

CO₂ ADSORPTION BY GRAPHENE: MECHANISMS, MODIFICATIONS, AND APPLICATIONS

*Siwoo Park

Safa Community School, Umm Suqeim Road, Al Barsha South 3rd, Dubai, United Arab EmiratesReceived 12th August 2025; Accepted 14th September 2025; Published online 23rd October 2025

Abstract

Rising CO₂ levels are accelerating global climate change at an unprecedented rate. Effective CO₂ capture and utilization technologies are critical now more than ever. Graphene has emerged as attractive solid adsorbents due to its exceptional surface area, modifiable electrical characteristics, and ease of chemical modification. This study analyzes the interactions between graphene and CO₂, both physical and chemical, and explores how the structure can be modified to improve capture capacity. We begin by introducing adsorption within the broad scope of carbon capture methodologies, including chemical absorption and solid adsorption. The discussion subsequently focuses on the inherent characteristics of graphene that facilitate these mechanisms, followed by techniques for functionalization and heteroatom doping that create active binding sites and enhance charge distribution. Application sections categorize graphene materials into physisorption, chemisorption, and hybrid systems, illustrating how hierarchical porosity and tailored surface chemistry jointly improve performance under realistic flue gas and ambient conditions. Finally, we highlight emerging opportunities for CO₂ utilization and discuss the remaining challenges of stability, regeneration energy, and scalability. Together, these insights position graphene-based sorbents as a vital bridge between high-capacity laboratory materials and the next generation of practical carbon capture technologies.

Keywords: Carbon capture and storage, 2D Nanomaterials, Graphene, Chemical functionalization, Doping, Climate change.

INTRODUCTION

Climate change has emerged as an urgent global issue with its consequences intensifying rapidly. Without significant mitigation effort, the world faces fatal irreversible environmental disruptions. According to a projection by the Intergovernmental Panel on Climate Change (IPCC), global average temperature is expected to increase by 3.7 degree celsius and increase average sea level by 3.8 m until 2100 if current emission trends persist. Such changes will inevitably result in droughts, excessive heat waves, floods, wildfires, and tropical storms. These impacts underscore the urgency of implementing effective technologies to curb greenhouse gas (GHG) emissions. Among various GHGs such as carbon dioxide (CO₂), methane (CH₄), and nitrous oxide (N₂O), CO₂ is a main contributor representing for about 64% of the total greenhouse effect. Human activities such as fossil fuel combustion, deforestation, and industrial processes have dramatically increased atmospheric CO₂ concentration from ~280 ppm before industrialization to over 410 ppm today. Annual CO₂ emissions have risen from 20.5 Gt in 1990 to 33.4 Gt in recent years, a 63% increase over three decades. These statistics emphasize the urgent need for innovative and scalable carbon mitigation technologies. Several strategies have been proposed to reduce CO₂ emissions, including coal-renewable energy development, improved efficiency of coal-fired plants, and carbon capture and storage (CCS). While transitioning to renewable energy remains essential, it alone cannot achieve the rapid reductions required within the next few decades. The IPCC projects that CCS could contribute up to 19% of the total emission reduction needed by 2050. (Figure 1) This highlights CCS as a crucial transitional technology that can be integrated into existing fossil-fuel-based infrastructures while renewable systems continue to expand.

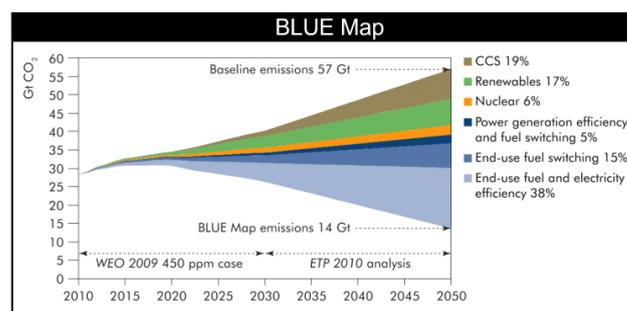


Figure 1. BLUE Map of CO₂ emissions projected by IPCC, IEA Report 2022

CCS processes are typically categorized into pre-combustion, post-combustion, oxy-fuel combustion routes, and direct air capture (DAC). Pre-combustion separates carbon monoxide and hydrogen from fossil fuels, post-combustion removes CO₂ from flue gas streams, and oxy-fuel combustion burns fuel in pure oxygen to produce CO₂ and water vapor, simplifying downstream capture. Also, DAC extracts CO₂ directly from air in ambient pressure, where its concentration of CO₂ is extremely low. Among these, post-combustion capture is the most widely applied in industry due to its compatibility with current energy systems. The chemical absorption process, particularly using amine-based solvents such as monoethanolamine (MEA), remains the dominant CO₂ capture mechanism for post-combustion capture. It achieves over 90% capture efficiency with high selectivity. However, its disadvantages, including large energy demand for solvent regeneration, high operational costs, and formation of harmful by-products, significantly limit large-scale deployment. As a result, solid adsorption technologies have gained increasing attention for their advantages of lower energy requirements, simpler regeneration, and elimination of liquid waste. In this context, graphene has emerged as a highly promising solid sorbent for CO₂ capture. Graphene is a two-dimensional hexagonal carbon lattice possessing exceptional properties

*Corresponding Author: Siwoo Park,
Safa Community School, Umm Suqeim Road, Al Barsha South 3rd, Dubai, UAE.

such as a theoretical surface area of $2630 \text{ m}^2 \text{ g}^{-1}$, thermal conductivity up to $5000 \text{ W m}^{-1} \text{ K}^{-1}$, and a Young's modulus of $\sim 1 \text{ TPa}$. It also exhibits excellent electrical conductivity, chemical stability, and mechanical strength, making it suitable for harsh process environments. Moreover, the surface of carbon lattice can be chemically engineered through functionalization, heteroatom doping, or nanocomposite hybridization. These modifications allow control of surface polarity, pore structure, and binding affinity toward CO_2 . These characteristics position graphene-based materials as next-generation sorbents capable of bridging the gap between conventional physisorption and chemisorption mechanisms (Figure 2).

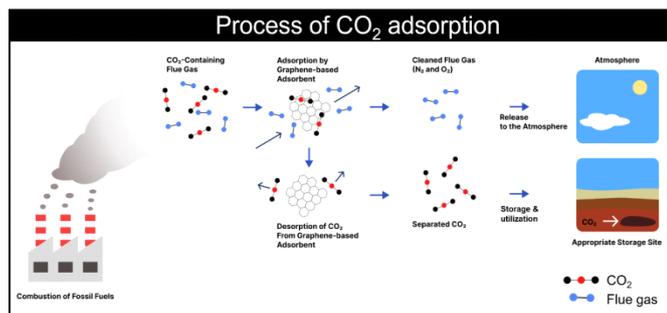


Figure 2. Overview of CO_2 capture by solid graphene adsorbent

This review explores the potential of graphene-based adsorbents for CO_2 capture. Following an overview of current CCS strategies and capture mechanisms, we examine characteristics and surface modification strategies of graphene as solid adsorbent. We then analyze recent developments in physisorption, chemisorption, and hybrid graphene systems and conclude with perspectives on CO_2 utilization and the role of graphene materials in advancing circular carbon technologies.

2. CO_2 Capture

The capture of carbon dioxide stands as a fundamental element in global decarbonization strategies, especially considering the persistent dependence on fossil fuels in power generation and heavy industry. Even with continuous progress in renewable energy systems, the huge extent of current fossil-based infrastructure guarantees that carbon capture technologies will be crucial for both transitional and long-term strategies aimed at achieving net-zero emissions. The primary difference between the several developed technological pathways is the location of the intervention within the combustion process.

2.1 Methods of CO_2 Capture

CO_2 capture technologies can be categorized based on the timing and location of the separation process in relation to fuel combustion. The three leading industrial methods include pre-combustion, post-combustion, and oxy-fuel combustion, while DAC is rising as an additional pathway for removing CO_2 from the atmosphere. Every method offers a distinct separation environment, placing varying requirements on the capture materials and process setups. In the pre-combustion, fossil fuels such as coal and natural gas undergo conversion, usually through pre combustion gasification or reforming, resulting in a synthesis gas (syngas) made up of CO and H_2 . A follow-up water-gas shift reaction transforms CO into CO_2 and H_2 . The CO_2 produced is captured at high pressure, typically between 20 and 50 bar, prior to combustion, while the refined H_2 serves

as a low-carbon fuel source. Due to elevated CO_2 partial pressures and the relatively dry, hot nature of gas streams, separation becomes more straightforward compared to flue gas, making physical solvents or pressure-driven adsorption methods viable options. In the process of post-combustion capture, carbon dioxide is separated from the flue gas following the combustion phase. Flue gas generally consists of approximately 10-15 vol% CO_2 , around 70-80% N_2 , some residual O_2 , water vapor, and trace impurities. Operating under conditions close to atmospheric pressure and within a moderate temperature range of 40-120 $^\circ\text{C}$ presents significant challenges, primarily due to the low partial pressure of CO_2 along with humidity and contaminants in the environment. Nevertheless, post-combustion capture presents an appealing option as it can be integrated into existing plants without the need to modify upstream combustion systems. Oxy-fuel combustion transforms the combustion process by utilizing oxygen or oxygen-enriched for burning fuels instead of air. The exhaust gas produced primarily consists of CO_2 and H_2O . By condensing the water, a high-purity CO_2 stream is obtained. The primary limitation lies in the requirement for an air separation unit to generate pure O_2 , which results in significant costs and energy penalties. Lastly, DAC aims to extract CO_2 from the surrounding atmosphere, which is approximately 400 ppm. Due to the exceptionally low CO_2 concentration, direct air capture requires materials that exhibit a remarkable affinity, selectivity, and minimal regeneration energy. While still in the early stages of commercial use, DAC is gaining traction as a valuable addition to point-source capture discussions.

2.2 CO_2 Capture Mechanisms

Post-combustion CO_2 capture must function in diluted gas streams (usually 10-15 vol% CO_2 in N_2), which are frequently humid and contain trace contaminants. This makes the separation difficult from a thermodynamic and kinetic standpoint. Chemical absorption and solid adsorption are the two main mechanisms for post-combustion carbon capture. Each of these processes has its own unique set of thermodynamic drivers, kinetic restrictions, and engineering consequences (Figure 3).

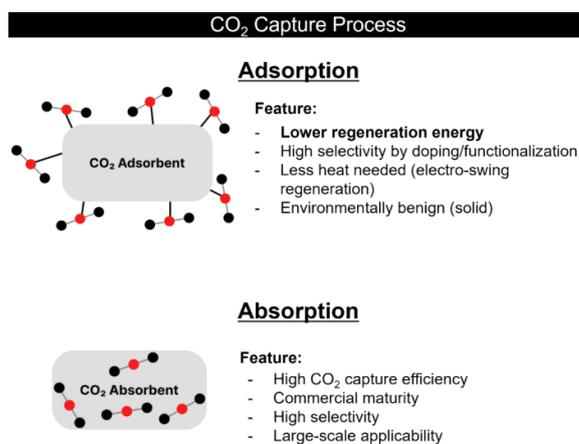


Figure 3. CO_2 adsorption versus absorption

2.2.1 Chemical Absorption

Chemical absorption, especially amine-based solvents (e.g., monoethanolamine) is the most established post-combustion CO_2 capture technology. In this process, CO_2 is first dissolved into the liquid phase and then chemically reacts with amines to form carbamates or bicarbonates. The method has been widely

adopted due to its high capture efficiency, often exceeding 90%, and its well-understood engineering design. Even with these benefits, the process is limited by inherent limitations. Regenerating solvents demands a considerable amount of thermal energy, usually between 3 and 4 GJ for every tonne of captured CO₂, leading to notable economic and environmental implications. Moreover, the oxidative and thermal breakdown of amines produces corrosive by-products like heat-stable salts, which require ongoing solvent replenishment and system upkeep. The substantial footprints of large equipment pose additional challenges to scalability. Recent advancements, such as blended amines, water-lean solvents, and ionic liquid systems, aim to minimize energy penalties and enhance stability. Despite this, the core thermodynamic challenge linked to bond-breaking in the regeneration process persists, driving the exploration of innovative capture methods.

2.2.2 Solid Adsorption

Solid adsorption stands out as an innovative alternative to solvent-based absorption, effectively capturing CO₂ directly on the surfaces of porous materials. This method circumvents problems related to solvent degradation and corrosion, while requiring less energy for regeneration via temperature or pressure swing processes. Solid sorbents can be designed into modular reactor, which allows integration into current industrial facilities. Nonetheless, their success relies on precisely customizing both physical and chemical properties to attain high capacity, swift kinetics, and consistent performance in practical use conditions. The adsorption mechanism can be categorized into two main types: physisorption and chemisorption. In physisorption, CO₂ attaches via weak van der Waals forces, quadrupole- π interactions, or induced dipole interactions with the surface of the adsorbent. The heats of adsorption typically range between 20 and 50 kJmol⁻¹, which allows the process both easily reversible and economically advantageous. However, the adsorption capacity decreases rapidly at higher temperatures and can be hindered by competing elements like H₂O. The use of physisorption is hindered to be utilized in humid or elevated temperature environment. In contrast, chemisorption depends on strong covalent or ionic interactions, often involving grafted amine groups, metal oxides, or heteroatom sites on carbon atoms. The interactions typically exhibit binding enthalpies exceeding 60 kJmol⁻¹, enhancing selectivity and performance under low CO₂ partial pressures. However, this also raises the energy needed for regeneration and limits desorption kinetics. Graphene and its derivatives hold a distinctive potential for advancing solid adsorption strategies forward. The ultra-thin, high-surface-area lattice offers numerous accessible adsorption sites, and the delocalized π -electron system enhances favorable quadrupole interactions with CO₂ molecules. Simultaneously, the surface chemistry of graphene can be modified by incorporating functional groups that contain nitrogen, boron, or oxygen. This dual property, which allows for both dispersive physical binding and variable chemical anchoring, provides graphene a versatile platform for addressing the trade-offs between capacity, selectivity, and regeneration energy that have traditionally limited solid adsorbents. In this study, we focus on how developments in graphene modification have resulted in materials that perform successfully across the physisorption-chemisorption spectrum, outperforming ordinary carbons and, in some cases, approaching the performance of sophisticated metal-organic frameworks.

3. Graphene

3.1 Carbon-based Nanomaterials

Carbon nanomaterials include a variety of dimensional shapes, ranging from zero-dimensional (0D) to two-dimensional (2D) porous structures. (Figure 4) This classification provides an important basis for material evaluation and highlights the structure-property interactions related to sorbent design.

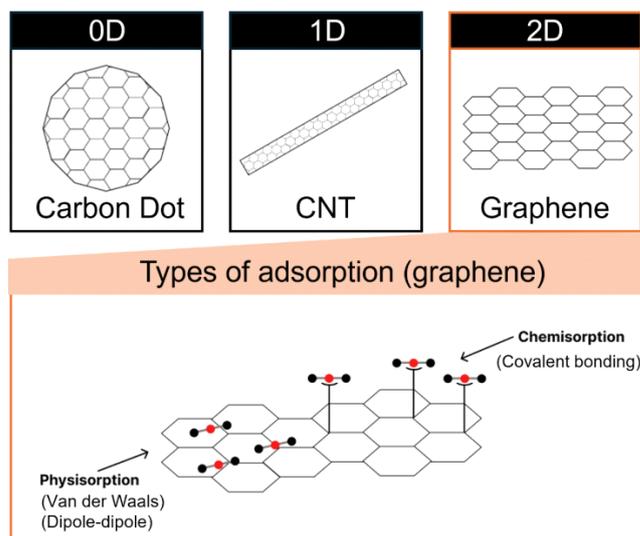


Figure 4. Types of carbon-based nanomaterials and distinct features of graphene as adsorbent

Zero-dimensional carbons, such as carbon quantum dots (CQDs), typically measure only a few nanometers in size, but possess a high density of edge sites and an extensive number of surface functional groups. Their internal pore capacity is limited; nevertheless, when included into larger porous matrices, CQDs may generate additional electronic interaction sites for CO₂, sometimes enhanced by heteroatom doping. Nitrogen-functionalized carbon quantum dots demonstrate improved selectivity due to intensified quadrupole- π interactions with carbon dioxide molecules. One-dimensional carbons, including carbon nanotubes (CNTs) and carbon nanofibers, offer linear transport channels distinguished by increased aspect ratios. Their intrinsic capacity for adsorption depends on preserving open, defect-laden channels while preventing nanomaterial aggregation. Chemical modifications such as nitrogen or oxygen doping effectively hinder aggregation and create crucial adsorption sites, hence enhancing CO₂ binding. While pristine carbon nanotubes exhibit limited CO₂ absorption, functionalized forms demonstrate significantly improved capacities and selectivity. Two-dimensional carbons, especially graphene and its variants, provide exceptional surface area per unit mass and significant chemical modification. However, the capacity of graphene sheets to restack as a result of π - π interactions hinders pore accessibility and reduces the surface area available for adsorption. Introducing defects, gaps, or micropores via activation techniques enable graphene to accomplish exceptional adsorption capabilities. Graphene oxide (GO), distinguished by its oxygen-containing functional groups, and reduced graphene oxide (rGO), which has partially restored conductivity, are often explored as more feasible substitutes for pristine graphene.

3.2 Characteristics of Graphene

Graphene is an atomically thin, two-dimensional sheet of sp^2 -hybridized carbon atoms arranged in a honeycomb lattice. It is a fundamental building block of graphite, in which graphene layers stack through weak van der Waals interactions. This single-layer graphene possesses an extraordinary intrinsic carrier mobility ($>200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and high thermal conductivity ($3000\text{--}5300 \text{ W m}^{-1} \text{ K}^{-1}$). Graphene also combines remarkable mechanical properties, including a Young's modulus of $\sim 1 \text{ TPa}$ and intrinsic tensile strength around 130 GPa . These exceptional electronic, thermal, and mechanical attributes, together with its low density and optical transparency, have established graphene as a cornerstone of modern nanomaterials research. For CO_2 adsorption, the delocalized π -electron network and the exceptionally high theoretical surface area ($\sim 2630 \text{ m}^2 \text{ g}^{-1}$) of graphene are the primary advantages. These features enable strong quadrupole- π interactions with CO_2 molecules and provide edge and defect sites for additional binding. However, the tendency of pristine graphene to restack through π - π interactions restricts accessible surface area and pore volume, leading to the widespread adoption of graphene oxide (GO) and reduced graphene oxide (rGO) as practical alternatives. GO provides several oxygenated functional groups that prevent restacking and facilitate additional modifications, while rGO merges enhanced conductivity with defect sites appropriate for dopant integration. The atomically thin structure ensures rapid diffusion and efficient heat dissipation, while chemical modification such as nitrogen doping enhances CO_2 affinity via basic pyridinic and pyrrolic sites. To fully utilize graphene as a high-performance CO_2 adsorbent, it is crucial to strike an ideal balance between porosity, conductivity, and defect density.

3.3 Functionalization Strategies

Although pristine graphene has an exceptional theoretical surface area and adjustable electrical properties, its inherent CO_2 adsorption capability is limited due to weak chemical affinity and a propensity for sheet restacking. To overcome these challenges, researchers have developed a broad range of functionalization strategies. These modifications introduce specific binding sites, stabilize porous frameworks, and create synergistic interactions for improved CO_2 uptake. Functionalization approaches can be classified into covalent modifications, non-covalent functionalization, incorporation of amine groups, and hybridization with polymers or inorganic materials. One of the most effective strategies is amine functionalization, which directly targets the acid-base chemistry between basic amine sites and acidic CO_2 molecules. Amine-incorporated GO and reduced rGO composites have consistently shown improved adsorption capacities and selectivity over nitrogen. For example, Li et al. demonstrated that polyethyleneimine (PEI)-modified GO achieved CO_2 capture as high as 4.2 mmol g^{-1} at 25°C , while also exhibiting strong CO_2/N_2 selectivity under flue gas conditions. Similarly, Pokhrel et al. reported that aminosilane-functionalized GO incorporated into ZIF-8 composites enhanced CO_2 adsorption by $\sim 36\%$ compared with non-functionalized GO. In addition, the presence of water vapor further boosted CO_2 capture performance by increasing amine- CO_2 interactions. These findings highlight how hydrophilic functional groups can improve CO_2 adsorption under gas stream with moisture conditions. Beyond amine chemistry,

oxygenated functional groups intrinsic to GO (e.g., hydroxyl, epoxy, and carboxyl groups) already provide polar anchoring sites for CO_2 . However, their density and distribution can be modified through chemical treatments. Dreyer et al. have shown that controlled oxidation increases hydrophilicity and defect density, which in turn creates favorable sites for both physisorption and chemisorption pathways. Yet, excessive oxidation introduces structural fragility and reduces conductivity in which underscoring the need for balance in functionalization. Non-covalent functionalization has also emerged as an attractive strategy for preserving π -conjugated structure in graphene while enhancing CO_2 affinity. Ionic liquids (ILs) and surfactant molecules can be adsorbed onto graphene surfaces to provide external binding sites. For instance, Tamilarasan et al. demonstrated that IL-modified rGO composites improved CO_2 adsorption kinetics and maintained reversibility over repeated cycles. Such systems utilize charge transfer and electrostatic stabilization while avoiding the loss of conductivity associated with covalent treatments. Another important direction is hybrid functionalization with polymers and porous materials. Embedding graphene into polymer matrices such as polyaniline, polypyrrole, or resorcinol-formaldehyde resins provides high surface area and microporosity, while the graphene skeleton stabilizes the framework. Ouyang et al. demonstrated that melamine-resorcinol-formaldehyde/GO composites exhibited CO_2 adsorption of over 5 mmol g^{-1} at 298 K and 500 kPa , alongside a CO_2/N_2 selectivity of 58 at low pressures. This hybridization strategy combines textural design with chemical affinity, producing composites that are both highly selective and regenerable. Functionalization can also be extended through inorganic hybridization, in which metal oxides or MOFs are anchored on graphene sheets to synergistically combine surface basicity with high surface area. For example, Ruhaimi et al. prepared graphene/MgO composites where the basic MgO sites enhanced CO_2 chemisorption, while the graphene substrate ensured structural stability and dispersibility. Various functionalization strategies illustrate that graphene in CO_2 adsorption is not limited to its intrinsic surface area but is augmented when paired with chemical functionalization.

3.4 Heteroatom Doping

Doping graphene with heteroatoms introduces active binding sites on carbon lattice in graphene, thereby improving CO_2 adsorption capacity. By substituting carbon atoms with heteroatoms such as nitrogen, boron, sulfur, or phosphorus, the charge distribution and local chemical potential of the π -electron system can be precisely tuned, disrupting the perfect sp^2 symmetry of the lattice and creating active sites that selectively interact with CO_2 molecules through Lewis acid-base interactions and dipole-quadrupole coupling. Compared to graphene, doped graphene provides stronger and more specific binding affinities, higher selectivity, and improved adsorption capacity under flue gas conditions. Lewis basicity of pyridinic and pyrrolic nitrogen sites generated by nitrogen doping introduces localized electron-rich regions that can interact favorably with the acidic CO_2 molecule. Nitrogen atoms can be incorporated into graphene in several configurations pyridinic, pyrrolic, graphitic (quaternary), and oxidized N. Pyridinic and pyrrolic N atoms, positioned at defect edges or five-membered rings, serve as strong Lewis bases that preferentially adsorb acidic CO_2 molecules via charge transfer or hydrogen bonding. In contrast, graphitic N

enhances overall conductivity and charge delocalization but provides weaker direct adsorption affinity. Boron doping provides a complementary effect by introducing electron-deficient sites within the graphene lattice. These sites act as Lewis acidic centers that attract CO₂ via dipole–quadrupole interactions. Boron atoms replace carbon in the hexagonal network, lowering local electron density and generating positively polarized centers. This results in stronger electrostatic attraction and enhanced chemisorption capability, as well as improved CO₂ selectivity under low partial pressures. Sulfur doping typically introduces thiophene-like or sulfoxide functional groups that polarize adjacent carbon atoms, facilitating CO₂ interaction through induced dipole forces. Meanwhile, phosphorus, with its larger atomic radius and five valence electrons, expands the lattice and introduces localized lone-pair electrons that enhance Lewis basicity. These dopants not only modify the chemical potential landscape but also improve wettability and stability under humid conditions of flue gas applications. Co-doping of N with S or P often creates dual-active sites with complementary acid–base properties, balancing adsorption strength and regeneration efficiency. Overall, heteroatom doping strategies highlight the versatility of graphene as a CO₂ adsorbent by controlling its charge distribution and surface polarity. Researchers have succeeded in bridging the gap between the weak interactions of physisorption and the high specificity of chemisorption. The ability to tailor both the chemistry and the pore architecture makes doped graphene systems uniquely positioned among carbonaceous adsorbents.

4. APPLICATION

4.1 Physisorption

Physisorption of CO₂ on graphene-based materials has been extensively studied as a low-energy, reversible capture process that benefits from rapid adsorption–desorption kinetics. Ghosh et al. measured the CO₂ adsorption of graphene derived from different carbon precursors. The graphene prepared via exfoliation of GO and from nanodiamond conversion achieved capacities of 7.8 and 8.6 mmol g⁻¹, respectively, at -78 °C and 1 bar. These adsorptions were fully reversible, with no hysteresis upon desorption, confirming the dominance of weak van der Waals interactions. Density functional theory (DFT) calculations further supported this observation, estimating binding energies around -59 kJ mol⁻¹, consistent with physisorption. Mishra et al. employed infrared spectroscopy to examine CO₂ adsorption on graphene obtained via hydrogen-induced exfoliation of GO. The emergence of an asymmetric CO₂ stretching band at 2324 cm⁻¹ provided direct spectroscopic evidence of physisorption. The graphene exhibited a maximum adsorption capacity of 12 mmol g⁻¹ under industrially relevant conditions (11 bar and 100 °C), highlighting the promise of graphene for post-combustion CO₂ capture. Meng et al. expanded this concept by producing graphene nanoplatelets with hierarchical porosity through vacuum exfoliation of GO. Their materials displayed a broad pore distribution, including supermicropores (~1.2 nm) and meso/macropores up to 90 nm, enabling high-pressure uptake of 56.4 mmol g⁻¹ at 25 °C and 30 bar. To overcome the intrinsic limitation of low microporosity in pristine graphene, several groups engineered pore structures within graphene sheets. Ning et al. synthesized graphene nanomeshes (GNMs) using MgO-templated CVD, achieving surface areas above 2000 m² g⁻¹ with uniformly distributed ~1 nm supermicropores. These GNMs exhibited

outstanding CO₂ capture capacities of 36.5 mmol g⁻¹ at 1 °C and 31 bar, far exceeding conventional graphene-based sorbents. Hybrid composites have also proven effective in optimizing physisorption behavior through synergistic interfacial interactions. For instance, Varghese et al. fabricated UV-treated HKUST-1/GO hybrids, where moderate UV exposure increased CO₂ uptake by 45%, achieving capacities up to 5.14 mmol g⁻¹ at 25 °C and 1 bar, and as high as 9.5 mmol g⁻¹ at 0 °C. The improvement stemmed from enhanced textural integration between the MOF's pore architecture and the GO substrate, alongside favorable adsorption thermodynamics under low-temperature conditions. Taken together, these results highlight the versatility of graphene-based adsorbents for physisorption-driven CO₂ capture. The combination of increased surface area with modified porous structures facilitates performance at both high pressure and near-ambient conditions, while the dangling CO₂ allows for easy desorption. Nonetheless, due to weak interaction of van der Waals, hybrid approaches such as incorporating of chemisorptive components or heteroatom dopants are likely critical to establishing steady operation in practical flue gas conditions.

4.2 Chemisorption

Unlike physisorption, which depends on weak van der Waals forces, chemisorption introduces specific chemical interactions between CO₂ molecules and functional groups attached to graphene surface. These stronger interactions, typically ionic or covalent, provide enhanced selectivity and uptake under ambient pressures. However, reversibility to desorb CO₂ is difficult because required energy for regeneration is high. Consequently, chemisorption strategies involving functionalization, heteroatom doping, and hybridization have been studied to increase CO₂ uptake capacity, while reducing regeneration energy.

Xiao et al. synthesized reduced GO/N-porous carbon (rGO/NPC) composites through KOH activation and thermal treatment of glucose–dicyandiamide precursors. Incorporation of nitrogen significantly enhanced CO₂ binding strength. The optimized material (rGO/NPC) achieved an equilibrium capacity of 5.77 mmol g⁻¹ at 298 K and 500 kPa, which are nearly three times that of its nitrogen-free control (2.08 mmol g⁻¹). Temperature-dependent isotherms indicated an exothermic process, with adsorption decreasing from 5.77 to 4.24 mmol g⁻¹ as the temperature increased from 298 to 323 K.

Ouyang et al. developed melamine–resorcinol–formaldehyde resin/graphene oxide (MR/GO) composites, exploring how activation temperature and GO content affect CO₂ uptake. Their optimal sample, GO (0.25)/MR-500, recorded capacities of 2.27 mmol g⁻¹ at 100 kPa and 5.21 mmol g⁻¹ at 500 kPa. Notably, the material exhibited rapid adsorption kinetics, reaching 90 % of saturation within 2 min, and showed high CO₂/N₂ selectivity (58 at 20 kPa). The adsorbent maintained 98 % of its initial capacity even after five adsorption–desorption cycles. The synergistic combination of nitrogen-rich resin precursors and conductive GO backbones allowed the adsorbent with both high diffusivity and abundant Lewis-basic sites. Amine functionalization is the most direct chemical treatment to improve CO₂ affinity. Pokhrel et al. functionalized 3-aminopropyltriethoxysilane (APTES) onto GO and achieved ~36 % higher CO₂ uptake than pristine GO under 1 atm and 30 °C. Also, pre-adsorbed water increased capacity by ~33 %, as

hydrogen-bonded water molecules facilitated the formation of carbamate and bicarbonate species. Functionalization and doping can transform graphene from a weakly physisorptive platform into high-performance CO₂ sorbent. Nitrogen and amine groups contribute specific binding sites, metal oxides introduce additional basic sites, and the intrinsic conductivity of graphene facilitates regeneration. The balance of these factors such as binding strength, reversibility, and stability determines the chemisorption-based CO₂ capture capacity.

4.3 Hybrid

The pursuit of efficient CO₂ capture has increasingly focused on materials that combine the complementary benefits of physisorption and chemisorption within a single adsorbent. Physisorption enables rapid, low-energy, and reversible uptake, while chemisorption enhances binding strength and selectivity through surface functionalities and heteroatom incorporation. Graphene, with its high surface area, tunable surface chemistry, and hierarchical porosity, offers a unique foundation for combining these two mechanisms within a single framework.

Yang et al. demonstrated a representative hybrid system based on layered double hydroxide (LDH)/graphene oxide (GO) composites doped with alkali metal nitrates [(Li_{0.3}Na_{0.18}K_{0.52})NO₃]. The introduction of nitrate ions provided Lewis basic sites that facilitated chemisorptive interactions, while the GO layers maintained a high surface area for physisorption. The optimized composite achieved an uptake of 3.13 mmol g⁻¹ at 30 mol % nitrate loading, nearly an order of magnitude higher than the undoped LDH/GO (0.35 mmol g⁻¹). However, further increases beyond 40 mol % caused a decline in capacity, attributed to pore blockage and hindered diffusion pathways. This delicate trade-off exemplifies how structural tuning must balance the density of chemical binding sites with the preservation of open porosity critical to physisorptive transport.

Ren et al. reported distinct method to incorporate CuBTC and ionic liquids (ILs) into graphene aerogels (GAs). While pristine GAs adsorbed only 0.34 mmol g⁻¹ of CO₂ at 298 K, adding CuBTC and ILs boosted uptake to 3.71 mmol g⁻¹ and improved CO₂/N₂ selectivity. The MOF provided well-defined adsorption sites, ILs contributed polar domains for chemisorption, and the GA network ensured rapid gas diffusion. Such hierarchically structured composites exemplify the synergy between physical porosity and chemical affinity that characterizes effective hybrid adsorbents.

Heteroatom-doped graphene carbons also demonstrate intrinsic hybrid behavior. N- and S-doped graphene hybrid from polymer precursors such as polypyrrole and polyaniline yielded microporous structures (>1200 m² g⁻¹) with basic heteroatom sites. These materials achieved CO₂ uptakes exceeding 4 mmol g⁻¹ at ambient pressure, combining dispersive physisorption within pores and chemisorptive binding at dopant sites. Collectively, such studies underscore that optimizing pore structure and dopant concentration enables graphene-based hybrids to deliver high capacity, strong selectivity, and excellent cyclic stability while remaining amenable to regeneration and scalable synthesis.

5. UTILIZATION OF ADSORBED CO₂

Adsorbing CO₂ into valuable products is a crucial connection between carbon capture and sustainable management. Researchers have investigated the conversion of CO₂ into methanol, formic acid, or methane by hydrogenation. Methanol is a liquid fuel and an industrial precursor. Graphene-based catalysts have been included into these systems to facilitate CO₂ activation and maintain the stability of intermediates. This is due to their extensive surface area and ability to modify their electrical characteristics. Simultaneous progress in electrochemical CO₂ reduction, often driven by renewable electricity, demonstrates the potential of functionalized or doped graphene to act as a conductive scaffold and provide active sites for the selective conversion into CO, hydrocarbons, or alcohols. These strategies demonstrate potential for converting collected CO₂ into valuable products while also reducing the energy expenses associated with conventional desorption–conversion cycles. Adsorbed CO₂ can be utilized in mineralization and polymerization procedures that provide stability and prolonged utility, in addition to serving as fuels. Mineralization processes, resulting from natural geochemical reactions, produce stable carbonates when CO₂ interacts with alkaline oxides or silicates. Graphene composites have shown the capability to accelerate these reactions. Concurrently, CO₂ copolymerization with epoxides has progressed to provide polycarbonates, an important class of sustainable polymers, with graphene-based catalysts improving both efficacy and durability. These methodologies demonstrate that graphene-based adsorbents enhance capture efficiency and facilitate the subsequent utilization of CO₂, thereby positioning adsorption as a crucial component of a circular carbon economy.

6. CONCLUSION

Graphene adsorbents present a compelling alignment of materials tunability, and process demands in CO₂ capture. Physisorption-oriented graphene with microporosity and minimal functionalization deliver fast kinetics and low regeneration energy, making them ideally suited for post-combustion streams under moderate humidity. In contrast, functionalization or heteroatom doping enables chemisorptive interactions that improve selectivity and uptake at low CO₂ partial pressures. Hybrid designs that co-locate ultramicropores for dispersive binding with basic sites for chemical anchoring can optimize working capacity over realistic swing windows, balancing reversibility and binding strength. In conclusion, graphene-based adsorbents have evolved into a highly tunable class of materials whose pore architecture and surface chemistry can be engineered across the physisorption–chemisorption spectrum. When paired with rigorous testing such as multi-component humid isotherms, long-cycle stability, energy balances, structured design, and integration with downstream utilization, graphene materials hold realistic potential to compete with established adsorbents and enable decarbonization pathways. The path forward demands bridging the gap between equilibrium metrics and system-level performance, ensuring graphene transforms into impactful CO₂ mitigation.

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