Supporting Information for

Penta-BCN monolayer with high specific capacity and mobility as a compelling anode material for rechargeable batteries

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1 A detailed description of the eleven inequivalent adsorption sites

The 11 adsorption sites can be described as following: H site is the center of a pentagon; T_1 site is the top of the bottom N atom (N_b); T_2 site is the top of the top N atom (N_t); T_3 site is the top of the bottom B atom (B_b); T_4 site is the top of the top B atom (B_t); T_5 site is top of the carbon atom; S_1 site is the bridge of the midpoint of C-N_b bond; S_2 site is the bridge of the midpoint of C-N_t bond; S_3 site is the bridge of the midpoint of C-B_b bond; S_4 site is the bridge of the midpoint of C-B_t bond; S_5 site is the bridge of the B_t-N_t bond.

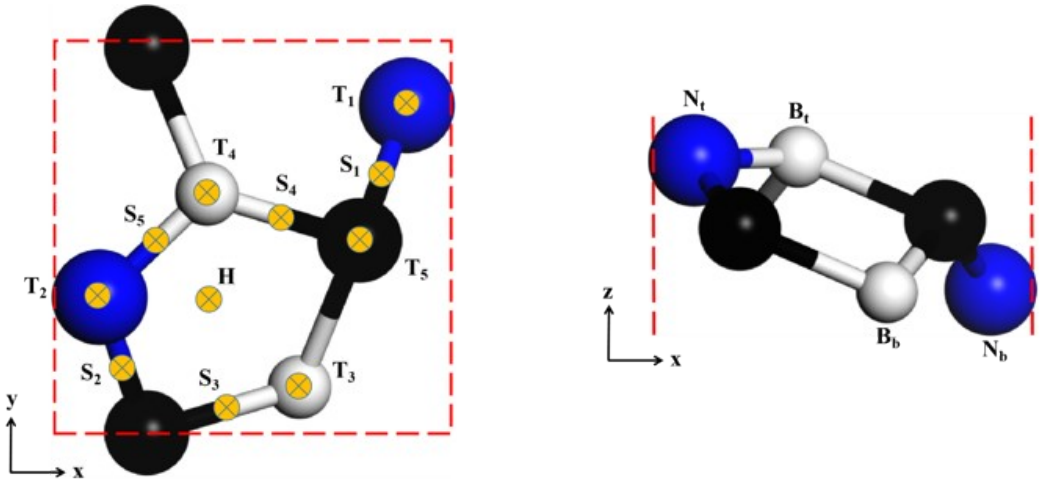

Fig. S1. Top and side views of the optimized unit cell of penta-BCN. Eleven inequivalent adsorption sites for anchoring Li/Na atom are marked.

2 Discussion of average adsorption energy

To explain why we only consider the average adsorption energy to study the adsorption properties, here, we take the case of Li adsorption as an example. To evaluate the adsorption energy for nth Li atom, several processing formulas are applied to study the variations of adsorption energies with the number of adsorbed Li atoms. The calculation details about the adsorption energy with different methods can be defined as follows. (a) The first method is the same to that in manuscript, for all the adsorbed Li atoms, the average adsorption energy E_{ave} can be defined as:

$$E_{ave} = \frac{E_{tot} - E_{sup} - nE_{Li}}{n} \quad (1)$$

here, the E_{tot} and E_{sup} are the total energies of the Li inserted and pristine penta-BCN monolayer. E_{Li} is the energy per atom in the bulk Li and integer n represents the adsorbed Li atoms on the penta-BCN monolayer. (b) The second method, the adsorption energy of nth Li atom E_n can be defined as:

$$E_n = E_{ntot} - E_{(n-1)tot} - E_{Li} \quad (2)$$

here, the E_{ntot} and $E_{(n-1)tot}$ are the total energies of the Li-BCN system with n and n-1 Li atoms. (c) The third method, the adsorption energy of 2nth Li atom E_{2n} can be defined as:

$$E_{2n} = E_{2ntot} - E_{2(n-1)tot} - 2E_{Li} \quad (3)$$

here, the E_{2ntot} and $E_{2(n-1)tot}$ are the total energies of the Li-BCN system with 2n and 2(n-1) Li atoms. (d) The fourth method, the adsorption energy of 3nth Li atom E_{3n} can be defined as:

$$E_{3n} = E_{3ntot} - E_{3(n-1)tot} - 3E_{Li} \quad (4)$$

here, the E_{3ntot} and $E_{3(n-1)tot}$ are the total energies of the Li-BCN system with 3n and 3(n-1) Li atoms. (e) The fifth method, the adsorption energy of nth Li atom E_{4n} can be

defined as:

$$E_{4n} = E_{4ntot} - E_{4(n-1)tot} - 4E_{Li} \quad (5)$$

here, the E_{4ntot} and $E_{4(n-1)tot}$ are the total energies of the Li-BCN system with 4n and 4(n-1) Li atoms.

The calculation results are shown in the Fig. S2(a), and the adsorption energies calculated by the formula (1)-(5) all have the same evolution trend: the fluctuation amplitude gradually decreases with the increase of adsorption concentration. However, the adsorption energy values obtained by different formulas are different at the same concentration. The fluctuation degree of the curves calculated by formula (2)-(5) are all relatively large, and it increases with the increase of sampling interval during calculation. That is to say, these results are not reliable. From the statistical point of view, the more densely the data is sampled and the average is taken into account, the more convincing the reflected tend is, such as the Eave curve calculated by formula (1). In addition, a large amount of Li ions are often adsorbed onto the anode material simultaneously during the actual operation processes of the batteries. Coincidentally, formula (1) is just satisfying such requirements, and in many reported studies[1–3], this formula is also used to conduct adsorption performance. Therefore, we employ the more convincing Eave to measure the interactions.

Similarly, we use the same analysis idea for the calculation of open circuit voltage. The open-circuit voltages (V_{OCV}) was only defined as (6)[1,4,5]:

(6)

where the E_{X_1} and E_{X_2} are the total energy of different Li-BCN systems, which loading X_1 and X_2 numbers of metal atoms. And e is the elementary charge. From this formula we can get that the differences in OCV results are mainly depended on the value of X_2—X_1. Taking the adsorption case of Li as an example, different data sampling intervals are taken for the denominator in formula (6) and the obtained results are shown in Fig. S2(b). As known to all, the open circuit voltage obtained at a certain moment or specific number of ions is meaningless. In practical application, the average open circuit voltage can more reflect the suitability of candidate material. At present, the open circuit

voltage performances in Fig. S2(b) all meet the threshold of ideal anode materials (0.1~1 V)[6]. In addition, many reported works have also applied the average open circuit voltage.[7,8]

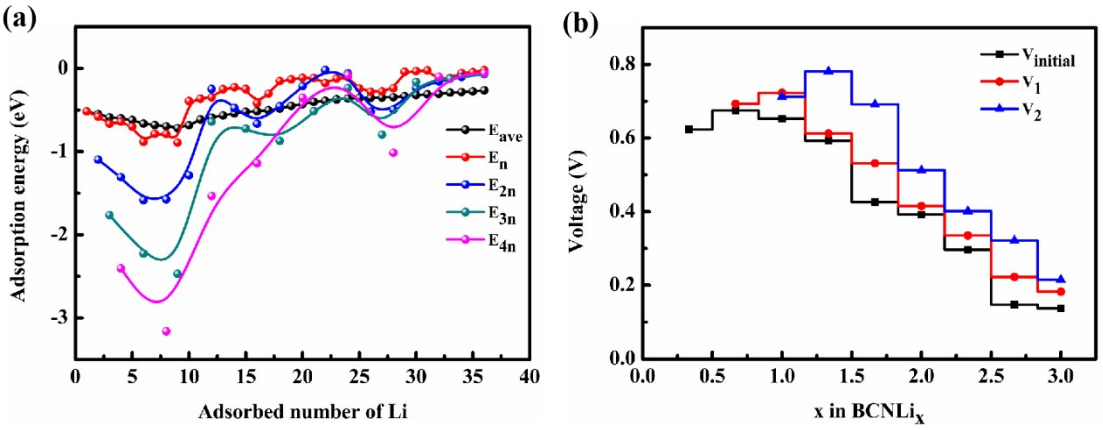

Fig. S2 (a) The variations of adsorption energy with the number of adsorbed Li atoms obtained by applying different processing methods. (b) The variations of open circuit voltage with the adsorption concentration of Li which are obtained by applying different processing methods. Black curve is the initial one mentioned in manuscript. Red and blue curves correspond to sampling intervals of 2 and 4.

Table S1 The adsorption height of the nth layer (d_n) corresponding to the adsorption cases of Li and Na. Length unit is Å.

Name	d_1	d_2	d_3	d_4
Li	1.87	2.76	4.75	6.42
Na	2.38	4.89	7.30	8.26

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