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## Current advancements in CO<sub>2</sub> capture using graphene-based materials

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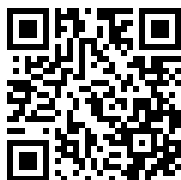
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### REVIEW ARTICLE

### ABSTRACT



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In 2023, global CO<sub>2</sub> emissions were 37.4 billion tonnes and a 1.1% increase compared to 2022. Although most countries try to decarbonize their economies, oil and gas supplied 52% of the world's energy needs in 2021, and by 2050 it will be 47%. Therefore, in the future, oil and gas will still account for a considerable percentage of the energy sector. However, the continuous release of CO<sub>2</sub> into the atmosphere at this rate can result in severe environmental problems. One of the promising approaches to address this issue is CO<sub>2</sub> capture. This captured CO<sub>2</sub> can then be stored underground or used to produce commercially valuable products. In recent years, graphene-based materials have gained attention in CO<sub>2</sub> capture due to their interesting properties, such as high thermal stability and durability. This review focuses mainly on recently published articles on carbon capture using graphene-based materials.

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### 1. Introduction

Graphite is one of the well-known carbon allotropes and consists of *sp*<sup>2</sup> hybridized carbons arranged in six-membered hexagonal rings [1,2]. Graphite is a layered structure, and hundreds and thousands of carbon layers are arranged on top of each other [3]. A single layer of graphite is known as graphene. It was first extracted by Andre Geim and Konstantin Novoselov in 2004, and they won the Nobel Prize in 2010 for their discovery [4]. These two scientists removed some flakes from graphite using scotch tapes, and they observed that some of the flakes were thinner than the others, so they kept removing the flakes until they obtained flakes that were only one atom thick. Although graphite and graphene have the same atomic arrangement, changing the number of layers can change the properties of the material (*i.e.*, it has been observed that decreasing the number of layers increases the electrical conductivity and thermal conductivity) [5-7]. Graphene has been emerging as a potential candidate for CO<sub>2</sub> capture in recent years due to interesting properties such as high surface area, high thermal stability, higher mechanical strength, high chemical stability, high selectivity, *etc.* [8,9]. However, the production of single-layer graphene is difficult and expensive; therefore, most studies are conducted using graphene

derivatives such as graphene oxide (GO) and reduced graphene oxide (RGO). In addition, it is well known that it is possible to incorporate other elements, such as nitrogen and sulfur, into graphene layers to synthesize different graphene derivatives. Researchers around the world have suggested different materials for CO<sub>2</sub> capture, such as zeolites, alumina-based compounds, metal-organic frameworks, metal oxides, *etc.* [10]. However, most of these materials have several drawbacks, such as high cost, limited adsorption capacity, poor thermal stability, *etc.* [11,12]. Existing CO<sub>2</sub> capture technologies can be classified into two types: (i) carbon capture and storage and (ii) carbon capture and utilization [13]. The first step of carbon capture is the separation of CO<sub>2</sub> from gas streams. Several techniques are used, such as solid adsorbents, solvent absorption, cryogenic separation, and metal-organic frameworks [11]. The most commonly used technique is the use of amine solutions (*i.e.*, monoethanolamine/MEA) to separate CO<sub>2</sub> [14]. However, most of these techniques have not met the expected carbon capture efficiency and are energy-intensive and expensive [15,16]. Therefore, it is necessary to optimize existing carbon capture technologies to make this technology more feasible. This review focuses on recent advances in CO<sub>2</sub> capture using graphene-based materials.

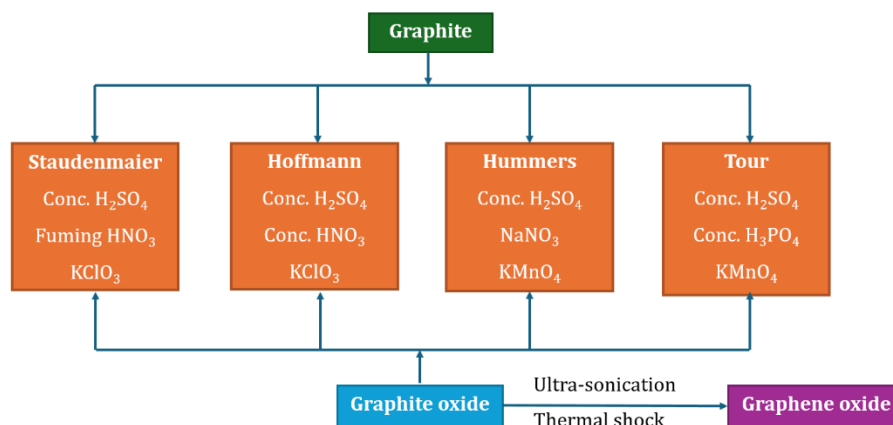


Figure 1. Various liquid phase processes which are used to synthesize graphene oxide from raw graphite.

## 2. Synthesis of graphene and graphene derivatives

Graphene can be synthesized using different methods such as chemical vapor deposition, electrochemical exfoliation, epitaxial growth, liquid phase exfoliation, mechanical exfoliation, and chemical exfoliation [17]. Out of these techniques, the most popular technique is liquid phase exfoliation due to its cost-effectiveness [18]. In liquid-phase exfoliation, intercalating agents are introduced into the graphite sheets, and these sheets are exfoliated. One of the popular liquid-phase exfoliation techniques is the modified Hummers method [19]. In the modified Hummers method, graphite is oxidized using an oxidizing agent in an acidic medium, and as a result, it will introduce oxygen functional groups (*i.e.*, epoxy groups, hydroxyl groups, and carboxylic acid groups) into the graphite layer. The introduction of oxygen functional groups increases the interlayer distance between graphite layers; hence, these graphite oxide layers can be separated by sonication, thermal shock, shearing, *etc.* [18]. Separated graphene oxide layers can be dissolved in water or any other suitable solvent due to the presence of functional oxygen groups [20]. Graphene oxide can be further reduced to form reduced graphene oxide using different reducing agents such as ascorbic acid (vitamin C), hydrazine hydrate, resveratrol, chitosan, polyethyleneimine, sodium borohydride, bovine serum albumin, green tea polyphenols, *etc.* [21,22]. As shown in Figure 1, there are some other liquid phase exfoliation methods, such as the Staudenmaier, Hofmann, and Tour methods [23]. Graphene oxide is commercially available now in different forms (*i.e.*, paste, powder, films, *etc.*), and there are leading suppliers such as Matexcel, CD bioparticles, Alfa Chemistry, ACS material, *etc.*

## 3. Graphene-based materials for CO<sub>2</sub> capture

An, L. *et al.* synthesized different types of nitrogen-rich porous carbons derived from graphene and tested them for their CO<sub>2</sub> adsorption capacity [24]. These nitrogen-rich porous carbons were synthesized using graphene oxide as the precursor. Graphene oxide was synthesized using the modified Hummers method and reacted with urea to incorporate nitrogen atoms, and this nitrogen-doped graphene was subjected to KOH activation [21]. Instead of urea, there are some other compounds that can be used as nitrogen doping agents, such as NH<sub>3</sub>, ammonium salts, and nitric acid. KOH activation was carried out at different KOH concentrations and different activation temperatures to form different N-rich porous carbons [24]. As shown in Equations 1-4, during the KOH activation process, carbon atoms in nitrogen-doped graphene oxide react with KOH to form CO<sub>2</sub>, CO, H<sub>2</sub>, and carbonates [25]. This will create new voids in the structure and,

as a result, will give rise to new micropores and mesopores; hence, it will significantly increase the surface area of BET (Brunauer, Emmett, and Teller) [26]. Micropores have a pore size of up to 2 nm, and pores in the range of 2 to 50 nm are classified as mesopores [27].



Most of the studies have conducted BET surface area analysis (Brunauer-Emmett-Teller) and total pore volume analysis using nitrogen adsorption isotherms to study the textural properties of graphene-based materials. Nitrogen adsorption-desorption isotherms are graphs that are generated by measuring the volume of nitrogen adsorbed and desorbed onto a surface with respect to the relative pressure [28]. In the BET model, monolayer coverage is considered and uses the linear region of nitrogen isotherms, usually at relative pressures in the range of 0.05-0.35; this relative pressure range can also change depending on the type of adsorbent [29]. As shown in Figure 2, An, L. *et al.* measured the BET surface area of nitrogen-doped porous carbons derived from graphene in the relative pressure range  $P/P_0 = 0.04-0.32$  [22].

Equation 5 represents the BET equation arranged in the  $y = mx + c$  format:  $V$  is the adsorbed gas quantity,  $P_0$  is the saturation pressure of the adsorbate,  $P$  is the equilibrium pressure of the adsorbate,  $V_m$  is the monolayer adsorbed gas volume,  $C$  is the BET constant,  $E_1$  is the heat of adsorption for the first layer, and  $E_L$  is the heat of vaporization. Equations 6-9 represent how to calculate the  $V_m$  using the slope and the intercept of the plot.

$$\frac{1}{V\left(\frac{P_0}{P}-1\right)} = \frac{C-1}{V_m C} \left(\frac{P}{P_0}\right) + \frac{1}{V_m C} \quad (5)$$

$$C = \exp\left(\frac{E_1 - E_L}{RT}\right) \quad (6)$$

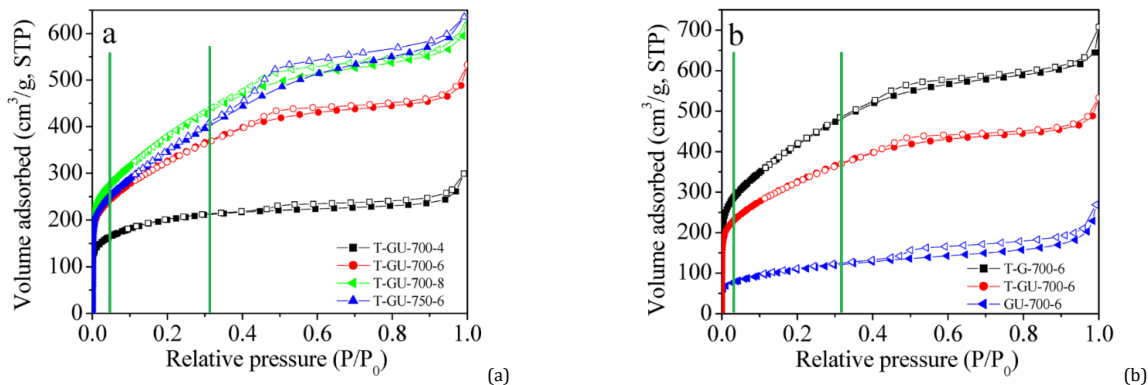
$$\text{Slope} = \frac{C-1}{V_m C} \quad (7)$$

$$\text{Intercept} = \frac{1}{V_m C} \quad (8)$$

$$V_m = \frac{1}{\text{slope} + \text{intercept}} \quad (9)$$

**Table 1.** CO<sub>2</sub> adsorption of T-GU-700-6, M90\_0.5 and activated-RGO-950.

Material	Surface area (m <sup>2</sup> /g)	Total pore volume (cm <sup>3</sup> /g)	CO <sub>2</sub> uptake at 1 atm (mmol/g)	Isosteric heat of adsorption (kJ/mol)	Reference
T-GU-700-6	1032.00	0.82	2.40	3.24	[24]
M90_0.5	328.00	1.35	2.10	-	[31]
Activated-RGO-950	1315.98	1.07	2.45	3.36	[33]

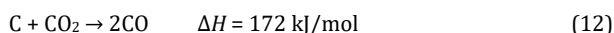
**Figure 2.** Nitrogen adsorption isotherms of nitrogen-doped porous carbons derived from graphene and the full green lines represent the approximate region that can be used for BET calculations (images are adapted with permission from An, L. et al. Copyright (2019) American Chemical Society) [24].

After evaluating the monolayer adsorbed gas volume ( $V_m$ ), then Equations 10 and 11 can be used to get the BET surface area, where  $S_t$  is the total surface area of sample material,  $N$  is the Avogadro number,  $V$  is the molar volume of the adsorbed gas,  $s$  (0.162 nm<sup>2</sup>) is the cross-sectional area of an adsorbed nitrogen molecule and  $a$  is the mass of the sample [30].

$$S_t = \frac{V_m N s}{v} \quad (10)$$

$$S_{BET} = \frac{S_t}{a} \quad (m^2/g) \quad (11)$$

An, L. et al. reported that the thermal shocking process and KOH activation can increase the surface area of graphene-based materials [22]. However, increasing the temperature above a certain threshold level and continuously increasing the KOH concentration can decrease the surface area of graphene-based materials [22]. Politakos, N. et al. synthesized monolithic nanostructures from graphene oxide and reported that reduction of graphene oxide to a certain threshold level can increase the BET surface area of monolithic nanostructures [31]. Monoliths are structures consisting of pores that resemble a honeycomb [32]. Chowdhury, S. et al. synthesized graphene-based adsorbents using graphite as the starting material [33]. Graphite powder was used to synthesize graphene oxide, and graphene oxide was thermally reduced to form reduced graphene oxide. The reduced graphene oxide was physically activated by heating it to temperatures of 750, 850, and 950 °C in the presence of CO<sub>2</sub> flow (1000 mL/min) [30]. This physical activation process works in the same way as KOH activation, where CO<sub>2</sub> reacts with carbons in reduced graphene oxide sheets and produces gaseous species, as shown in Equation 12 [30]. As a result, it will increase the porosity of the material.

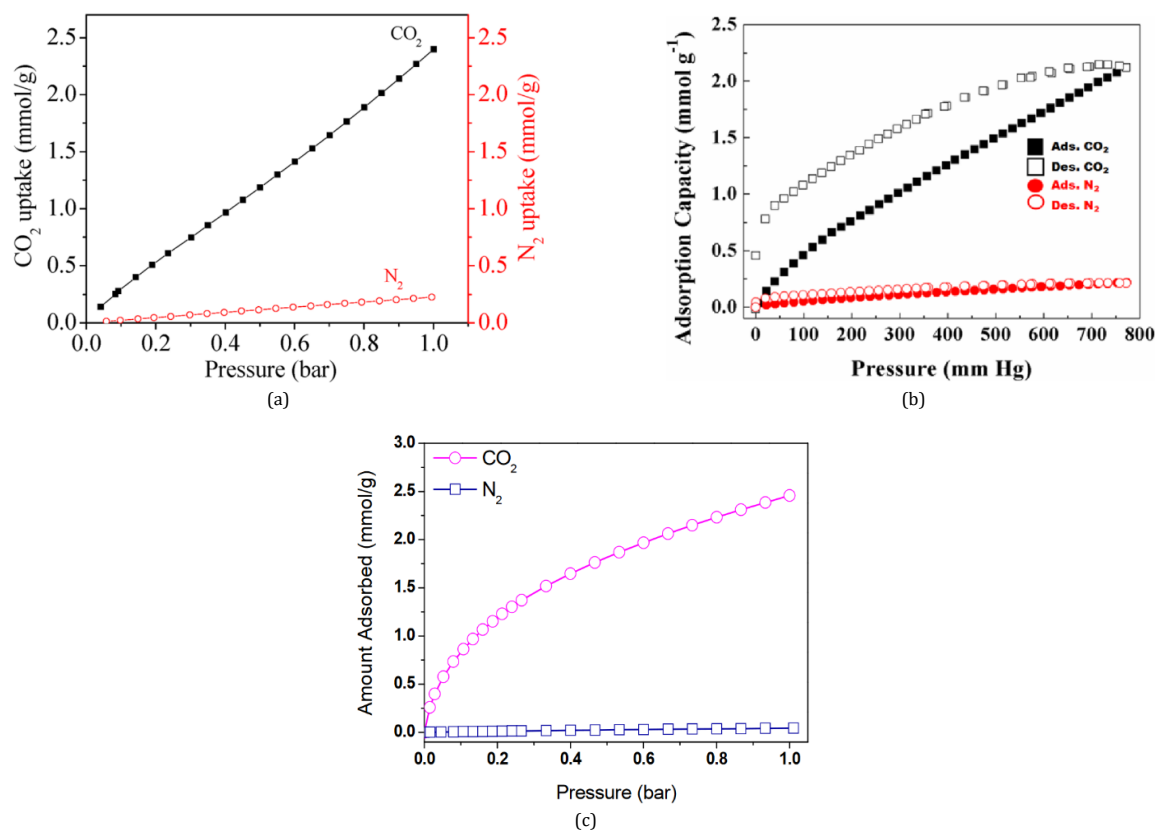


An, L. et al., measured the CO<sub>2</sub> adsorption of different types of nitrogen-doped graphene synthesized under various conditions, and the highest CO<sub>2</sub> adsorption was observed in T-GU-700-6 [22]. T-GU represents the graphene oxide that reacted with urea after the thermal shock process, 700 is the activation temperature in Celsius during the KOH activation process, and 6 represents the ratio of KOH/T-GU [22]. Politakos, N. et al., synthesized different monolithic nanostructures under

various conditions and found that the highest CO<sub>2</sub> adsorption was seen in M90\_0.5 [28]. M represents the monolithic nanostructure, 90 represents the temperature in Celsius at which the monolith was synthesized, and 0.5 represents the ratio of GO/ASA (Ascorbic acid/Vitamin C) ratio [28]. Studies by Chowdhury, S. et al. have shown that the highest CO<sub>2</sub> adsorption was seen for activated-RGO-950 [30]. 950 represents the temperature in Celsius at which the reduced graphene oxide was activated. Table 1 shows the CO<sub>2</sub> adsorption capacity, surface area, and isosteric heat of adsorption of these materials [24,31,33].

By comparing the values of CO<sub>2</sub> uptake of all materials in Table 1, it is clear that increasing the surface area does not significantly affect the CO<sub>2</sub> uptake. The highest total pore volume was observed for M90\_0.5; however, CO<sub>2</sub> adsorption was lowest in M90\_0.5; therefore, it is not possible to suggest that increasing porosity increases CO<sub>2</sub> adsorption. Comparing the textural properties of different materials from different studies on CO<sub>2</sub> adsorption is complex; therefore, evaluating the effect of surface area and total pore volume on CO<sub>2</sub> adsorption needs more studies. It is well known that gas adsorption decreases with increasing temperature; this can be seen here by comparing CO<sub>2</sub> uptake values at 25 and 0 °C; with increasing temperature, CO<sub>2</sub> adsorption has decreased [34]. The isosteric heat of adsorption is another important property that needs to be considered when studying gas adsorption because if an adsorbent has a very high isosteric heat of adsorption, it is difficult to desorb the adsorbed gas. If it has a very low isosteric heat of adsorption, it will decrease the CO<sub>2</sub> adsorption; therefore, moderate values are preferred. For physisorption processes, the isosteric values are generally between 5-40 kJ/mol [35]. The isosteric heat of adsorption values of T-GU-700-6 and activated-RGO-950 suggest that this is a physisorption process; however, no isosteric heat of adsorption values were given for M90\_0.5.

An, L. et al., N. et al., and Chowdhury, S. et al. all have tested for the selectivity of CO<sub>2</sub> in the presence of N<sub>2</sub>. T-GU-700-6, M90\_0.5, and activated-RGO-950 had higher selectivity for CO<sub>2</sub> in the presence of nitrogen, as shown in Figure 3. However, none of these studies measured the selectivity of CO<sub>2</sub> in real flue gas conditions because the flue gas contains other species, such as O<sub>2</sub>, SO<sub>2</sub>, NO<sub>x</sub>, and H<sub>2</sub>O [36].



**Figure 3.** (a) CO<sub>2</sub> and N<sub>2</sub> adsorption of T-GU-700-6, (b) CO<sub>2</sub> and N<sub>2</sub> adsorption desorption of M90\_0.5, and (c) CO<sub>2</sub> and N<sub>2</sub> adsorption of activated-RGO-950 (Images are adapted with permission from An, L. et al. Copyright (2019) American Chemical Society Politakos, N. et al. Copyright (2020) American Chemical Society and Chowdhury, S. et al Copyright (2016) American Chemical Society) [24,31,33].

In order to use these materials in the industry, their stability and recyclability must be tested. An, L. et al., studied the cycle stability of T-GU-700-6 for five cycles and observed that there was only a 5% decrease in CO<sub>2</sub> adsorption compared to the first cycle [22]. Politakos, N. et al. studied the cycle stability of M90\_0.5 for five cycles and reported that after each cycle there was a decrease in CO<sub>2</sub> adsorption; however, this decrease in CO<sub>2</sub> adsorption has slowly reduced with each cycle, suggesting that these monoliths are slowly stabilizing with respect to CO<sub>2</sub> adsorption [28]. Chowdhury, S. et al., tested activated-RGO-950 for ten cycles and observed that there was no loss in CO<sub>2</sub> adsorption after ten cycles. Despite recent advances in developing novel materials derived from graphene for CO<sub>2</sub> capture, none of these materials have been commercially used so far. Therefore, more testing and research is needed to commercialize these graphene-based materials for CO<sub>2</sub> capture.

#### 4. Conclusion

Different types of graphene-based materials were synthesized by An, L. et al., Politakos, N. et al., and Chowdhury, S. et al., and CO<sub>2</sub> adsorption of all these materials was measured. Out of these studies, the highest CO<sub>2</sub> adsorption was seen for activated-RGO-950 at 25 °C and 0 °C. T-GU-700-6, M90\_0.5 and activated-RGO-950 had higher CO<sub>2</sub> selectivity in the presence of nitrogen; however, all of these materials must be tested in real flue gas conditions. T-GU-700-6, M90\_0.5, and activated-RGO-950 have shown the cycle stability and isosteric heat of adsorption values of T-GU-700-6 and activated-RGO-950 have shown that CO<sub>2</sub> adsorption is a physisorption process.

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#### Disclosure statement

Conflict of interest: The authors declare that they have no conflict of interest. Ethical approval: All ethical guidelines have been adhered.

#### CRedit authorship contribution statement

Conceptualization: Madushan Dhammika Gunarathna; Methodology: Madushan Dhammika Gunarathna, Nimeshi Aviddika Abeysinghe; Investigation: Madushan Dhammika Gunarathna, Ashan Sithija Wickramaarachchi; Resources: Madushan Dhammika Gunarathna, Ashan Sithija Wickramaarachchi, Nimeshi Aviddika Abeysinghe; Data Curation: Madushan Dhammika Gunarathna, Ashan Sithija Wickramaarachchi, Nimeshi Aviddika Abeysinghe, Polegodage Dilushi Sureka Ruwan Kumari; Writing - Original Draft: Madushan Dhammika Gunarathna; Writing - Review and Editing: Madushan Dhammika Gunarathna, Ashan Sithija Wickramaarachchi, Nimeshi Aviddika Abeysinghe, Polegodage Dilushi Sureka Ruwan Kumari; Visualization: Madushan Dhammika Gunarathna, Ashan Sithija Wickramaarachchi, Nimeshi Aviddika Abeysinghe, Polegodage Dilushi Sureka Ruwan Kumari; Supervision: Madushan Dhammika Gunarathna; Project Administration: Madushan Dhammika Gunarathna.

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