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Graphene Oxide–Based Thin-Film Composite Membranes for CO_2 Separation

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22nd August 2025

Acknowledgements

We would like to begin by expressing our profound gratitude to **Imperial College London** for providing the collaborative environment and essential resources that enabled this work. Our appreciation also goes to the **KAUST Academy** for granting us this unique opportunity through the WE Spark program, and to the **Women and Energy Associati**on as well **as King Abdullah City for Atomic and Renewable Energy (K.A.CARE)** for their constant encouragement and invaluable support.

We extend our sincere gratitude to **Dr. Niladri Banerjee** for his continuous guidance and invaluable feedback, which greatly shaped the direction and quality of this study.

We are especially thankful to **Zeyou,** whose tireless efforts in conducting AFM scans, guiding data analysis, and providing consistent support were fundamental to the success of this project. His patience in addressing questions and resolving challenges made a remarkable impact on both the progress and the quality of our work.

Finally, we would like to express our appreciation to **Dr. Shanglong Ning** for preparing the graphene samples and generously sharing his expertise in the transfer process.

Abstract

Saudi Arabia, as a major energy producer, faces growing challenges in reducing CO_2 emissions from energy-intensive sectors such as power generation and petrochemicals. Achieving the Kingdom's Vision 2030 targets requires efficient carbon capture solutions, with membrane technologies offering a scalable and energy-efficient pathway. In this context, graphene oxide (GO)–based thin-film composite (TFC) membranes are attracting increasing attention. This study explores the effect of GO deposition on silicon substrates as a preliminary step toward developing GO–TFC membranes. Atomic force microscopy (AFM) and Gwyddion analysis showed that GO layers introduced wrinkle-like corrugations and nanoscale features, in contrast to the more irregular bare silicon surface. Line profile and Fast Fourier Transform (FFT) analyses confirmed the emergence of periodic wrinkles ordered in GO/Si. These findings demonstrate that GO deposition enhances nanoscale surface organization and periodicity, underscoring its potential to improve TFC membrane performance for CO_2 separation applications.

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1. Introduction

The growing urgency to mitigate global warming and achieve net-zero emissions has accelerated the global search for efficient and sustainable solutions. Although the transition toward renewable energy and improved efficiency has reduced reliance on hydrocarbons, certain industrial sectors continue to depend heavily on fossil fuels, making Carbon Capture, Utilization, and Storage (CCUS) a pivotal component in global decarbonization strategies (Rowaihy et al., 2025). However, the large-scale deployment of CCUS remains constrained by economic and technical challenges, particularly in the capture stage, which is widely recognized as the most energy-intensive and costly phase (Fetisov et al., 2023). Addressing these challenges requires tailored solutions that reflect the industrial structure and energy profile of each country.

In the Kingdom of Saudi Arabia (KSA), this challenge is particularly pronounced due to its role as one of the world's largest energy producers. Balancing energy security, economic growth, and environmental sustainability has become a national priority under the Vision 2030 framework. Recent assessments show that the majority of Saudi Arabia's CO_2 emissions originate from energy-intensive sectors. As illustrated in Figure 1, the electricity generation sector is the largest contributor, emitting nearly 188 MTPA in 2022. Other significant sources include desalination, petrochemicals, refineries, and cement production, while the iron & steel and ammonia industries contribute to a lesser extent (DNA, 2016; Hamieh et al., 2022; Rowaihy et al., 2025) This distribution underscores the pressing need for targeted decarbonization pathways in Saudi Arabia, with CCUS expected to play a pivotal role alongside renewable energy expansion and efficiency improvements.

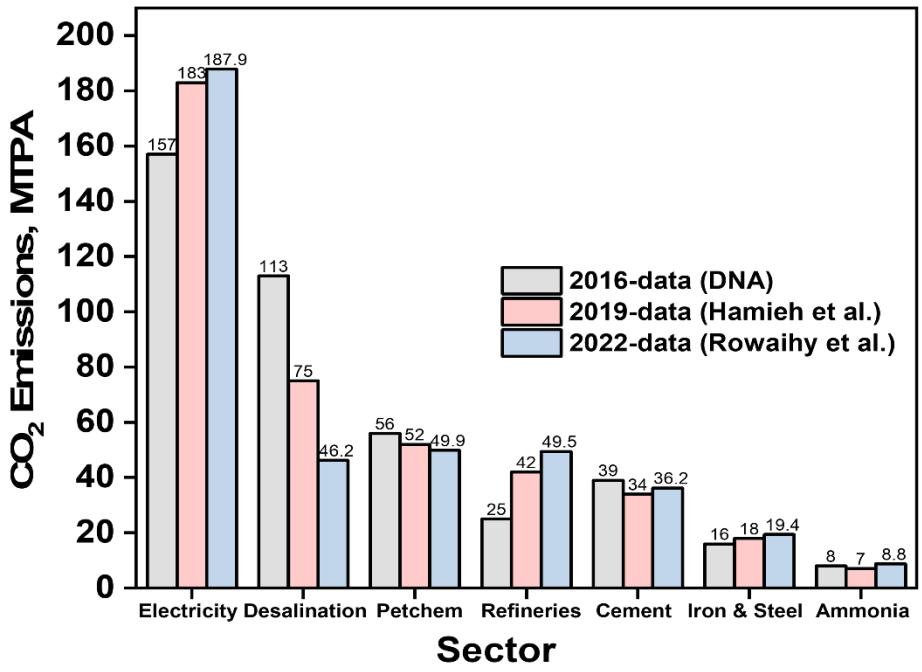


Figure 1. CO_2 emissions from major industries in Saudi Arabia.

1.1 Global CO_2 Capture Pathways

Several CO_2 capture pathways have been developed to address the challenges associated with large-scale deployment of CCUS. These are commonly classified into pre-combustion capture, oxy-fuel combustion capture, post-combustion capture (PCC), and direct air capture (DAC). Each pathway can be supported by a range of separation techniques, including liquid absorption, solid adsorption, membrane separation, and cryogenic separation. The overall classification of these systems is illustrated schematically in Figure 2 (Saenz Cavazos et al., 2023).

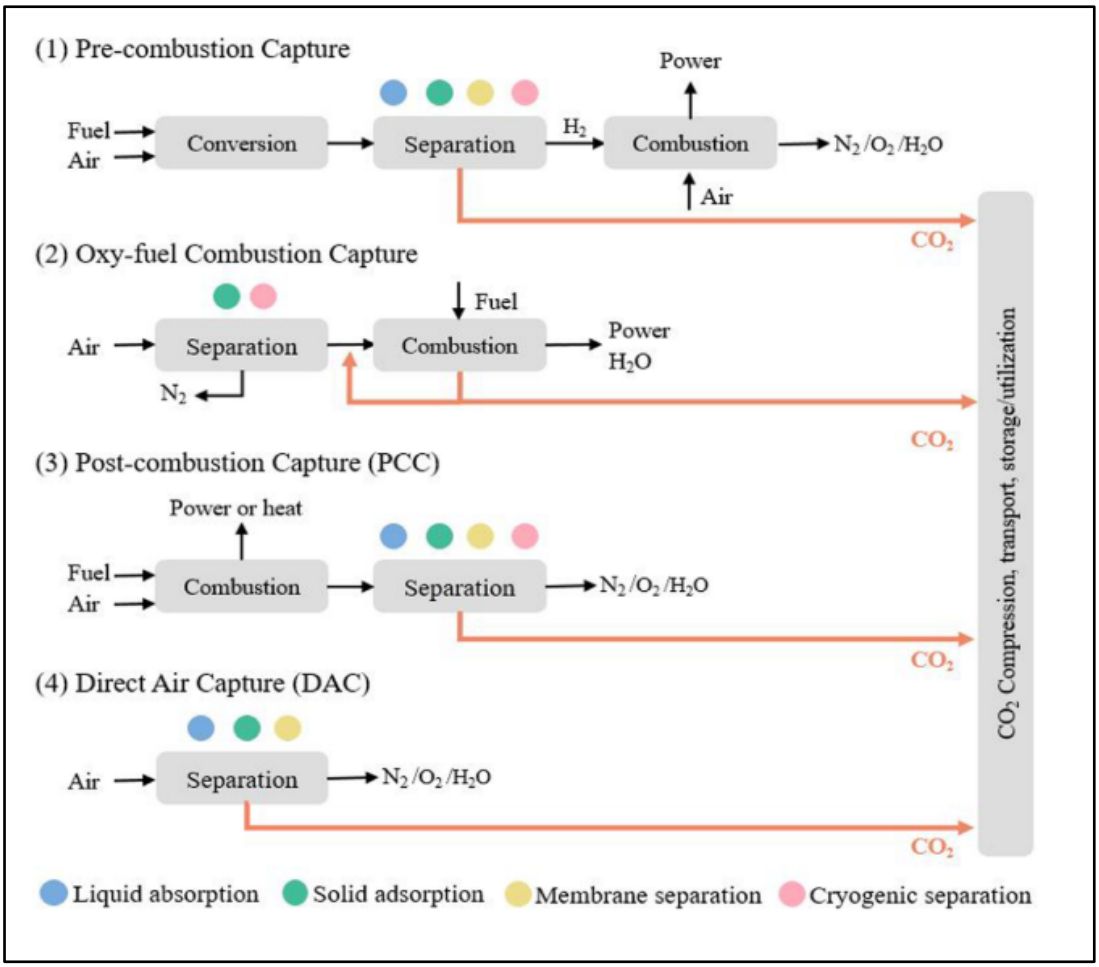


Figure 2. Carbon capture systems include pre-combustion, oxy-fuel combustion, post-combustion, and direct air capture with possible separation technologies (Saenz Cavazos et al., 2023) .

1.2 Overview of Current Technologies

Several approaches have been developed for CO_2 capture and utilization, each with distinct mechanisms and levels of technological readiness. These include afforestation, direct air capture (DAC), cryogenic processes, and membrane-based separation, as summarized in Table 1. While afforestation relies on biological storage of CO_2 in biomass, DAC and cryogenic processes employ advanced sorbents and cryogenic fluids, respectively, for CO_2 extraction from ambient air or industrial streams. Among these options, membrane separation has gained increasing attention due to its modularity, scalability, and relatively lower energy demand compared to conventional capture systems.

Table 1. Overview of carbon dioxide removal options in the literature.

Technology	Process / Key Feature	Applications	TRL	Ref.
Afforestation	Stores CO_2 in biomass	–	9	(Odeh et al., 2025)
Direct Air Capture (DAC)	Chemical / solid sorbents	Ambient air, power plants	6–7	(Saenz Cavazos et al., 2023)
Cryogenic Capture	Cryogenic fluids (e.g., N_2)	H_2 production, power plants	6–7	(Aneesh & Sam, 2023)
Membrane Separation	Thin-film composite membranes (TFC) with MOFs, GO	Gas purification, H_2 separation	5–7	(Liu et al., 2019)

TRL: Technology Readiness Level. basic research (TRL 1–3), applied R&D (TRL 4–6), or nearing commercialized (TRL 7–9).

1.2.1 Membrane Separation Technologies

Membrane separation is emerging as a promising alternative for CO_2 capture owing to its ability to selectively separate gases without the need for extensive thermal input. A particularly attractive design is the thin-film composite (TFC) membrane, which integrates a selective active layer with a porous support to achieve both high permeability and selectivity (Liu et al., 2019).

1.2.2 Thin-Film Composite (TFC) Membranes

TFC membranes are typically composed of three layers: a dense selective polyamide film responsible for gas separation, a porous support layer that provides mechanical stability, and a substrate that reinforces the structure Figure 3. Recent advancements have incorporated novel nanomaterials, such as metal–organic frameworks (MOFs) and graphene oxide (GO), into the selective layer to enhance performance. These modifications have enabled TFC membranes to demonstrate excellent potential in gas purification and hydrogen separation, as well as CO_2 capture from flue gas streams (Liu et al., 2019). Despite their advantages, challenges remain regarding long-term stability, fouling resistance, and cost-effective large-scale fabrication.

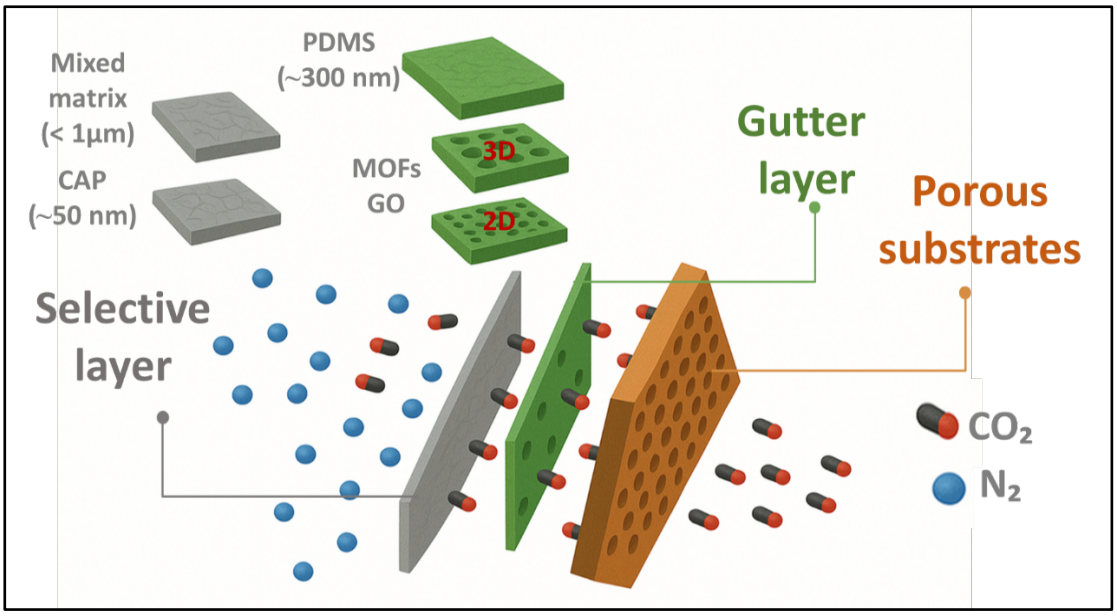


Figure 3. Three-layer structure of TFC membranes for CO_2 separation.

1.2.2.1 Graphene Oxide

Graphene oxide (GO) has been successfully used in many studies to improve the gas separation performance of resultant membranes due to its high aspect ratio, layered structure, which can provide gas transport channels with different path tortuosity for different gases (Choi et al., 2021). There are many different methods to produce nanomaterials, which can be classified into two classes: bottom-up methods and top-down methods.

2.Preparation Methods of Graphene Oxide

Graphene oxide (GO) can be synthesized through various techniques that are generally categorized into two main approaches: bottom-up and top-down.

2.1 Bottom-up methods

The bottom-up methods involve assembling carbon atoms or small molecules into a graphene structure. Examples of these include chemical vapor deposition (CVD) and epitaxial growth. Although these methods can produce high-quality graphene, they are less commonly used for GO due to their complexity, high energy consumption, and elevated cost (Gutiérrez-Cruz et al., 2022).

2.2 Top-down methods

In contrast, top-down methods are more widely applied for GO production. These approaches rely on breaking down bulk graphite into individual layers through chemical, mechanical, or electrochemical processes. Among them, the Hummers' method and its modified versions are the most frequently used. This method involves oxidizing graphite using strong oxidizing agents (e.g., $KMnO_4$, $NaNO_3$, and concentrated H_2SO_4), which introduces abundant oxygen-containing functional groups onto the graphene sheets. The oxidized graphite can then be exfoliated into single- or few-layer GO nanosheets by ultrasonication or mechanical stirring. The advantage of the Hummers' method lies in its scalability, relatively low cost, and ability to produce GO with tunable oxygen functionalities. These oxygen groups not only improve the hydrophilicity and dispersibility of GO in aqueous and polymeric media but also facilitate subsequent chemical modifications, making GO highly attractive for membrane applications (Gutiérrez-Cruz et al., 2022). As illustrated in Figure 4, In this study, graphene oxide (GO) was prepared using the top-down approach, specifically a modified Hummers' method.

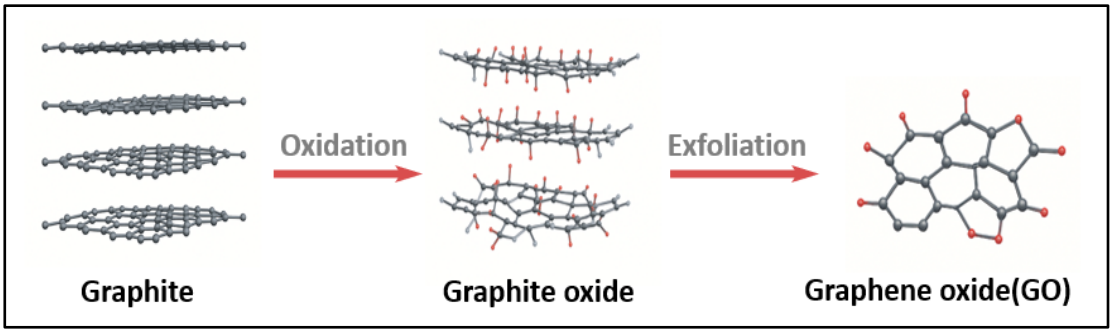


Figure 4. Top-down synthesis of graphene oxide from graphite.

3 Objectives of the Study

The aim of this study is to investigate the potential of GO-based thin-film composite (TFC) membranes for CO_2 capture. In this initial phase, we focused on the characterization of graphene oxide (GO) by performing atomic force microscopy (AFM) scans on GO deposited over a silicon substrate. This step provides preliminary insights into the surface morphology and highlights the challenges of achieving uniform nanosheet distribution, which are critical for future membrane development.

4. Atomic Force Microscopy (AFM)

Atomic force microscopy (AFM) was performed using the setup shown in Figure 5(a). AFM is a high-resolution imaging technique that enables the investigation of surface morphology at the nanoscale without the need for a vacuum environment or conductive coating, making it suitable for a wide range of materials.

The working principle of AFM is illustrated in Figure 5(b). A sharp probe tip, typically made of silicon or silicon nitride, is mounted on a flexible cantilever. As the probe scans across the surface, interatomic forces cause deflections of the cantilever. A laser beam reflected from the top of the cantilever onto a position-sensitive photo detector records these deflections, which are then converted into topographical data. AFM can operate in several modes, including contact mode, non-contact mode, and tapping mode (Goksu et al., 2009)

In this study, tapping mode was employed, where the cantilever oscillates near its resonant frequency and intermittently touches the surface. This approach reduces lateral forces and minimizes damage to both the sample and the tip, while still providing accurate surface height information.

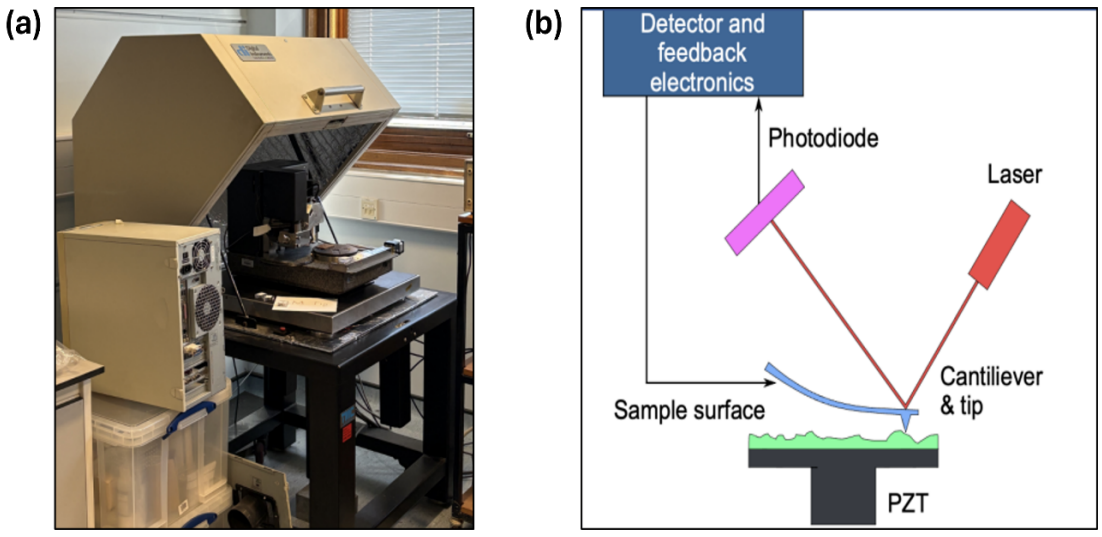


Figure 5. (a) The AFM used to take our data (b) The mechanism of AFM, showing how the tip scans across the sample surface and reflects corresponding amounts of the laser beam for measurements.

4.1 Gwyddion

An application called Gwyddion, that analyzes AFM height data, may import the raw data obtained from AFM scans. The first step was image leveling by

subtracting a polynomial background to remove scanner tilt and artifacts. This correction is essential for exposing nanoscale features on the surface.

Subsequently, noise reduction was applied using filtering tools, and the scans were visualized in both 2D and 3D modes. Line profiles were extracted along selected scan lines to measure wrinkle height, amplitude, and spacing. In addition, FFT analysis was performed to study the periodicity and orientation of wrinkle patterns, enabling comparison between GO/Si and bare Si surfaces.

Gwyddion provided statistical surface parameters, including roughness values and height distribution histograms, which were used to quantitatively compare the morphological differences between the two samples.

4.2 Data Collection

The raw images were obtained using Atomic Force Microscopy (AFM). Scans of both bare silicon and GO-coated silicon substrates were collected under identical measurement conditions. The images were uploaded into Gwyddion software for further processing, including flattening, extraction of line profiles, and generation of 3D topography views for subsequent analysis.

In addition to AFM measurements, previously reported Raman spectroscopy data for GO and graphite (Wong et al., 2017) were consulted. These data provide structural insights (such as D, G, and 2D bands) that allow correlation of surface morphology with vibrational properties.

4.3 Data analysis

The processed AFM images were analyzed to evaluate the effect of GO deposition on the silicon surface morphology. As shown in Figure 6 (a) and (b), the bare silicon surface exhibits an uneven and less-defined texture, whereas the GO-coated surface displays wrinkle-like corrugations and bright domains associated with local height differences. The 3D topography views in Figure 6 (c) and (d) further highlight these variations, revealing increased roughness for GO/Si compared to the relatively flatter but disordered bare Si substrate.

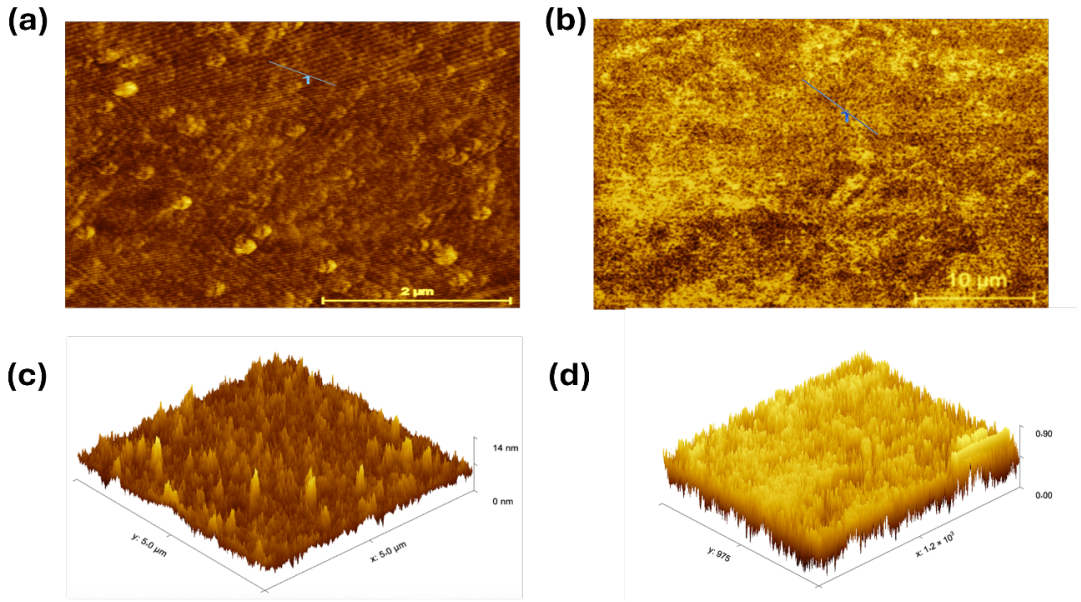


Figure 6. (a) and (b) AFM topography images of (a) GO/Si and (b) Si, with selected line profiles indicated in blue for further analysis. The corresponding 3D surface topographies are shown in (c) and (d), respectively.

The line profile analysis (Figure7 (a) and (b)) quantitatively supports these observations. The GO-coated surface demonstrates more pronounced height variations (FWHM ≈ 61.7), while the bare silicon surface shows lower fluctuations (FWHM ≈ 17.7).

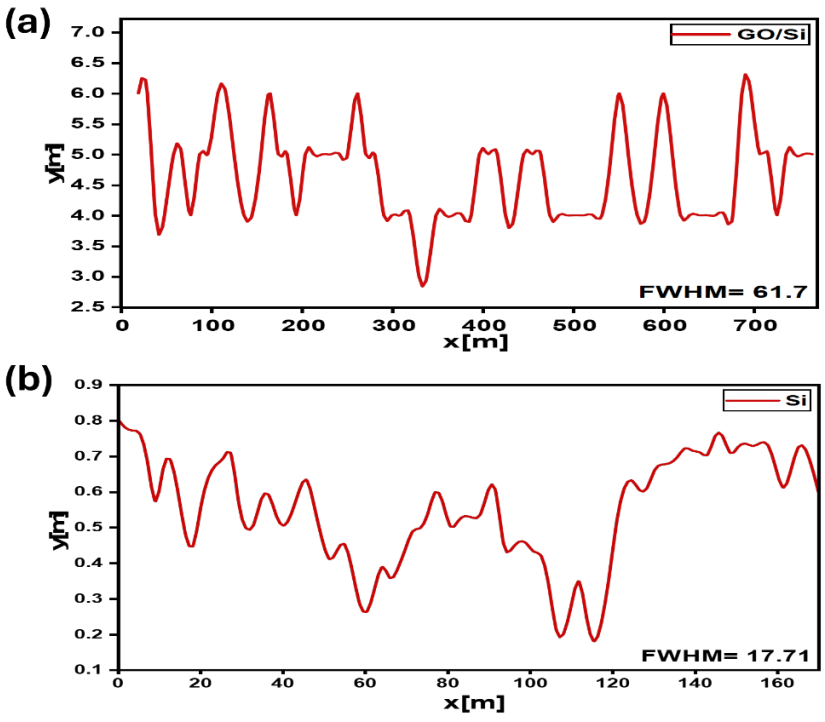


Figure 7. (a) and (b) Line profiles of GO/Si and Si surfaces with FWHM values.

Further insights are obtained from FFT analysis of the line profiles. As shown in Figure 8 (a), the FFT spectrum of GO/Si reveals several strong peaks at low wavenumbers, indicating periodic wrinkle-like patterns with different characteristic wavelengths. In contrast, the FFT of bare Si (Figure 8(b)) exhibits fewer and weaker peaks, reflecting the absence of distinct periodicity. This suggests that GO deposition induces ordered nanoscale corrugations, while the bare silicon surface remains largely disordered.

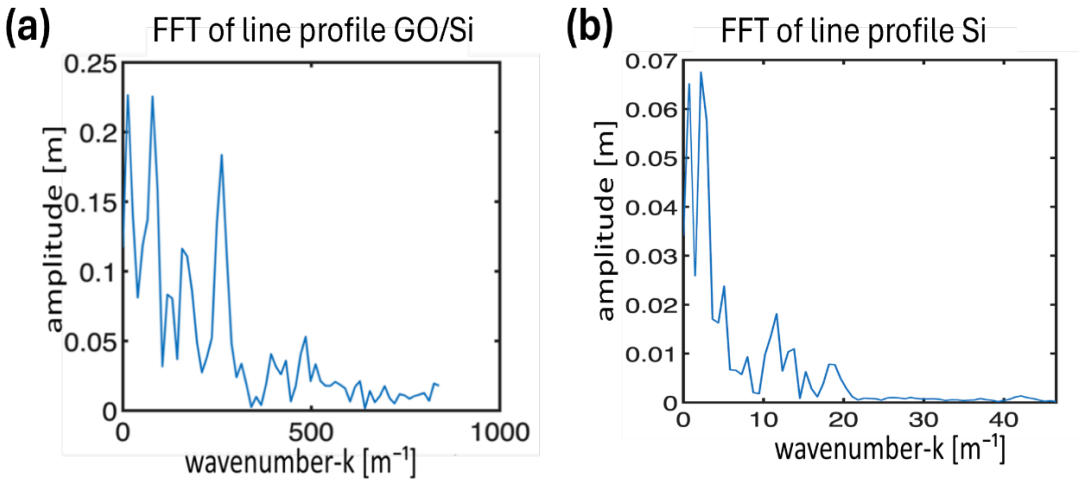


Figure 8 . FFT analysis of AFM line profiles for (a) GO on silicon and (b) bare silicon, showing differences in periodic surface features. The x-axis represents the position along the line, while the y-axis corresponds to the height of each point.

These findings are further supported by the statistical roughness parameters in Table 2. Although the bare Si surface shows higher RMS roughness (0.124) and mean roughness (0.101) compared to GO/Si (0.083 and 0.065, respectively), the GO/Si sample exhibits a greater maximum peak height (0.348 vs. 0.261), highlighting the formation of elevated wrinkle structures. The scan line discrepancy is also higher for GO/Si (0.136 vs. 0.062), consistent with the presence of additional nanoscale features introduced by the GO layer.

Table 2. Statistical Quantities of GO on silicon and silicon obtained from AFM analysis

Parameter	GO/Si	Si
RMS roughness (Sq)	0،0831278	0،124342
Mean roughness (Sa)	0،0654780	0،101240
Maximum peak height (Sp)	0،347922	0،261099
Maximum pit depth (Sv)	0،244235	0،413411
Maximum height (Sz)	0،592157	0،674510
Scan line discrepancy	0،135744	0،0619167

Finally, Raman spectroscopy data reported previously (Wong et al., 2017) and illustrated in Figure 9 reveal a higher D/G intensity ratio for GO compared to graphite, reflecting increased structural disorder. This observation aligns with our AFM and FFT results, which demonstrate that GO deposition introduces nanoscale corrugations and enhances local height variations.

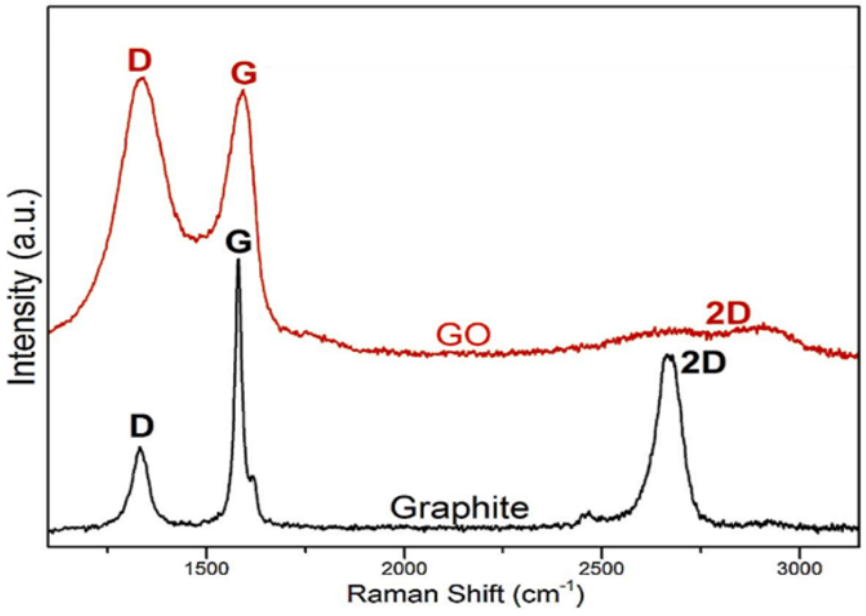


Figure 9. Raman spectra of graphite (black) and graphene oxide (red), showing the characteristic D, G, and 2D bands. The increase in the D/G intensity ratio in GO indicates structural disorder and successful oxidation.

5. Conclusion

This study demonstrated the significant influence of graphene oxide deposition on the nanoscale morphology of silicon substrates. AFM imaging combined with Gwyddion analysis revealed that the GO-coated surface exhibited pronounced wrinkle-like corrugations and higher peak features compared to the bare silicon surface, which remained relatively disordered. Line profile and FFT analyses confirmed the emergence of ordered periodic wrinkles in GO/Si, whereas bare Si showed limited periodicity.

Furthermore, statistical parameters supported these findings, with GO/Si showing higher peak heights and scan line discrepancies, despite bare Si exhibiting greater average roughness values. These results highlight that GO deposition modifies not only the roughness but also the periodic organization of the surface, paving the way for enhanced membrane performance in CO_2 separation applications.

Overall, the integration of AFM characterization and Fourier-domain analysis provided robust evidence that GO-based thin-film composite membranes hold strong potential for advancing CO_2 capture technologies.

6. Future Work

Building upon the insights gained in this study, future research should aim to further deepen the understanding of GO–based thin-film composite membranes and their potential in CO_2 separation. Complementary structural analyses, performance testing, and optimization strategies will be essential to bridge the gap between laboratory observations and scalable applications. The following directions are proposed:

- Conduct additional surface and structural characterizations (e.g., XPS, XRD).
- Evaluate gas separation performance of the TFC–GO membranes under different conditions.
- Optimize membrane fabrication parameters to improve stability and selectivity.

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