.274	1.185	0.273	-10.253	-3.532	-10.252	-3.532	4.730	1.824
	2.655	1.409	0.535	1.409	0.535	-10.799	-3.046	-10.799	-3.046	5.213	1.748

Table. S9 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of ScN₄, TiN₄, VN₄.

SACs	Adsorb	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
Species	Species	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
ScN ₄	H*	0.372	0.008	0.372	0.008	-8.572	-7.896	-8.572	-7.896	2.499	0.728
	O*	3.105	0.059	3.099	0.059	-10.565	-5.393	-10.572	-5.381	8.058	6.087
	OH*	1.630	0.046	1.630	0.046	-12.701	-5.312	-12.701	-5.312	6.397	3.168
TiN ₄	H*	0.523	0.058	0.499	0.047	-10.325	-1.288	-11.710	-1.594	3.331	0.917
	O*	3.802	0.131	3.802	0.131	-13.214	-17.275	-13.214	-17.266	9.795	7.341
	OH*	1.733	0.128	1.699	0.126	-10.315	-3.194	-10.366	-3.325	5.887	3.178
VN ₄	H*	0.373	0.045	0.362	0.030	-7.140	-3.381	-8.124	-5.255	2.300	0.659
	O*	4.008	0.132	3.895	0.050	-13.894	-4.584	-14.271	-3.535	10.51	7.720
	OH*	1.630	0.046	1.630	0.046	-12.701	-5.312	-12.701	-5.312	6.397	3.168

Table. S10 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of Pt Bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bonding information for each bond.

Adsorb	Number	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
Species	of bond	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	3-1	0.792	0.069	0.791	0.069	-7.650	-1.745	-7.647	-1.750		
Hollow (hcp)	3-2	0.835	0.074	0.837	0.075	-7.618	-1.815	-7.607	-1.816	6.070	4.495
	3-3	0.836	0.073	0.837	0.073	-7.634	-1.698	-7.629	-1.697		
H*	1-1	1.610	0.049	1.610	0.049	-5.511	-1.209	-5.511	-1.216	4.198	3.121
O*	1-1	3.768	1.755	3.717	0.755	-11.081	-1.248	-10.698	-0.787	8.873	4.974
OH*	1-1	2.328	0.864	2.327	0.862	-9.484	-0.876	-9.487	-0.875	6.530	2.929
OOH*	2-1	2.070	0.649	2.070	0.648	-11.094	-1.424	-11.095	-1.424		
Bridge	2-2	2.095	0.655	2.096	0.655	-11.087	-1.600	-11.079	-1.599	9.404	5.723

Table. S11 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of Ni Bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bonding information for each bond.

Adsorb Species	Num.	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H* Hollow (hcp)	3-1	0.758	0.064	0.762	0.069	-6.580	-1.286	-6.542	-1.389		
	3-2	0.761	0.069	0.768	0.075	-6.561	-1.385	-6.519	-1.584	5.418	4.157
	3-3	0.757	0.063	0.761	0.070	-6.580	-1.284	-6.540	-1.401		
H*	1-1	1.323	0.126	1.344	0.133	-4.538	-1.243	-4.497	-1.688	3.415	2.408
O* Hollow (hcp)	3-1	1.814	0.627	1.831	0.363	-11.104	-1.113	-10.830	-1.497		
	3-2	1.824	0.632	1.840	0.361	-11.102	-1.118	-10.830	-1.520	10.771	7.962
	3-3	1.797	0.617	1.815	0.357	-11.102	-1.103	-10.826	-1.495		
O*	1-1	2.559	1.178	2.609	0.674	-9.766	-1.206	-9.316	-1.312	6.855	3.316
OH* Bridge	2-1	1.523	0.633	1.513	0.377	-9.725	-1.236	-9.584	-1.453		
	2-2	1.538	0.639	1.527	0.384	-9.750	-1.234	-9.610	-1.431	7.504	4.068
OH*	1-1	2.148	0.960	2.156	0.717	-8.596	-1.072	-8.398	-1.099	5.895	2.627
OOH* Hollow (hcp and fcc)-1	6-1	1.858	0.671	1.885	0.376	-11.041	-1.102	-10.705	-1.528		
	6-2	1.491	0.688	1.483	0.414	-10.634	-1.585	-10.491	-1.969		
	6-3	1.849	0.666	1.876	0.372	-11.039	-1.097	-10.704	-1.525	14.033	13.190
	6-4	1.632	0.617	1.645	0.275	-10.914	-1.096	-10.614	-1.492		
	6-5	1.308	0.597	1.297	0.313	-10.406	-1.644	-10.266	-2.109		
	6-6	1.480	0.685	1.471	0.412	-10.666	-1.595	-10.527	-1.980		
OOH* Hollow (hcp and fcc)-2	6-1	1.814	0.627	1.831	0.367	-11.096	-1.189	-10.823	-1.588		
	6-2	1.450	0.662	1.438	0.395	-10.652	-1.622	-10.547	-2.043		
	6-3	1.442	0.647	1.432	0.393	-10.586	-1.646	-10.470	-2.060	14.185	13.430
	6-4	1.422	0.640	1.412	0.387	-10.627	-1.653	-10.513	-2.072		
	6-5	1.814	0.627	1.831	0.367	-11.096	-1.189	-10.823	-1.588		
	6-6	1.823	0.632	1.840	0.371	-11.097	-1.192	-10.825	-1.584		

Table. S12 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of Ag Bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bonding information for each bond.

Adsorb Species	Num.	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H* Hollow (hcp)	3-1	0.630	0.130	0.630	0.130	-6.890	-0.950	-6.890	-0.950		
	3-2	0.639	0.139	0.639	0.139	-6.854	-1.037	-6.854	-1.037	5.034	3.000
	3-3	0.631	0.131	0.631	0.131	-6.888	-0.951	-6.888	-0.951		
H*	1-1	1.140	0.225	1.140	0.225	-5.402	-0.701	-5.402	-0.701	3.464	1.830
O* Hollow (hcp)	3-1	1.160	0.486	1.161	0.488	-9.846	-1.052	-9.844	-1.053		
	3-2	1.193	0.508	1.193	0.510	-9.880	-1.045	-9.878	-1.046	8.143	4.071
	3-3	1.168	0.490	1.168	0.492	-9.851	-1.053	-9.850	-1.054		
O*	1-1	1.825	0.827	1.825	0.827	-10.131	-0.704	-10.131	-0.704	5.984	1.996
OH* Bridge	2-1	1.258	0.613	1.258	0.613	-8.780	-1.205	-8.780	-1.205	4.540	1.289
	2-2	1.268	0.621	1.268	0.621	-8.785	-1.213	-8.785	-1.213		

Table. S13 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of Cu Bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bonding information for each bond.

Adsorb Species	Num.	SpinUpW		SpinDownW		SpinUp ϵ_{C}		SpinDown ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
H*	3-1	0.645	0.135	0.645	0.135	-6.967	-1.258	-6.967	-1.260		
Hollow	3-2	0.648	0.143	0.648	0.143	-6.946	-1.293	-6.946	-1.293	5.093	3.053
(hcp)	3-3	0.645	0.135	0.645	0.135	-6.967	-1.263	-6.967	-1.260		
H*	1-1	1.065	0.227	1.065	0.227	-5.064	-1.116	-5.064	-1.116	3.207	1.676
O*	3-1	1.514	0.633	1.514	0.633	-10.455	-1.244	-10.455	-1.244		
Hollow	3-2	1.529	0.646	1.529	0.646	-10.445	-1.257	-10.445	-1.257	9.513	5.291
(hcp)	3-3	1.515	0.634	1.515	0.634	-10.455	-1.243	-10.455	-1.243		
O*	1-1	1.747	0.887	1.747	0.887	-8.766	-1.367	-8.766	-1.367	5.311	1.721
OH*	2-1	0.855	0.406	0.855	0.406	-10.769	-1.401	-10.769	-1.401		
Bridge	2-2	1.421	0.629	1.421	0.629	-10.509	-1.028	-10.509	-1.028	8.277	3.707
O*	3-1	1.064	0.451	1.064	0.451	-11.072	-0.996	-11.072	-0.996		
Hollow	3-2	0.818	0.415	0.818	0.415	-10.635	-1.392	-10.635	-1.392	7.874	3.271
(hcp)	3-3	1.142	0.522	1.142	0.522	-10.641	-1.160	-10.641	-1.160		

Table. S14 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in CrN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	1.431	1.553	3.952	3.000	-1.828	0.115
	1.631	1.572	3.930	3.000	-1.891	0.050
	1.831	1.613	4.029	3.002	-2.145	0.235
	2.031	1.632	3.862	3.000	-1.981	0.048
	2.231	1.656	3.841	3.000	-1.999	0.079
	2.431	1.688	3.825	3.000	-2.000	0.137
	4.031	4.007	3.959	3.000	-0.041	-0.013
	6.031	6.013	3.956	3.000	-0.025	0.000
	1.919	1.628	3.876	3.000	-1.967	0.042
O*	1.814	1.633	3.816	2.000	-5.069	0.284
	2.014	1.640	3.807	2.000	-5.149	0.240
	2.214	1.587	3.782	0.000	-5.456	0.425
	2.414	1.599	3.780	0.000	-5.507	0.462
	2.614	1.610	3.780	0.000	-5.515	0.538
	2.814	1.623	3.779	0.000	-5.497	0.684
	2.278	1.654	3.796	2.000	-5.232	0.250
	2.465	1.903	3.823	3.000	-3.299	0.107
OH*	2.565	1.915	3.816	3.000	-3.311	0.132
	2.665	1.923	3.813	3.000	-3.310	0.163
	2.765	1.935	3.811	3.000	-3.311	0.203
	2.865	1.952	3.802	3.000	-3.306	0.259
	3.065	1.988	3.796	3.000	-3.273	0.393
	3.265	2.041	3.790	3.000	-3.197	0.560
	2.380	1.893	3.829	3.000	-3.286	0.089

Table. S15 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in MnN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	1.831	1.550	3.221	2.000	-1.548	0.041
	1.931	1.654	3.264	3.136	-1.689	0.191
	2.031	1.663	3.289	3.267	-1.718	0.199
	2.131	1.704	4.155	3.593	-1.779	0.248
	2.231	1.716	4.234	3.647	-1.772	0.242
	2.431	1.773	4.490	3.784	-1.729	0.232
	4.031	4.014	3.217	4.012	-0.028	-0.006
	6.031	6.013	3.213	4.008	-0.009	0.000
	2.183	1.542	3.219	2.000	-1.536	0.033
O*	2.114	1.694	4.253	3.000	-4.734	0.226
	2.214	1.702	4.290	3.000	-4.739	0.226
	2.314	1.712	4.373	3.000	-4.730	0.230
	2.414	1.720	4.455	3.000	-4.704	0.238
	2.514	1.727	4.530	3.000	-4.670	0.250
	2.614	1.736	4.618	3.000	-4.616	0.263
	2.201	1.701	4.286	3.000	-4.738	0.224
OH*	2.465	1.963	4.474	3.967	-3.215	0.152
	2.565	1.977	4.543	3.975	-3.203	0.143
	2.665	1.984	4.592	3.982	-3.183	0.146
	2.765	1.921	4.816	4.000	-3.024	0.007
	2.865	1.926	4.814	4.000	-3.023	-0.006
	3.065	1.952	4.811	4.020	-3.034	0.008
	3.265	1.981	4.813	4.079	-3.035	0.061
	2.445	1.960	4.440	3.966	-3.219	0.154

Table. S16 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in FeN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	1.431	1.467	2.352	-1.000	-1.882	0.387
	1.631	1.498	1.963	1.000	-1.587	0.053
	1.831	1.529	1.958	1.000	-1.607	0.089
	2.031	1.687	1.948	1.001	-1.513	0.140
	2.231	1.678	-1.943	1.000	-1.547	0.230
	2.431	1.680	-1.941	1.000	-1.568	0.391
	4.031	4.013	1.970	1.000	-0.037	-0.003
	6.031	6.014	1.967	0.998	-0.012	0.000
	1.608	1.495	1.962	-1.000	-1.582	0.051
O*	1.814	1.655	-1.927	2.000	-4.447	0.372
	2.014	1.664	2.232	2.000	-4.656	0.503
	2.214	1.678	-2.133	2.000	-4.642	0.486
	2.414	1.686	-1.844	2.000	-5.007	0.929
	2.614	1.710	-2.183	2.000	-4.781	0.810
	2.814	1.724	3.844	4.000	-4.857	0.637
	4.014	3.935	1.964	3.304	-0.401	-0.003
	2.168	1.674	-1.919	2.000	-4.580	0.418
OH*	2.065	1.891	-1.917	2.990	-2.970	0.373
	2.265	1.922	1.906	2.997	-3.167	0.557
	2.465	1.946	1.910	-3.000	-3.193	0.621
	2.665	1.874	3.862	5.000	-3.312	0.579
	2.865	1.903	3.836	3.121	-3.254	0.640
	3.065	1.923	3.836	3.231	-3.272	0.697
	6.065	5.990	1.963	1.754	-0.068	0.000
	2.462	1.918	1.904	2.997	-3.167	0.556

Table. S17 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in CoN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	1.431	1.420	1.004	0.000	-1.942	0.043
	1.631	1.445	1.003	0.000	-1.966	0.057
	1.831	1.475	1.000	0.000	-1.972	0.123
	2.031	1.519	0.998	0.000	-1.955	0.235
	2.231	1.558	0.996	0.000	-1.926	0.396
	2.431	1.608	0.993	0.000	-1.863	0.602
	4.031	4.011	1.005	0.000	0.007	0.006
	6.031	6.014	1.006	0.000	-0.010	0.000
	1.527	1.430	1.004	0.000	-1.955	0.041
O*	2.214	1.658	2.884	3.000	-5.000	1.283
	2.814	1.693	3.156	3.000	-4.840	1.175
	2.914	1.701	2.893	3.000	-5.040	1.444
	3.014	1.708	2.888	2.999	-5.050	1.546
	3.114	1.715	2.890	3.000	-5.060	1.665
	3.214	1.723	3.151	3.000	-4.876	1.612
	4.014	3.940	1.008	5.005	-0.460	0.000
	2.428	1.672	3.063	3.000	-4.881	1.114
OH*	2.465	1.831	0.901	3.942	-3.557	1.516
	2.665	1.855	0.896	3.881	-3.787	1.715
	2.865	1.890	2.887	3.774	-3.311	1.269
	2.965	1.905	2.895	3.751	-3.329	1.329
	3.065	1.925	2.894	3.722	-3.344	1.400
	3.165	1.946	2.887	3.659	-3.351	1.475
	3.265	1.972	2.887	3.586	-3.351	1.564
	2.580	1.850	-1.082	3.892	-3.766	1.697

Table. S18 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in NiN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	1.431	1.417	1.996	1.000	-1.936	1.118
	1.631	1.441	1.979	1.000	-1.959	1.101
	1.831	1.468	-1.779	1.000	-2.001	1.145
	2.131	1.516	1.963	1.000	-1.979	1.213
	2.231	1.549	1.596	1.000	-2.044	1.329
	2.431	1.588	1.909	1.000	-1.972	1.376
	4.031	4.014	0.000	-1.000	-0.035	0.000
	6.031	6.015	0.000	-1.000	-0.008	0.000
	1.641	1.449	2.000	1.000	-1.867	1.009
O*	2.214	1.730	-1.942	2.250	-3.629	1.312
	2.314	1.736	1.939	2.308	-3.684	1.355
	2.414	1.738	1.937	2.346	-3.708	1.385
	2.514	1.748	1.933	2.378	-3.755	1.452
	2.614	1.753	1.932	2.397	-3.777	1.508
	2.814	1.798	1.927	2.312	-3.787	1.643
	2.335	1.735	1.938	2.325	-3.697	1.369
OH*	2.565	1.962	0.001	1.044	-2.536	1.028
	2.665	1.975	1.934	1.044	-2.762	1.290
	2.765	1.988	1.931	1.044	-2.775	1.345
	2.865	2.008	1.929	1.038	-2.783	1.411
	2.965	2.024	1.926	1.034	-2.794	1.493
	3.065	2.049	1.923	1.026	-2.795	1.581
	3.265	2.096	1.917	1.016	-2.776	1.783
	2.208	2.014	-1.759	1.001	-2.617	1.147

Table. S19 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in CuN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	2.031	1.623	0.000	1.649	-1.477	0.791
	2.131	1.519	0.000	0.000	-1.673	1.110
	2.231	1.652	0.000	1.432	-1.566	0.866
	2.331	1.684	0.000	-1.349	-1.613	0.933
	2.431	1.536	0.798	0.000	-1.737	1.178
	2.531	1.714	0.000	-1.243	-1.654	1.046
	4.031	4.013	1.020	-2.000	-0.040	0.000
	6.031	6.013	1.020	-2.001	-0.012	0.000
	2.051	1.630	0.000	1.608	-1.499	0.809
O*	2.614	1.996	0.000	2.385	-2.907	0.858
	2.714	2.024	0.000	2.394	-2.928	0.900
	2.614	1.961	0.000	2.332	-2.955	0.896
	2.814	1.758	0.495	1.133	-3.774	1.664
	2.914	2.095	0.000	0.047	-2.878	0.970
	3.014	2.102	0.000	2.396	-2.918	1.049
	2.457	1.983	0.000	2.393	-2.865	0.781
OH*	2.065	1.994	0.000	1.248	-2.349	0.720
	2.265	2.007	0.000	-1.227	-2.431	0.718
	2.465	2.033	0.000	1.204	-2.501	0.756
	2.665	2.070	0.000	-1.181	-2.551	0.828
	2.865	1.955	0.000	-0.986	-2.721	1.257
	3.065	1.960	-0.029	0.730	-2.721	1.420
	3.265	2.321	-1.029	1.131	-2.068	0.735
	2.494	2.038	0.000	1.201	-2.512	0.763

Table. S20 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in FeN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
H*	2.331	1.566	0.000	0.000	-2.135	0.613
	2.431	1.574	0.000	0.000	-2.184	0.641
	2.531	1.585	0.000	0.000	-2.244	0.694
	2.631	1.599	0.000	0.000	-2.305	0.766
	2.731	1.612	0.000	0.000	-2.349	0.833
	2.831	1.621	0.000	0.000	-2.384	0.904
	4.031	4.012	0.000	-1.000	-0.038	-0.001
	6.031	6.013	0.000	-1.000	-0.016	0.000
	2.439	1.578	0.000	0.000	-2.202	0.658
O*	2.814	1.843	0.000	-0.526	-3.726	0.740
	2.914	1.844	0.000	-0.488	-3.808	0.834
	3.014	1.850	0.000	0.476	-3.855	0.910
	3.114	1.856	0.000	-0.481	-3.886	0.986
	3.214	1.865	0.000	-0.440	-3.970	1.137
	3.314	1.876	0.000	-0.409	-4.000	1.261
	2.747	1.840	0.000	0.588	-3.660	0.688
OH*	2.755	1.894	0.000	0.000	-3.241	0.540
	2.855	1.904	0.000	0.000	-3.290	0.601
	2.955	1.914	0.000	0.000	-3.325	0.666
	3.055	1.925	0.000	0.000	-3.365	0.749
	3.155	1.939	0.000	0.000	-3.395	0.838
	3.255	1.953	0.000	0.000	-3.433	0.954
	3.355	1.972	0.000	0.000	-3.443	1.063
	2.655	1.888	0.000	0.000	-3.192	0.494

Table. S21 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in ScN₄, TiN₄, VN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent.

SACs Species	Adsorb Species	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy
ScN ₄	H*	1.957	1.000	0.000	-1.168
	O*	1.831	0.000	0.000	-4.342
	OH*	2.009	0.000	0.000	-3.134
TiN ₄	H*	1.864	0.000	1.000	-1.771
	O*	1.682	0.999	0.000	-6.349
	OH*	2.000	0.999	0.987	-3.477
VN ₄	H*	1.953	2.764	2.000	-2.292
	O*	1.635	3.000	3.000	-6.625
	OH*	2.009	2.114	2.000	-4.104

Table. S22 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in Pt bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bond, e magnetic moment and energy information for each bond. Mag represents the magnetic moment, which is divided into the magnetic moment of Pt Bulk slab during adsorption process (Mag of slab), and the total magnetic moment of Pt Bulk slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent.

Adsorb Species	Num.	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy
H*	3-1	1.870			
Hollow (hcp)	3-2	1.847	0.000	2.126	-3.036
	3-3	1.845			
H*	1-1	1.548	0.000	0.334	-3.124
O*	1-1	1.823	0.000	2.178	-4.227
OH*	1-1	1.986	0.000	0.964	-2.734
OOH* Bridge	2-1	2.012			
	2-2	2.008	0.000	0.157	-5.687

Table. S23 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in Ni bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bond, e magnetic moment and energy information for each bond. Mag represents the magnetic moment, which is divided into the magnetic moment of Pt Bulk slab during adsorption process (Mag of slab), and the total magnetic moment of Pt Bulk slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent.

Adsorb Species	Num.	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy
H* Hollow (hcp)	3-1	1.693			
	3-2	1.694	40.362	39.658	-3.242
	3-3	1.694			
H*	1-1	1.462	40.363	39.938	-2.692
O* Hollow (hcp)	3-1	1.844			
	3-2	1.842	40.381	40.559	-6.153
	3-3	1.847			
O*	1-1	1.768	40.308	41.215	-4.770
OH* Bridge	2-1	1.931			
	2-2	1.927	40.359	40.430	-3.504
OH*	1-1	1.824	40.369	40.587	-3.017
OOH* Hollow (hcp and fcc)-1	6-1	1.837			
	6-2	1.921			
	6-3	1.838	40.391	41.044	-9.454
	6-4	1.897			
	6-5	1.982			
	6-6	1.924			
OOH* Hollow (hcp and fcc)-2	6-1	1.846			
	6-2	1.933			
	6-3	1.936	40.387	40.755	-9.597
	6-4	1.942			
	6-5	1.846			
	6-6	1.844			

Table. S24 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in Ag bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bond, e magnetic moment and energy information for each bond. Mag represents the magnetic moment, which is divided into the magnetic moment of Pt Bulk slab during adsorption process (Mag of slab), and the total magnetic moment of Pt Bulk slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent.

Adsorb Species	Num.	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy
H* Hollow (hcp)	3-1	1.917			
	3-2	1.914	0.000	0.000	-2.220
	3-3	1.916			
H*	1-1	1.661	0.000	0.000	-1.743
O* Hollow (hcp)	3-1	2.176			
	3-2	2.164	0.000	-0.048	-4.084
	3-3	2.173			
O*	1-1	1.982	0.000	0.000	-2.987
OH* Bridge	2-1	2.148			
	2-2	2.145	0.000	0.000	-2.186

Table. S25 Relevant data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in Cu bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bond, e magnetic moment and energy information for each bond. Mag represents the magnetic moment, which is divided into the magnetic moment of Pt Bulk slab during adsorption process (Mag of slab), and the total magnetic moment of Pt Bulk slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent.

Adsorb Species	Num.	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy
H* Hollow (hcp)	3-1	1.728			
	3-2	1.728	0.000	0.000	-2.724
	3-3	1.728			
H*	1-1	1.511	0.000	0.000	-2.127
O* Hollow (hcp)	3-1	1.918			
	3-2	1.915	0.000	0.000	-5.209
	3-3	1.918			
O*	1-1	1.888	0.000	0.000	-2.560
OH* Bridge	2-1	2.108			
	2-2	1.933	0.000	0.000	-5.552
O* Hollow (hcp)	3-1	2.036			
	3-2	2.129	0.000	0.001	-5.835
	3-3	2.028			

Table. S26 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of FeN_4 .

Adsorb Species	Height	Spin Up W		Spin Down W		Spin Up ϵ_{C}		Spin Down ϵ_{C}		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
NH_2^*	2.119	1.812	0.598	1.703	0.114	-6.719	-0.966	-6.351	-1.341	4.871	2.804
	2.319	1.735	0.590	1.610	0.108	-6.564	-0.956	-6.236	-1.322	4.704	2.647
	2.519	1.635	0.577	1.496	0.097	-6.393	-0.957	-6.104	-1.319	4.502	2.458
	2.719	1.505	0.553	1.351	0.083	-6.205	-0.990	-5.945	-1.344	4.246	2.220
	2.819	1.315	0.510	1.153	0.062	-5.922	-1.029	-5.697	-1.430	3.869	1.896
	3.119	1.534	0.639	1.815	0.089	-8.726	-2.324	-6.014	-0.822	5.085	2.621
	6.152	0.004	0.001	0.004	0.000	-3.179	-3.665	-1.757	-4.423	0.149	0.007
	2.211	1.793	0.603	1.676	0.115	-6.661	-0.966	-6.308	-1.371	4.822	2.751
CH_3^*	2.119	1.438	0.327	1.372	0.111	-5.189	-1.981	-4.700	-1.633	3.839	2.372
	2.319	1.387	0.326	1.316	0.104	-5.103	-2.012	-4.603	-1.666	3.737	2.272
	2.519	1.314	0.322	1.238	0.093	-4.958	-2.014	-4.457	-1.667	3.582	2.136
	2.719	1.214	0.298	1.152	0.079	-4.847	-1.983	-4.317	-1.703	3.403	1.989
	2.919	1.046	0.322	1.302	0.119	-7.736	-3.172	-4.647	-0.983	3.909	1.906
	3.119	1.008	0.220	1.271	0.107	-7.476	-3.757	-4.463	-0.911	3.759	1.951
	6.152	0.005	0.000	0.003	0.000	-5.725	-10.162	-11.229	-12.254	0.261	0.008
	2.130	1.411	0.324	1.345	0.107	-5.156	-2.003	-4.669	-1.661	3.792	2.324

Table. S27 Relevant data for BASED and ICOHP. The area weight of bonding orbital area and anti-bonding orbital (W_{Bond} and $W_{\text{Anti-Bond}}$), center value of bonding orbital area and anti-bonding orbital ($\epsilon_{\text{C-bond}}$ and $\epsilon_{\text{C-Antibond}}$), and value of Q data are obtained by BASED theory, and the last column is the data for -ICOHP, which all base on the adsorption process of RuO₂ Bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bonding information for each bond.

Adsorb Species	Num.	Spin Up W		Spin Down W		Spin Up ϵ_c		Spin Down ϵ_c		Q	-ICOHP
		Bond.	Anti.	Bond.	Anti.	Bond.	Anti.	Bond.	Anti.		
O*	1	3.916	1.414	3.965	0.398	-10.641	-0.732	-10.024	-0.887	9.100	6.069
O*	2	4.065	0.723	4.032	1.128	-10.323	-0.767	-10.564	-0.657	9.265	6.245
O*	3	3.696	1.057	3.725	0.588	-9.923	-0.632	-9.599	-0.743	8.575	5.776
O*	4	3.962	1.259	3.995	0.461	-9.943	-0.483	-9.477	-0.708	8.843	6.237
O*	5	0.001	0.000	0.000	0.000	-3.111	-15.944	-4.675	-3.138	0.059	0.001
O*	6	4.029	0.831	3.990	1.111	-10.299	-0.654	-10.569	-0.534	9.209	6.077

Table. S28 Relevant validation set data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in FeN₄. Mag represents the magnetic moment, which is divided into the magnetic moment of SACs slab during adsorption process (Mag of slab), and the total magnetic moment of SACs slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent. Relative Energy represents the energy of the SAC slab during the adsorption process relative to the SAC slab without adsorption (at 6 Å), positive value represents instability, while negative value represents it is more stability.

Adsorb Species	Height	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy	Relative Energy
NH ₂ *	2.119	1.872	0.006	1.000	-2.690	-2.659
	2.319	1.899	-0.008	1.000	-2.713	-2.682
	2.519	1.934	1.889	1.000	-2.189	-2.157
	2.719	1.982	1.960	1.000	-2.126	-2.094
	2.819	2.060	-1.925	1.000	-2.081	-2.049
	3.119	1.943	4.217	5.000	-2.798	-2.766
	6.152	5.996	1.968	3.000	-0.032	0.000
	2.211	1.882	-0.068	1.000	-2.773	-2.741
CH ₃ *	2.119	1.975	2.216	1.000	-2.276	-1.859
	2.319	2.000	1.961	1.000	-1.906	-1.489
	2.519	2.035	1.889	1.000	-1.924	-1.507
	2.719	2.076	1.960	1.000	-1.868	-1.452
	2.919	2.054	3.858	4.953	-2.013	-1.596
	3.119	2.084	4.223	4.972	-2.095	-1.678
	6.152	6.008	1.968	2.997	-0.417	0.000
	2.130	1.986	1.941	1.000	-1.911	-1.495

Table. S29 Relevant validation set data of Bond length, Magnetic moment and Energy. The bond length, magnetic moment, and energy information obtained during the calculation of adsorption process in RuO₂ bulk. Num. represents the serial number of different bonds in the adsorbent. Due to some adsorption methods causing the adsorbed atoms to bond with multiple active center atoms, it is necessary to calculate the bond, e magnetic moment and energy information for each bond. Mag represents the magnetic moment, which is divided into the magnetic moment of Pt Bulk slab during adsorption process (Mag of slab), and the total magnetic moment of RuO₂ Bulk slab with adsorbent during adsorption process (Mag of ado). Adsorption Energy represents the adsorption energy of the active center atom on the adsorbent.

Adsorb Species	Num.	Bond length	Mag of slab	Mag of Ado.	Adsorption Energy
O*	1	1.761	3.201	0.000	-6.229
O*	2	1.751	5.910	-0.620	-5.890
O*	3	1.814	7.314	0.573	-5.724
O*	4	1.773	12.028	-3.135	-5.881
O*	5	6.199	5.116	-0.206	-0.589
O*	6	1.753	6.960	2.186	-5.701

Table. S30 Adsorbent energy data. The energy data and temperature correction data for various adsorbents.

Adsorbents species	E_{ado^*} (eV)	Magnetic moment (μB)
H*	-1.10229	1
O*	-1.50645	2
OH*	-7.93965	1.0004
OOH*	-13.2976	1

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