Can all nitrogen-doped defects improve the performance of graphene anode materials for lithium-ion batteries?

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1. Electronic density of state for the N-doped graphene nanosheets

The electronic densities of state (DOS and PDOS) for the nine N-doped graphene nanosheets are listed in Figs. S1-S3. In Figs. S1 and S2, as well as Figs. S3e and S3f, the occupation numbers in the spin-up and spin-down states are equal, showing that the N$_1$, N$_2^{AA}$, N$_2^{AB}$, N$_1$V$_1$, pyridinic N$_3$V$_1$, and pyridinic N$_4$V$_2$ defects in graphene are nonmagnetic. However, from Figures S3a-d, we know that the pyrrolic N$_3$V$_1$ and pyridinic N$_2$V$_2$ are magnetic.

![Fig. S1](image)

**Fig. S1** (a) DOS for N$_1$, (b) PDOS for N$_1$, (c) DOS for N$_2^{AA}$, (d) PDOS for N$_2^{AB}$, (e) DOS for N$_2^{AB}$ and (f) PDOS for N$_2^{AB}$ conformations. The arrows denote spin-up (↑) and spin-down (↓) states.
2. Magnetic moment variant in the optimization process for pyridinic-N$_3$V$_1$ defects

Evolutions of energy and magnetic moment in the process of optimization for pyridinic N$_3$V$_1$ defect in graphene are demonstrated in Fig. S4. From Figure S4 one can see that the stable structure of the pyridinic N$_3$V$_1$ defect in graphene is nonmagnetic state. Both methods (DMol$^3$ and CASTEP) predict that the magnetic state for the pyridinic N$_3$V$_1$ defect in graphene is unstable. In the CASTEP calculation, the ultrasoft pseudopotentials generated with the atomic valence configurations of C(2$s^2$2$p^2$) and N(2$s^2$2$p^3$)
were used and the vdW dispersion correction was included in the density functional theory calculations.

![Energy and Magnetic Moment Graphs](image)

**Fig. S4** Evolutions of energy and magnetic moment in the process of optimization for pyridinic N\textsubscript{3}V\textsubscript{1} defect: (a) energy from DMol\textsuperscript{3}, (b) magnetic moment from DMol\textsuperscript{3}, (c) energy from CASTEP and (d) magnetic moment from CASTEP.

3. **Formation energy of lithium adsorbed on N-doped graphene**

The optimized stable structures, formation energies and the heights of lithium atom from the base plane of graphene for the N\textsubscript{1}, N\textsubscript{2}\textsuperscript{AA}, N\textsubscript{2}\textsuperscript{AB} and N\textsubscript{2}\textsuperscript{AF} configurations are presented in Figs. S5-S8, respectively.
Fig. S5 Geometries, formation energies and heights of lithium atom to the base plane of graphene for different adsorption sites on the N$_1$ defect at 0 K. The small gray, middle-sized blue and large pink balls represent carbon, nitrogen and lithium atoms, respectively.
Fig. S6 Optimized geometries, formation energies and adsorption heights of one lithium atom on different sites of the $N_2^{AA}$ defect at 0 K. The small gray, middle-sized blue and large pink balls represent carbon, nitrogen and lithium atoms, respectively.
Fig. S7 Optimized geometries, formation energies and adsorption heights of one lithium atom on different sites of the $N_{2}^{AB}$ defect at 0K. The small gray, middle-sized blue and large pink balls represent carbon, nitrogen and lithium atoms, respectively.
Fig. S8 Optimized geometries, formation energies and adsorption heights of one lithium atom on different sites of the $\text{N}_2^{\text{AH}}$ defect at 0K. The small gray, middle-sized blue and large pink balls represent carbon, nitrogen and lithium atoms, respectively.