





Article

Absorption Behavior of Graphene Nanoplates toward Oils and Organic Solvents in Contaminated Water

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Abstract: In this work, graphene nanoplates (GNPs) were successfully prepared via direct chemical exfoliation from natural graphite. The properties of prepared GNPs were investigated using scanning electron microscopy and X-ray diffraction. The resultant GNPs had a low bulk density of 0.015 g mL⁻¹, C content of 99.6%, a lateral diameter of 5–15 μm, and thickness of <15 nm, respectively. The absorption capacity of the GNPs for a range of oils and organic solvents including pump oil, polydimethylsiloxane oil (PDMS), dioctyl phthalates (DOP) oil, olive oil, bean oil, mazut oil, xylene, ethanol, and ethylene glycol (EG) was evaluated. The effects of the sorption conditions such as the absorption time (sorption kinetics), temperature, and aging manner (static, stirring, and ultrasonic) on the absorption capability of the GNPs were also investigated. It could be seen that the GNPs revealed extremely high absorption capacities for oil and organic solvents, reaching 21–42 times their own weight only after 2 min of absorption at room temperature, which were found to be much higher than those shown by various commercially available absorbents.

Keywords: absorbent; graphene; oil absorption; organic solvent absorption; water treatment

1. Introduction

Oil spillage and organic solution effluents, which are the waste products of many process industries such as the plastic, textiles, pulp and paper, mining, and coating industries, have created serious environmental and ecological problems [1–3]. Therefore, it is crucial to develop novel methods for the effective treatment of oil and organic solvents in industrial wastewater. Various approaches, including

the use of sorbent materials [4,5], dispersants [6], burning [7], bioremediation [8], and mechanical collection [9], have been employed for removing organic solvents and oils from wastewater. A good material as an absorbent should exhibit high absorption capacity, cost-effectiveness, high selectivity, low density, environmental friendliness, and good recyclability. Carbon-based materials such as activated carbon and carbon nanotube sponges [10,11] are the most widely used sorbent materials for the treatment of oils and organic solvents in contaminated wastewater [12]. However, the development of a simple and cost-effective method for efficient and safe absorption of organic solvents and oils is rather a challenging task.

Over the past few years, graphene, one of the carbon materials with a two-dimensional (2D) structure, has gained immense scientific and industrial attention owing to its wide range of applications in the area of physics, chemistry, and engineering [10,11,13–17]. Three general routes have been employed to fabricate graphene: mechanical peeling, epitaxial graphene growth, and solution-based reduction of graphene oxide. Graphene possesses many excellent properties such as remarkable thermal and electrical conductivities, high surface area, and mechanical stiffness. Graphene scaffolds exhibit excellent electrical properties, which are essential for various applications such as batteries, sensors, and conducting polymer composites for the application of solar cells [18–21]. Moreover, owing to its hydrophobicity and highly specific surface area, graphene is considered as a promising material for the removal of oils and organic solvents from wastewater. As evident from recent reports, various graphene-based absorbents have been reported to remove oils and organic solvents from wastewater [22,23]. For example, the hydrothermal method was used to prepare spongy graphene from graphene oxide, which demonstrated high absorption capacity for organic solvents and oils [11,24]. In another study, an efficient and recyclable porous reduced graphene oxide absorbent was tested for removing oils and organic solvents from wastewater [25]. Furthermore, graphene can also form composites with other materials such as metals, metal oxides, and polymers to produce high-performing absorbents for the removal of oils and organic solvents from wastewater [10,11,26]. However, most of the graphene-based absorbents, especially for oils and solvents absorption, have been fabricated only at the laboratory scale. Furthermore, the oil and solvent absorption behavior of graphene prepared by direct chemical exfoliation of natural graphite has not been studied till now. Therefore, in order to realize its practical applications as an absorbent, it is imperative to investigate the oil and organic solvent absorption properties of graphene nanoplates fabricated by a new, scalable approach.

Herein, a study on the oil and organic solvent absorption properties of graphene nanoplates (GNPs) prepared using a new approach is reported. A range of oils and organic solvents including ethanol, xylene, polydimethylsiloxane oil (PDMS), dioctyl phthalates (DOP), ethylene glycol (EG), pump oil, bean oil, mazut oil, and olive oil, which are commonly used in industries, are tested in this study. The effects of the absorption conditions such as the temperature, time, and absorption method (static and dynamic) on the absorption capacity of the GNPs are also investigated.

2. Materials and Methods

2.1. Materials

Graphite was purchased from VNgraphene. Ethanol (C_2H_5OH), xylene (C_8H_{10}), ethylene glycol (EG), polydimethylsiloxane (PDMS), olive oil, bean oil, pump oil, DOP oil, and mazut oil were obtained from Van Minh company Ltd., Hanoi, Vietnam. The obtained graphite was dried at 100 °C for 4 h to remove the moisture from the surface. The other reagents were used as received.

2.2. Synthesis of GNPs

GNPs were synthesized by adapting a protocol used in a previous work (Figure 1) [27]. In a typical reaction, certain amount of graphite flakes was added in 240 mL sulfuric acid (98%) in a 1000-mL reactor and stirred for 10 min. Then, sodium persulfate ($Na_2S_2O_8$) was gradually added into the reaction mixture and further stirred for 3 h at room temperature. The resulting product was filtered

without quenching by using a glass sintered filter. The resulting GNPs were rinsed with dry acetone (3×30 mL) and 30 mL of water to remove any residual reactants. Finally, the GNPs were dried at 60°C in air.

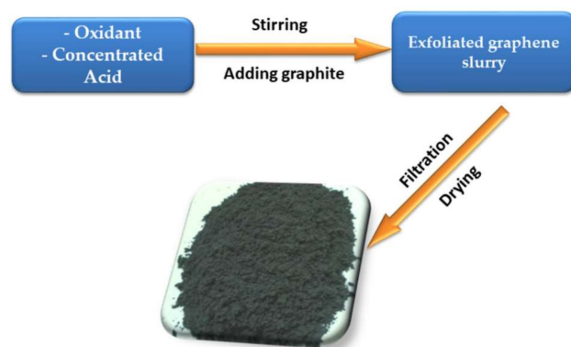


Figure 1. Schematic of the graphene nanoplate (GNP)-fabricating route by the direct chemical exfoliation of graphite.

2.3. Absorption of Oils and Organic Solvents

The testing procedures for absorption capability of GNPs for oil and organic solvents were carried out following previous work [28]. Nine different types of organic solvents and oils including ethanol, xylene, PDMS, EG, DOP oil, pump oil, bean oil, mazut oil, and olive oil were used in this study. Batch absorption tests were carried out at room temperature by adding 0.03 g of the GNP powder to a 50 mL beaker containing 10 mL of the oils/organic solvents. The mass of the GNPs was weighed prior to the absorption tests. After 2 min of absorption at room temperature, the absorbed GNPs were removed and weighed immediately.

2.4. Absorption Kinetics

The oils and organic solvents absorption kinetics of the GNPs were recorded as a function of absorption time [28]. Batch desorption was carried out by adding 0.03 g of the GNP powder to a 50-mL beaker containing 10 mL of mazut oil or EG. The absorption processes were carried out at room temperature, under static conditions for 2, 5, 10, 30, 60, 90, and 120 min, respectively. The mass of the GNPs was weighed before and after the absorption process to evaluate their absorption capacity as a function of time.

2.5. Effect of Temperature

Batch absorption was carried out by adding 0.03 g of the GNP powder to a 50-mL beaker containing 10 mL of mazut oil. The absorption experiments were carried out under the static condition for 5 and 60 min at various temperatures: 25, 45, and 60°C [28]. The mass of the GNPs was measured before and after the absorption to evaluate their oil and organic solvent absorption capacities as a function of temperature.

2.6. Effect of Aging Conditions

Batch absorption was carried out by adding 0.03 g of the GNP powder to a 50-mL beaker containing 10 mL of mazut oil. The absorption process was carried out at room temperature for 5 min under different aging conditions: static, stirring, and ultrasonic. The mass of the GNPs was measured before and after the absorption to evaluate their oil and organic solvent absorption capacities.

2.7. Recyclability and Recoverability

Ethanol and xylene were used to evaluate the recyclability and recoverability of the GNPs [28]. Batch desorption tests were carried out at room temperature by adding 0.03 g of the GNP powder to a

50-mL beaker containing 10 mL of ethanol or xylene for 5 min. The mass of the GNPs was measured before and after the absorption tests. The absorbed GNPs were dried at 100 °C for 4 h to completely remove the organic solvents. The “absorbing and drying” process was repeated five times.

3. Results and Discussion

The typical nature of graphite flakes with a thick lateral structure is shown in Figure S1a. The GNPs prepared in this study showed a low bulk density of 0.015 g mL^{−1}, C content of 99.6%, lateral diameter of 5–15 µm, and thickness of <15 nm [27]. Figure 2a shows the optical images of the GNPs. These images show that the GNPs were extremely light and highly porous. Figure 2b shows the XRD patterns of the GNPs and natural graphite. Unlike graphite, which showed an XRD peak at 25–28°, the GNPs showed a broad XRD peak at 26.5° corresponding to their less orderly structure. The less orderly structure of the GNPs was further confirmed by scanning electron microscopy (SEM) images (Figure 2c,d, Figures S1b and S2). It can be observed from Figure 2c,d that the obtained GNPs showed a creased and wrinkled morphology with 10–30 microns in lateral diameter and less than 20 nm in thickness [11].

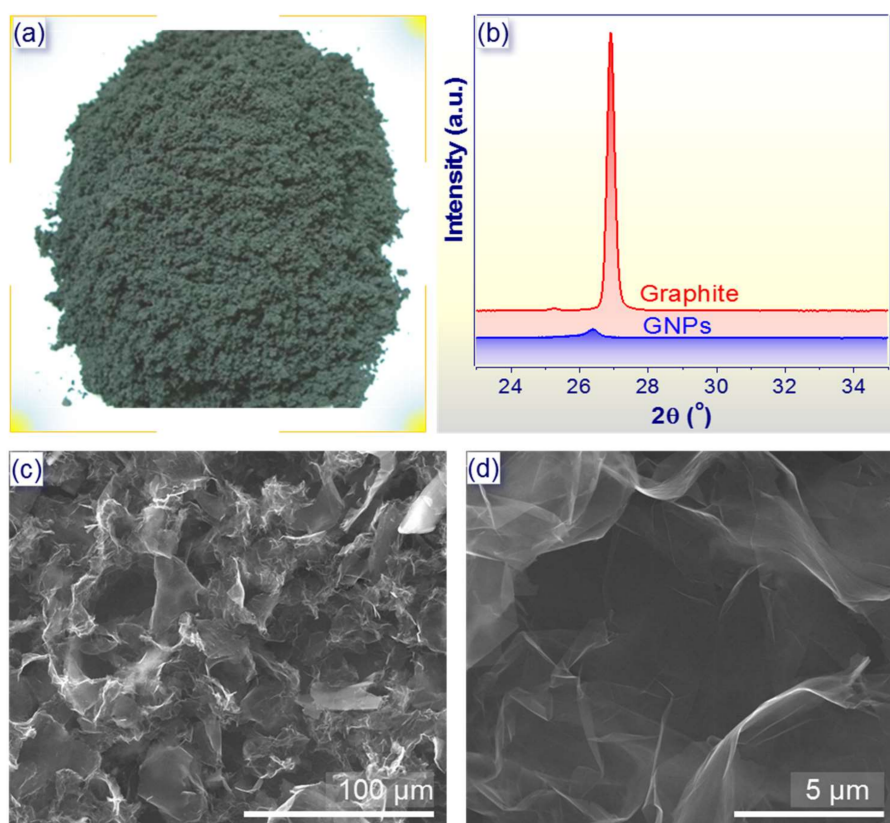


Figure 2. (a) Optical image of the resultant graphene products, (b) XRD patterns of natural graphite and the obtained GNPs, low- (c) and high-magnification, and (d) SEM images of the GNPs.

Figure 3a shows the optical images of the absorption process with the oil/water mixture before and after the introduction of the GNPs. It can be observed from the figure that the GNPs effectively absorbed the pump oil from water. When the GNPs were added to the pump oil/water mixture, the brown color of the mixture quickly and completely disappeared within a few minutes, indicating that the pump oil was effectively absorbed by the GNPs. This clearly suggests that GNPs can potentially be used as selective absorbents for the removal of organic solvents and oils from wastewater.

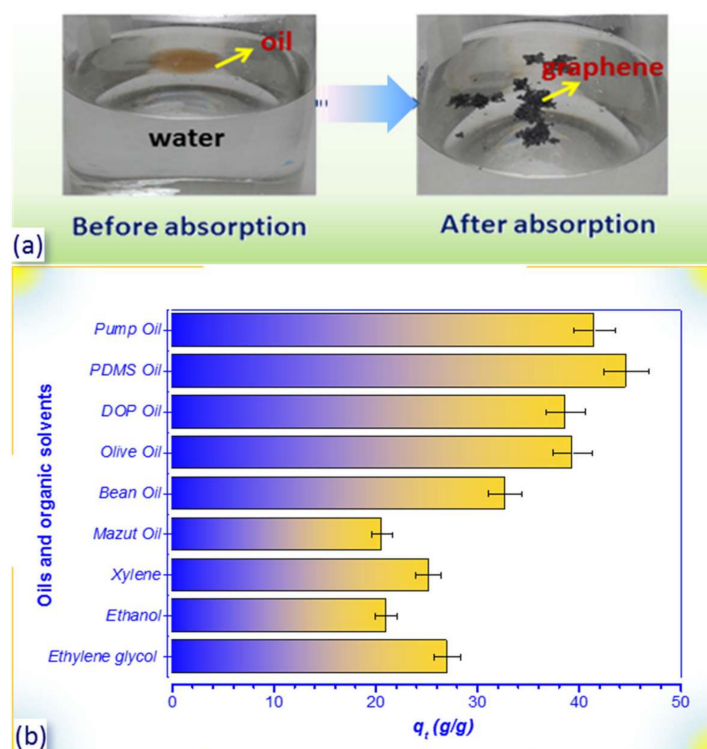


Figure 3. (a) Optical images of the pump oil/water mixture before and after the absorption by the GNPs and (b) absorption capacities of the GNPs towards various oils and organic solvents after 2 min of absorption at room temperature.

In order to demonstrate the absorption capability of GNPs toward organic solvents and oils, the absorption capacities (q_e) of the GNPs for selected oils and organic solvents including pump oil, polydimethylsiloxane oil, DOP oil, dioctyl phthalate oil, olive oil, bean oil, mazut oil, xylene, ethanol, and EG were evaluated. The absorption capacity was calculated according to Equation (1)

$$q_t = \frac{m_t - m_0}{m_0} \quad (1)$$

where q_t is the absorption capacity (g/g), m_0 is the weight of the GNPs before the absorption (g), and m_t is the weight of the GNPs after the absorption.

The absorption capacities of the GNPs for several oils and organic solvents are shown in Figure 3b. The GNPs showed absorption capacities ranging from 21 to 42 times their own weight only after 2 min of absorption at room temperature. This is much higher than the absorption capacities shown by other common commercial oil absorbents such as sepiolite [29], polydimethylsiloxane [29], corn stalk [30], polypropylene [31], butyl rubber [32], modified polyurethane sponge [33], and CNF/carbon foam [11]. The absorption capacities of the GNPs were affected by the density, viscosity, and surface tension of the oils and organic solvents [34]. Furthermore, the organic solvents loaded on GNPs could be easily removed by heating at 100 °C, and the oils loaded on GNPs could be removed by washing with acetone followed by heating at 100 °C. This indicates that the GNPs could be reused many times for the absorption of the organic solvents and oils.

The absorption kinetics and absorption behavior of mazut oil and EG on the surface of the GNPs were investigated. Figure 4 shows the absorption data of mazut oil and EG at different time intervals at room temperature. In the case of EG, the absorption process quickly reached equilibrium within 30 min. On the other hand, in the case of mazut oil, more than 1 h was required to reach the

equilibrium absorption state. The absorption kinetics of mazut oil and EG could be described using the pseudo-second-order model (Equation (2)), as follows:

$$q_t = \frac{Kq_e^2 t}{1 + Kq_e t} \quad (2)$$

where q_t (g/g) is the amount of mazut oil or EG absorbed on the GNPs at time t (min), q_e (g/g) is the amount of mazut oil or EG absorbed on the solid phase at equilibrium, and K is the absorption rate constant (g/g.h). The absorption kinetic values are listed in Table 1. The experimental data fitted well with the correlation coefficients of 0.894 and 0.8416 for mazut oil and EG, respectively. This indicates that the absorption process was chemical in nature and was accompanied by electron exchange between the GNPs and the oils and organic solvents [10]. The maximum absorption capacities of the GNPs for mazut oil and EG at equilibrium were calculated to be 61.35 and 50.35 g/g, respectively, which are much higher than the absorption capacities of these liquids obtained after 2 min of absorption.

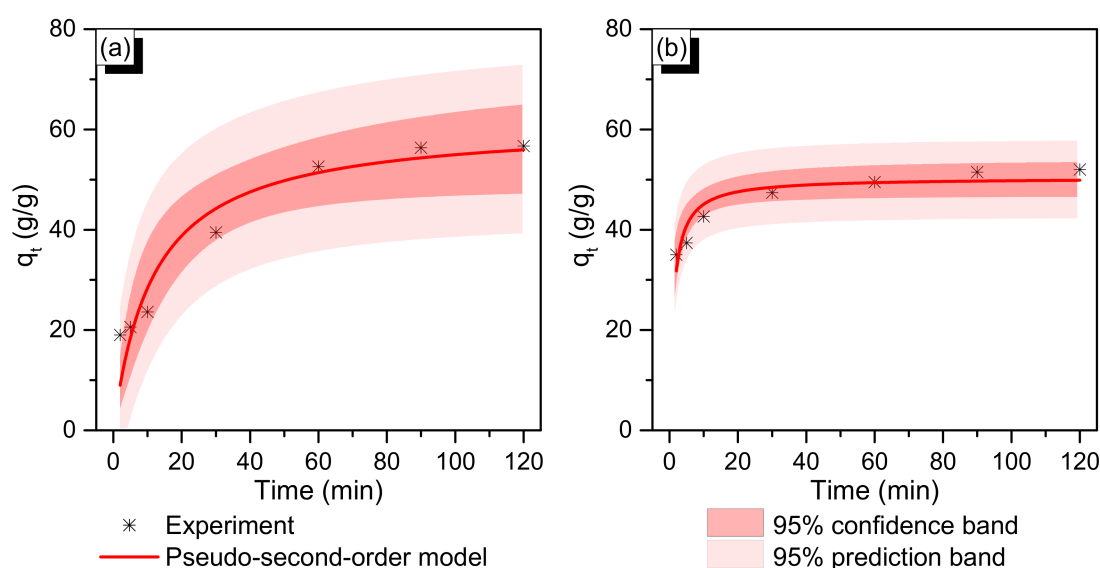


Figure 4. Absorption kinetics of (a) mazut oil and (b) ethylene glycol (EG) on the GNPs.

Table 1. Kinetics parameters for the absorption of mazut oil and EG on the GNPs.

Kinetic Parameters	Mazut Oil	Ethylene Glycol (EG)
q_e (g/g)	61.358 ± 5.046	50.356 ± 1.433
K (h^{-1})	$0.001 \pm 5.574 \times 10^{-4}$	0.017 ± 0.004
Reduced Chi-Sqr	31.214	7.412
R-Square (R^2)	0.912	0.868
Adj. R^2	0.894	0.842

The effect of temperature on the absorption capacity of the GNPs was also investigated. Experiments were carried out using mazut oil, for 2 min at various temperatures: 25, 45, and 60 °C (Figure 5a). The absorption capacity increased significantly from ~21 g/g at 25 °C to ~57 g/g at 45 °C. However, a further increase in temperature to 60 °C resulted in a negligible decrease in the absorption capacity (55 g/g). This indicates that 45 °C was the optimum temperature for the absorption process. The effect of the stirring and ultrasonic aging conditions on the absorption capacities of the GNPs was also investigated in this study. Figure 5b shows the absorption capacities of the GNPs for mazut oils at room temperature, after 2 min under the three different aging conditions: static, stirring, and ultrasonic. It can be observed from Figure 5b that stirring and ultrasonic aging promoted the absorption process of mazut oil on the GNPs. The fastest absorption was observed under the ultrasonic

aging condition. However, the differences in the absorption capacities under the three conditions were found to be negligible, indicating that the static condition can be utilized to remove oils and organic solvents from wastewater.

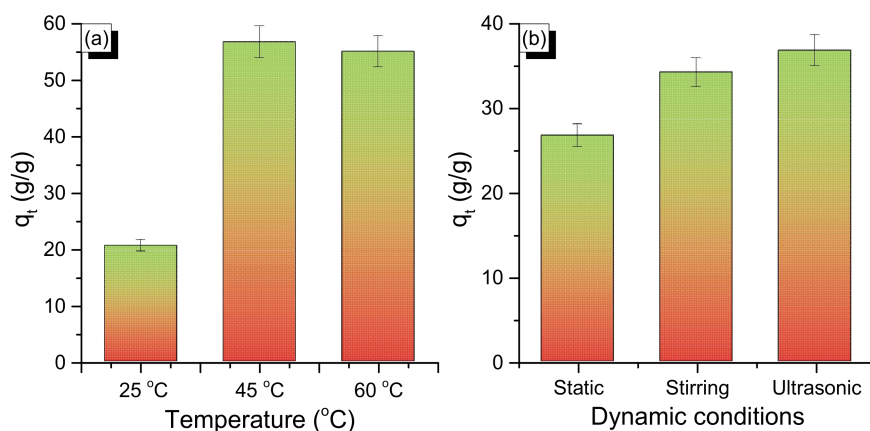


Figure 5. (a) Effect of temperature and (b) dynamic conditions (static, stirring, and ultrasonic) on the absorption capacity of the GNPs for mazut oil.

In order to realize the practical applications of GNPs for the removal of oils and organic solvents from wastewater, it is imperative to evaluate their recyclability and recoverability. Figure 6 shows the recyclability of the GNPs for the absorption of ethanol and xylene up to five cycles. The GNPs showed a negligible decrease in the removal efficiency (less than 4%) after five cycles, indicating high durability for the absorption of oils and organic solvents. The absorbed oils and organic solvents on the GNPs could also be easily recovered by washing with acetone and heating, respectively [35].

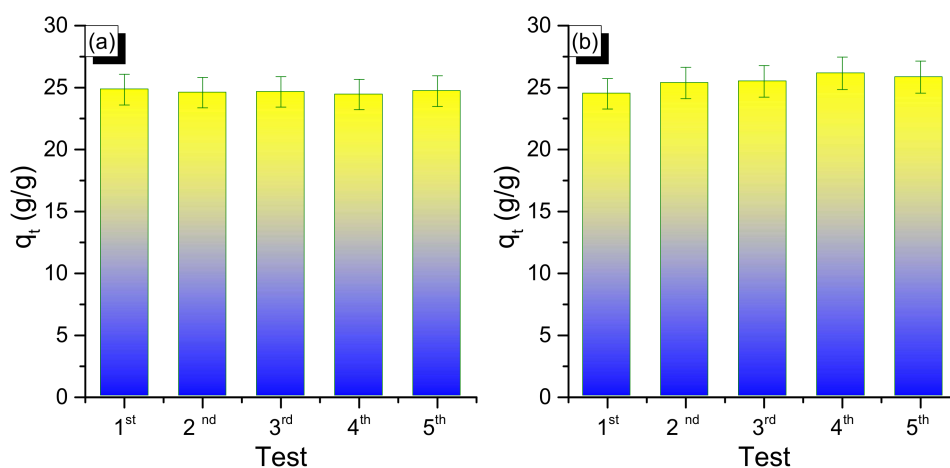


Figure 6. Recyclability of GNPs for the absorption of (a) ethanol and (b) xylene.

4. Conclusions

The absorption behavior of GNPs prepared by chemically exfoliating natural graphite towards different oils and organic solvents was successfully investigated. The resultant GNPs had a low bulk density of 0.015 g mL^{-1} , C content of 99.6%, lateral diameter of 5–15 μm , and thickness of <15 nm. The GNPs exhibited excellent oil and organic solvents absorption capacities, i.e., 21–42 times their own weight only after 2 min of absorption at room temperature. The GNPs showed higher absorption capacities than the commercial absorbents. The temperature and the dynamic conditions significantly affected the absorption behavior of the GNPs. The GNPs also demonstrated remarkable recyclability and recoverability, as oil-loaded GNPs can be extracted facilely via mechanical collection and reused for

next oil spill treatment. The approach proposed in this study to fabricate GNPs suggests that graphene can be used for efficient oil-spill treatment and for treating oil- and organic solvent-contaminated water.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2071-1050/11/24/7228/s1>, Figure S1: SEM images of natural graphite and graphene nanoplatelets, Figure S2: TEM images of graphene nanoplatelets.

Author Contributions: Conceptualization—D.D.L., D.D.N., T.A.N., and H.P.N.T.; Methodology—D.D.L., D.D.N., and X.X.; software D.D.N. and T.A.N.; Investigation—T.A.N., T.H.P.N., H.D.N., and T.T.N. (Tham Thi Nguyen); data curation—D.A.N. and T.D.D.; writing—original draft—D.D.L. and T.A.N. (Tung Thanh Nguyen); writing—reviewing scientific contents and editing—E.R.R., S.W.C., H.P.N.T., H.T.T., and D.D.N.

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