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Hydrogen Peroxide Electrochemical Synthesis on Hybrid Double-Atoms (Pd-Cu) doped N vacancy g-C_3N_4: A Novel Design Strategy for Electrocatalysts Screening

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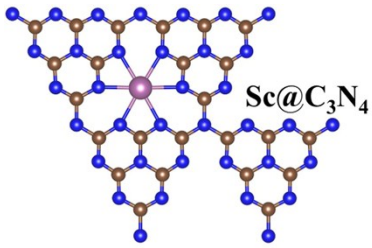
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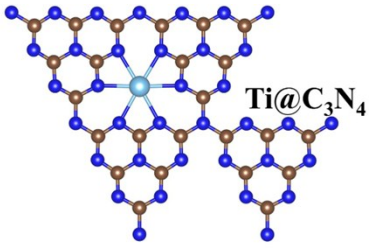
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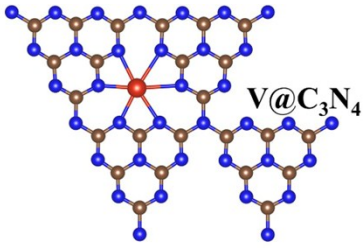
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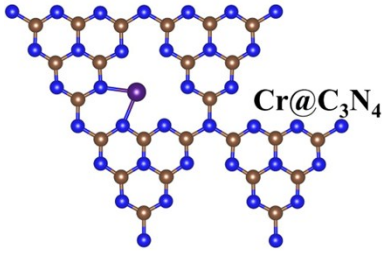
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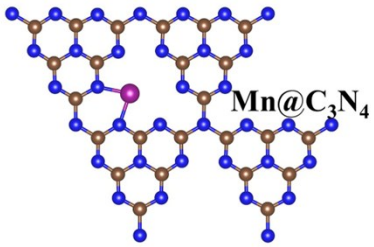
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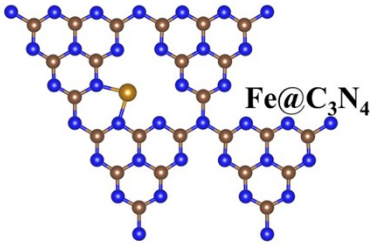
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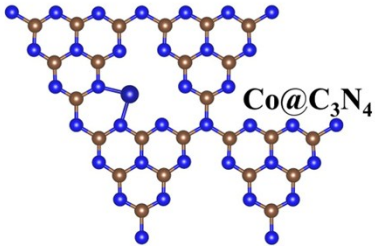
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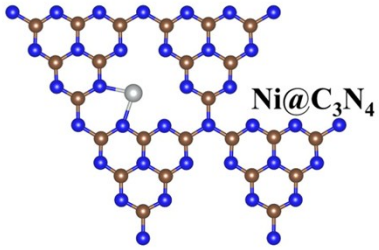
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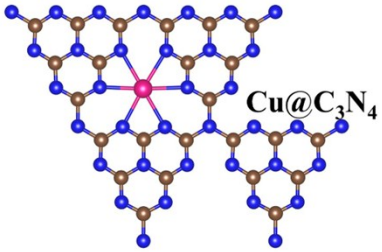
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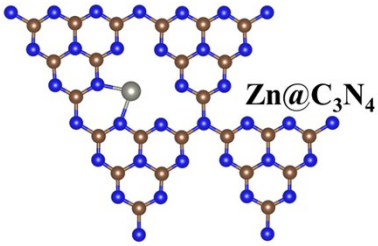
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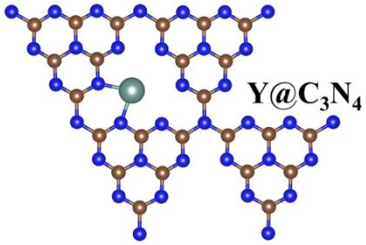
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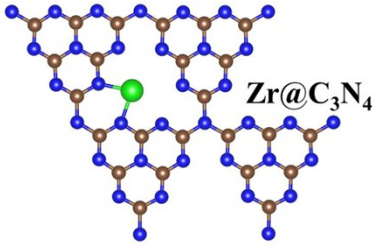
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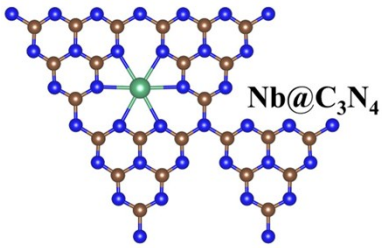
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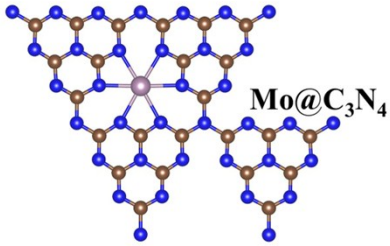
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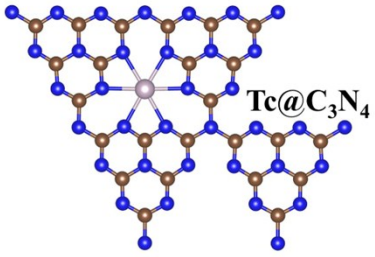
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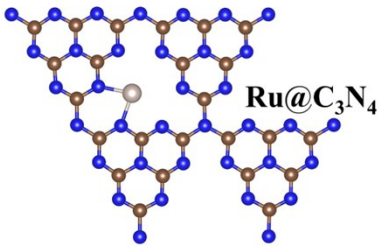
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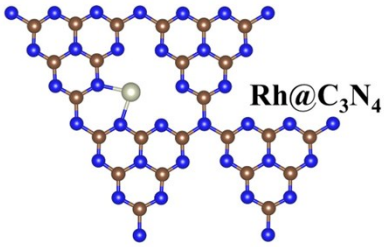
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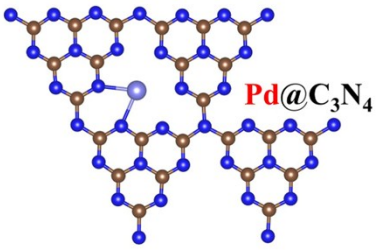
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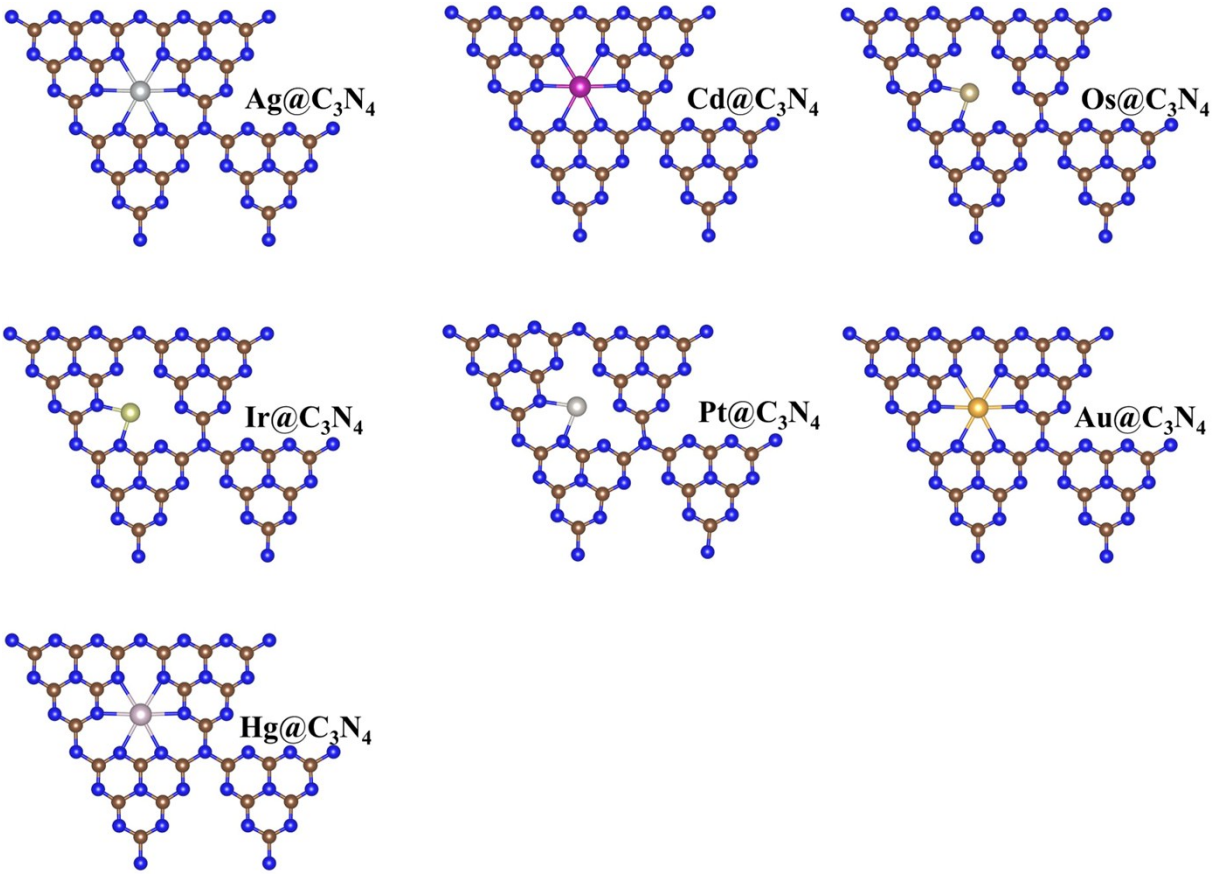


Fig. S1 The optimized structure of 25 single-atom embedded in g-C_3N_4 monolayer.

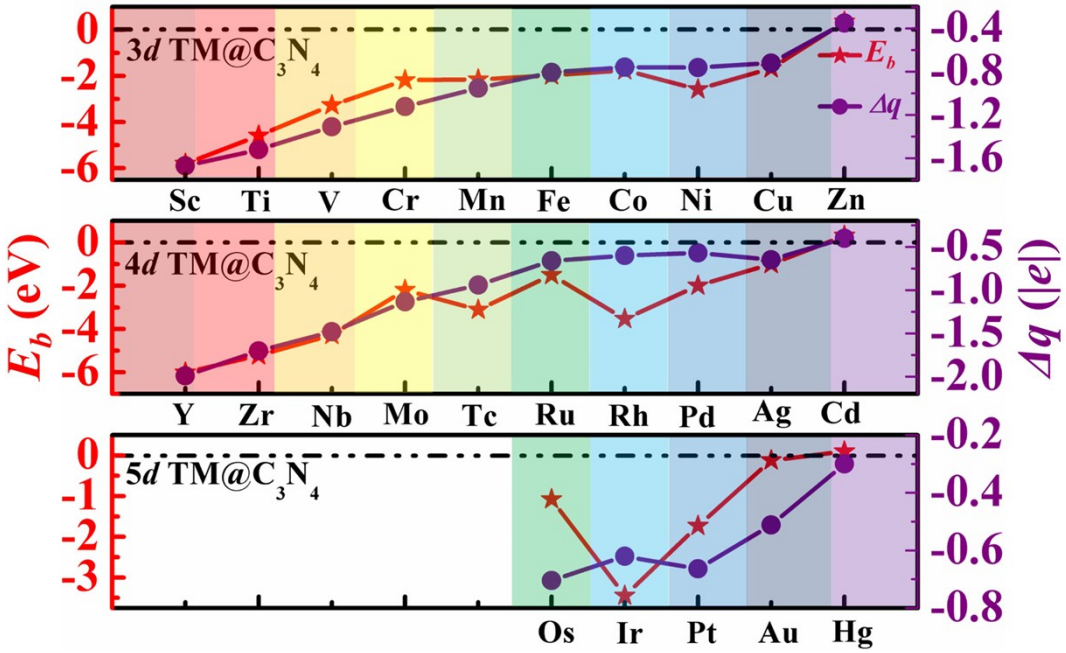


Fig. S2 The binding energies (E_b) for 25 single atoms embedded in g-C_3N_4 monolayer and the number of electrons lost Δq ($|e|$) by the metal atoms (sign '−' represents the loss of electron).

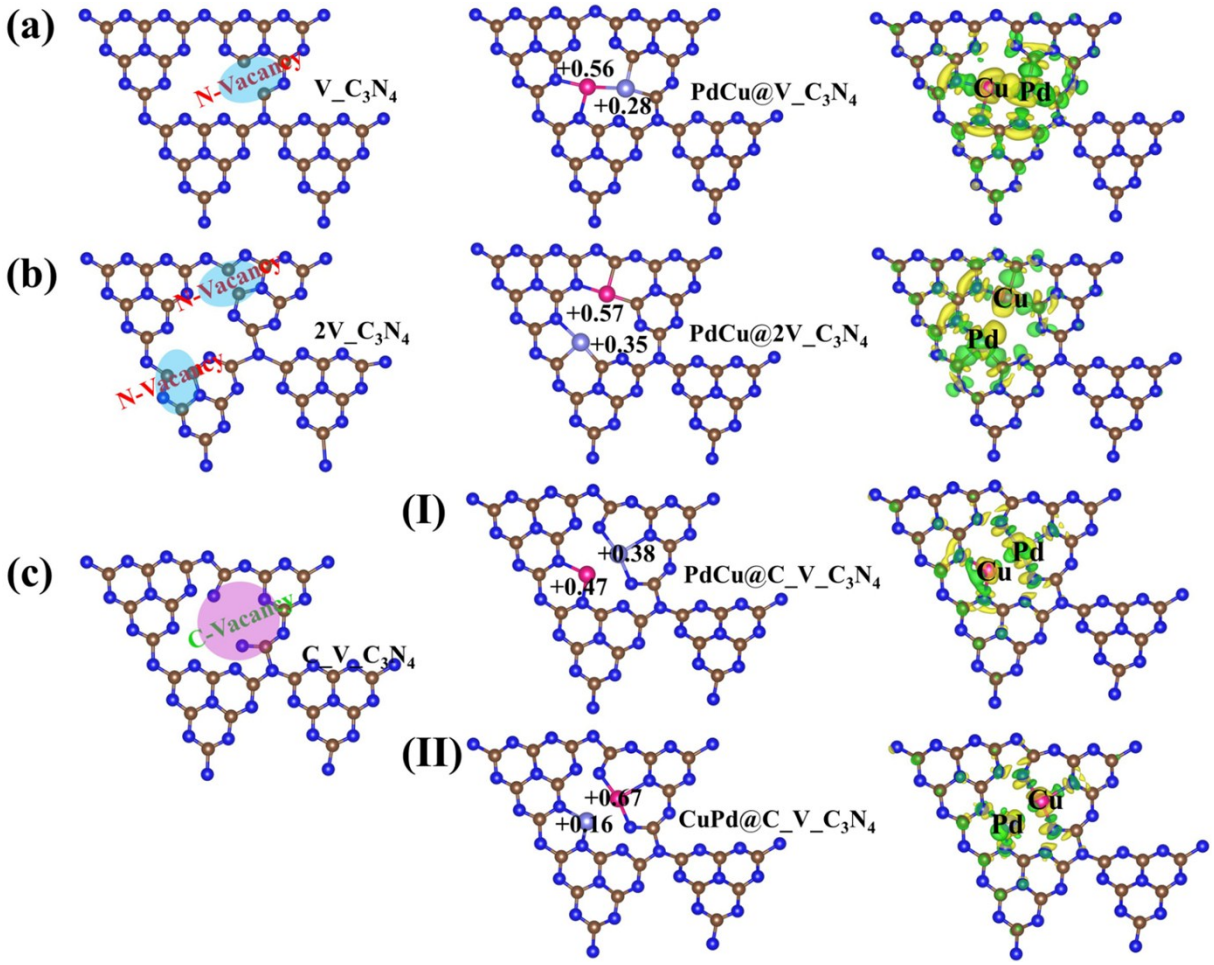


Fig. S3 The optimized structure of Pd-Cu hybrid double-atoms embedded in the single N vacancy ($PdCu@V\_C_3N_4$) and double N vacancies ($PdCu@2V\_C_3N_4$) and single C vacancy ($PdCu@C\_V\_C_3N_4$ and $CuPd@C\_V\_C_3N_4$) in the g-C_3N_4. And the corresponding Charge density difference (CDD) values for $PdCu@V\_C_3N_4$, $PdCu@V\_C_3N_4$, $PdCu@C\_V\_C_3N_4$ and $CuPd@C\_V\_C_3N_4$. The isosurface value is taken as 0.005 e $Å^{-3}$.

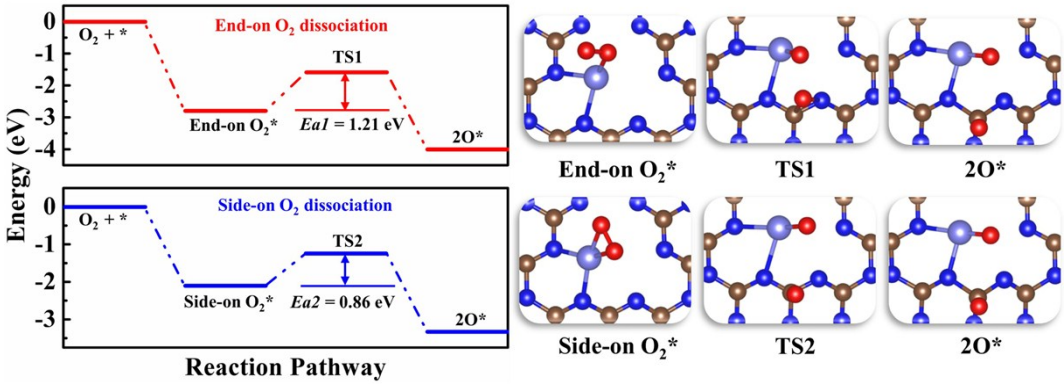
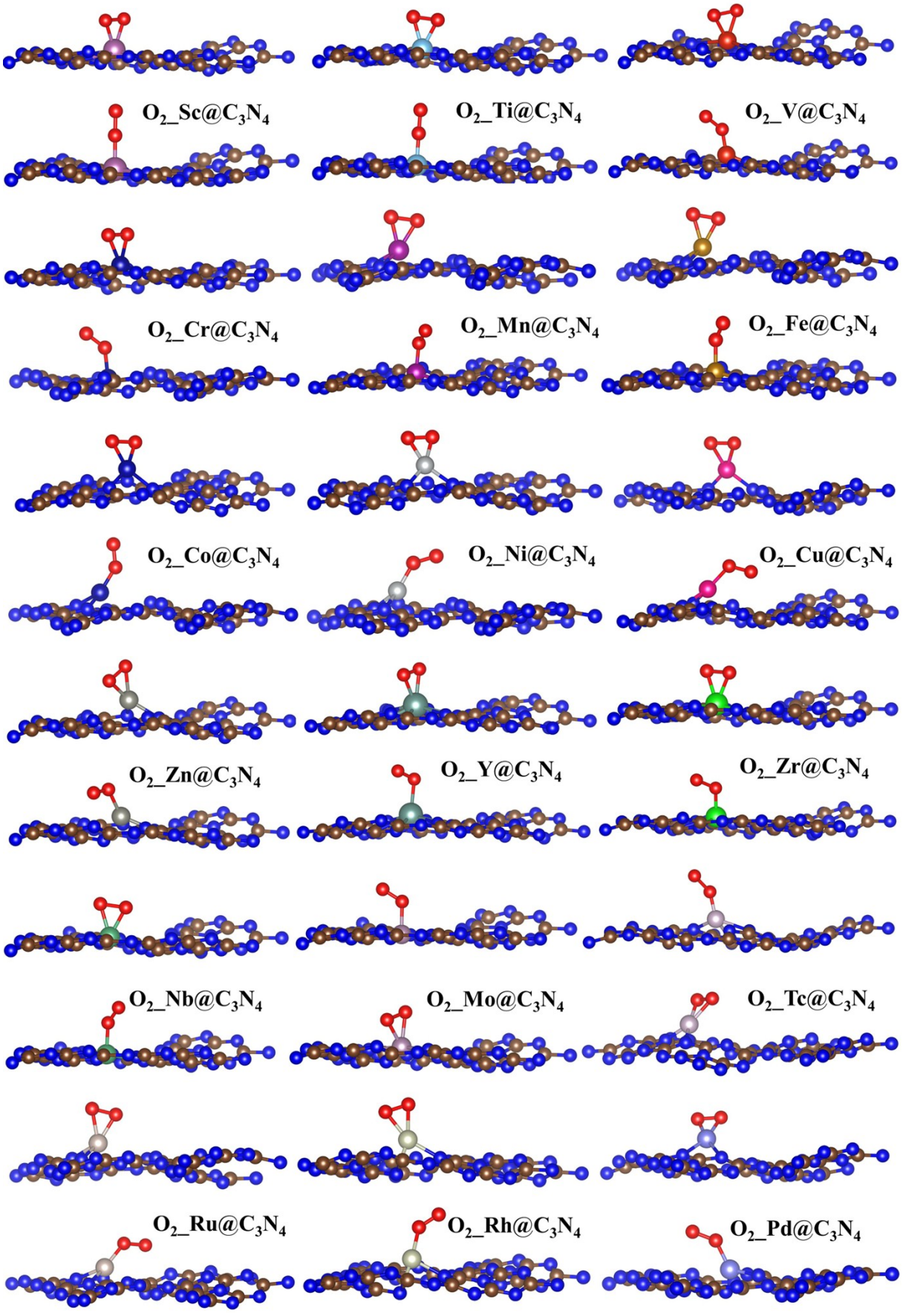


Fig. S4 Energy and profile of optimized structures for the O_2 dissociation by end-on and side-on configurations on the Pd@C_3N_4.


O2_Sc@C3N4
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O2_Cr@C3N4
O2_Mn@C3N4
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O2_Co@C3N4
O2_Ni@C3N4
O2_Cu@C3N4
O2_Zn@C3N4
O2_Y@C3N4
O2_Zr@C3N4
O2_Nb@C3N4
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O2_Pd@C3N4

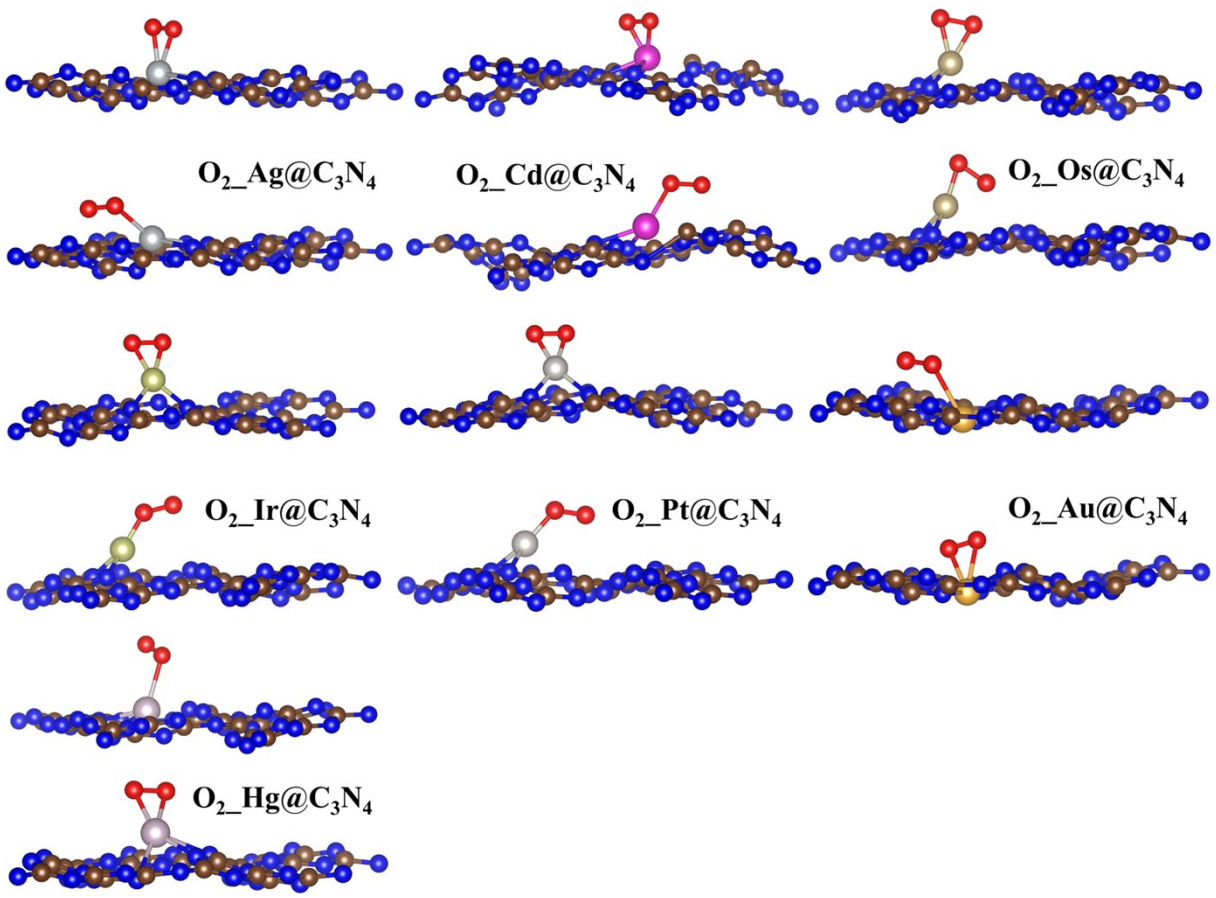

Fig. S5 Optimized structures of O_2 molecule on 25 M@C_3N_4 for side-on and end-on configuration.

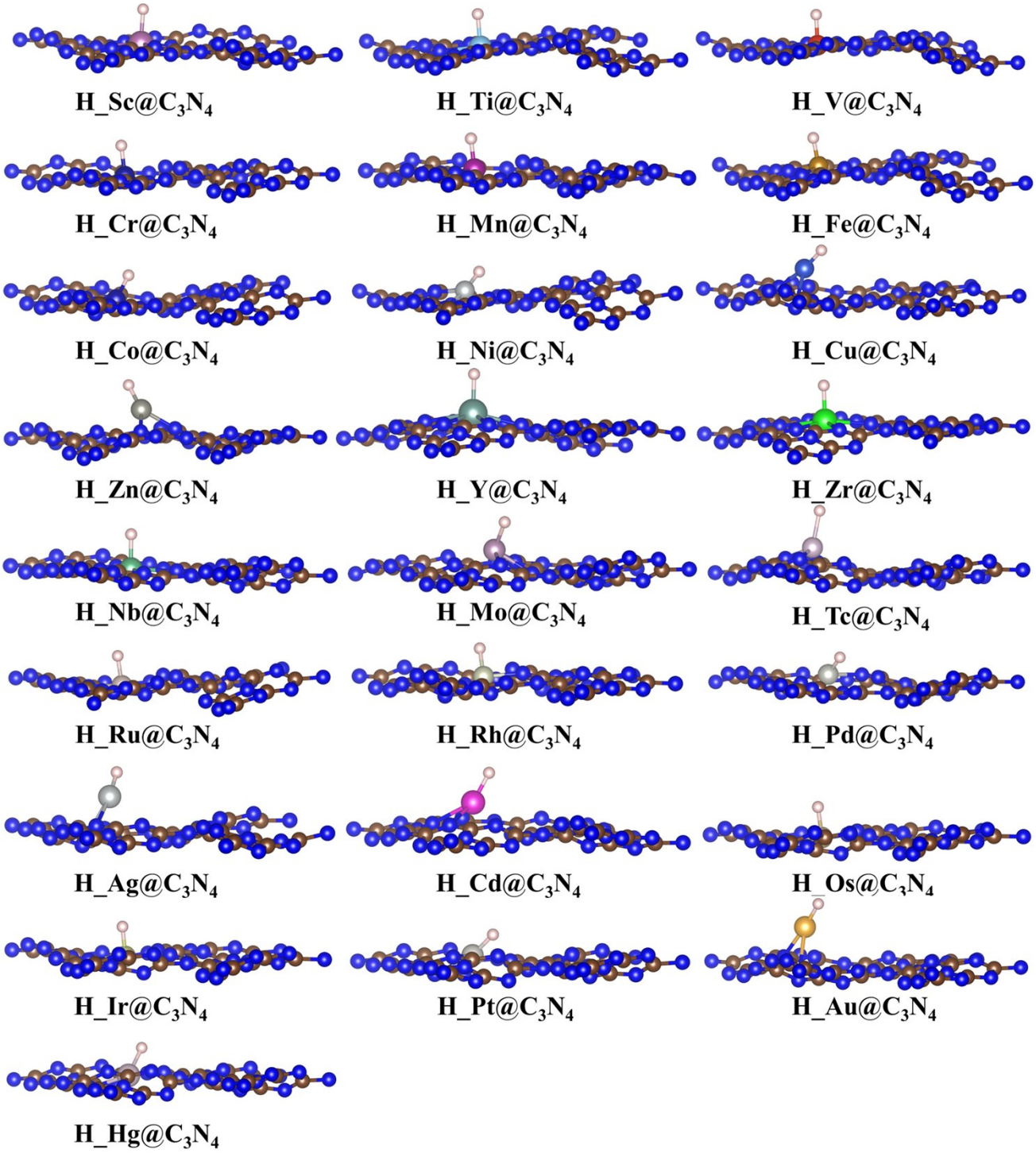

Fig. S6 Optimized structures of H proton on 25 $M@C_3N_4$ configuration.

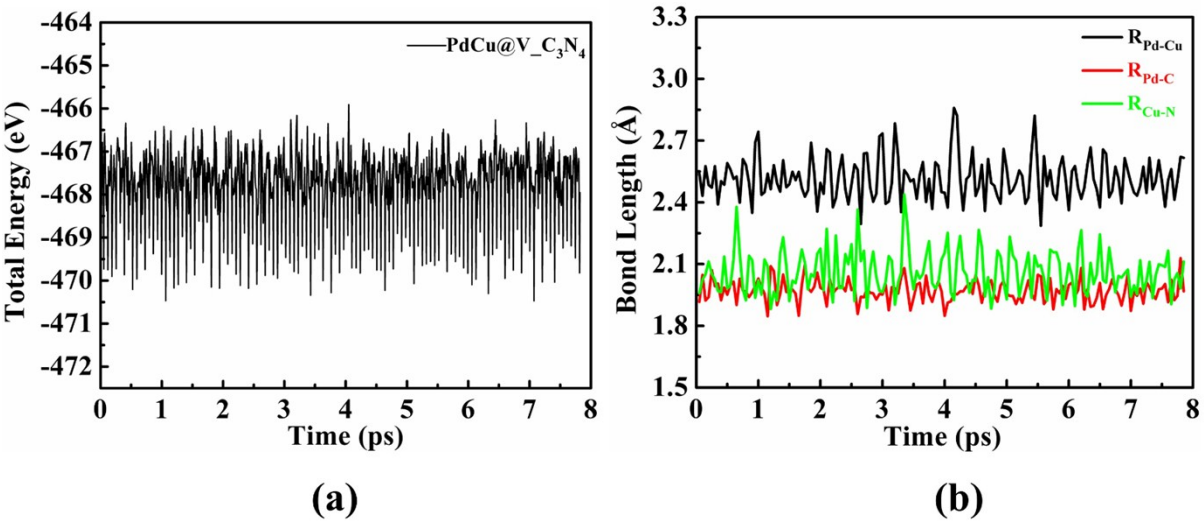

Fig. S7 (a) The total energy and (b) the bond length of Pd-Cu, Pd-C, and Cu-N of PdCu@V_C_3N_4 by performing the AIMD simulation at 500.0 K after 7.85 ps.

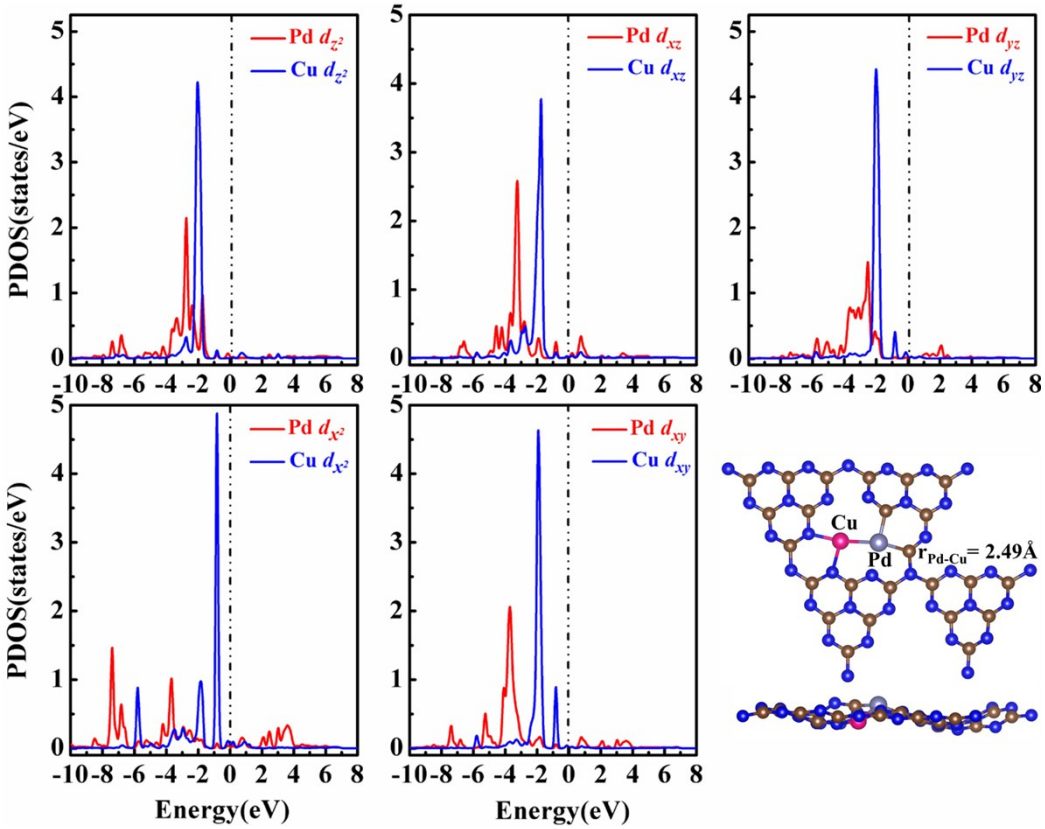

Fig. S8 The PDOS of *d*-orbitals on the Pd-Cu atom-embedded N vacancy g-C_3N_4 monolayer.

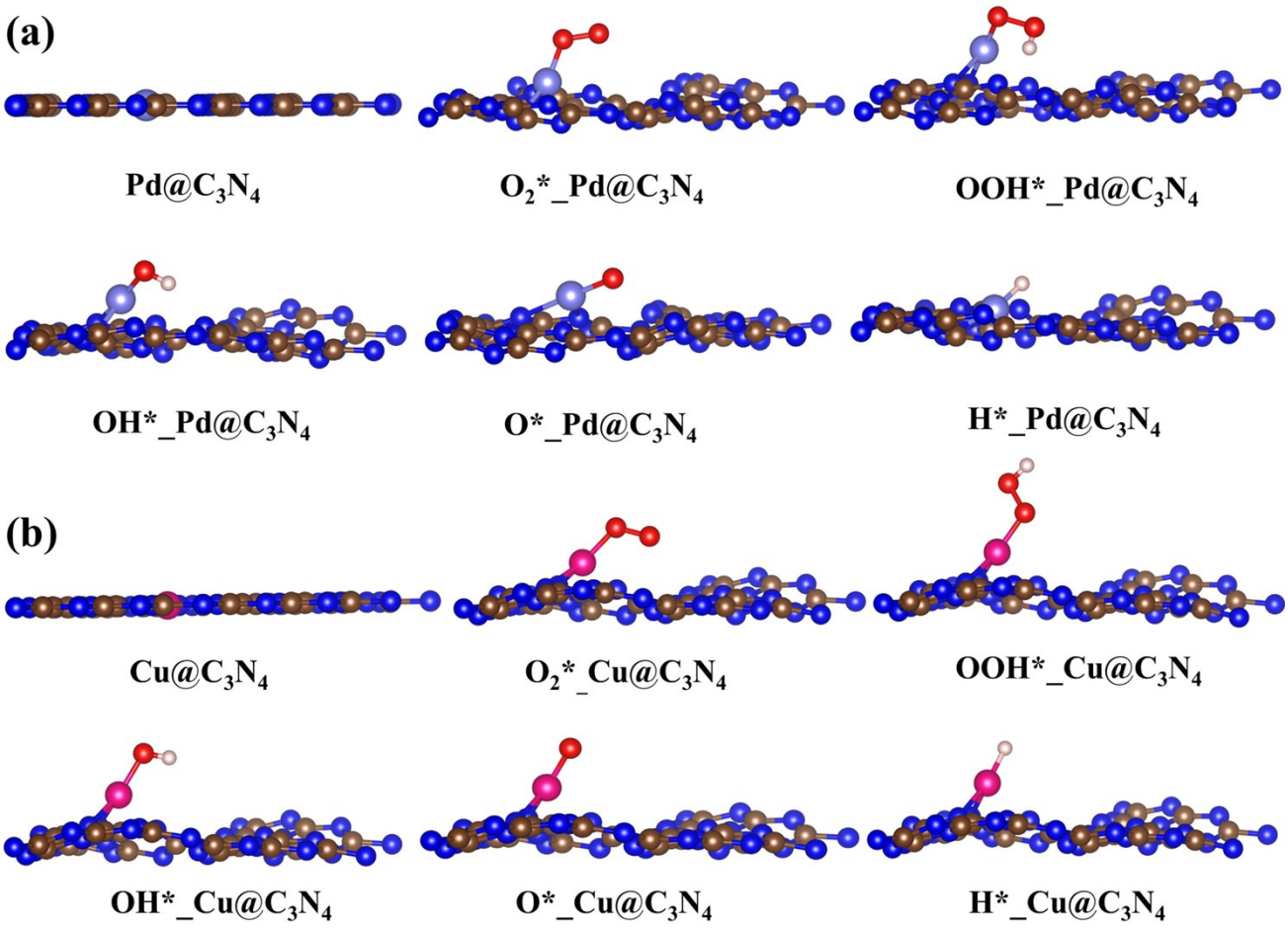

Fig. S9 Optimized structures of O_2, OOH, OH, O and H intermediates on (a) Pd@C_3N_4 and (b) Cu@C_3N_4.

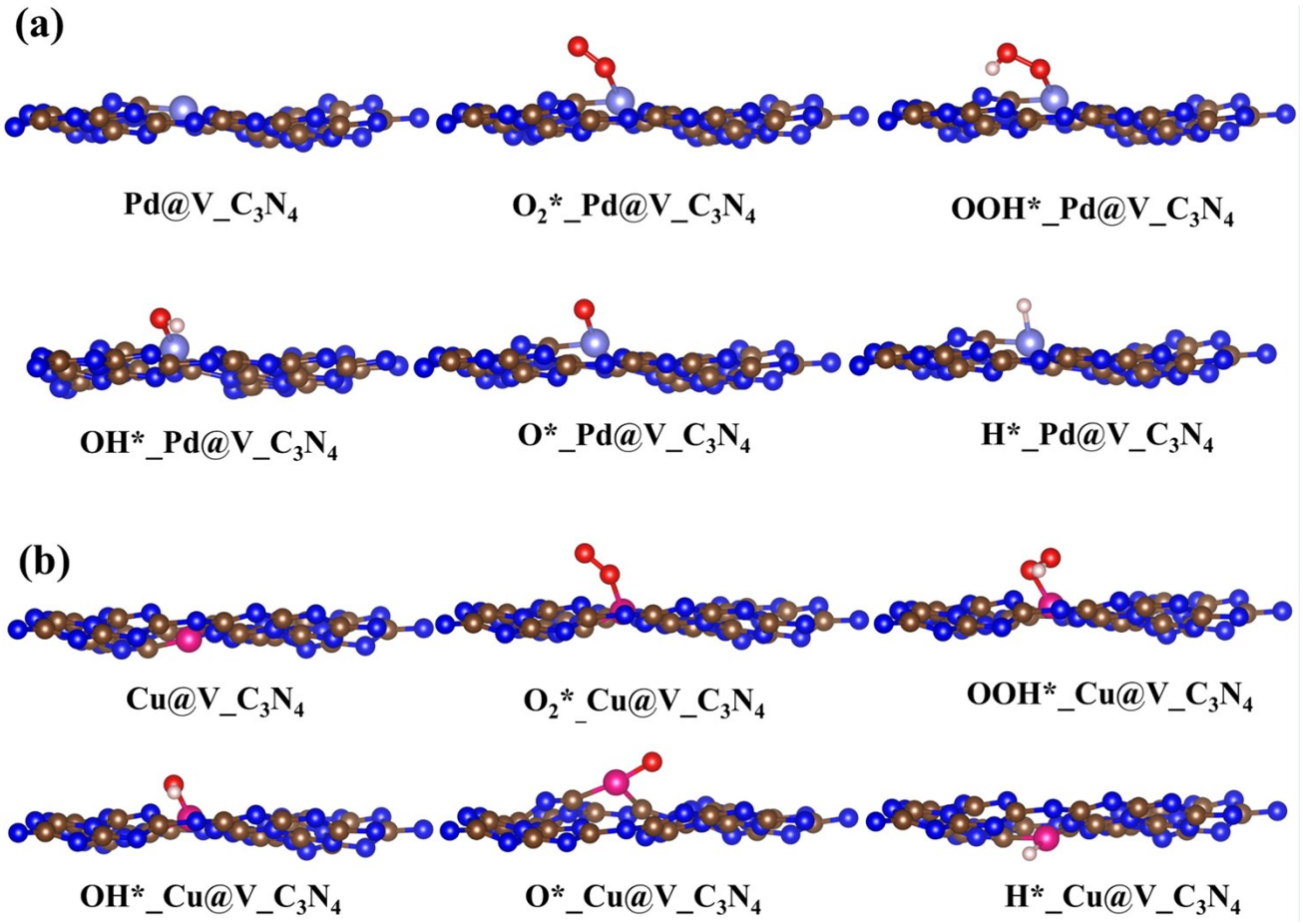

Fig. S10 Optimized structures of O_2, OOH, OH, O and H intermediates on (a) Pd@V_C_3N_4 and (b) Cu@V_C_3N_4.

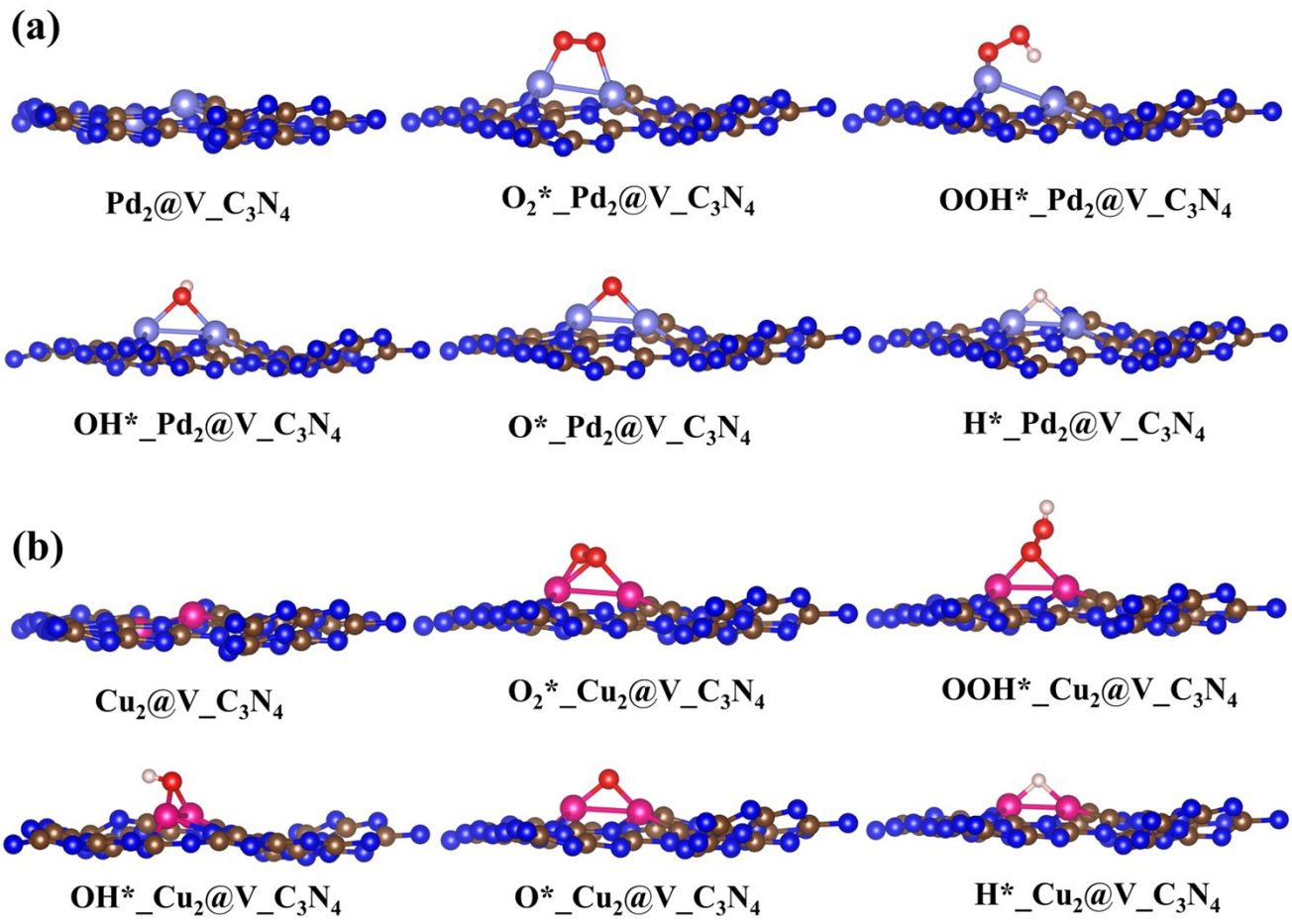

Fig. S11 Optimized structures of O_2, OOH, OH, O and H intermediates on (a) Pd_2@V_C_3N_4 and (b) Cu_2@V_C_3N_4.

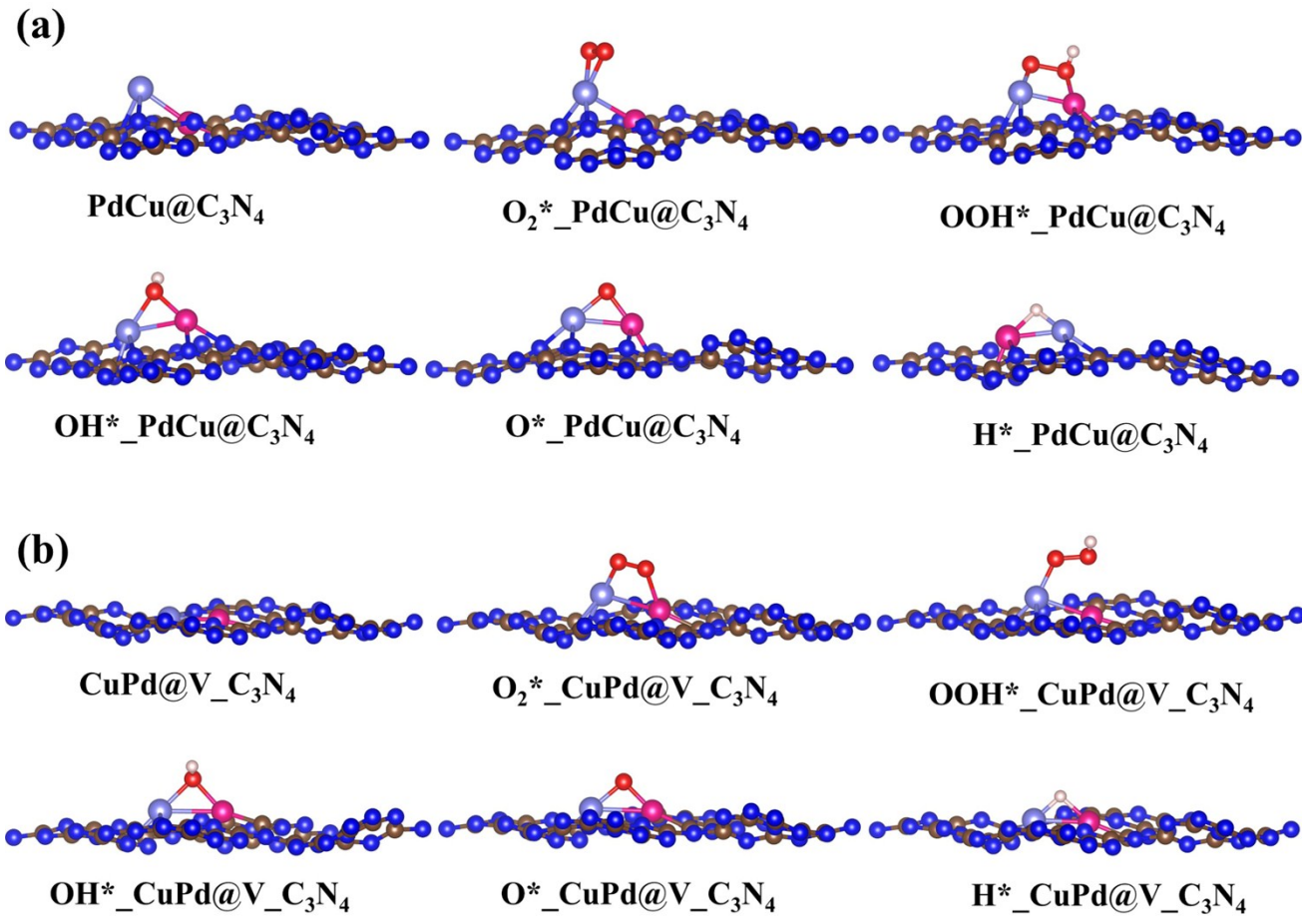

Fig. S12 Optimized structures of O_2, OOH, OH, O and H intermediates on (a) PdCu@V_C_3N_4 and (b) CuPd@V_C_3N_4.

Table S1. Zero-point energy (*ZPE*), enthalpy (*H*) and entropy correction (-*TS*) of adsorbents used to covert electron into free energy. The temperature is set to 298.15 K.

$Pd@C_3N_4$				**$Cu@C_3N_4$**			
Adsorbates	*ZPE*	*H*	*-TS*	Adsorbates	*ZPE*	*H*	*-TS*
$*O_2$	0.1364	0.0744	-0.1482	$*O_2$	0.1275	0.088	-0.188
*OOH	0.4552	0.0875	-0.1798	*OOH	0.4224	0.0828	-0.1795
*OH	0.3608	0.0484	-0.0925	*OH	0.3227	0.0726	-0.1687
*O	0.0615	0.0374	-0.0702	*O	0.0485	0.0471	-0.1112
*H	0.2183	0.0071	-0.0094	*H	0.165	0.0178	-0.0267
$Pd@V\_C_3N_4$				**$Cu@V\_C_3N_4$**			
Adsorbates	*ZPE*	*H*	*-TS*	Adsorbates	*ZPE*	*H*	*-TS*
$*O_2$	0.1297	0.0865	-0.1821	$*O_2$	0.1335	0.0842	-0.1840
*OOH	0.4554	0.0747	-0.1428	*OOH	0.4376	0.0797	-0.1627
*OH	0.3657	0.0460	-0.0803	*OH	0.3545	0.0493	-0.0885
*O	0.0510	0.0424	-0.0861	*O	0.0595	0.0395	-0.0782
*H	0.1746	0.0128	-0.0188	*H	0.1942	0.0094	-0.0131
$Pd_2@C_3N_4$				**$Cu_2@C_3N_4$**			
Adsorbates	*ZPE*	*H*	*-TS*	Adsorbates	*ZPE*	H	*-TS*
$*O_2$	0.1488	0.0661	-0.1225	$*O_2$	0.1447	0.0634	-0.1102
*OOH	0.4400	0.0902	- 0.1717	*OOH	0.4371	0.0913	-0.1825
*OH	0.3569	0.0452	-0.0723	*OH	0.3812	0.0400	-0.0697
*O	0.0768	0.0268	-0.0433	*O	0.0763	0.0274	-0.0458
*H	0.2054	0.0042	-0.0054	*H	0.2027	0.0058	-0.0078
$PdCu@V\_C_3N_4$				**$CuPd@V\_C_3N_4$**			
Adsorbates	*ZPE*	*H*	*-TS*	Adsorbates	*ZPE*	*H*	*-TS*
$*O_2$	0.1419	0.0715	- 0.1415	$*O_2$	0.1396	0.0718	-0.1384
*OOH	0.4401	0.0801	-0.1435	*OOH	0.4286	0.0729	-0.1378
*OH	0.3633	0.0469	-0.0881	*OH	0.3753	0.0406	-0.0662
*O	0.0753	0.0282	-0.0480	*O	0.0779	0.0256	-0.0403
*H	0.1917	0.0073	-0.0102	*H	0.2133	0.0038	-0.0047

$PdCu@C_3N_4$			
Adsorbates	*ZPE*	*H*	*-TS*
$*O_2$	0.1482	0.0606	-0.1047
*OOH	0.3855	0.0393	-0.0717
*OH	0.3891	0.0380	-0.0651
*O	0.0739	0.0285	-0.0480
*H	0.2046	0.0056	-0.0075