

**Table S1**

Calculated binding energy ( $E_b$ ), cohesive energy ( $E_{coh}$ ), the distance ( $d$ ) between g- $C_3N_4$  and  $C_3N$  layer for different stacked patterns.

	AA1	AA2	AB1	AB2	AB3
$E_b$ (eV)	-1.021	-1.166	-1.212	-1.143	-1.188
$E_{coh}$ (eV)	-2.965	-2.978	-2.982	-2.976	-2.980
$d$ (Å)	3.259	2.890	2.988	2.968	2.969

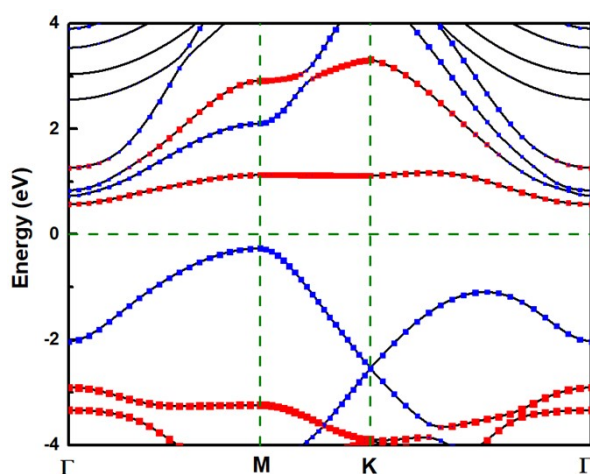


Fig.S1 Projected band structure of g- $C_3N_4/C_3N$  vdWH at HSE06 level.

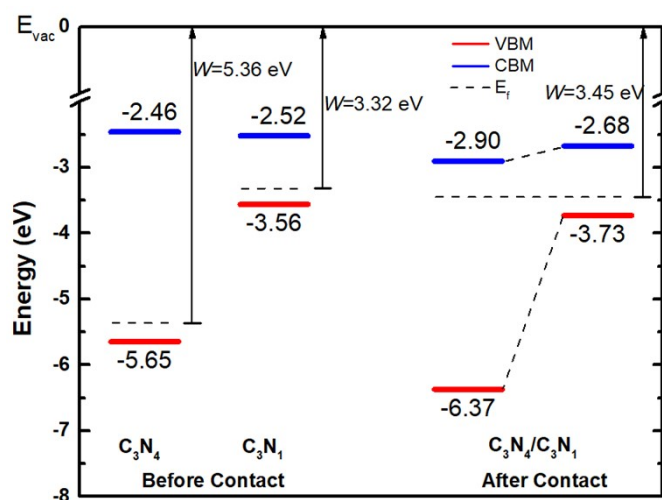


Fig.S2 Band alignments of g- $C_3N_4/C_3N$  vdWH at HSE06 level.

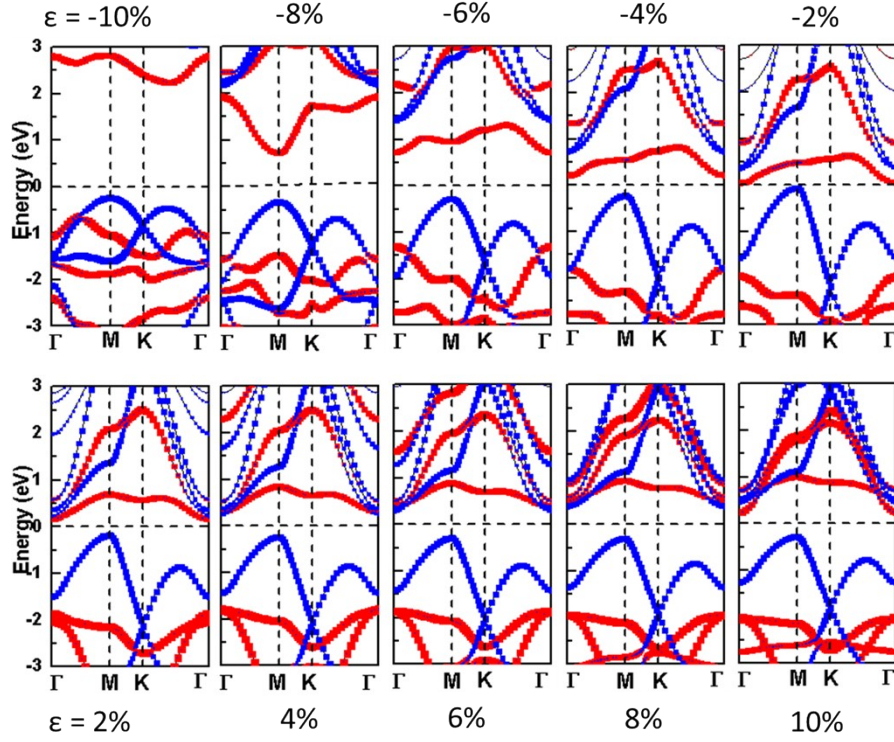


Fig.S3 Projected band structures of g-C<sub>3</sub>N<sub>4</sub>/C<sub>3</sub>N vdWH under the in-plane biaxial strains.

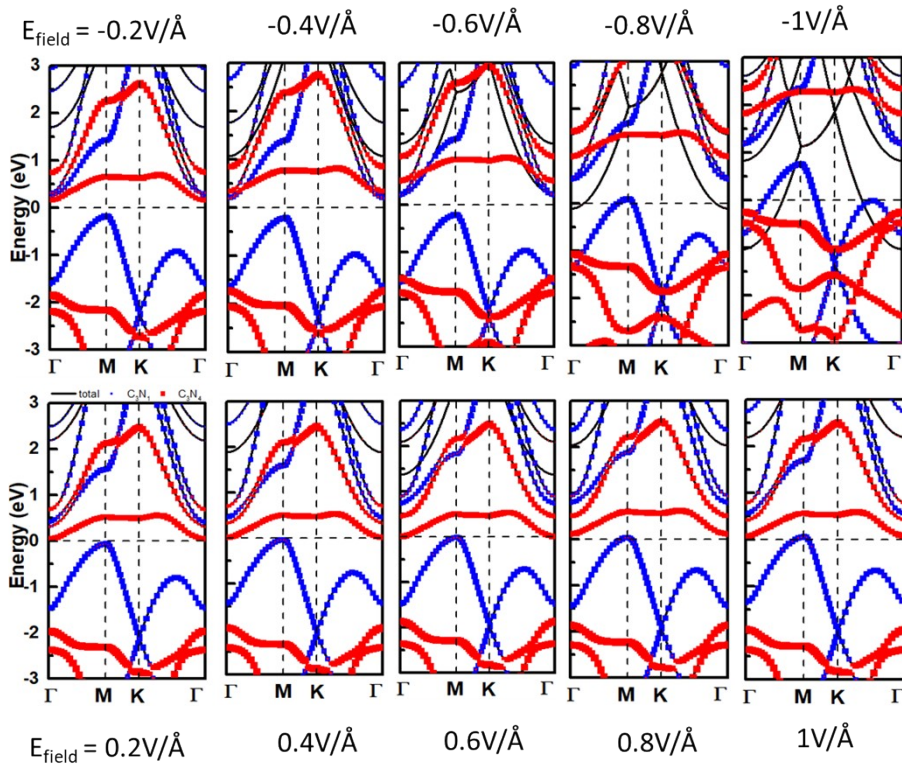


Fig.S4 Projected band structures of g-C<sub>3</sub>N<sub>4</sub>/C<sub>3</sub>N vdWH under different  $E_{\text{field}}$ .

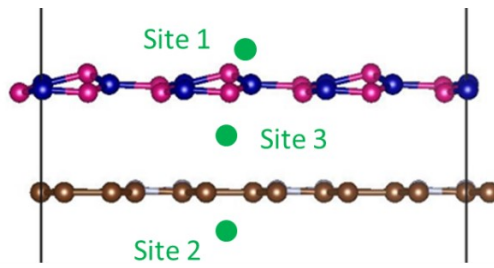


Fig.S5 Three sites of Ti atom at g-C<sub>3</sub>N<sub>4</sub>/C<sub>3</sub>N vdWH.

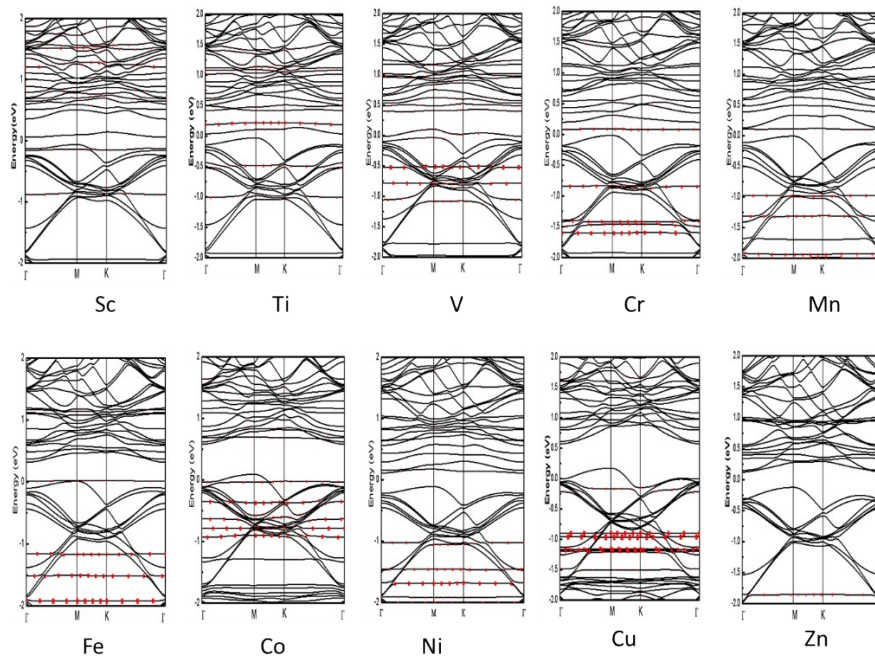


Fig.S6 Band structures of metal atoms adsorbed on g-C<sub>3</sub>N<sub>4</sub> surface of g-C<sub>3</sub>N<sub>4</sub>/C<sub>3</sub>N vdWH.

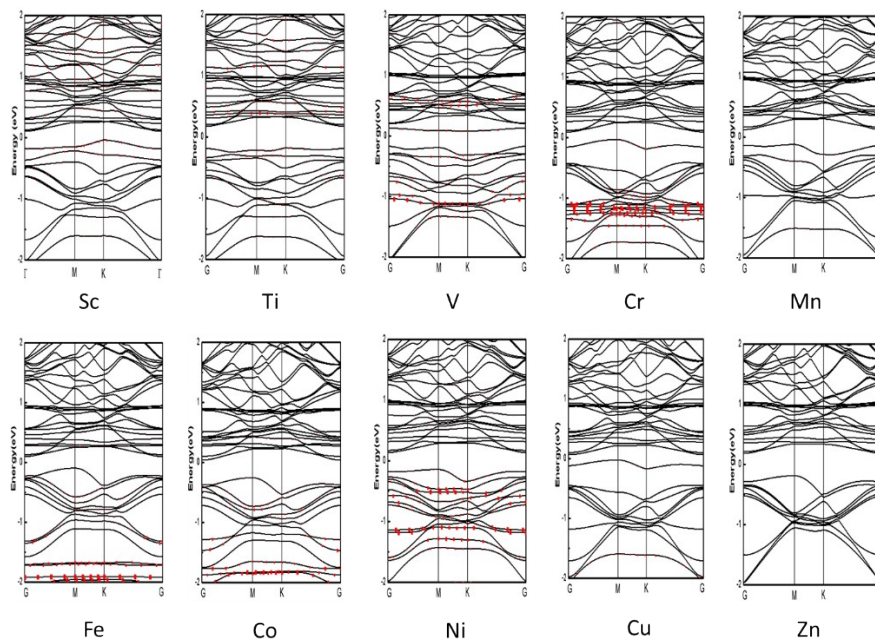


Fig.S7 Band structures of metal atoms adsorbed on  $C_3N$  surface of  $g-C_3N_4/C_3N$  vdWH.

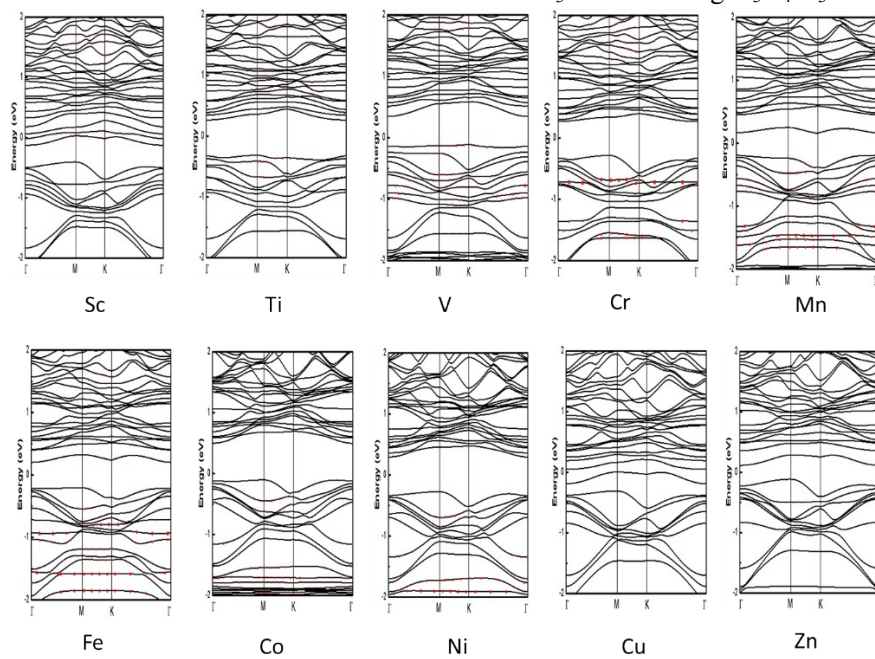


Fig.S8 Band structures of metal atoms embedded into the interlayer of  $g-C_3N_4/C_3N$  vdWH.