

Effects of N-doped concentration in graphene on CO₂ adsorption

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ABSTRACT

Solving the problem of the environment related to CO₂ emission is an urgent and challenging task to prevent the escalation of climate change, which is due to increasing energy demand from fossil fuels and industrial activities. Using graphene to capture CO₂ gas is of great interest to mitigate global warming, where CO₂ capture by doped graphene has been shown to significantly improve the CO₂ adsorption capacity compared to that of pure graphene. In particular, N-doped graphene is a unique structure that was suggested for CO₂ capture by the adsorption phenomenon. However, there is no research available to clarify nitrogen doping concentration in graphene on CO₂ adsorption. Therefore, this work has been devoted to elucidating the problem using the density functional theory method considering van der Waals interaction. We showed that increasing the doping content of nitrogen by 3.1, 6.3, 9.4, and 12.5 % will increase the CO₂ adsorption energy, monotonically for N content below 9.4 % and significantly for 12.5 %. The most favorable adsorption configuration of the CO₂ molecule should be parallel to the surface of the N-doped graphene. The structure of the substrate at 12.5 % N distorted upon the CO₂ adsorption; therefore, the substrate is more active for CO₂ capture. The physical meaning underlying the interaction of the N@graphene system is physisorption, which is due to the contribution of the N *p_z* state and the CO₂ *p_z* state along the surface normal of the substrate. Increasing the N-doping content shifts down the unoccupied states of the N *p_z* orbital in the conduction band across the Fermi level to the valence band. Hence, the population of the occupied state at the Fermi level increases as increasing the N-doping concentration, leading to the stronger interaction of the CO₂ molecule with the N@graphene substrate. The results should be useful for the rational design of suitable N@graphene substrates for CO₂ capture and storage applications.

Key words: Toxic gas, Capture, Doping, Two-dimensional materials, Density functional theory, Physical properties

INTRODUCTION

Capturing and storing CO₂ by adsorption is a viable method attracting much attention to control the amount of CO₂ released into the environment, which is the cause of climate change. Finding suitable adsorbents for efficient and low-cost CO₂ capture is challenging for researchers. Various CO₂ adsorbents such as zeolites¹, amine-enriched carbon sorbent², hydrotalcite-like compound³, molecular sieves⁴, metal-organic frameworks⁵, carbon-based materials⁶, porous graphene and nitrogen-doped graphene⁷⁻¹⁰ have been investigated.

Graphene is not only a transparent material with a large surface area, high mechanical strength, and high carrier mobility but also has a good surface structure for the CO₂ adsorption sites⁷. Doping graphene with N could offer high CO₂ adsorption capacity¹⁰. The N atoms in the graphene structure enhanced the interaction energies¹⁰. Many approaches have been used to fabricate N-doped graphene^{11,12}. Experimental results showed that CO₂ adsorption capability significantly increased when doping nitrogen in

the pyridinic and pyrrolic forms⁹. Theoretical studies have demonstrated that graphitic-doping nitrogen is more stable than the pyridinic and pyrrolic forms in the free-standing graphene layer¹³. Generally, the literature has analyzed the CO₂ adsorption on the N@graphene system^{14,15}. However, no studies are available to clarify the influences of the N concentration on CO₂ adsorption. Therefore, we investigated the CO₂ adsorption on the N@graphene system based on density function theory calculations with van der Waals interaction by analyzing the geometric structures and physical properties of the N@graphene system. Graphitic nitrogen concentrations vary from 3.1, 6.3, and 9.4, to 12.5 %.

METHOD

In the present work, we analyzed the geometric structure, total energy, and electronic properties of CO₂ on the N@graphene using the van der Waals density functional theory method integrated into the VASP code¹⁶⁻²⁰. The exchange-correlation energy was considered by using the Perdew-Burke-Ernzerhof gen-

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eralized gradient approximation (GGA-PBE)²¹⁻²³. The valence electron-ion interactions were described by using the projector-augmented-wave method^{24,25}. We used Monkhorst-Pack's special k-point sampling technique with a $3 \times 3 \times 1$ k-point grid for structure optimization and total energy calculation²⁶. The plane-wave cutoff energy was 400 eV. We used the Gaussian smearing of order 0 with the width of 0.1 eV for a better convergence speed of the calculations. The N@graphene system was modeled using the 4×4 unit cell with a vacuum space of 18 Å in a perpendicular direction to the surface of the N@graphene system. All-atom positions of the N@graphene/CO₂ system were fully relaxed during the geometry optimization with the upper criterion of 0.001 eVÅ^{-1} for interatomic forces.

To evaluate the adsorption capacity of CO₂ on the N@graphene, we used the following formula:

$$E_a = E_{sub+CO_2} - (E_{sub} + E_{CO_2}), \quad (1)$$

where, E_{sub+CO_2} is the total energy of N@graphene/CO₂ system. E_{sub} and E_{CO_2} are total energies of clean substrate and isolated CO₂, respectively.

RESULTS AND DISCUSSION

Substrates of N@graphene

We optimized the possible structure of the N@graphene substrates according to the nitrogen content of 3.1, 6.3, 9.4, and 12.5 % by substituting one, two, three, and four nitrogen atoms into the graphene unit cell, respectively. The N-doping content in graphene varying from 3.1 to 9.4 % has been investigated previously for other applications^{27,28}. We focused only on the close arrangement of the N atoms in the unit cell because the far-away dispersion will approach the case of the single N atom doping²⁸. The optimized structures were listed in Figure 1, where, N₁, N₂, N₃, and N₄ are the denotation for the substitution of one, two, three, and four N atoms, respectively. The A, B, C, and D symbols represented the position of the N atoms in the graphene unit cell. For doping one N atom (3.1 %), all possibilities are equivalent and denoted as N₁. For doping two and more N atoms, we considered four structures (N_{2AA}, N_{2AB}, N_{2AC}, and N_{2AD}) for 6.3 %, two structures (N_{3AAB} and N_{3ABB}) for 9.4 %, and four structures (N_{4AABD}, N_{4AABB}, N_{4AABC}, and N_{4AADD}) for 12.5 %. For each percent of doping, there are many possible arrangement of the N atoms. However, for the selected substrates, we have shown in the following parts that we had found the simple rule

of the adsorption energy of CO₂ molecule on the N content. We also optimized the unit cell size for each N content; however, they remain almost the same as that of the pure graphene.

CO₂ molecule on N@graphene substrates

We investigated CO₂ adsorption on the optimized N@graphene substrates by initially placing the CO₂ molecule with parallel and incline configurations with a distance of 3 Å to the surface. We first performed a structural optimization, then calculated CO₂ adsorption energy. Table 1 showed that all the calculated values of adsorption energy are negative. The more negative adsorption energy, the more stable the configuration of the CO₂ adsorption will be. The favorable order of the CO₂ adsorption is N_{2AA} > N_{2AC} > N_{2AD} > N_{2AB} for 6.3 %, N_{3ABB} ≈ N_{3AAB} for 9.4 %, and N_{4AADD} > N_{4AABB} > N_{4AABD} > N_{4AABC} for 12.5 % N. The favorable order of the CO₂ adsorption on the N₂ substrates is similar to that of water on the N@graphene²⁸. Therefore, the most favorable adsorption configuration of the CO₂ molecule was found on the N₁, N_{2AA}, N_{3ABB}, and N_{4AADD} substrates for the N-doping percentage of 3.1, 6.3, 9.4, and 12.5 %, respectively. Shown in Figure 2 is the most favorable adsorption configuration for each N-doping content.

The CO₂ molecule in the most favorable adsorption configuration locates almost parallel to the substrates' surface, see Figure 2. The average bond length to the surface is about 3.19 Å for the N₁, N₂, and N₃ substrates. However, the bond length becomes shorter than 3.0 Å for the N₄ substrate. The configuration, adsorption energy, and bond length to the N₁ slab of the CO₂ molecule are in good agreement with the study¹⁵. Two oxygen atoms on top of the N atoms offer the most favorable adsorption configuration of CO₂ on N_{2AA}. For the N_{3ABB} slab, the C atom of the CO₂ molecule on the hollow site of the benzene ring and one oxygen atom on the top of the C atom nearest to one N atom in the substrate should be the most favorable configuration. For the N_{4AADD} substrate, the C-O bond of CO₂ is parallel to the N-N bond in the substrate, while the left oxygen atom of CO₂ is on the hollow site of the benzene ring.

Figure 3 shows the dependence of the CO₂ adsorption energy on the N-doped concentration in graphene. The adsorption energy monotonically decreases, and the adsorption strength increases as the doping content increases. With doping content 3.1, 6.3, and 9.4 %, the behavior is linear; however, it becomes significantly more negative for 12.5 % N. This behavior of

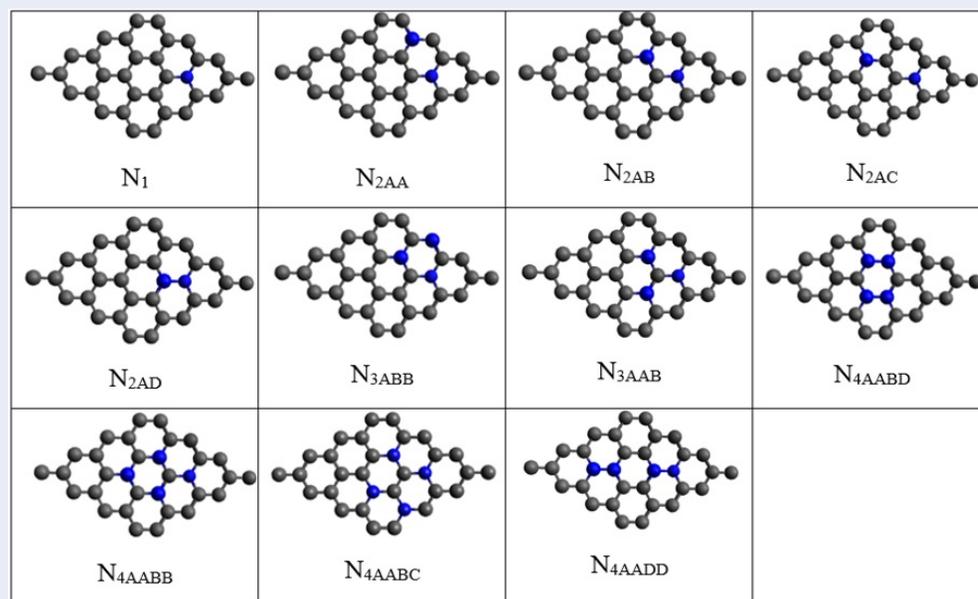


Figure 1: The substrate models of the graphitic N@graphene N_1 , N_2 , N_3 , and N_4 with a nitrogen content of 3.1, 6.3, 9.4, and 12.5 %, respectively. The nitrogen atom (blue) and the carbon atom (gray).

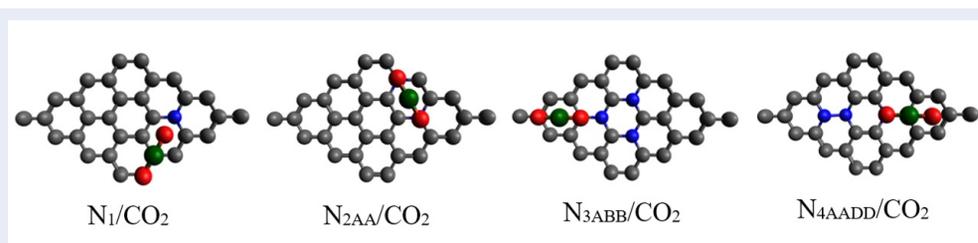


Figure 2: The most favorable adsorption configuration of CO_2 on the N@graphene substrates with nitrogen atom content of 3.1, 6.3, 9.4, and 12.5 % in that order. The carbon atom of graphene (gray), the nitrogen atom (blue), the oxygen atom (red), and the C atom of CO_2 molecule (green).

Table 1: CO_2 adsorption energy is E_a (eV), and $d_{gas-sub}$ (Å) is the average bond length of the CO_2 molecule with the substrate surface

N-doping content (%)	E_a (eV)				$d_{gas-sub}$ (Å)
3.1	-0.312				3.17
	N_1				N_1
6.3	-0.325	-0.316	-0.322	-0.310	3.21
	N_{2AA}	N_{2AD}	N_{2AC}	N_{2AB}	N_{2AA}
9.4	-0.338	-0.337			3.19
	N_{3ABB}	N_{3AAB}			N_{3ABB}
12.5	-0.343	-0.324	-0.326	-0.549	2.96
	N_{4AABB}	N_{4AABC}	N_{4AABD}	N_{4AADD}	N_{4AADD}

the adsorption energy on the N-doping content can be explained when considering the changes in the geometry and electronic properties.

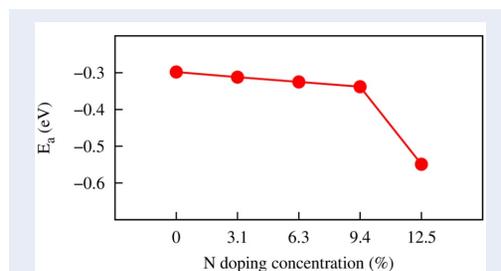


Figure 3: Adsorption energy of CO₂ molecule versus N doping content in graphene.

Analyzing the substrate's structure before and after the CO₂ adsorption showed that no significant changes have been found for the N-doped content from 3.1 to 9.4 %. When the N-doped content increased to 12.5 %, the geometrical structure of the substrate had a buckling shape around the CO₂ adsorption site compared with the structure of the clean substrate, see Figure 4. Figure 4b shows that the A1 and D2 atoms moved up about 0.01 Å; while A2 and D1 moved down about 0.02 Å in comparison to the isolated substrate structure Figure 4a. The total undulating distance is about 0.03 Å.

Electronic properties of N@graphene/CO₂ system

The interaction between CO₂ and the graphene substrate can be explained via the Bader charge analysis, as shown in Table 2. We found that the O and C atoms of CO₂ molecule always accumulate and donate charge, respectively. Also, the N and C atoms of the graphene substrate gain and lose charge in that order. The result is that the CO₂ molecule gain the negative charge from the graphene substrates. We also find that the charge exchange is small for 3.1, 6.3, and 9.4 % N, while it is significant for 12.5 % N. This behavior is consistent with that of adsorption energy analyzed above.

To understand which electronic states of CO₂ and the N@graphene dominate the interaction, we analyzed the density of states, as visualized in Figure 5.

As the N-doping content increases, the unoccupied state of the N p_z orbital in the conduction band shift down across the Fermi level to the valence band. The higher the N doping content, the more the occupied state of N p_z will become. Therefore, the population

of the occupied state at the Fermi level increases, leading to a stronger interaction of the CO₂ molecule with the substrates because the occupied states of the substrates will attract the unoccupied p_x , p_y , and p_z states of CO₂. Upon increasing the N content, these states of CO₂ move closer to the Fermi level, leading to a stronger interaction with the occupied state of N p_z . Especially, for the N content of 3.1, 6.3, and 9.4 %, the unoccupied states of the CO₂ molecule do not cross the Fermi level. Therefore, the charge exchange is ignorable, while it is significant due to the unoccupied state of CO₂ p_z crossing the Fermi level for the N doping content of 12.5 %. Thereby, the interaction of the CO₂ molecule with the substrates is the electrostatic attraction for 3.1, 6.3, and 9.4 %, and the charge exchange for 12.5 % N. This finding agrees with the analysis of Bader's charge above. We can state that the CO₂ p_z state becoming occupied is the reason for the charge exchange of the N@graphene system and significantly enhanced the CO₂ adsorption energy for 12.5 % N. As observed in Figure 6, the charge clouds of the N atoms and the oxygen atoms exhibit the shape of p_z orbitals (look like bowling pin along the surface normal).

CONCLUSION

We have elucidated the influences of the N concentration in graphene on CO₂ adsorption by the van der Waals density functional theory approach. Our results showed that the p_z orbital state of the doped nitrogen shifted down to increase the occupied states at the Fermi level upon the increase of N doping. These occupied states of the N atoms enhanced the interaction with the CO₂ unoccupied states to increase the CO₂ adsorption energy. Particularly, the unoccupied state of CO₂ p_z orbital partially becoming the occupied state lying in the top region of the valence band increased the charge exchange and significantly enhanced the adsorption energy of CO₂ for 12.5 % N over linear region 0.0, 3.1, 6.3, and 9.4 % N. The findings contribute to the knowledge of the interaction between the N@graphene and CO₂ for applications in CO₂ capture and sensors.

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COMPETING INTERESTS

The authors declare that they have no competing interests.

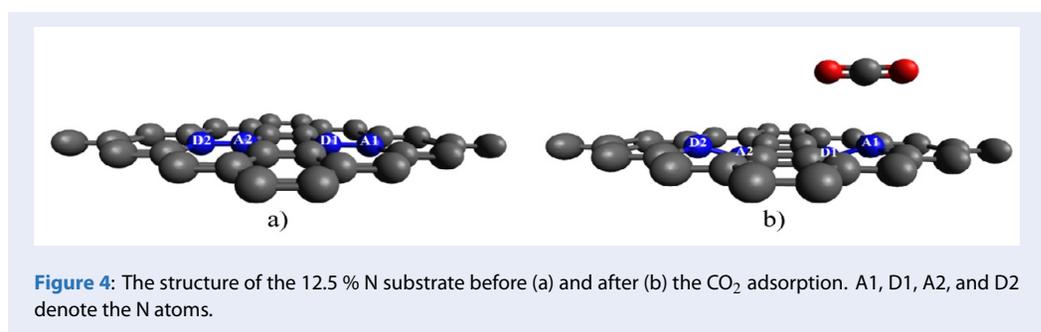
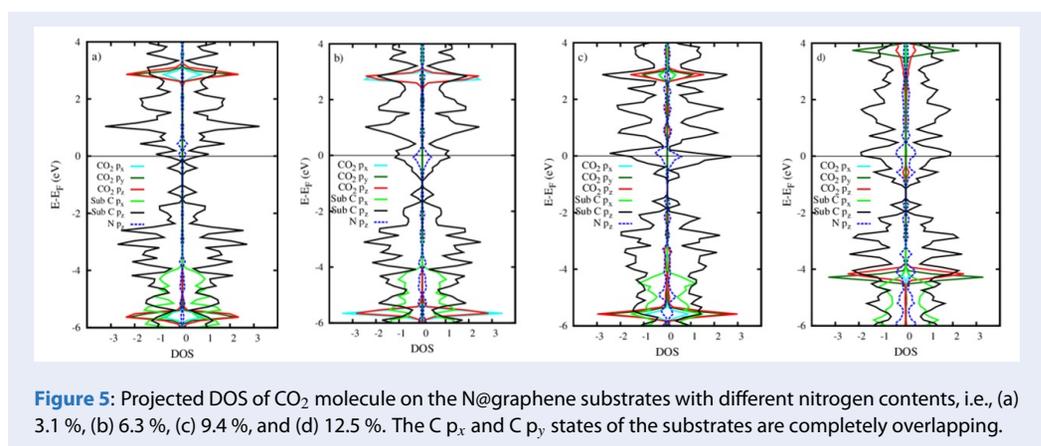


Table 2: Bader charge (e^-) for CO₂ adsorbed on N@graphene: (+) charge gain, (-) charge loss

N content (%)	3.1	6.3	9.4	12.5
2O	4.015	4.013	4.017	4.167
C	-4.000	-4.000	-4.000	-4.000
CO ₂	0.015	0.013	0.017	0.167
All	2.621	5.343	8.288	10.764
All C _{substrate}	-2.636	-5.356	-8.305	-10.931
Substrate	-0.015	-0.013	-0.017	-0.167



AUTHORS' CONTRIBUTIONS

Ong Kim Le: investigation, visualization, formal analysis, writing manuscript, reviewing and editing

Viorel Chihaiia: resources, supervision, reviewing and editing

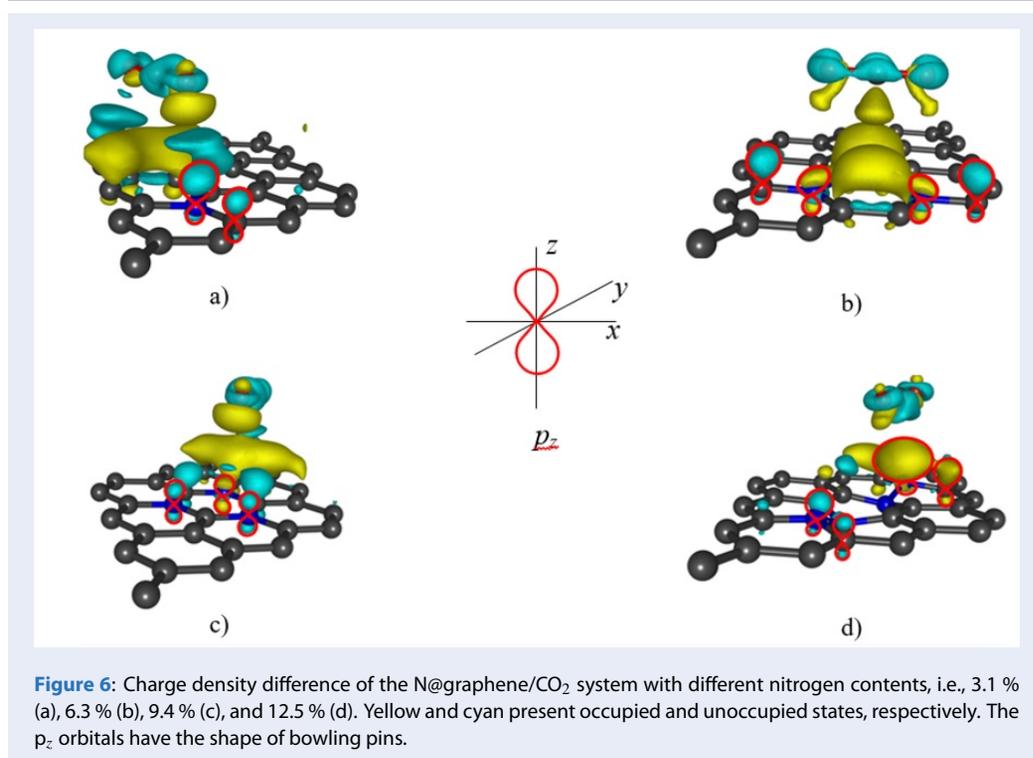
Do Ngoc Son: Conceptualization, formal analysis, resources, supervision, validation, writing manuscript, reviewing and editing

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Ảnh hưởng nồng độ pha tạp N trong graphene lên hấp phụ CO₂

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TÓM TẮT

Giải quyết vấn đề môi trường liên quan đến phát thải CO₂ là nhiệm vụ cấp bách và thách thức nhằm ngăn chặn sự leo thang của biến đổi khí hậu, do nhu cầu năng lượng từ nhiên liệu hóa thạch và các hoạt động công nghiệp ngày càng tăng. Sử dụng graphene để bắt giữ khí CO₂ rất được quan tâm để giảm thiểu sự nóng lên toàn cầu, trong đó việc thu giữ CO₂ bằng graphene pha tạp đã được chứng minh là cải thiện đáng kể khả năng hấp phụ CO₂ so với graphene nguyên chất. Đặc biệt, graphene pha tạp N là một cấu trúc độc đáo được đề xuất để thu giữ CO₂ bằng hiện tượng hấp phụ. Tuy nhiên, hiện chưa có nghiên cứu nào làm rõ ảnh hưởng nồng độ pha tạp nitơ trong graphene đối với sự hấp phụ CO₂. Do đó, công trình này được dành để làm sáng tỏ vấn đề bằng cách sử dụng phương pháp lý thuyết phiếm hàm mật độ có xem xét tương tác van der Waals. Chúng tôi đã chỉ ra rằng, việc tăng hàm lượng nitơ pha tạp lên 3,1, 6,3, 9,4 và 12,5% sẽ làm tăng năng lượng hấp phụ CO₂, một cách đơn điệu đối với hàm lượng N dưới 9,4% và rất đáng kể đối với 12,5%. Cấu hình hấp phụ thuận lợi nhất của phân tử CO₂ là song song với bề mặt của graphene pha tạp N. Cấu trúc của chất nền ở 12,5% N bị biến dạng khi hấp phụ CO₂; do đó, là chất nền có mức độ hoạt hóa cao hơn để thu giữ CO₂. Ý nghĩa vật lý làm cơ sở cho tương tác của hệ N@graphene chính là sự hấp phụ vật lý, do sự đóng góp của trạng thái N p_z và trạng thái CO₂ p_z dọc theo phương pháp tuyến bề mặt của chất nền. Việc tăng hàm lượng pha tạp N làm dịch chuyển các trạng thái không chiếm giữ của quỹ đạo N p_z trong vùng dẫn vượt qua mức Fermi sang vùng hóa trị. Do đó, mật độ trạng thái bị chiếm ở mức Fermi tăng lên khi tăng nồng độ pha tạp N, dẫn đến tương tác mạnh hơn của phân tử CO₂ với chất nền N@graphene. Các kết quả thu được sẽ hữu ích cho việc thiết kế các chất nền N@graphene phù hợp cho các ứng dụng thu giữ và lưu trữ CO₂.

Từ khóa: Khí độc, Bắt giữ, Pha tạp, Vật liệu hai chiều, Lý thuyết phiếm hàm mật độ, Tính chất vật lý

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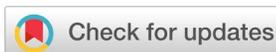
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