

Carbon nanolayers modified with boron atoms as a basis for devices with ionic conductivity: theoretical study

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Abstract: The paper discusses the possibility of realizing ionic conductivity in graphene-like layers containing impurity boron atoms. Currently, the search for new batteries is an urgent task. This is due to a number of disadvantages of modern materials, in particular, lithium-ion batteries are characterized by easy flammability, combustibility and the formation of dendritic structures with repeated use. Quantum-chemical studies of the vacancy migration process identified with the ion movement on the nanolayer surface have been performed and the effect of substituting B atoms on the phenomenon under study has been determined. The results of computer modeling of the defect movement on the surface with nanolayers containing a different number of impurity atoms – from pure graphene to the so-called “borophene”, i.e., a nanolayer consisting entirely of boron atoms are presented. It is established that with an increase in boron atoms impurity, the height of the potential barrier decreases, which makes the realization of ionic conductivity in boron-containing nanolayers more effective than in pure carbon ones. The direct relationship between the concentration of impurity boron atoms in the nanolayer and the height of the potential barrier has been unambiguously determined. The results presented in this article were obtained using the molecular cluster model and the DFT calculation method with the exchange-correlation functional B3LYP (valence-split basis set 6-31G).

Keywords: boron impurity; density functional theory; graphene; ionic conductivity; molecular cluster model; surface defect; quantum chemical studies.

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Углеродные нанослои, модифицированные атомами бора, как основа для приборов с ионной проводимостью: теоретические исследования

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Аннотация: Обсуждается возможность реализации ионной проводимости в графеноподобных слоях, содержащих примесные атомы бора. В настоящее время поиск новых элементов питания является актуальной задачей. Это связано с рядом недостатков современных материалов, в частности, литий-ионных аккумуляторов, таких как легкая воспламеняемость, горючесть и образование дендритных структур при многократном использовании. Выполнены квантово-химические исследования процесса миграции вакансии, отождествляемые с движением иона по поверхности нанослоя и определено влияние замещающих атомов В на изучаемое явление. Представлены результаты компьютерного моделирования перемещения дефекта по поверхности нанослоев, содержащих различное количество примесных атомов – от чистого графена до так называемого «борофена», то есть нанослоя, целиком состоящего из атомов бора. Установлено, что при увеличении примесных атомов бора происходит уменьшение высоты потенциального барьера, что делает реализацию ионной проводимости в боросодержащих нанослоях более эффективной, чем в чистых углеродных. Однозначно определена прямая зависимость между концентрацией примесных атомов бора в нанослое и высотой потенциального барьера.

Результаты, излагаемые в данной статье, получены с использованием модели молекулярного кластера и расчетного метода DFT с обменно-корреляционным функционалом B3LYP (валентно-расщепленный базисный набор 6-31G).

Ключевые слова: графен; ионная проводимость; дефект поверхности; модель молекулярного кластера; борная примесь; теория функционала плотности; квантово-химические исследования.

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1. Introduction

It is impossible to imagine modern society without modern power elements, especially those based on lithium-ion batteries. In terms of its electronic energy characteristics, such as capacitance and output voltage, lithium is one of the most efficient power sources. But at the same time, the repeated use of batteries leads to the growth of dendritic structures in the electrolyte solution. In addition, the electrolyte solution itself is highly flammable and combustible. Therefore, energy consumption requires new materials with greater capacitance and unique conductive properties. One of the most popular solutions is the use of so-called solid electrolytes, in which ionic conductivity is realized [1–4]. Recently, various applications of carbon and boron nanotubes as devices with ionic or proton conductivity have become very popular among researchers [5–8]. But in addition to them, there is also a carbon nanolayer – graphene, which also exhibits a number of properties that are interesting from the point of view of practical use, making it possible to predict the presence of ionic conductivity in it.

Graphene is a sp^2 -hybridized nanolayer material with a two-dimensional structure, exhibiting useful properties such as efficient thermal conductivity, electrical conductivity, mechanical strength, and ease of functionalization. These properties have led to significant attention being paid to graphene in the fields of energy, environmental protection, and biomedicine [9–12]. Its chemical neutrality when used as a catalyst and its metallic conductivity greatly limits the commercial application of pure graphene. However, efforts are being made to unlock the potential of graphene and related materials by modifying its structure to control its physicochemical properties. Doping graphene with another element can significantly change the electronic and physicochemical properties of the primary nanostructure. Changing the properties of graphene by carrying out substitution reactions leads to the loss of free electrons, as described in [13, 14]. Boron is

one of the most preferred elements for substitution reactions in carbon nanostructures [15]. To obtain graphene with impurity boron atoms, we used the method of catalytic vapor deposition from the gas phase [16], which previously proved to be excellent in the case of tubular nanostructures [15].

The reaction of replacing carbon atoms with boron in graphene is successful due to its close size to carbon and their close proximity in the periodic table. It is interesting that B in the hexagonal carbon framework increases holes with charge carriers due to its p-type properties and changes the band gap, which makes it possible to control the electronic properties of the nanolayer [17].

But the synthesis of graphene modified with boron atoms remains a difficult industrial task. In this regard, there are few practical works on studying the properties of this nanomaterial. For example, the research team led by Khan was one of the first to report and investigate B-doped graphene materials [18]. Park's group described graphene modified with B and investigated its electrochemical properties [19]. Tirumal has demonstrated that B-doped graphene obtained using a hydrothermal process exhibits significant electrochemical performance for batteries with a high capacitance value [20]. In addition, Yang's group demonstrated B-modified graphene as an electrode material for solar cells [21]. The discussed works, as well as the studies of the Lee group [22], show that graphene modified with boron atoms has the potential to be used as batteries or electrodes for solar batteries.

Thus, the use of graphene nanolayers modified with boron atoms can act as a power element, but in order to more accurately elucidate the physical principles underlying this mechanism, it is necessary to conduct theoretical studies to establish the dependence of the properties of nanolayers on the content of various concentrations of impurity boron atoms in them. This work is devoted to this problem.

To conduct a model experiment, we carried out a theoretical study using the density functional method of the mechanisms of ionic conductivity in nanolayers containing different contents of impurity

boron atoms. During the experiment, various concentrations of substituting atoms were considered in order to more accurately elucidate the features of the mechanism under study dependence on the concentration of B. Quantum-chemical calculations covered objects from pure graphene nanolayers to fully boron ones, which made it possible to formulate the final conclusion about the type of phenomena and regularities under study.

2. Materials and Methods

The density functional theory method is based on the principle of calculating the main characteristics of various chemical systems using the electron density functional $n(r)$. This function takes into account three variables responsible for the ground state and the excitation spectrum of the molecule. The first scientists who established the possibility of introducing such a functional for a multielectron system at zero temperature were Kohn and Hohenberg [23]. But the principle of setting such functionals was not covered in their work. Constructing such a functional in a practical way was first managed by Kohn and Sham (the Kohn–Sham) [24].

It is important to consider the derivation of the Kohn–Sham equation. The first step is to determine the form of the functional for the average energy, taking into account the above theorems:

$$\begin{aligned} E[n] &= \langle \Psi[n] | (\hat{T} + \hat{U} + \hat{V}_{ext}) | \Psi[n] \rangle = T + U + V_{ext} = \\ &= T_S + V_H + V_{ext} + (T - T_S + U - V_H). \end{aligned}$$

In this equation, the last term is responsible for the contribution of the exchange-correlation energy:

$$V_{XC} = (T - T_S + U - V_H).$$

This expression includes four terms, the pair wise difference of which adds up to the indicated energy value. The first difference is between the kinetic energies of interacting and free particles, and the second is between the energies of the Coulomb interaction and Khartri.

For greater certainty, let's rewrite the Kohn–Sham functional indicating the functional dependence of the terms before moving on to specific calculations:

$$E_{KS}[n] = T_S[n] + V_H[n] + V_{ext}[n] + V_{XC}[n].$$

To carry out the variation, we set the appropriate ratios:

$$\begin{aligned} \frac{\delta E_{KS}}{\delta \Psi_{i\sigma}(r)} &= \frac{\delta T_S}{\delta \Psi_{i\sigma}(r)} + \\ &+ \left[\frac{\delta V_H}{\delta n(r)} + \frac{\delta V_{ext}}{\delta n(r)} + \frac{\delta V_{XC}}{\delta n(r)} \right] \frac{\delta n(r)}{\delta \Psi_{i\sigma}(r)} = 0; \\ \frac{\delta T_S}{\delta \Psi_{i\sigma}(r)} &= -\frac{1}{2} \nabla^2 \Psi_{i\sigma}(r), \frac{\delta n(r)}{\delta \Psi_{i\sigma}(r)} = \Psi_{i\sigma}(r). \end{aligned}$$

The introduction of the Lagrange multiplier (denoted below as $\varepsilon_{i\sigma}$) sets the normalization condition. Considering all the above operations, we can write the Kohn–Sham equation:

$$-\frac{1}{2} \nabla^2 \Psi_{i\sigma}(r) + v_{KS}(r) \Psi_{i\sigma}(r) = \varepsilon_{i\sigma} \Psi_{i\sigma}(r).$$

The following formula defines the form of the Kohn–Sham potential:

$$v_{KS}(r) = v_{ext}(r) + v_H(r) + v_{XC}(r);$$

$$v_H(r) = \int dr' \frac{n(r')}{|r-r'|};$$

$$v_{XC}(r) = \frac{\delta V_{XC}}{\delta n(r)};$$

$$n(r) = \sum_{i\sigma} |\Psi_{i\sigma}(r)|^2.$$

The Kohn–Sham equation coincides in form with the one-particle Schrödinger equation, which describes the behavior of a particle in a self-consistent potential. The key difference from this equation is the allowance for the exchange-correlation potential and eigenvalues $\varepsilon_{i\sigma}$.

As mentioned above, nanolayers can act as materials in which ionic conductivity can be realized [25]. Figure 1 shows images of some of the studied nanolayers to illustrate the mutual arrangement of B and C atoms [26]. To test the implementation of ionic conductivity in nanolayers, we simulated the movement of a vacancy over their surface. The process of vacancy migration in nanolayers was modeled as follows: one of the atoms was removed from the surface until the moment of complete detachment and the formation of a vacancy defect [5]. After that, a step-by-step (after 0.01 nm) approach of the nearest neighboring atom to the place of vacancy localization took place. That is, the atom and the vacancy actually changed places, which can be identified with the movement of the vacancy over the surface of the

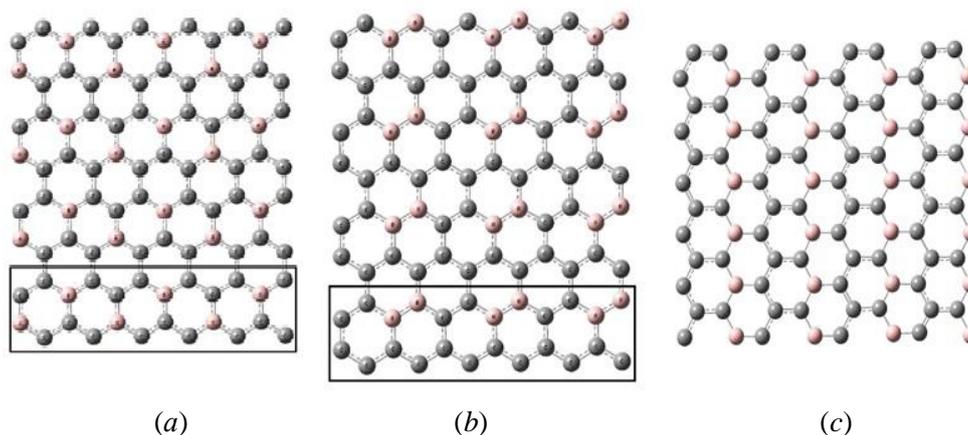


Fig. 1. Fragments of boron-carbon nanolayers with different concentrations of boron atoms:
a, b – the relative position of boron and carbon atoms in BC₃ graphene nanolayers of types A and B;
c – a variant of the atomic ordering of B and C atoms in a fragment of BC₅ nanolayer

nanolayer. For the movement of the atom, two main paths were chosen, which correspond to two nonequivalent chemical bonds in the nanolayer. Figure 2 shows the directions of motion of nanolayer atoms to the location of the vacancy. When moving, the atom deviates from the plane of the two-dimensional nanostructure, by giving it two degrees of freedom during optimization. The positions of the rest of the surroundings of the vacancy defect were optimized to analyze the geometry of the system. When performing quantum chemical calculations, the DFT method was used within the framework of the B3LYP functional and the 6-31G basis. Such modeling ensured the correspondence of the movement of a vacancy over the nanolayer surface due to the replacement of its location by a neighboring surface atom. Due to the emerging redistribution of the electron density and the fact that the environment of the vacancy turned out to be charged due to uncompensated chemical bonds, its movement can be identified with the movement of an ion over the surface of a graphene nanolayer with impurity boron atoms.

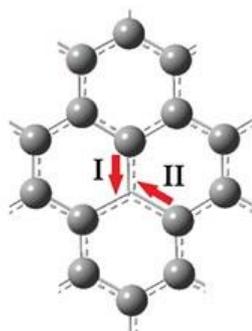


Fig. 2. The main directions of the movement of atoms to the vacancy on the surface of a graphene boron-containing nanolayer

To determine the role of impurity boron atoms in the processes under study, quantum chemical calculations were performed for graphene nanolayers with different contents of impurity boron atoms: BC; BC₃; BC₅; B₃C; B₅C.

3. Results and Discussion

In the course of studying the theoretical model, when a boron impurity appeared in nanolayers, it was found that an increase in the concentration of substituting B atoms also increases the band gap. This may be due to the redistribution of the electron density with the appearance of impurity B atoms. According to the band gap width, the considered nanomaterials belong to semiconductors.

As a result of performing model experiments according to the mechanism described above, the energy curves of these processes were obtained. The analysis of the energy curves showed that when a defect moves over the surface, potential barriers appear on the energy curves, which are located approximately in the middle of the bond between neighboring atoms for both considered nanolayers. The atom, leaving the equilibrium position, which corresponded to 0 on the abscissa axis, moved along the chemical bond connecting it and the location of the vacancy. At the same time, at a distance of 0.08 nm for the boron nanolayer and 0.09 nm for the carbon nanolayer, it reached the peak of the potential barrier, identified with the activation energy of these processes. For the carbon layer, the barrier height was 8.7 eV; for the boron layer, it was 1.25 eV. Thus, it can be argued that vacancy migration over the surface of the boron layer can be realized, in contrast to the case of the carbon layer, where the potential barrier height is too high. Thus, when studying the

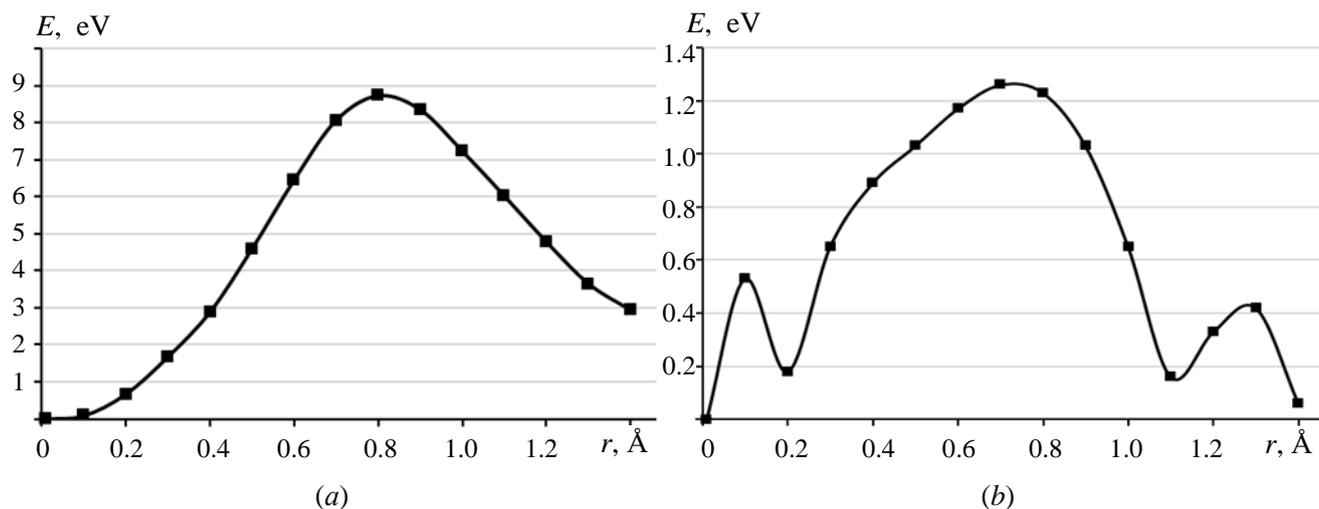


Fig. 4. The energy curve showing the features of the vacancy migration process r is the distance between the atoms of the nanolayer, which actually corresponds to the distance between the vacancy and the atom closest to it: a – for the carbon layer; b – for the boron nanolayer

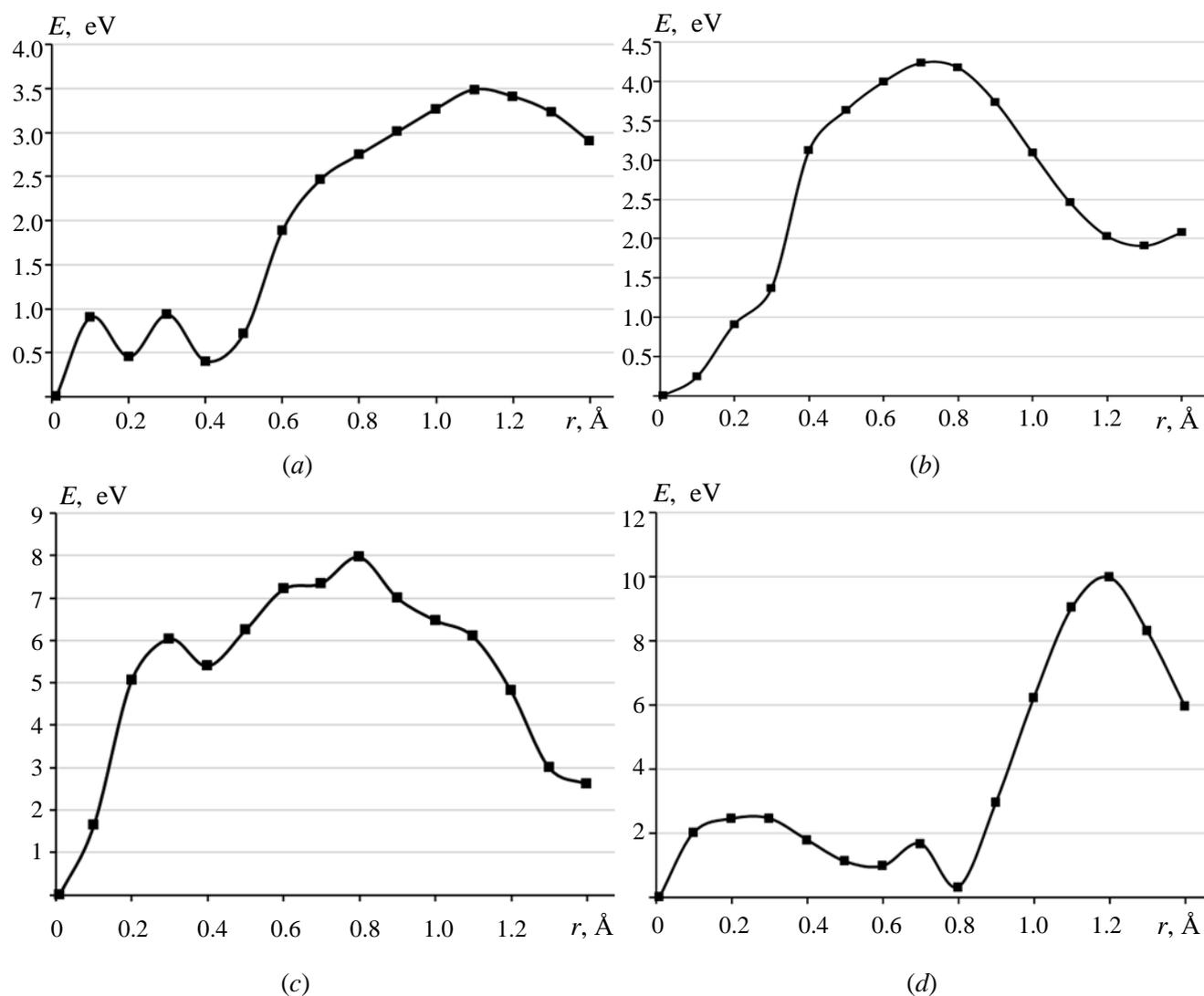


Fig. 5. The energy curve showing the features of the migration process of vacancies: a – in the BC_5 layer when the vacancy moves from the carbon atom to the place of the boron atom; b – in the BC_5 layer when the vacancy moves from the B atom to the place of the C atom; c – in the B_3C layer when the vacancy moves from the C atom to the place of the B atom; d – in the B_5C layer when the vacancy moves from the B atom to the place of atom B

boron layer, it was concluded that the height of the potential barrier makes it possible to implement ionic conductivity in this nanostructure. It should be noted that, due to the high energy of the potential barrier in the carbon layer, the implementation of the vacancy migration mechanism in it is impossible.

Next, the mechanism of vacancy migration was studied in carbon nanolayers with different percentages of boron in them. Nanolayers with different concentrations of impurity boron atoms, from the minimum in the BC₅ structure to the maximum B₅C, were considered in order to evaluate the effect of B atoms on the implementation of ionic conductivity in them, since a pure graphene nanolayer showed the impossibility of this type of conductivity even in a semi-metallic nanostructure.

When studying nanolayers with different concentrations of boron for the migration of vacancies in them, the following results were obtained. A potential barrier is observed for the BC₅ nanolayers (when a vacancy moves from a boron atom to a carbon atom, it is 4.25 eV, and when a vacancy moves from C to B 3.4 eV), in the B₃C layer when a vacancy moves from a carbon atom to the place of a boron atom (the height of the energy barrier during vacancy migration in this layer is 8 eV). But at the same time, it should be noted that large energy expenditures are needed to overcome these barriers, and, therefore, it can be argued that it is rather problematic to implement the migration of vacancies in them.

4. Conclusions

In the course of a model experiment, various options for a vacancy movement over the surface of boron-carbon nanolayers were considered, since the formation of a vacancy defect at the site of boron or carbon atoms is not chemically equivalent. This is due to the appearance of a non-equilibrium charge distribution, i.e., when a carbon atom is removed, the vacancy is a positively charged ion, and when a boron atom is removed, it acts as a negative charge carrier. This nonequivalence is seen when considering a vacancy moving from a carbon atom to a boron atom over the surface of the B₃C nanolayer, since in this case a high potential barrier of 8 eV is observed. In all other cases, the introduction of boron atoms in the form of an impurity to the carbon atoms of the nanolayer has a positive effect on the migration process, since it reduces the height of the potential barrier to zero in the case when the concentration of impurity boron atoms exceeds 25 %. At a lower concentration of B atoms, a potential barrier is observed for any of the considered pathways of

vacancy migration. Moreover, the activation energies identified with the height of the potential barrier are approximately equal, as shown in the table. The barrierless movement of a vacancy over the surface of the nanolayer can be explained by the fact that, due to the mixed boron-carbon composition, a nonequilibrium distribution of electron density arises, which is concentrated at carbon atoms, but can also be redistributed under the influence of external and internal factors. At the same time, such local regions can create their own local fields, which will attract or repel a vacancy, depending on the sign of the charge on the atom, contributing to its effective movement. The uniform electron density in pure nanostructures, on the contrary, hinders the implementation of this mechanism.

Thus, it can be concluded that the introduction of impurity boron atoms into carbon nanolayers leads to an improvement in its conducting properties when ionic conductivity is realized.

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6. Conflict of interests

The authors declare no conflict of interest.

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