

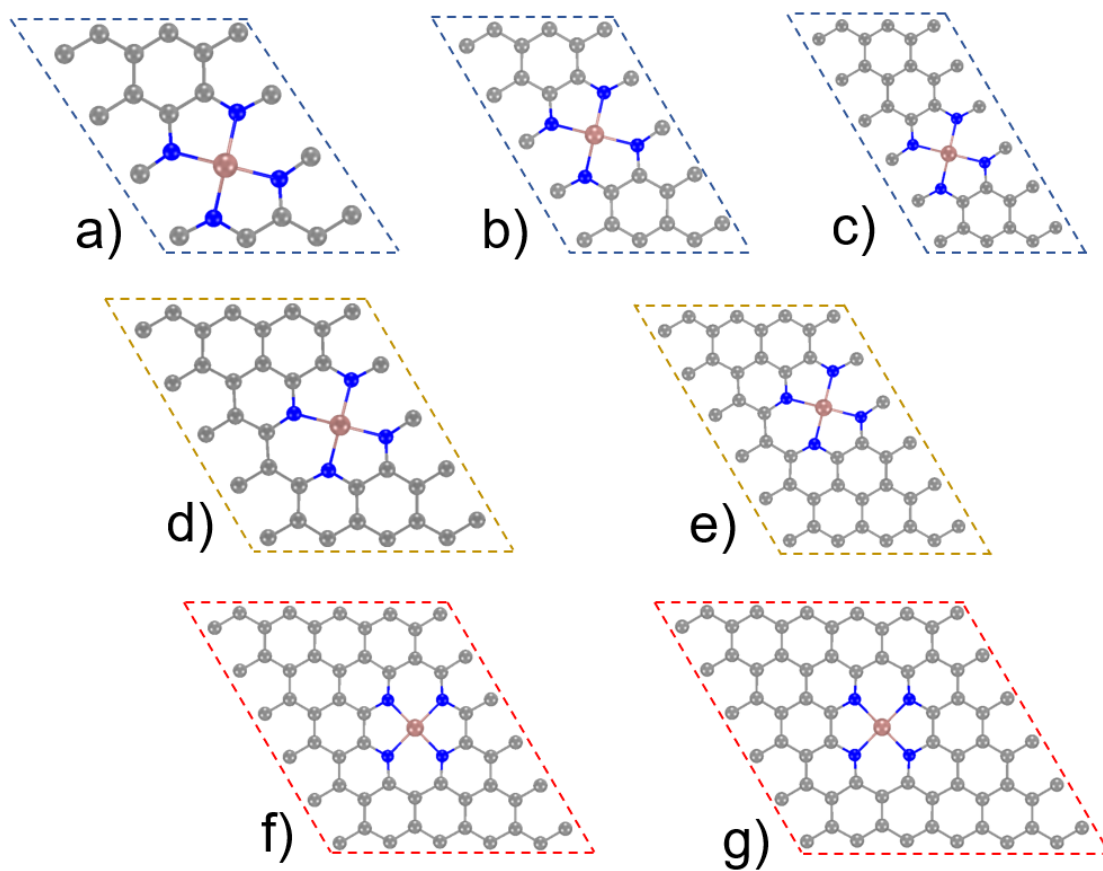
# Unravelling the 2e- ORR activity induced by distance effect on main-group metal InN<sub>4</sub> surface based on first principles

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**Figure S1** The doping structure of InN<sub>4</sub> on the surface of two-dimensional graphene in different unit cells. (a) InN<sub>4</sub>-3X<sub>4</sub>; (b) InN<sub>4</sub>-3X<sub>5</sub>; (c) InN<sub>4</sub>-3X<sub>6</sub>; (d) InN<sub>4</sub>-4X<sub>5</sub>; (e) InN<sub>4</sub>-4X<sub>6</sub>; (f) InN<sub>4</sub>-5X<sub>6</sub>; (g) InN<sub>4</sub>-6X<sub>6</sub>; . And gray-black, blue and maroon, are C atoms, N atoms and In atoms, respectively.

**Table S1** The doping energy of In atom in different supercells.

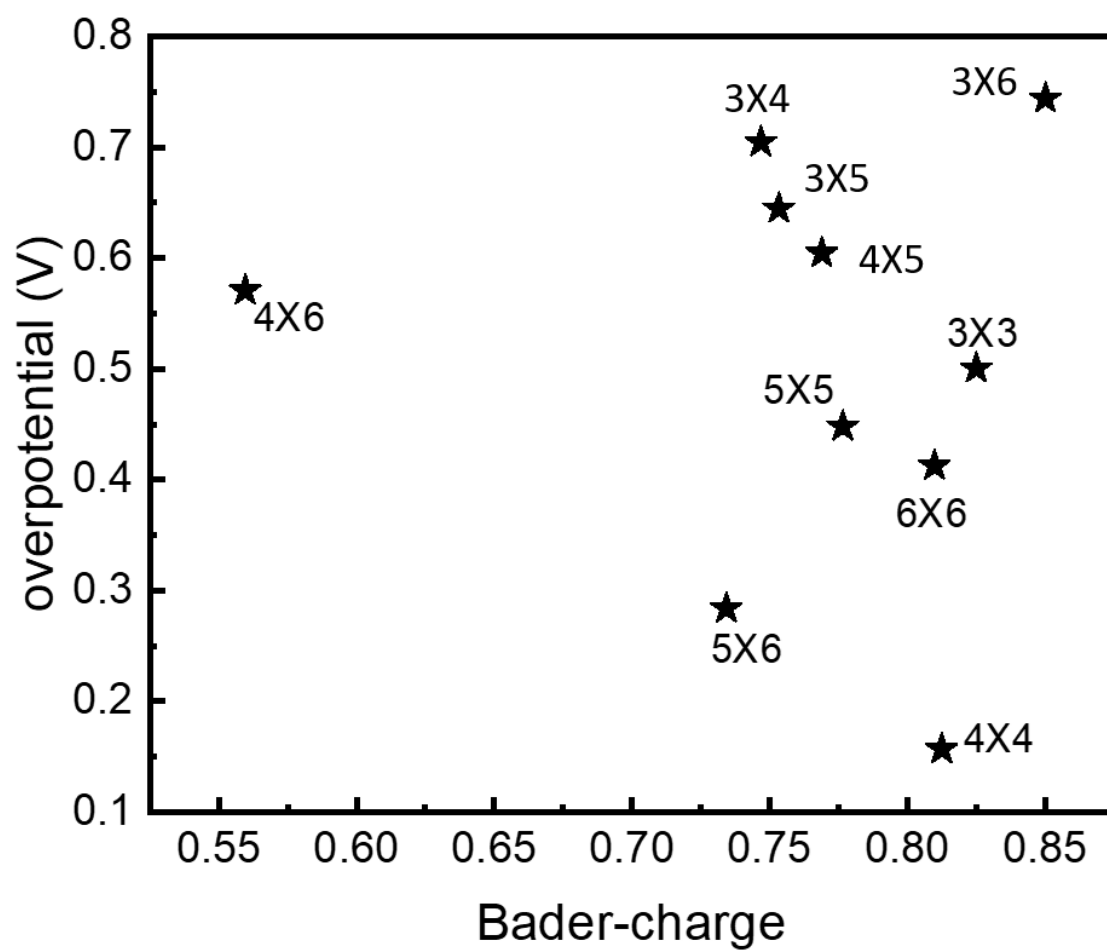
	3X3	3X6	4X4	6X6
$E_{\text{doping}}$ (eV)	-3.40	-3.53	-3.81	-3.45

**Table S2** The adsorption energy of In atom on graphene in different supercells.  $E_{\text{doping}}$  is defined as  $E_{\text{adsorption}} = E_{\text{In-G}} - E_{\text{In}} - E_{\text{G}}$ , where  $E_{\text{In-G}}$ ,  $E_{\text{In}}$  and  $E_{\text{G}}$  are the energy of the graphene with In atoms adsorbed on its surface (In-G), In atom and graphene, respectively.

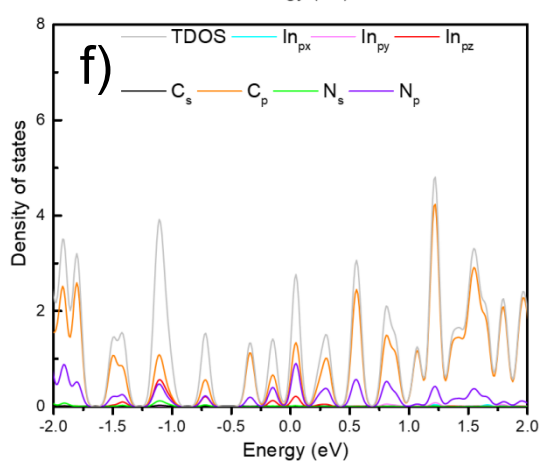
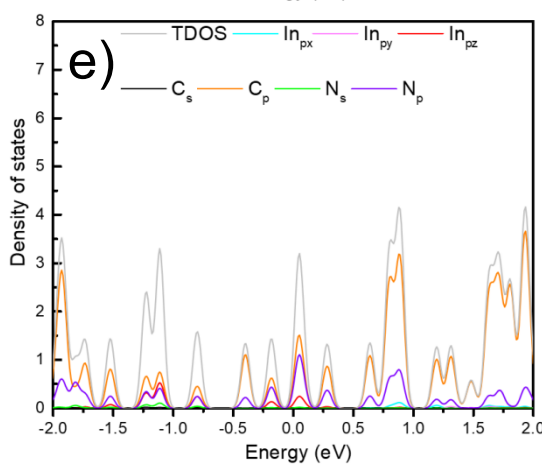
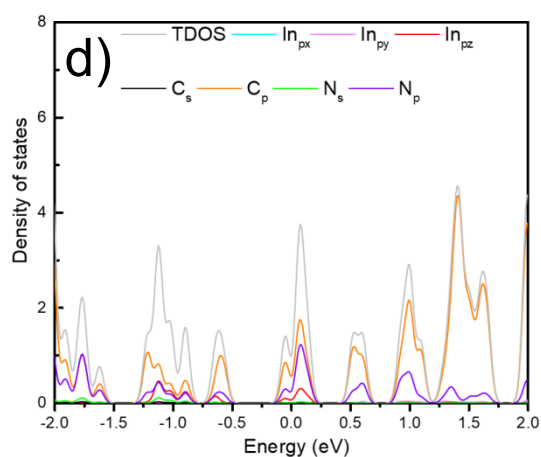
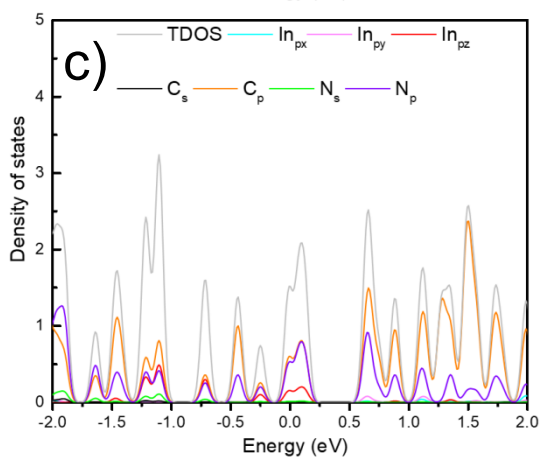
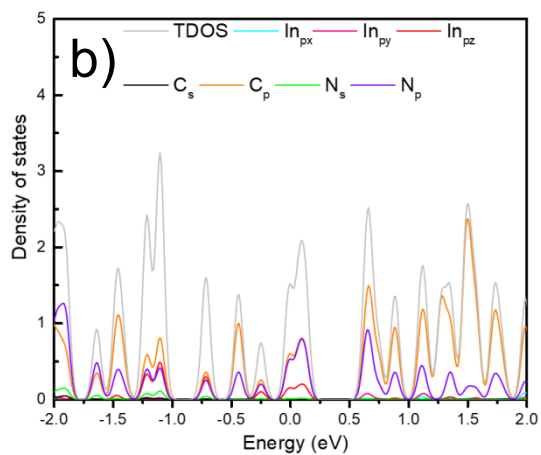
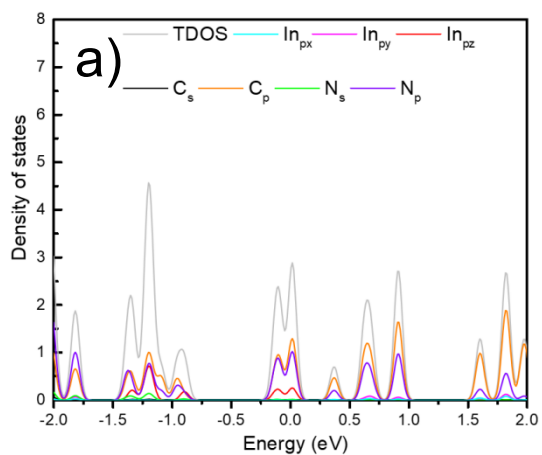
	3X3	3X6	4X4	5X5
$E_{\text{adsorption}}$ (eV)	-0.96	-1.38	-1.15	-1.34

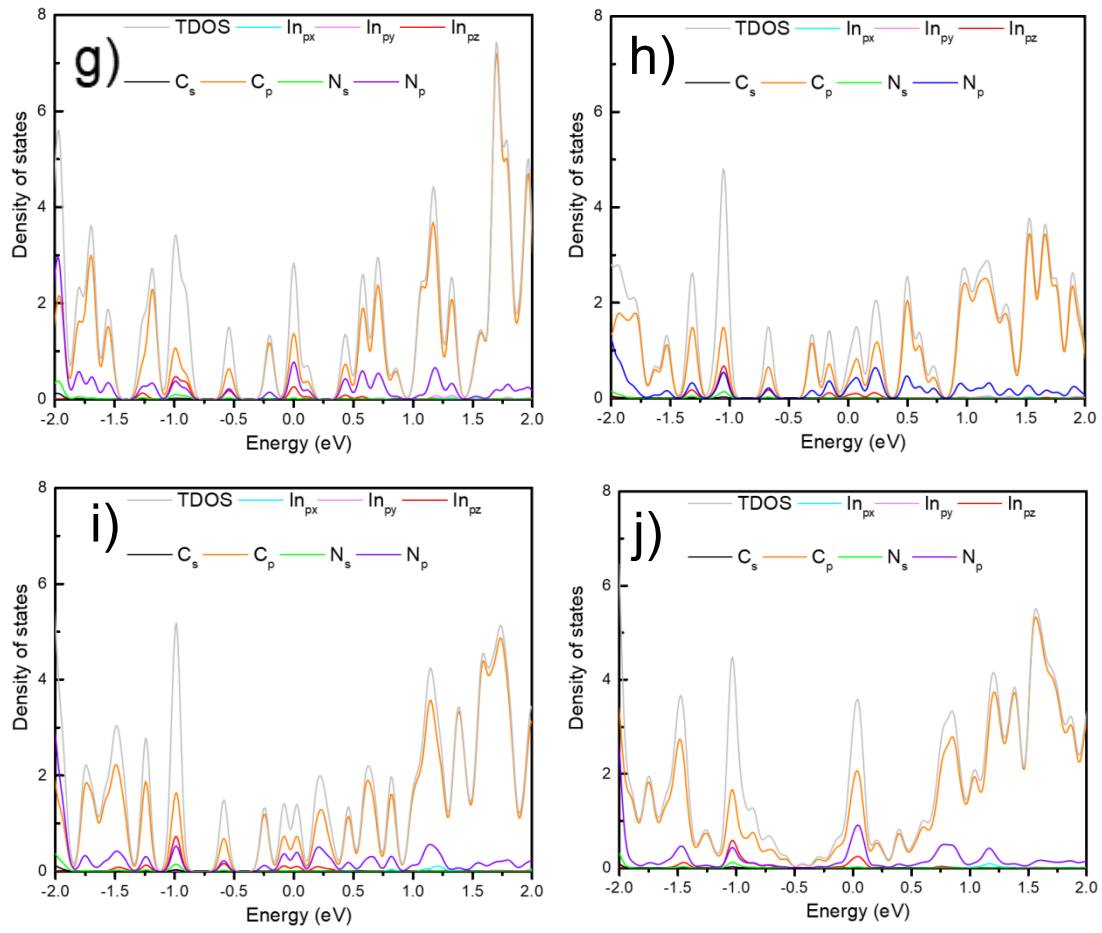
**Table S3** The relative energy of  $\text{InN}_4$  calculated by using different convergence threshold.

	cut-off energy (eV)	EDIFF (eV)	EDIFFG	$\Delta E$ (eV)
1	400	$10^{-4}$	-0.05	0
2	400	$10^{-5}$	-0.02	-0.06
3	400	$10^{-7}$	-0.01	-0.06
4	450	$10^{-4}$	-0.05	-0.06
5	500	$10^{-4}$	-0.05	-0.09

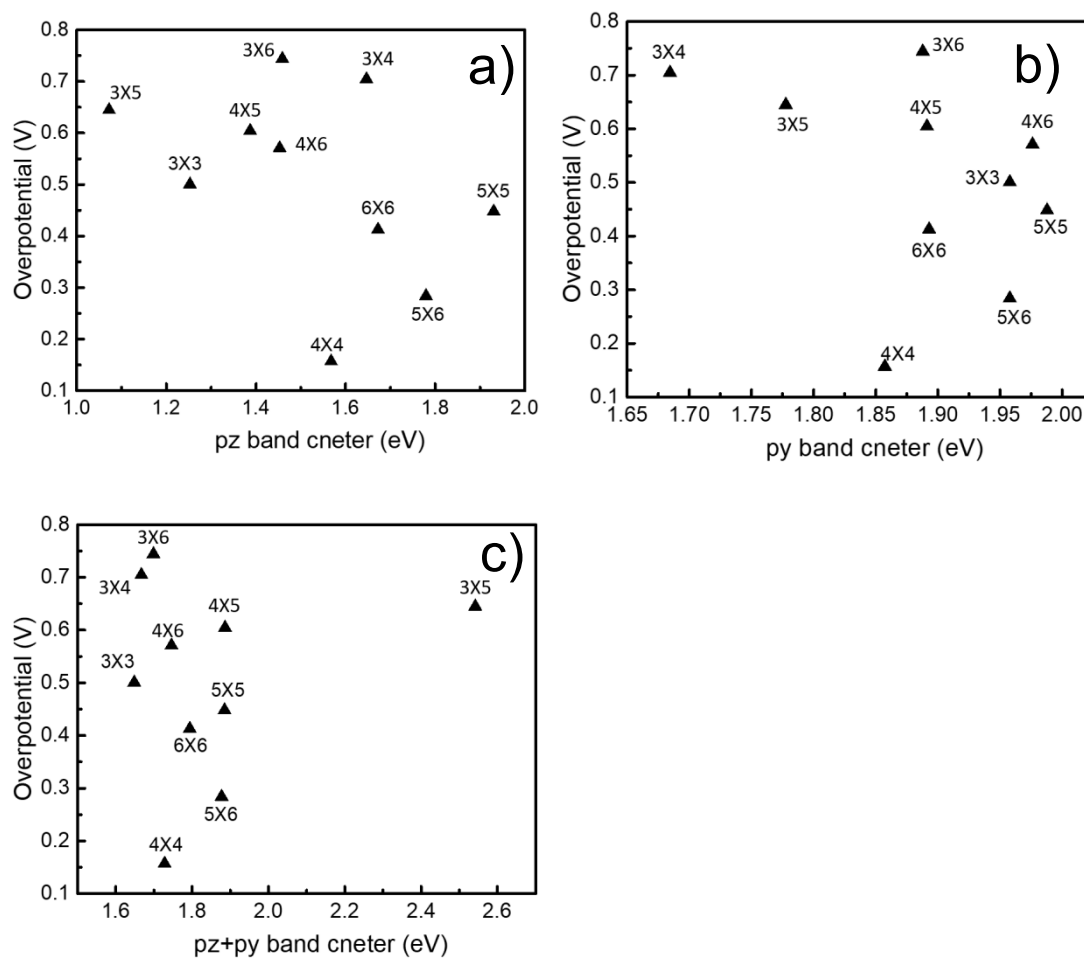


**Figure S2** The relationship between  $2e^-$  ORR catalytic activity and bader charge.

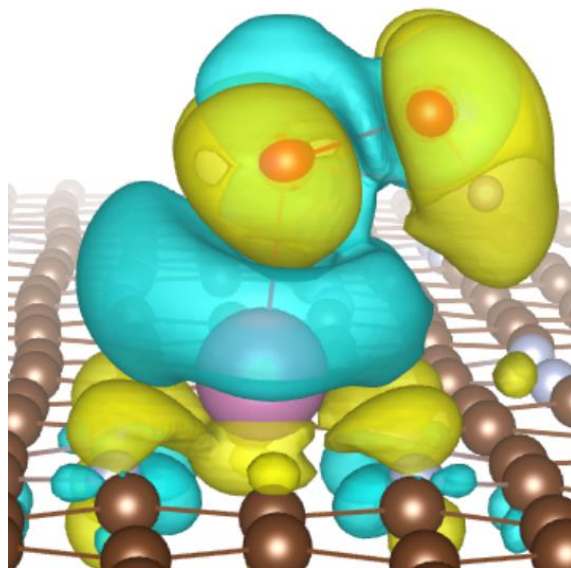




**Figure S3** The distribution of density of states (DOS) of InN<sub>4</sub> in different supercells. (a) 3X3; (b) 3X4; (c) 3X5; (d) 3X6; (e) 4X4; (f) 4X5; (g) 4X6; (h) 5X5; (i) 5X6; (j) 6X6.

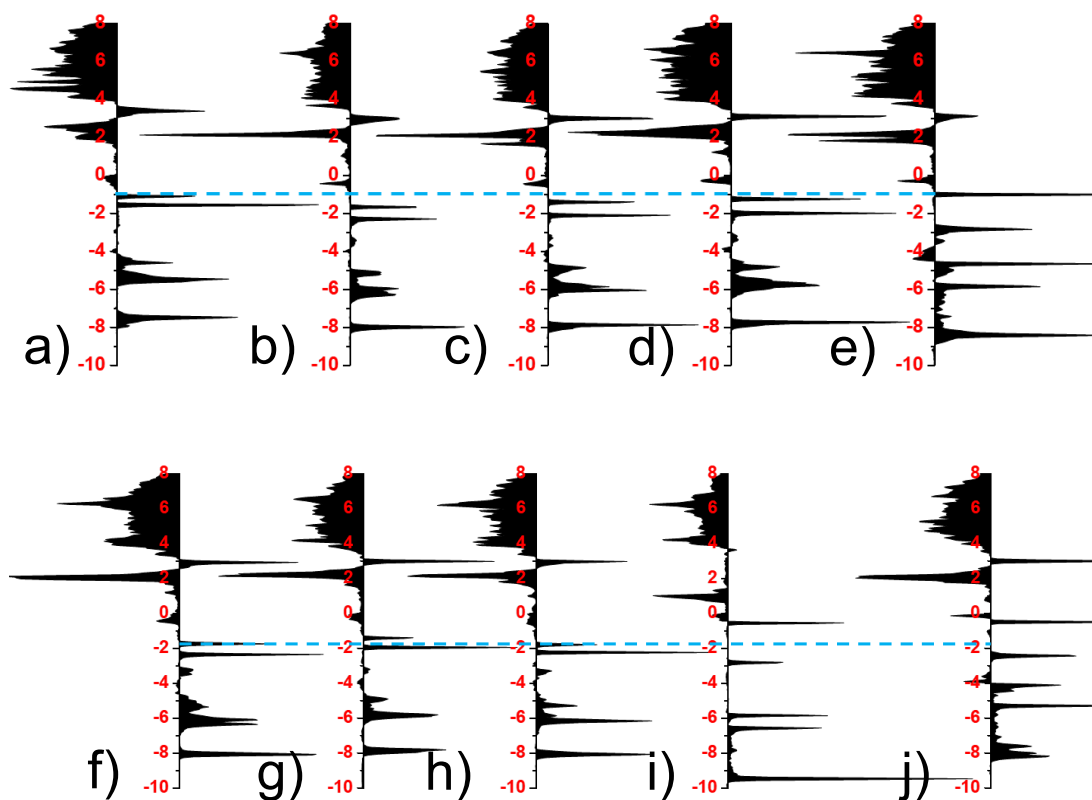


**Figure S4** The relationship between band center value and  $2e^-$  ORR overpotential. (a) pz orbit center; (b) py orbit center; (c) The center of the superposition of the pz orbital and the py orbital.

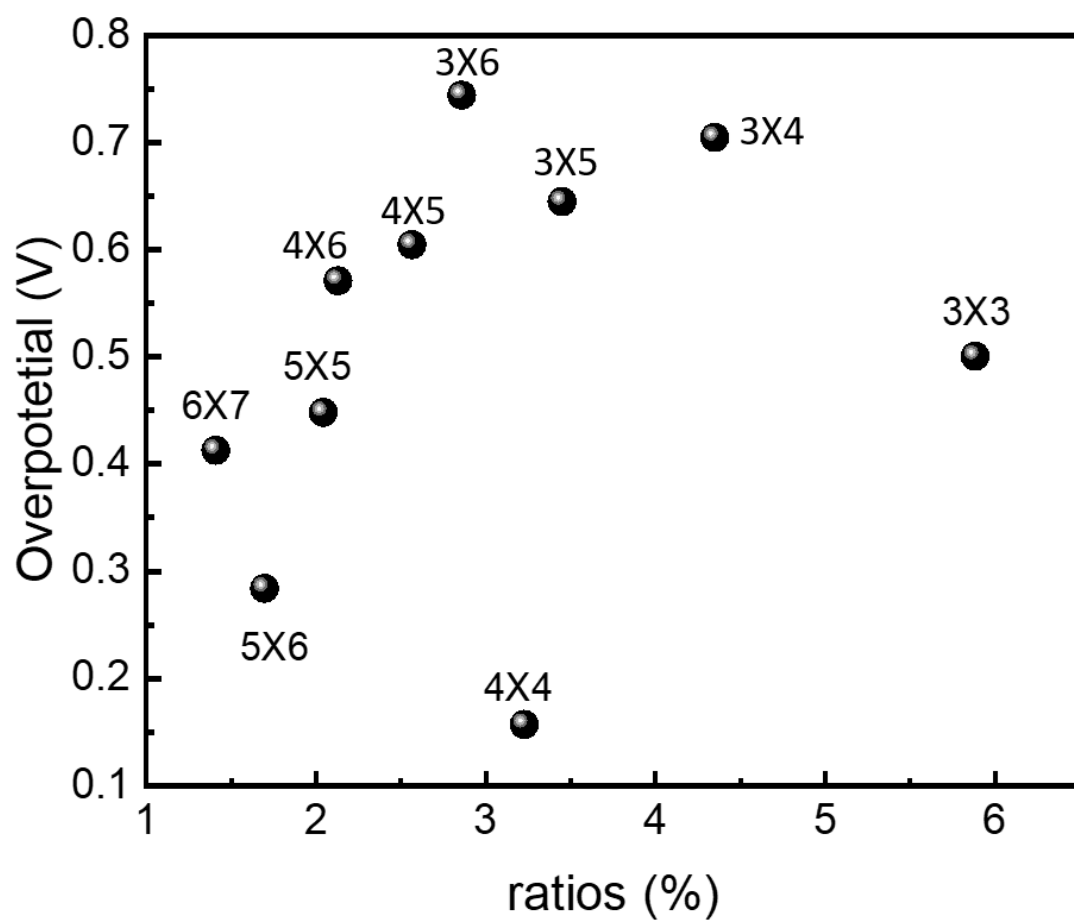


**Figure S5** The difference charge distribution of the surface of InN<sub>4</sub> (4X4) with adsorbing OOH. Yellow is the electron accumulation area and cyan is the electron deficient area.

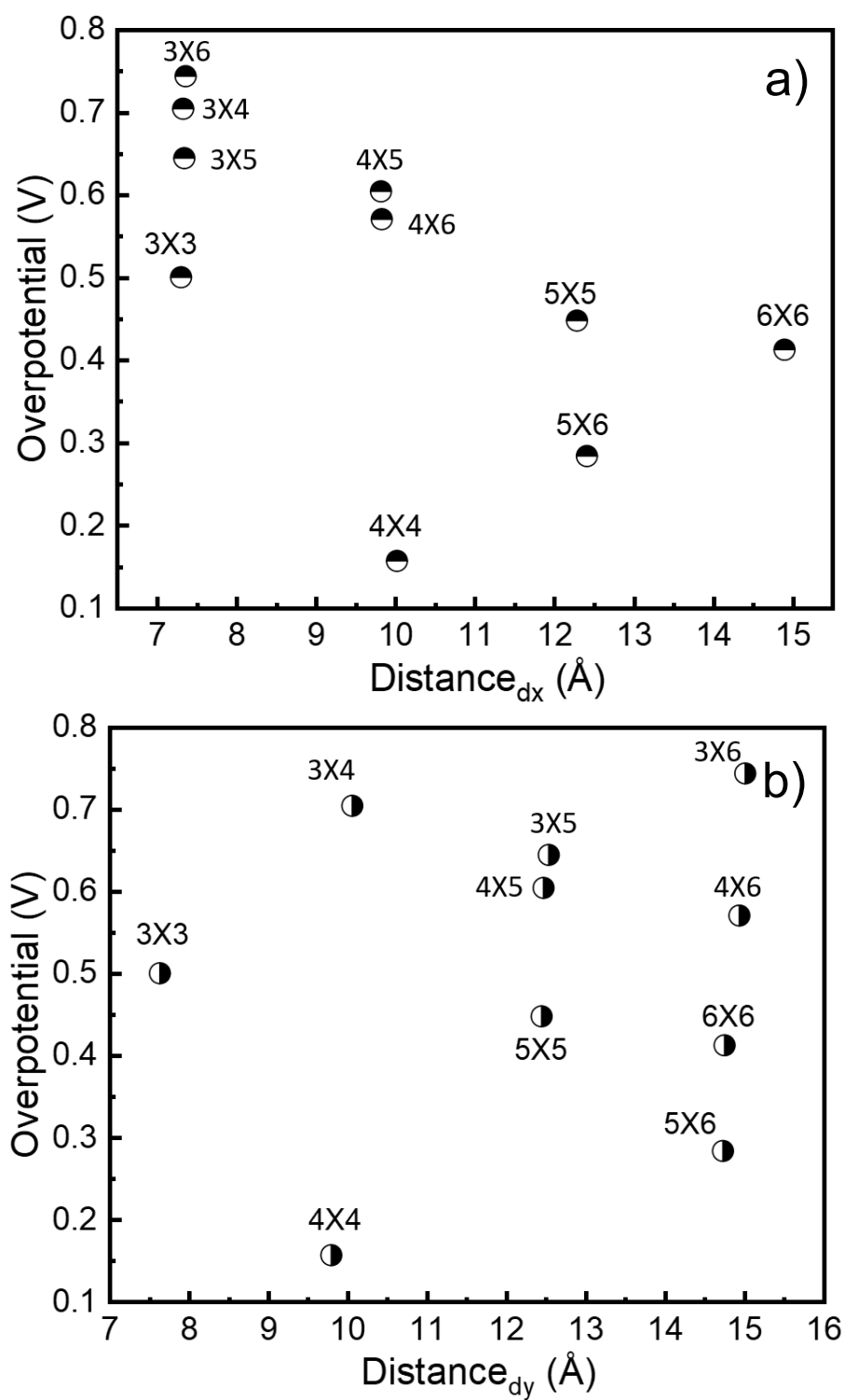




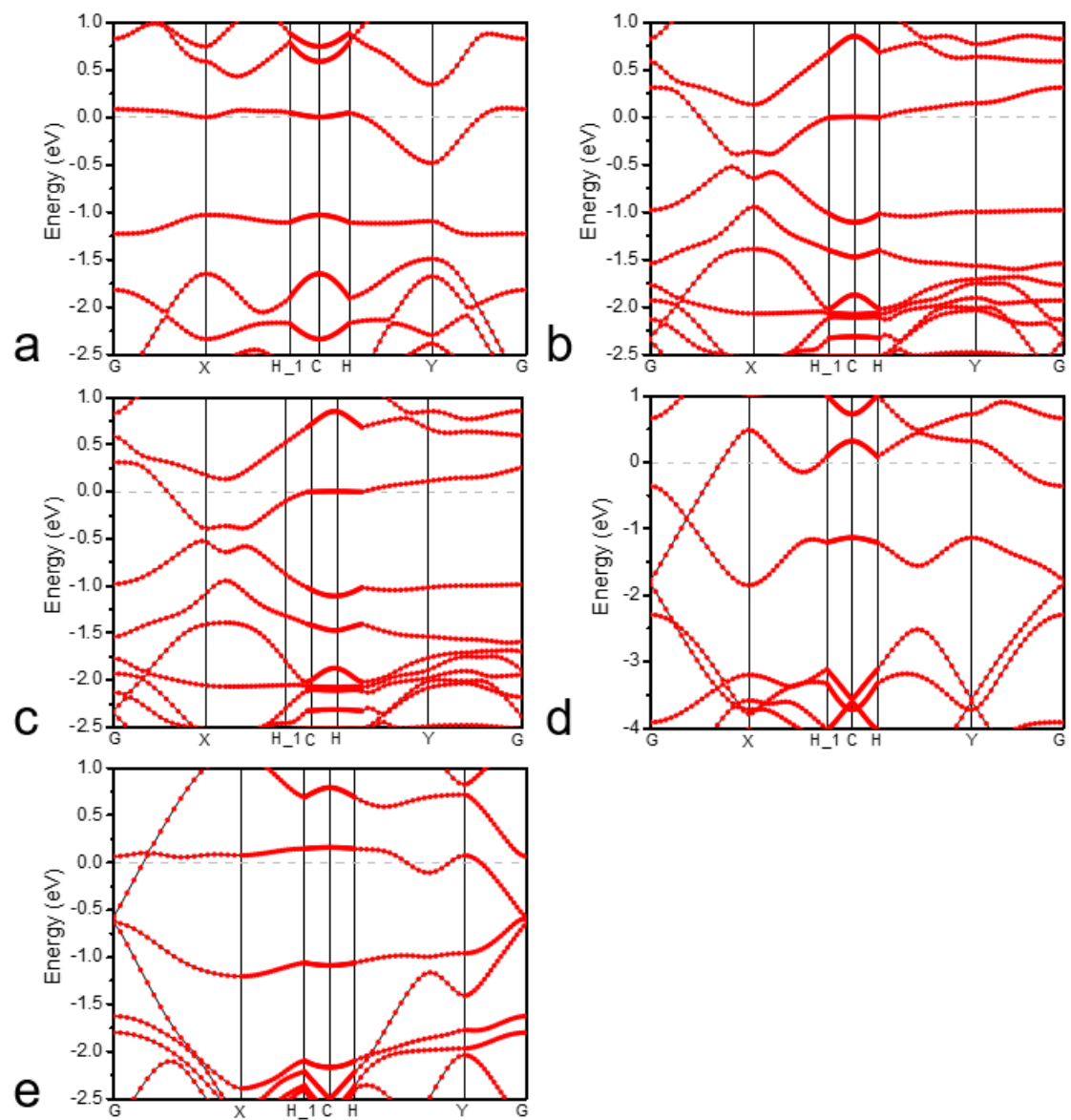
**Figure S6** The COHP distribution of In-O bonds in different unit cells of InN<sub>4</sub>. The left side is the antibonding component, and the right side is the bonding part (the abscissa is COHP). (a) InN<sub>4</sub>-3X3; (b) InN<sub>4</sub>-3X4; (c) InN<sub>4</sub>-3X5; (d) InN<sub>4</sub>-3X6; (e) InN<sub>4</sub>-4X4; (f) InN<sub>4</sub>-4X5; (g) InN<sub>4</sub>-4X6; (h) InN<sub>4</sub>-5X5; (i) InN<sub>4</sub>-5X6; (j) InN<sub>4</sub>-6X6.



**Figure S7** The relationship between the loading ratio of main group metal In atoms and the overpotential of  $2e^-$  ORR



**Figure S8** The relationship between the loading ratio of main group metal In atoms and the overpotential of  $2e^-$  ORR. (a) The spacing of In atoms in the horizontal direction ( $dx$ ); (b) The spacing of In atoms in the vertical direction ( $dy$ ).



**Figure S9** The band structure of InN<sub>4</sub> and the weights of the  $p_z$  orbitals in (a) 4X4 (b) 5X6 (c) 6X6 (d) 3X3 (e) 3X6 supercells. The density of the band points reflects the weight of the  $p_z$  orbitals.