

**Investigation and Characterization of the Optical, Thermal,
and Adsorption Properties of Carbon Aerogels for CO₂
Capture Applications**

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Abstract

Climate change driven by rising atmospheric CO₂ concentrations has intensified the need for efficient, regenerable solid sorbents. Carbon aerogels are promising candidates due to their ultra-high porosity, low density, and tunable structure, yet their coupled optical, photothermal, and CO₂ adsorption behaviour as monolithic sorbents remains insufficiently characterized. This thesis investigates a disc-shaped carbon aerogel for CO₂ capture to clarify how its radiative and thermal properties govern adsorption performance and photothermal regeneration.

Carbon aerogels were characterized using UV–Vis–NIR and FTIR spectroscopy to measure reflectance, transmittance, and absorptance across the solar and thermal-infrared ranges. Mid-wave infrared thermography under a xenon arc lamp (1 Sun irradiance) quantified transient heating and cooling. Dynamic breakthrough experiments with a 11% CO₂ / 89% N₂ mixture at 25 °C, followed by photothermal desorption at 110 °C, were performed to determine CO₂ capacity and regeneration performance.

Optical measurements showed negligible transmittance and low reflectance from the ultraviolet through the mid-infrared, corresponding to broadband absorptance generally above 0.9 and high effective emissivity in the thermal-imaging band. Under 1 Sun illumination, the surface temperature rose from ~28 °C to ~64 °C within 1–2 min and cooled rapidly back toward ambient once the light source was removed, indicating efficient radiative heating and low thermal mass. Breakthrough measurements yielded a net CO₂ adsorption capacity of 1.88 mmol g⁻¹ and a photothermal desorption capacity of 1.14 mmol g⁻¹, with good reproducibility but partial hysteresis between adsorption and desorption.

Overall, the results demonstrate that carbon aerogels can combine meaningful CO₂ uptake with favourable photothermal characteristics, making them strong candidates for radiatively driven temperature-swing adsorption processes. The work provides a quantitative baseline for future material design, further characterization, and reactor-level optimisation of carbon-aerogel-based CO₂ capture systems.

To My Brave and Honorable Compatriots

Light Prevails Over Darkness

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1 Introduction

Global efforts to mitigate climate change have intensified interest in carbon capture technologies. Carbon dioxide (CO₂) capture and storage is increasingly recognized as essential for decarbonizing hard-to-abate sectors and meeting climate targets. In this context, solid adsorbents have emerged as promising alternatives to conventional liquid amine scrubbers, offering the potential for lower regeneration energies and avoidance of issues like solvent corrosion and degradation [1-3]. Advanced porous materials, including zeolites, metal–organic frameworks (MOFs), porous carbons, and hybrid polymers, are being widely researched for CO₂ capture applications [4]. Among these, aerogels have gained attention due to their extraordinary porosity and tunability [5,6]. Aerogels are ultralight materials derived from a sol–gel process followed by careful drying to preserve a 3D nanoporous network [7,8]. They typically exhibit >90% porosity and enormous surface area (often >800 m²·g⁻¹) [5], which are highly advantageous for gas adsorption. Crucially, the aerogel framework's composition can be tailored (organic, inorganic, or hybrid) and functionalized, allowing strategic modifications for improved CO₂ affinity [9,10].

Carbon aerogels, obtained by carbonizing organic aerogel precursors (e.g., resorcinol–formaldehyde or biopolymers) in an inert atmosphere, are a particularly intriguing subclass. By virtue of their composition, carbon aerogels can be electrically conductive, intrinsically hydrophobic, and mechanically robust, which is attractive for cyclic adsorption processes and for operation under humid conditions where water fouling can degrade performance [11]. In addition, carbon aerogels are typically black and behave as broadband absorbers with negligible reflectance or transmittance across the UV–visible and infrared spectrum [12] (since reflectance (R), absorptance (A), and transmittance (T) satisfy $R + A + T = 1$, very low R and T imply $A \approx 1$). For

solar-driven photothermal CO₂ capture, this is beneficial because the attainable sorbent temperature is governed by an energy balance between absorbed solar power and losses by convection and thermal radiation; therefore, high solar absorptance (roughly over the wavelength range 0.3–2.5 μm) directly increases the heating rate and peak temperature. However, efficient solar heating also favors low thermal emissivity at longer wavelengths (mid-IR), because radiative losses scale with emissivity and become increasingly important as temperature rises. In the ideal case, a photothermal sorbent behaves as a spectrally selective absorber, combining high solar absorptance with low thermal emittance/emissivity to suppress reradiation losses. This optical selectivity requirement motivates the present work: beyond confirming broadband solar absorption and low thermal conductivity (which localize heat and accelerate temperature swing) [13], it is also essential to quantify the aerogel's infrared optical response relevant to emissivity and radiative losses, and to evaluate how these coupled optical–thermal characteristics translate into photothermal regeneration for CO₂ capture.

1.1 Motivation

Climate change driven by rising carbon dioxide (CO₂) levels has created an urgent need for effective carbon capture solutions. Atmospheric CO₂ concentration has climbed from about 280 ppm in pre-industrial times to around 420 ppm as of the early 2020s, a nearly 50% increase caused largely by fossil fuel emissions [14]. This unprecedented rise in potent greenhouse gas is fueling global warming, ocean acidification, and other climate impacts. International accords such as the Paris Agreement call for rapid emissions reductions and carbon-neutral targets by mid-century (2050) to limit warming to 1.5–2 °C. Achieving these goals will require not only emissions cuts but also gigaton-scale removal or capture of CO₂ from major sources.

Carbon capture and storage (CCS) has emerged as a key mitigation strategy, but current deployment falls far short of what is needed. At present, CCS and related technologies capture only on the order of 50 million tons of CO₂ per year – about 0.1% of global CO₂ emissions. In contrast, climate models consistent with 1.5 °C warming scenarios indicate that around 1 billion tons per year of CO₂ capture will be required by 2030, and several billions of tons by 2050 [15]. This stark gap highlights a pressing motivation for research into advanced CO₂ capture technologies and materials that can be deployed at scale to bridge toward climate targets.

1.2 Research Goals and Gaps in Current Literature

Given the above context, this thesis is motivated by the gap in current literature regarding carbon aerogels for CO₂ capture. Most prior studies have focused on silica-based or polymer-based aerogels with chemical modifications, while pure carbon aerogel materials have not been investigated in the same depth. The distinct properties of carbon merit a dedicated study to evaluate how effective they are in capturing CO₂ and under what conditions they excel. The overarching goal of this research is therefore to evaluate whether and how a carbon aerogel's unique properties make it an effective CO₂ adsorbent, by systematically characterizing the optical properties of materials and performance in CO₂ capture. We aim to develop a comprehensive understanding of the relationships between the carbon aerogel's physicochemical characteristics and its CO₂ adsorption behavior, which will help determine how it benchmarks against other sorbents and how it might be further improved.

In pursuit of this goal, a multi-faceted experimental approach is employed. A prefabricated monolithic carbon aerogel is examined using a suite of analytical techniques to characterize its optical and thermal properties, and gas adsorption performance. By correlating these findings, we

address the research questions: How do the optical and thermal features of the carbon aerogel influence its capacity and selectivity for CO₂? And can the material's high porosity and other attributes be leveraged to make it a competitive CO₂ capture sorbent? Ultimately, this work seeks to fill the knowledge gap about carbon aerogels by providing data and insights that can guide future design or application of such materials in carbon capture technologies.

1.3 Literature Review

Conventional CO₂ Capture Technologies: Industrial CO₂ capture is currently dominated by liquid amine scrubbing, a chemical solvent process that can effectively separate CO₂ from flue gases. However, amine-based systems suffer significant drawbacks, including high regeneration energy penalties, solvent degradation, and corrosion issues. The thermal energy required to strip CO₂ from amine solutions can consume a large fraction of a power plant's output, making the process energetically and economically costly. Furthermore, amine solvents gradually degrade and can cause equipment corrosion, leading to high operational and maintenance demands [16]. Alternative separation methods like membrane-based CO₂ capture and cryogenic distillation have been explored. Membranes offer a compact solution but often face problems such as fouling and limited selectivity, while cryogenic methods demand excessive energy for cooling and are generally impractical for large-scale scrubbing [1]. These challenges with conventional approaches underscore the need for more efficient, robust, and cost-effective CO₂ capture materials and processes.

Solid Sorbents for CO₂ Capture: In contrast to liquid solvents, adsorption-based CO₂ capture on solid sorbents is attracting extensive interest as an energy-efficient and modular approach [17]. Solid adsorbents can in principle be regenerated with lower energy input and avoid many issues

of liquid solvents. A diverse array of solid materials has been investigated for CO₂ adsorption, including porous oxides like zeolites and silica, metal–organic frameworks (MOFs), covalent organic frameworks (COFs), inorganic oxides that chemisorb CO₂ (e.g. solid alkali metal carbonates), and a wide range of carbon-based sorbents [4]. Among these, carbonaceous adsorbents are particularly promising due to their chemical stability, low-cost precursors, and highly tunable structure [2]. Many forms of engineered carbon have been studied for CO₂ uptake from activated carbons and charcoals to carbon nanotubes, graphene derivatives, and carbon aerogels, all of which typically offer high surface areas and porosity, providing abundant adsorption sites for CO₂ molecules [11]. Carbon-based materials also tend to be thermally stable and resistant to degradation, which is advantageous for repeated adsorption–desorption cycling in industrial conditions [18].

Aerogels for CO₂ Capture: Within the family of porous materials, aerogels represent a unique class of ultra-porous, lightweight sorbents that has gained increasing attention for CO₂ capture [7]. An aerogel is a rigid, foam-like material derived from a gel in which the liquid component has been replaced by air while preserving the gel's framework. Typically, aerogels are formed by a sol–gel polymerization of precursors (organic or inorganic), followed by a drying process, such as supercritical CO₂ drying or freeze-drying, that carefully removes the solvent without collapsing the pore structure [7,19]. The result is a monolithic material with extremely low density and high porosity. Aerogels consist of a three-dimensional interconnected network of nanometer-sized particles or polymer chains, with a pore volume fraction often above 90% [3]. Early aerogels were silica-based (invented in the 1930s), but modern aerogels encompass a variety of compositions including metal oxides, organic resins, biopolymers, and carbon (usually obtained by pyrolysis of

organic polymer aerogels), as well as hybrid or composite aerogels that combine multiple components[11].

Several key physicochemical properties make aerogels highly attractive for CO₂ capture applications:

- Ultra-high porosity and low density: Aerogels are often ~95–99% empty space. Densities as low as 0.1–0.3 g/cm³ have been reported, meaning most of the material's volume is accessible pore space. This yields an enormous internal pore volume for gas adsorption. The extremely low bulk density is also beneficial for weight-sensitive applications such as aviation or direct air capture (DAC) units where minimizing sorbent weight is important [20, 21].
- High specific surface area: Owing to their nanostructured porosity, aerogels can achieve surface areas on the order of 600–1000 m² per gram. Such high surface area correlates with a high concentration of adsorption sites available for binding CO₂ molecules. In general, a larger surface area provides more active sites for physisorption, typically leading to higher CO₂ uptake capacity [20, 21].
- Hierarchical pore structure with mesopores: Aerogels often have a pore size distribution centered in the mesopore range (~20–50 nm) [19]. These mesopores facilitate fast gas diffusion into and through the material, mitigating the slow kinetics that purely microporous adsorbents (pores < 2 nm) can suffer [19]. Many aerogels also contain micropores within their structure, which contribute strongly to adsorption capacity, while the mesopores provide rapid transport pathways – yielding a beneficial combination of high capacity and fast kinetics [21].

- Tunable surface chemistry: The composition and surface functionality of aerogels can be readily tailored either by incorporating dopants/functional monomers during synthesis or by post-synthesis modifications. The open pore network and high surface area of aerogels make it feasible to decorate their internal surfaces with functional groups such as amines, alkali carbonates, ionic liquids, or other moieties that selectively bind CO₂ [21, 22]. This flexibility in chemical modification allows aerogels to be optimized for greater CO₂ affinity or selectivity. For instance, amine-functionalized aerogels can capture CO₂ via reversible chemisorption (carbamate formation), greatly enhancing uptake especially at low CO₂ partial pressures [23].
- Thermal and optical properties: Owing to their nanoscale porosity, aerogels, especially those made of carbon, often exhibit very low thermal conductivity and unique optical behavior such as high infrared absorptance [21,24]. Aerogels have very low thermal conductivity, which makes them excellent thermal insulators. While this low conductivity can hinder heat transfer in conventional temperature swing adsorption (TSA), it becomes advantageous in photothermal or other localized-heating TSA systems because it helps retain the heat generated within the sorbent, reducing thermal losses to the surroundings. High IR absorption (in carbon aerogels) means they can be efficiently heated by infrared radiation. These traits can be exploited to rapidly heat the sorbent via IR or sunlight in a TSA process, enabling faster or more energy-efficient regeneration of the material [24].

Aerogels, with their combination of ultra-porosity, high surface area, tunable chemistry, and favorable heat/mass transfer properties, have shown considerable promise for CO₂ capture. Indeed, recent reviews highlight that aerogels allow tailoring of textural characteristics and surface chemistries to achieve high CO₂ uptake capacities and selectivity [21]. Researchers have been

actively developing modified aerogels to improve performance, for example by introducing amine functional groups, creating composite/hybrid aerogels, and optimizing pore structures [23].

Amine-Functionalized and Modified Aerogels: A common strategy to enhance CO₂ capture in aerogels is amine functionalization of the surfaces. This can be achieved by impregnating the aerogel with polyamines or grafting aminosilanes and other amine-containing molecules onto the pore walls [23, 25]. The amine groups act as reactive sites that chemically bind CO₂ and forming carbamate or bicarbonate species, thereby dramatically increasing the CO₂ capacity and selectivity, especially at low partial pressures. Numerous studies have pursued this approach. For example, Keshavarz et al. reported that loading a silica aerogel with ~80 wt% polyethylenimine (PEI) resulted in a CO₂ uptake of about 6.1 mmol CO₂ per gram at 25 °C [2]. Similarly, Bahadi et al. noted in a recent review that amine-modified aerogels consistently show improved uptake and preferential CO₂ adsorption over N₂ [11]. In one innovative design, a “liquid-in-aerogel” composite was created by incorporating tetraethylenepentamine (TEPA) along with a tertiary amine carrier into silica aerogel particles; this composite achieved ~5.62 mmol/g CO₂ capacity at 85 °C (15% CO₂), with excellent CO₂/N₂ selectivity and stable performance over multiple cycles [26]. Another study functionalized a fly-ash-derived silica aerogel with aminopropyltriethoxysilane (APTES), yielding ~4.95 mmol/g CO₂ uptake at 70 °C and over 94% capacity retention after 10 adsorption–desorption cycles [27]. Beyond amines, researchers have also explored inorganic and ionic modifiers; for instance, impregnating aerogels with alkali metal carbonates to capture CO₂ as solid carbonates, or attaching ionic liquid-based and amino-acid-based functional groups to aerogel frameworks [28, 29]. These modifications aim to introduce stronger chemisorptive interactions for CO₂ while leveraging the aerogel’s porous structure as a support.

Hybrid and Carbon Aerogel Composites: Rather than modifying an aerogel with small molecules, another route is to combine aerogels with other high-performance adsorbent phases. Hybrid aerogel composites have demonstrated impressive results. For example, Zhao et al. fabricated composite material by integrating a copper-based MOF (HKUST-1) into an aramid nanofiber-derived aerogel matrix. The resulting hybrid monolith had a hierarchical pore structure and achieved a CO₂ capacity of ~7.3 mmol/g at 1 bar, with a remarkable CO₂/N₂ selectivity of ~40:1 [30, 31]. In another study, a cellulose-derived carbon aerogel was grafted with PEI, producing a sorbent that captured ~5.6 mmol/g CO₂ from a 15% CO₂ flue-gas mixture (at ~40 °C) with a very high separation factor (~113 for CO₂/N₂) [32]. These examples illustrate that aerogels, especially carbon-based ones, can serve as versatile scaffolds: their porous network can accommodate functional additives or dispersed active phases, yielding composites that greatly boost CO₂ uptake capacity, selectivity, and cycling stability under realistic conditions [33].

Despite the substantial progress in aerogel research for carbon capture, it is noteworthy that much of the literature to date has been dominated by silica aerogels and their amine-modified variants. In contrast, comprehensive studies focusing specifically on pure carbon aerogels for CO₂ capture are comparatively scarce. Carbon aerogels derive from pyrolyzing organic aerogel precursors, and they retain the open nanostructure of the parent aerogel while forming an all-carbon framework [34]. This imbues carbon aerogels with a set of advantageous properties, including hydrophobic surfaces, electrical conductivity, and greater thermal and chemical stability, that are not found in silica or polymer aerogels [35]. Such properties could be highly beneficial for CO₂ adsorption; for example, hydrophobicity can prevent water interference, and high thermal stability allows thermal swing regeneration at higher temperatures. However, the adsorption characteristics of carbon aerogels, and how their intrinsic pore architecture and surface chemistry influence CO₂ uptake,

remain relatively under-explored in the literature. There is a clear knowledge gap in understanding and optimizing carbon aerogels as CO₂ sorbents, especially in comparison to the more extensively studied classes like MOFs or zeolites.

1.4 Thesis Objectives

Building on the identified gaps and goals, the specific objectives of this thesis are as follows:

1. Characterize the Optical Properties of Carbon Aerogels (FTIR and UV–Vis Spectroscopy):
Quantitatively measure the carbon aerogel’s optical reflectance, transmittance, and absorbance across a broad spectrum. Using Fourier-transform infrared (FTIR) and UV–Vis–NIR spectroscopy, the aerogel’s ability to absorb light/infrared radiation will be assessed. This will reveal if the aerogel’s black carbon framework and porous structure lead to exceptionally high absorption as hypothesized.
2. Characterize the Photo-thermal Behavior of Carbon Aerogels (Infrared Thermography):
Assess the photothermal response and heat-transfer characteristics of the carbon aerogel using infrared thermal imaging. By monitoring the surface temperature of the aerogel under light-induced heating from the xenon arc lamp, IR thermography reveals how efficiently the material absorbs light and converts it into heat, as well as whether temperature gradients form across the surface. These measurements directly relate to photothermal-driven temperature swing adsorption processes, where a sorbent that heats rapidly under illumination and retains heat due to its low thermal conductivity can facilitate more energy-efficient CO₂ desorption (regeneration).

3. Evaluate CO₂ Adsorption Capacity and Selectivity: Determine the carbon aerogel's CO₂ uptake capacity and its selectivity for CO₂ over N₂ under relevant conditions. Gas adsorption experiments will be conducted at near-ambient temperature and pressure using a CO₂/N₂ mixture to simulate flue gas. The objective is to measure how much CO₂ the aerogel can adsorb (in mmol CO₂ per g) and to calculate metrics like CO₂/N₂ selectivity and cyclic working capacity. We hypothesize that the aerogel's high surface area and microporosity will enable a significant CO₂ capacity (on the order of a few mmol/g) even without chemical functionalization. However, since the carbon surface may be largely non-polar, the selectivity for CO₂ over N₂ may rely primarily on physical adsorption forces unless specific surface groups are present. By comparing the results to literature benchmarks for similar sorbents, the aerogel's efficacy will be assessed. Multiple adsorption-desorption cycles will also be performed to evaluate the material's regenerability and stability over repeated use.

Through these objectives, the present study seeks to establish a strong integration between material characterization and performance evaluation, thereby developing a comprehensive understanding of the functional behavior of the synthesized carbon aerogel as a CO₂ adsorbent. Accomplishing these aims will generate a detailed case study on carbon aerogels for CO₂ capture, elucidating the relationships between their structural, chemical, and photothermal properties and their overall adsorption performance.

1.5 Thesis Outline

This thesis is organized into several chapters, as outlined below:

- Chapter 1 – Introduction: Introduces the research problem and motivation, provides background and a literature review on CO₂ capture and aerogel sorbents, identifies the gaps in current knowledge, and states the research goals, objectives, and thesis structure.
- Chapter 2 – Background and Literature Review: Reviews relevant literature in greater depth, including the science of CO₂ adsorption, detailed discussion of various CO₂ capture materials, and prior studies on carbon aerogels. This chapter establishes the theoretical and experimental context for the thesis and further substantiates the need for the present research.
- Chapter 3 – Experimental Methodology: Describes the methods employed in this study, focusing on the characterization of carbon aerogel samples. It details each experimental technique, FTIR and UV–Vis spectroscopy, infrared thermography, and CO₂ adsorption measurements, along with the corresponding equipment, procedures, and data analysis methods.
- Chapter 4 – Results and Discussion: This chapter presents and interprets the experimental findings of the thesis in an integrated manner. It first reports the optical spectra of the carbon aerogel, thermal imaging observations, and quantitative CO₂ adsorption performance, with data summarized in figures and tables alongside baseline analyses. These results are then discussed in the context of the research objectives, examining how the measured optical and thermal properties relate to CO₂ uptake behaviour, and comparing the performance with literature values. The chapter evaluates whether the initial hypotheses are supported, analyzes the advantages and limitations of the carbon aerogel for CO₂ capture, and proposes plausible explanations for the observed trends in the data.

- Chapter 5 – Conclusions and Future Work: Summarizes the key conclusions of the research, highlighting to what extent the carbon aerogel met the expectations as an effective CO₂ sorbent. The chapter also discusses the implications of the results for carbon capture applications and provides recommendations for future work. Possible extensions of the research and remaining challenges are identified to guide subsequent investigations.

By following this structure, the thesis logically progresses from fundamental background, through experimental work, to analysis and conclusions. The goal is to thoroughly document how the study was conducted and to convincingly address the research questions posed, thereby contributing new knowledge on the use of carbon aerogels in CO₂ capture.

2 Background

2.1 Global Climate Crisis and the Imperative of CO₂ Mitigation

Industrial activity since the Industrial Revolution has caused a huge rise in heavy pollution, mainly from burning fossil fuels like coal, oil, and natural gas. Reducing atmospheric CO₂ levels is now one of the world's most urgent environmental challenges. Research consistently identifies CO₂ as the primary human-made greenhouse gas driving global warming and climate change. This warming has direct, visible effects: steadily increasing global temperatures and more frequent, intense extreme weather. This includes heatwaves, droughts, blizzards, and heavy rainstorms [36]. Studies indicate human actions have already raised global temperatures by about 1°C compared to pre-industrial times, likely between 0.8°C and 1.2°C. By 2020, the average atmospheric CO₂ concentration reached 412.5 parts per million (ppm), the highest level ever recorded and roughly 50% higher than in the mid-1700s [37].

Unfortunately, ongoing global economic growth, fast industrialization, and rising populations mean CO₂ emissions will keep increasing. If current trends continue, global warming is expected to reach 1.5°C within 30 years [36]. The 2022 IPCC report warns that immediate action to reduce climate change is critical. They state greenhouse gas emissions must peak by 2025 at the latest, leading to a 43% cut by 2030. Without this, average global temperatures could rise 3.2°C and CO₂ levels could hit 570 ppm by 2100. Meeting the Paris Agreement goals requires major changes to energy systems. Industry, responsible for 25% of CO₂ emissions from energy and processes especially in cement, chemical, iron, and steel production, needs significant innovation. Key steps include wider use of electricity, better energy efficiency, careful control of fossil fuel use, and increasing renewable energy sources like hydrogen, biofuels, solar, and nuclear power. While these

long-term strategies are important, they can't quickly reduce humanity's carbon footprint alone. Cutting CO₂ from fossil fuels and fully developing clean energy technologies need large investments and considerable time [36-38].

Because of this, global efforts in recent decades have focused on developing ways to capture CO₂ directly from industrial sites before it enters the atmosphere. Carbon Capture, Utilization, and Storage (CCUS) are seen as especially vital for moving toward low-carbon energy. CCUS includes technologies like Carbon Capture and Storage (CCS), Carbon Capture and Utilization (CCU), and Bioenergy with CCS (BECCS). Within the CCUS process, capturing the CO₂ typically makes up 70–90% of the total operating costs. This makes capture technology a key research area for improving CCUS and reaching net-zero emissions. There are three main approaches to capturing CO₂, chosen based on how the CO₂ is produced, operating conditions, waste gas composition, and other process details: pre-combustion capture, post-combustion capture, and oxy-fuel combustion. Each method uses different CO₂ separation techniques, which are the core of capture. These techniques include absorption, membranes, cryogenic separation, chemical looping combustion (CLC), calcium looping (CaL), and adsorption [39].

2.2 Overview of CO₂ Capture Technologies

Carbon capture and storage (CCS) represent a critical pathway for mitigating greenhouse gas emissions. However, deploying CO₂ capture demands substantial supplementary infrastructure, leading to significant system costs and increased energy consumption. The principal capture methodologies are post-combustion, pre-combustion, and oxy-fuel combustion [40]. These differ fundamentally: pre-combustion involves high-pressure separation of CO₂/H₂ mixtures; post-

combustion primarily addresses low-pressure CO₂/N₂ separation; and oxy-fuel combustion focuses on CO₂/H₂O separation [41].

Post-combustion capture extracts CO₂ directly from flue gases. In contrast, pre-combustion integrates with advanced low-carbon systems like Integrated Gasification Combined Cycle (IGCC) plants using oxy-fuel combustion, where pure oxygen significantly reduces carbon output [42]. Capturing CO₂ from sources like ethanol plants (high concentration) or thermal power stations (low concentration) incurs considerable energy penalties. Integrating capture technology typically lowers plant thermal efficiency, though heat recovery and regeneration methods can partially offset this loss [43]. Global variations also exist in how CO₂ emissions correlate with economic growth across developed and developing regions [44].

2.2.1 Pre-combustion Capture

This approach captures carbon before conversion processes. It is particularly suited to IGCC plants. Here, fuel undergoes gasification under heat, pressure, steam, and controlled oxygen to produce syngas. Carbon is removed from this syngas prior to combustion and power generation. Operating conditions typically involve high pressure (2–7 MPa), temperatures of 200–400°C, and elevated CO₂ concentrations (15–60% vol., dry) [45]. These conditions facilitate separation, making pre-combustion attractive for fertilizer and hydrogen production. While IGCC plants without CCS have higher electricity costs than pulverized coal plants, analyses show natural gas-fired combined cycle plants with pre-combustion capture achieve around 80% CO₂ capture efficiency [45, 46]. Applications extend to coal gasification, ammonia synthesis, and gas separation. Key separation technologies include physical/chemical solvent absorption and adsorption using porous organic frameworks [47]. Emerging options include membranes, hydrate separation, cryogenics, sorbent-enhanced reactions, and membrane reactors [45]. In ammonia

production, CO₂ co-produced with hydrogen via steam reforming is typically purified using absorption techniques before synthesis, removing residual ammonia. Physical solvents offer lower regeneration energy penalties than chemical solvents, especially under high operating pressures. While porous organic framework membranes show promising CO₂ selectivity and uptake, their industrial deployment remains unrealized [47, 48].

2.2.2 Oxy-fuel Combustion

Oxy-fuel combustion stands out among CCS technologies due to its adaptability for retrofitting existing plants or integrating into new builds. Two primary implementation pathways exist: Air Separation Units (ASU) and Ion Transport Membrane Reactors (ITMR). ASUs separate oxygen from air cryogenically or via membranes, both energy-intensive processes, for conventional combustion. Conversely, ITMR systems combine oxygen separation and fuel combustion within a single unit [49]. Recent developments propose Oxygen Transport Membrane Reactors (OTMRs), where specialized membranes integrated into the combustor separate oxygen directly at operational temperatures (650–950°C). Membrane materials under investigation include lanthanum cobaltite perovskites, modified perovskites, brownmillerites, thin dual-phase membranes, dual-phase metal-carbonate membranes, and ceramic-metal composites. Future oxy-combustion concepts include syngas production in ITMs, oxy-combustion integrated power plants using Oxygen Transport Reactors (OTRs), liquid fuel combustion integration, and third-generation systems [50, 51].

2.2.3 Post-combustion Capture

Post-combustion is the most viable retrofit option for existing power infrastructure. Multiple separation techniques are applicable, including adsorption, absorption, cryogenics, and membranes [52]. These generally fall into biological, physical, or chemical categories. Biological

methods currently exhibit lower absorption capacity and efficiency compared to alternatives, limiting their use alongside established chemical absorption [53]. Physical absorption removes CO₂ from flue gas using organic solvents without chemical reaction. Membrane separation offers a potentially low-cost, low-energy solution but requires advances in permeability. Cryogenic capture and liquefaction present an innovative route, producing liquid or solid CO₂ [54]. Chemical methods encompass adsorption, absorption, and chemical looping. Among these, chemical absorption, particularly mature amine scrubbing, offers high efficiency and relatively low costs [54, 55]. A key energy consideration for coal plants is the ~0.11 MWh per metric ton of CO₂ required to compress captured gas to pipeline pressure (150 bar). Economic optimization is crucial, weighing capture energy costs, capital expenditure, potential CO₂ leakage penalties, and rising carbon taxes [54, 56].

2.2.4 Direct Air Capture (DAC)

Direct air capture (DAC) is a key technological technique used in indoor and industrial decarbonization processes to remove carbon dioxide from the atmosphere. DAC uses sophisticated sorbents such metal-organic frameworks (MOFs), porous materials, and zeolites to remove CO₂ from ambient air. DAC systems are used in sustainable building design to further enhance indoor air quality control and green architecture concepts [57, 58].

The technological foundations of DAC trace back to mid-20th-century applications in submarines and spacecraft. Contemporary deployment has accelerated markedly, with approximately 130 facilities operational or under construction worldwide. Recent materials science advancements have refined sorbent selection, particularly organic-silica hybrid adsorbents whose temperature-dependent adsorption/desorption dynamics have been characterized extensively [58, 59]. The core objective remains concentrating dilute atmospheric CO₂ (~400 ppm) into purified streams suitable

for sequestration or utilization [60]. Commercial-scale DAC implementation uses two pathways: absorption using alkaline solutions and adsorption via amine-functionalized solid sorbents [61].

A fundamental operational constraint emerges from atmospheric dilution: capturing 1 kg of CO₂ necessitates processing approximately 1,300 m³ of air. This volumetric inefficiency drives substantial capital and operational expenditures, with current costs ranging from 150–1,500 CAD per metric ton of CO₂ removed [62]. Consequently, converting captured CO₂ into value-added commodities, such as carbon fibers, polymers, and carbonated concrete, has emerged as an economically critical pathway to offset capture expenses through integrated processing systems [62, 63].

2.3 Adsorption Process of CO₂

CO₂ adsorption is fundamentally a surface process determined by the material's physicochemical properties. This phenomenon involves distinct interactions between gas molecules and the adsorbent, categorized as either physisorption or chemisorption. Both pathways release thermal energy, confirming their exothermic nature [64].

2.3.1 Physisorption

Physisorption relies primarily on van der Waals forces and electrostatic interactions to bind CO₂ molecules within adsorbent pores. The associated adsorption enthalpy typically falls between -25 and -40 kJ/mol, values comparable to the heat of sublimation [65]. Due to this mechanism's reversibility, CO₂ can be released under controlled conditions by varying the pressure or temperature. At ambient conditions, physisorption dominates capture performance with gas uptake directly linked to pore structure [66]. The appropriate pore size varies greatly with operating

temperature and pressure, but micropores with a diameter of 0.33–1 nm often exhibits the best adsorption capability [65, 67].

2.3.2 Chemisorption

Chemisorption addresses the selectivity constraints of purely physical adsorption. Here, basic functional groups are grafted onto porous materials to chemically react with acidic CO₂ molecules [68, 69]. This mechanism prevails above 140°C, though both adsorption types coexist between 25–140°C. Reaction enthalpies are stronger, ranging from -60 to -100 kJ/mol depending on the specific chemistry of the functional groups [69].

2.3.3 Regeneration Strategies

Regenerating adsorbents includes releasing captured CO₂ through several established methods: pressure swing adsorption (PSA), temperature swing adsorption (TSA), vacuum swing adsorption (VSA), combined pressure-vacuum swing adsorption (PVSA), and electric swing adsorption (ESA) [70, 71]. Selection depends critically on the adsorbent's chemical and structural characteristics. TSA and PSA, developed in the 1960s, remain widely used. TSA typically operates by adsorbing at ~40°C and regenerating via heating to ~120°C; its operational simplicity makes it the preferred method for amine-functionalized adsorbents. PSA obtains desorption by lowering system pressure, while VSA applies sub-atmospheric vacuum after high-pressure capture [68]. ESA modulates adsorption cycles through electrical current manipulation [70]. Common adsorbents like activated carbons, MOFs, zeolites, activated alumina, and silica gel are typically regenerated using TSA or PSA. Although ESA offers potential cost advantages over TSA and VSA, it is restricted to electrically conductive materials [71]. For pre-combustion carbon capture, cyclic PSA emerges as particularly promising since the process gas stream is already pressurized following conversion reactions [68, 70].

Temperature swing adsorption (TSA) is widely used because it is operationally simple, but it faces well-known practical limitations for CO₂ capture: (i) a large fraction of the energy input can be consumed as sensible heat to warm the contactor, reactor walls, and “dead” thermal mass rather than directly weakening CO₂–surface interactions; (ii) heating and cooling rates are often constrained by heat-transfer resistances in packed beds, leading to long cycle times and reduced productivity; and (iii) temperature gradients and non-uniform heating can cause incomplete regeneration and reduced cyclic working capacity, especially in structured or low-conductivity sorbents. These limitations motivate alternative regeneration approaches that deliver heat more directly to the sorbent, including electrically driven heating, microwave/induction approaches, and photothermal or solar-assisted regeneration.

A growing set of studies specifically explores photothermal or solar-driven swing adsorption for CO₂ capture, in which illumination is used to generate localized heating at the sorbent and drive desorption. Wu et al. proposed solar thermal swing adsorption (STSA) using porous carbon monoliths, where CO₂ is adsorbed during “light-off” periods and released during “light-on” periods due to photothermal heating; they report a CO₂ capture rate up to 0.226 kgCO₂ kgcarbon⁻¹ h⁻¹ from 20% CO₂/80% N₂ under 1,000 W m⁻² illumination, and note that combining STSA with vacuum swing adsorption can further increase working capacity. This work highlights how carbon-based structured sorbents can couple high CO₂ affinity (via ultramicroporosity) with efficient light-to-heat conversion [150].

Related work has also targeted direct air capture (DAC) using photothermal regeneration of chemisorptive sorbents. Kataoka et al. [151] developed an amine-modified silica/carbon aerogel containing carbon black as a photothermal additive; after capturing CO₂ from air, LED irradiation at 2.0 kW m⁻² increased the material temperature to about 70 °C, enabling CO₂ desorption, and

reported photothermal desorption efficiencies approaching ~80% after optimizing carbon black loading. More recently, Leick et al. demonstrated a “photo-swing” concept for DAC using branched polyethylenimine (PEI) combined with TiN light absorbers and low-power LEDs (irradiances up to 420 mW cm^{-2}), applied to 400 ppm CO_2 streams; they report robust cycling over >90 cycles and photo-driven desorption approaching ~83–100% of thermally driven desorption (depending on humidity and conditions) [152].

Across these studies, photothermal regeneration is consistently motivated by the same bottlenecks that limit conventional TSA: heat-transfer inefficiencies, unnecessary heating of inactive thermal mass, and cycle-time penalties. However, the literature also shows that performance is strongly architecture- and mechanism-dependent: carbon monolith STSA emphasizes structured sorbents and productivity metrics under flue-gas-like CO_2 concentrations, while many DAC-focused photothermal studies emphasize amine chemistry and desorption fractions under very dilute CO_2 concentrations. Importantly, relatively few studies report a unified characterization linking broadband optical absorption/emissivity, transient temperature fields (including gradients), and dynamic breakthrough-based CO_2 uptake/regeneration for the same monolithic material. This gap motivates the integrated optical–thermal–adsorption framework developed in the present thesis.

2.4 Adsorption Mechanisms in Solid Sorbent Materials

The most frequent method of CO_2 capture in solid sorbents is physisorption, a physical process governed by weak van der Waals interactions between the material's surface and CO_2 molecules. Physisorption is reversible and ideal for regeneration-based applications since it does not change the electronic structure of the adsorbed molecules, unlike chemisorption, which creates chemical bonds. The efficiency of this mechanism is controlled by several factors, including the material's

surface area, pore architecture, degree of functionalization, and operational variables such as temperature and pressure [72]. CO₂ interacts with the surface of the adsorbent at the molecular level via weak intermolecular interactions. Due to its linear nature and unique quadrupole moment, CO₂ is especially sensitive to surfaces that are polar or electron rich. In porous sorbents, adsorption often proceeds through a pore-filling mechanism, in which CO₂ molecules condense into narrow pores under capillary forces [73].

The adsorption capacity of porous materials depends on their structural and chemical features. A high surface area is one of the most important attributes since it directly offers more active sites for CO₂ interaction [74]. Materials such as activated carbon, metal–organic frameworks (MOFs), and zeolites are exceptionally effective, often exhibiting surface areas exceeding 100 m²/g. The pore size and distribution also significantly influence performance, pores that are close in size to the kinetic diameter of CO₂ (~3.3 Å) can capture molecules more efficiently by size matching [75]. Surface functional groups, such as hydroxyls and amines, can strengthen hydrogen bonds or dipole–quadrupole interactions to further enhance adsorption. Adsorption efficiency is also influenced by operating parameters like temperature and pressure. Lower temperatures promote adsorption because they decrease molecular mobility and prolong the time that CO₂ remains bonded to the surface since physisorption is exothermic. Meanwhile, according to isotherm models such as Langmuir and Freundlich, raising pressure improves absorption by driving more CO₂ into the pores [76].

The heat of adsorption provides insight into how strongly CO₂ interacts with the sorbent. For physisorption, the heat of adsorption ranges between 15 and 50 kJ/mol; lower than chemisorption energies but adequately strong for effective capture while still allowing easy regeneration. Sorbents containing polar groups or highly microporous structures tend to exhibit higher

adsorption heats, indicating stronger molecular attraction. Selectivity is an equally critical consideration, especially in processes that involve gas separation, where CO₂ must be distinguished from other gases such as methane (CH₄) or nitrogen (N₂). The quadrupole moment of CO₂ makes it particularly sensitive to polarized or electron-rich surfaces, thereby promoting selective adsorption. Controlling pore size can also enhance selectivity; materials with narrow micropores can exclude larger gas molecules, giving preference to CO₂. Chemical modifications such as grafting amines onto MOFs or silica surfaces further improve selectivity by enabling stronger dipole–quadrupole interactions and hydrogen bonding. In addition, nitrogen doping has been found to increase both adsorption heat and selectivity, as reported in previous studies [75–78].

2.5 Criteria for Selecting Solid Sorbents for CO₂ Capture

Solid sorbents have gained significant attention as efficient and scalable technologies for CO₂ capture, offering advantages in selectivity, energy efficiency, and reusability. Choosing the right sorbent is essential for developing a system that is both effective and economically viable. Because selection involves several interrelated factors, it requires a comprehensive assessment that accounts for the composition of the gas stream, pore size distribution, thermal properties, and structural stability. Several studies emphasize the importance of tailoring sorbent characteristics to specific applications while considering thermodynamic performance and suitable support materials [79, 80].

2.5.1 Sorbent Capacity and Affinity

A primary criterion in sorbent selection is its ability to adsorb and desorb CO₂ efficiently. This depends on the material's surface area, porosity, and chemical composition. A larger surface area

provides more adsorption sites, while specific surface functionalization can enhance CO₂ affinity. Additionally, there must be a balance between adsorption capacity and regeneration efficiency to ensure consistent performance across multiple cycles [81, 82].

2.5.2 Kinetics and Reaction Rate

Adsorption and desorption kinetics of CO₂ have a direct impact on a material's suitability for large-scale applications. To increase process efficiency, fast sorption kinetics shorten residence time and boost throughput. The pore size distribution, internal diffusion pathways, and the reactivity of surface functional groups all affect the reaction rate, and these factors should all be optimized in material design [83].

2.5.3 Selectivity and Impurity Tolerance

An effective sorbent must capture CO₂ selectively even in the presence of other gases and impurities typical of industrial flue streams. High selectivity ensures that the separated CO₂ is of sufficient purity while reducing the need for additional purification stages. However, moisture often interferes with adsorption, particularly in materials like activated carbon and zeolites; therefore, most flue gas systems incorporate a pre-drying step before CO₂ adsorption [79, 81].

2.5.4 Thermodynamic and Chemical Stability

Durability and stability under repeated adsorption–desorption cycles are essential for long-term operation. The sorbent must resist thermal or chemical degradation while maintaining its adsorption performance. In industrial systems, this means sustaining efficiency under continuous operation without structural breakdown. Additionally, adsorbents should remain stable against oxidation and decomposition during regeneration steps. Functionalized materials, such as amine-grafted sorbents, must also retain their active groups throughout repeated use to preserve capacity [84].

2.5.5 Cost-Effectiveness and Scalability

The overall economic feasibility of CO₂ capture depends largely on the cost and scalability of the chosen sorbent. Low-cost raw materials, simple synthesis routes, and environmentally friendly production processes help reduce expenses and improve sustainability. For widespread adoption, the synthesis technique must be adaptable to large-scale manufacturing without significant environmental impact [81, 85].

2.5.6 Regeneration Energy Requirements

Economic efficiency is mostly determined by the amount of energy used during regeneration. Sorbents that require minimal energy input for CO₂ desorption help lower operational costs. Using waste heat or renewable energy to assist regeneration can further improve system sustainability and energy performance [81, 86].

2.5.7 Cyclic Stability

Sorbent lifetime and cycle time strongly influence cost and practicality. Cycle time is the amount of time needed for a whole adsorption–regeneration process, whereas lifetime is the amount of time the material retains an acceptable adsorption capacity until replacement is required. Longer cycles and stable performance reduce the frequency of replacement, minimizing operational costs. Hence, optimizing working capacity, stability, and cycle parameters is essential for advancing next-generation sorbents [60].

2.5.8 Environmental concerns

A comprehensive life cycle assessment (LCA) is necessary to completely understand the sustainability and environmental impact of sorbent production. Every step of the synthesis process is examined in detail in this evaluation, including the sources of raw materials, energy consumption, and waste management. It takes into consideration not just emissions of CO₂ and other greenhouse gases, but also more general environmental problems like eutrophication and the

impact on freshwater ecosystems. By using life cycle assessment (LCA), researchers and developers can identify areas where environmental harm can be minimized, resulting in more sustainable and environmentally friendly CO₂ capture solutions [84].

2.6 Sorbent Materials for CO₂ Capture

2.6.1 Carbon-Based Sorbents

Carbon materials remain at the forefront of CO₂ capture research because of their versatility and availability. They occur in several forms; activated carbon (AC), carbon nanotubes (CNTs), and carbon molecular sieves (CMS) [87]. Among these options, activated carbon stands out as particularly practical. It combines strong thermal stability with low production costs and notable adsorption strength. One remarkable advantage is that it can be synthesized from waste biomass, including agricultural residues or food waste, making the process both cost-effective and sustainable. This dual benefit explains its widespread use across industries ranging from water purification to flue-gas treatment. Significantly, activated carbon's performance and texture can be adjusted. Specific surface area, pore volume, and pore distribution are all influenced by the synthesis parameters and the choice of precursor material [88]. In order to establish the best adsorption sites for CO₂ molecules, researchers frequently modify these characteristics. By introducing stronger dipole–quadrupole interactions that speed up CO₂ capture, further functionalization, especially with nitrogen-containing groups, can further boost adsorption efficiency.

2.6.2 Porous Polymers

Over the past several years, the study of Porous Organic Polymers (POPs) has expanded rapidly, driven by their strong potential for efficient CO₂ capture. Many researchers have focused on improving the low-pressure CO₂ adsorption performance of these materials. Among the different

kinds of POPs, those containing azo linkages are particularly interesting because of their nitrogen-rich frameworks. Patel et al introduced nitrogen-enriched nanoporous polymers, known as azo-linked covalent organic polymers (azo-COPs), that belong to this class [89]. In these structures, the azo ($-N=N-$) groups interact strongly with CO_2 through dipole–quadrupole forces, resulting in a CO_2 uptake of 3.4 mmol g^{-1} at 263 K and 1 bar. The same azo functionality also repels nitrogen molecules, giving the material its N_2 -phobic nature. This selective interaction provides azo-COPs with excellent CO_2/N_2 separation efficiency at higher temperatures, making them promising candidates for targeted gas separation applications. Since azo-functionalized Microporous Organic Polymers (azo-MOPs) are usually produced by oxidative polymerization of aromatic amine monomers, similar synthesis techniques have been used to create them. The intrinsic microporosity of these frameworks, along with the strong interaction between azo functional groups and the quadrupole moment of CO_2 molecules, are key factors in improving their CO_2 adsorption capacity, as was previously mentioned. Azo-MOP-2 demonstrated the highest CO_2 uptake of any material reported, at 3.1 mmol g^{-1} at 273 K and 1 bar. Additionally, the ALP-1–4 series, characterized by substantial surface areas ranging from 862 to $1235\text{ m}^2\text{ g}^{-1}$, demonstrated an impressive CO_2 capture capacity of 5.37 mmol g^{-1} at 273 K and 1 bar, along with a remarkable CO_2/N_2 selectivity of 43 under identical conditions [88-90].

2.6.3 Covalent Organic Frameworks (COFs)

Covalent Organic Frameworks (COFs) represent another fast-evolving class of porous crystalline materials. Constructed from light elements such as hydrogen, boron, carbon, nitrogen, and oxygen, these frameworks are formed via reversible covalent bonds that yield orderly two- or three-dimensional structures. Their chemical precision produces materials that are crystalline, low in density, and exceptionally porous. Many structures for adsorption applications have been

developed since Yaghi et al. published the first boron-based COF [91]. COF-6 had the maximum CO₂ uptake, measuring 85 cm³ g⁻¹ at 273 K and 1 bar, according to a comparison of COF-1, COF-5, COF-6, COF-8, COF-10, COF-102, and COF-103 [92]. It is interesting to note that this capacity did not scale directly with surface area, indicating that surface chemistry and pore geometry are more important factors than size alone. A later development by El-Kaderi and co-workers introduced TDCOF-5, synthesized from 1,4-benzene boronic acid and hexahydroxy triptycene. With a surface area of about 2497 m² g⁻¹ and mesopores averaging 2.6 nm, the material demonstrated strong adsorption performance, roughly 2.1 mmol g⁻¹ at 273 K and 1 bar, and a heat of adsorption of 21.8 kJ mol⁻¹. Their boron–oxygen bonds, which can partially hydrolyze in humid environments and reduce durability, are these materials' main drawback [92, 93].

2.6.4 Metal–Organic Frameworks (MOFs)

Metal–Organic Frameworks (MOFs) have been widely studied over the past decade for their potential in carbon capture, utilization, and storage (CCUS). They serve both as sorbent materials and as catalysts [94]. The outstanding CO₂ adsorption capacity of MOFs is largely attributed to their high porosity and the presence of active metal centers within the framework. Among thousands of reported MOFs, MOF-74 and its analogues stand out for their performance. Its general formula, M(dobdc), consists of a metal ion (M) coordinated to H₄dobdc (2,5-dihydroxy-1,4-benzenedicarboxylic acid). This coordination produces a three-dimensional framework with one-dimensional hexagonal channels aligned along the c-axis. During synthesis, solvent molecules occupy the sixth coordination site of the metal ion. Removing these solvent molecules through solvent exchange and heating exposes open metal sites, which can strongly interact with CO₂ via Lewis acid–base interactions, enhancing adsorption capacity [95]. Because CO₂ uptake depends on the type of metal, Long and co-workers investigated MOF-74 variants containing Mn, Fe, Mg,

Zn, Co, and Cu. CO₂ adsorption ranged from 3 to 9 mmol g⁻¹, with isosteric heats of adsorption (Q_{st}) spanning 22.1–43.5 kJ mol⁻¹. The Mg-based MOF-74 showed the highest uptake, around 9 mmol g⁻¹, with a Q_{st} of approximately 44 kJ mol⁻¹. Despite these advantages, open metal sites are sensitive to moisture. Water molecules compete for the same binding sites, reducing the MOF's efficiency under humid conditions [95, 96].

HKUST-1, a copper-based MOF with a face-centered cubic structure, has square pores of about 9 × 9 Å and a BET surface area near 850 m² g⁻¹. Its dense array of active metal sites makes it another promising candidate for CO₂ capture. Choi et al. [97] reported that HKUST-1 adsorbs roughly 3.3 mmol g⁻¹ of CO₂. To improve selectivity, researchers functionalized the framework with amines. Degassing HKUST-1 at 150 °C for 16 hours exposed the Cu²⁺ Lewis acid centers, which were then grafted with ethylenediamine (ED). This modification increased CO₂/N₂ selectivity, though the total adsorption slightly decreased due to reduced porosity. A similar effect was observed with tetraethylenepentamine (TEPA) grafting [98]. Interestingly, incorporating ionic liquids into the framework nearly doubled CO₂ uptake compared to the pristine MOF at 0.15 bar and 305 K [99]. MIL-101, with a surface area of around 2400 m² g⁻¹, has also been explored for CO₂ capture, achieving an uptake of 1.5 mmol g⁻¹. Lawson et al. [100] modified MIL-101 with polyethyleneimine (PEI), which increased adsorption despite significantly reducing surface area. A mixed-metal version, MIL-101(Cr, Mg), displayed a slightly larger surface area (2747 m² g⁻¹) and adsorbed 1.9 mmol g⁻¹ of CO₂. When functionalized with 10 % PEI, its uptake increased further to 3.1 mmol g⁻¹, demonstrating the beneficial impact of post-synthetic modification.

2.6.5 Zeolite-Based Sorbents

Zeolites are another widely studied class of porous materials for CO₂ adsorption. These microporous aluminosilicates exhibit versatile physicochemical properties along with excellent

ion-exchange capabilities [101]. Zeolite 13X has a BET surface area of approximately $591 \text{ m}^2 \text{ g}^{-1}$ and a pore volume of $0.250 \text{ cm}^3 \text{ g}^{-1}$. Its microporous structure allows it to adsorb CO_2 up to 6.9 mmol g^{-1} . Various modifications have been applied to improve zeolite-based CO_2 capture. For example, impregnating polyethyleneimine into zeolite cages introduces CO_2 -philic sites, enhancing selective adsorption from air [102]. Similarly, Kim et al. [103] functionalized Zeolite Y with ethylenediamine, achieving a CO_2 uptake of 1.1 mmol g^{-1} over twenty consecutive temperature-swing cycles, even under humid conditions. β -Zeolite has also been explored for CO_2 separation from gases such as CH_4 , O_2 , and N_2 . Henry's law was used to model CO_2 adsorption and assess separation performance at different temperatures. Li and Tezel [104] observed that separation factors for gas pairs like CO_2/CH_4 , CO_2/N_2 , N_2/O_2 , and CH_4/N_2 decreased with increasing temperature. Micropore diffusion was identified as the main pathway for mass transfer during adsorption. NaX-zeolite is another notable material, which can be synthesized from rice husk ash (RHA), an abundant agricultural by-product [103, 105]. It offers CO_2 adsorption performance comparable to commercial adsorbents while being more cost-effective due to its sustainable production. In a related study, Muriithi et al. prepared NaX-zeolite from coal fly ash. The environmentally friendly synthesis route makes this material particularly attractive for large-scale CO_2 capture applications [106].

The most promising options for CO_2 capture among the various classes of porous materials are porous carbons. Their appeal lies in a unique combination of properties: they are inexpensive to produce, exhibit strong thermal and chemical stability, and possess a large surface area with a tunable pore network. In addition, their hydrophobic nature and low regeneration energy make them particularly practical for repeated adsorption–desorption cycles. An additional advantage is their sustainable origin; porous carbons can be derived from a wide variety of renewable and waste

biomass sources [107, 108]. Over the past several years, considerable research has focused on improving the structure and performance of porous carbons for CO₂ capture applications [107]. These materials can be engineered in numerous morphologies and particle sizes, from the nanoscale to the microscale, with pore dimensions that can be adjusted from micropores (<2 nm) to mesopores (2–50 nm) and macropores (>50 nm). Additionally, it is possible to precisely control the orientation and distribution of these pores. Graphitization, chemical functionalization, and heteroatom doping are surface modification techniques that have been shown to be very successful in improving adsorption characteristics. To improve the practical performance of porous carbons for CO₂ separation and storage, careful consideration of both the pore structure and surface chemistry is therefore essential [108, 109]. The carbonization and activation conditions, as well as the synthesis parameters during preparation, can be changed to tune the adsorption behavior of these materials. Nitrogen doping has been found to strengthen the affinity between CO₂ molecules and the carbon surface, thereby improving the overall adsorption capacity [110]. Measurements at room temperature (25 °C) and atmospheric pressure (1 bar) are frequently used as standard references, even though CO₂ uptake is usually reported under a variety of temperature and pressure conditions. Since higher pressures and lower temperatures typically result in greater CO₂ adsorption, this enables direct comparison between various studies [111].

2.7 Aerogel Materials for CO₂ Adsorption

Aerogels are a unique class of three-dimensional (3D) porous materials that can be synthesized from organic, inorganic, or hybrid molecular precursors through a combined sol–gel, solvent-exchange, and drying process [21]. The concept of aerogels dates to 1932, when Kistler first produced an aerogel using a sol–gel technique followed by supercritical drying [112]. These materials can be categorized according to their physical form (such as monoliths, powders, or

films), pore structure (microporous, mesoporous, or hierarchical), or chemical composition (organic, inorganic, or hybrid) [2, 21]. Aerogels have attracted substantial attention for CO₂ capture because of their outstanding characteristics; extremely low density, high porosity, large surface area, and the flexibility of the sol–gel synthesis route, which allows precise tuning of surface chemistry [21]. Numerous types of aerogels have been developed for this purpose, including hybrid systems like metal–organic framework (MOF) aerogels [31, 113], organic aerogels based on polymers [114] or biomass-derived materials [115], and inorganic variations like silica aerogels [116]. However, the restricted capacity of pure aerogels to adsorb CO₂ limits their practical application. To overcome this restriction, scientists have added functional groups, particularly amines, to aerogel surfaces to increase the affinity of CO₂ at low temperatures [115–117].

Amine-functionalized solid adsorbents can be fabricated using several approaches: wet impregnation, post-synthesis grafting, and direct condensation [118]. A notable example was reported by Jiang et al. [26], who designed liquid-in-aerogel porous composites for CO₂ capture. In their system, the functional liquid tetraethylenepentamine (TEPA) was confined within the pores of a SiO₂ aerogel while retaining permanent porosity. The thickness of the restricted layer may be changed from 10.9 to 19.5 nm by varying the liquid loading. Strong interactions between the liquid and solid components, superior mechanical robustness, and thermal stability were all demonstrated by this composite. It achieved an impressive CO₂ uptake of 5.44 mmol g⁻¹ at 75 °C under a 15 vol% CO₂ stream, along with rapid sorption kinetics and high amine utilization. Moreover, it exhibited long-term cycling stability and remarkable CO₂/N₂ selectivity, with a separation factor of 1182.68 at 1% humidity. [2, 21, 115, 119]. In general, amine-functionalized adsorbents consist of a porous support material integrated with amine groups. These supports, often derived from inorganic or organic precursors, provide the necessary structural framework and surface area.

Biopolymer-based supports such as cellulose, chitosan, and proteins have gained particular interest due to their sustainable source, biocompatibility, and low toxicity [23, 120]. Such materials show strong potential for direct air capture (DAC) applications, achieving CO₂ adsorption capacities of 44–66 mg CO₂ g⁻¹ under humid and low-concentration conditions (400–500 ppm CO₂). They also offer improved energy efficiency, requiring 20–30% less regeneration energy compared with conventional aqueous amine systems [23, 121]. By adding amine groups to their pore surfaces, aerogels' affinity for CO₂ can be further increased. Nitrogen atoms in amine groups function as Lewis bases, giving lone-pair electrons to CO₂ molecules to create interactions because CO₂ acts as a weak Lewis acid that may receive an electron pair. By generating dipole–quadrupole interactions between the CO₂ molecules and the polar nitrogen functions, this electron donation significantly raises the aerogel's total adsorption capacity [120–122].

2.7.1 Carbon Aerogel

Carbon aerogels are a distinct class of emerging porous carbon materials, typically produced by carbonizing organic aerogels that originate from polymeric or biomass-based precursors. Because of this synthesis route, they combine the advantageous characteristics of both porous carbons and aerogels. Thus, carbon aerogels have a high surface area, a three-dimensional interconnected network, uniform pore distribution, exceptional thermal and chemical stability, a very low density, and a controlled porosity structure. They are very versatile for a range of uses, such as gas adsorption, energy storage, catalysis, and thermal insulation, thanks to their distinct structural and physical characteristics [123]. Among these applications, CO₂ capture has become one of the most actively explored areas for carbon aerogels. Compared with other porous carbon materials, such as activated carbons, carbon aerogels offer several important advantages. Their low density, well-

defined and uniform pore structure, great mechanical properties, and superior cyclic stability make them particularly suitable for practical CO₂ adsorption systems [4, 123].

From a deployment perspective, carbon aerogels offer several practical advantages beyond equilibrium uptake. Their hierarchical porosity can be engineered to balance capacity with mass-transfer rates, and their monolithic/structured forms have the potential to reduce pressure drop relative to fine powders while avoiding some palletization steps required for other sorbents. Carbon frameworks are also generally chemically and thermally robust under typical post-combustion capture temperatures, and carbonaceous adsorbents are often reported to be less sensitive to humidity than many strongly hydrophilic adsorbents (e.g., certain zeolites), which is beneficial for real flue-gas operation. However, there are also important limitations affecting feasibility and scalability. Many carbon aerogels are intrinsically fragile/brittle, which can severely complicate machining, handling, and integration into industrial contactors without fracture or attrition. In addition, conventional resorcinol–formaldehyde-derived carbon aerogels often rely on solvent exchange and supercritical or freeze-drying, which can be energy- and time-intensive, and the use of resorcinol and long processing cycles can raise production cost—factors that have been repeatedly highlighted as barriers to large-scale manufacture [4, 125].

Accordingly, recent work has focused on pathways to improve manufacturability and robustness, such as ambient-pressure drying routes, fiber reinforcement/composites, bead/particle forms, and additive-manufacturing approaches that enable larger parts and tailored geometries. These strategies can significantly improve mechanical integrity and scalability but may introduce trade-offs, which must be evaluated alongside CO₂ working capacity and regeneration energy in process-relevant tests [123, 125].

2.7.2 Synthesis of Carbon Aerogels

The synthesis of carbon aerogels typically involves three key stages: gelation, drying, and carbonization [123]. As shown in Fig. 1a organic gels are first prepared through polymerization and crosslinking of molecular precursors such as resorcinol and aldehydes. Afterward, solvent exchange and supercritical CO₂ drying yield the organic aerogel, which is subsequently carbonized to produce the final carbon structure [124]. Among these, the gelation stage plays a decisive role in determining the structural and chemical characteristics of the resulting aerogel.

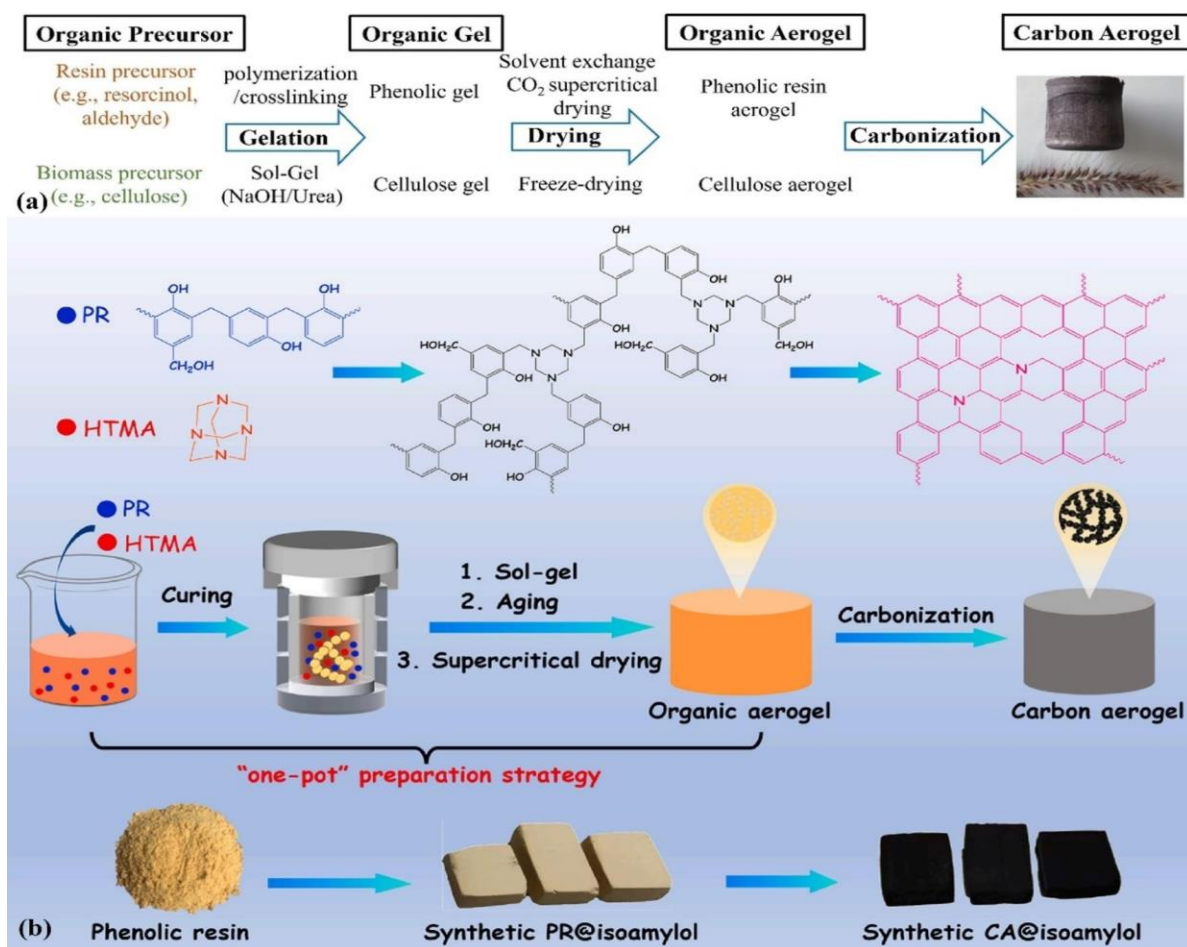


Figure 2.1.(a) Overview of the general synthesis route for carbon aerogels produced from organic precursors [116], and (b) schematic representation of the preparation process for phenolic resin-based aerogels and their corresponding carbon aerogels [126].

2.7.3 Gelation

The gelation process forms a three-dimensional polymer network through a sequence of addition and condensation reactions. Hydroxymethyl groups derived from aldehydes react with hydroxyl groups, leading to the formation of methylene and methylene-ether linkages that build the gel framework [127]. Both alkaline and acidic catalysts, such as Na_2CO_3 or HCl , are employed to facilitate nucleophilic and electrophilic reactions, respectively [128]. Beyond the classical resorcinol–formaldehyde system, alternative combinations such as melamine–formaldehyde and phenol–furfural have also been explored [129]. To enhance pore uniformity and structural integrity, templating agents including polymers, inorganic salts, or nanoparticles can be incorporated during polymerization [130, 131]. Biomass-derived precursors have recently attracted growing attention due to their abundance, sustainability, and environmental compatibility [132]. Interestingly, hydrated biomass sources (e.g., bacterial cellulose or watermelon) can directly yield aerogels without the conventional gelation step, as their intrinsic moisture and cellular porosity allow for the formation of 3D networks through sublimation [133].

2.7.4 Drying

Drying is a crucial step that removes solvents from the hydrogel while maintaining the porous framework. Common drying methods include supercritical, freeze, and ambient drying [134]. Supercritical drying produces highly uniform aerogels by minimizing surface tension and preventing pore collapse [135]. For instance, Barim et al. [136] achieved uniform RF aerogels using supercritical CO_2 drying at 138 bar and 50 °C. Freeze-drying offers a simpler and more cost-effective alternative, where frozen solvent sublimates under low pressure, avoiding capillary stress [137]. The freezing rate and precursor concentration strongly influence the final pore size distribution. Ambient drying, while scalable and inexpensive, often causes structural shrinkage due

to capillary forces [138]. To mitigate this, solvent exchange with low-surface-tension liquids or chemical crosslinking is applied to strengthen the framework [139, 140]. However, such modifications can introduce excessive solvent usage and environmental concerns, highlighting the need for greener and more efficient drying strategies [141].

2.7.5 Carbonization

The final carbonization stage transforms the organic aerogel into a carbon-based network through pyrolysis under an inert atmosphere (typically N₂ or Ar) at 500–2500 °C. This process eliminates volatile elements and develops a robust, conductive, and porous carbon framework [142]. The carbonization temperature significantly influences the material's texture: heating above 1200 °C may reduce surface area due to densification, whereas higher temperatures (>2000 °C) lead to graphitization and enhanced conductivity [143]. Additional activation treatments; using CO₂, steam, or alkaline agents like KOH can further tailor microporosity and surface area [144]. Despite the advantages, conventional sol–gel and supercritical methods remain energy-intensive and time-consuming [145]. Recent advances such as solvent-tuning and “one-pot” synthesis approaches have shown promise for producing lightweight, high-porosity carbon aerogels more rapidly and cost-effectively (Fig. 1b) [126, 146, 147].

Given the pressing global need for carbon neutrality, porous carbon materials, particularly carbon aerogels, have received growing attention due to their remarkable mechanical strength, thermal and chemical stability, and high CO₂ adsorption capacity, along with efficient regeneration capability. The preparation of carbon aerogels typically involves three main steps: sol–gel polymerization of molecular precursors, drying of the resulting organic gels, and subsequent carbonization. Carbonization plays a crucial role in forming a porous carbon network with large specific surface area and well-defined pore structures. Furthermore, through appropriate

modification and activation, these carbon aerogels can be tailored to function as high-performance CO₂ sorbents [4].

In the subsequent chapters, the focus will shift toward the methodology, results, and discussion of this study. The methodology chapter will describe in detail the materials, experimental procedures, and characterization techniques used to evaluate carbon aerogels for CO₂ adsorption. This will be followed by a presentation and analysis of the experimental results, where the optical, thermal, and adsorption properties of the materials are examined. Finally, the discussion section will interpret these findings in the context of existing literature, highlighting the key factors influencing CO₂ capture performance and the potential pathways for future research and material optimization.

3 Experimental Methodology

To fulfill the research objectives, the prefabricated carbon aerogel was characterized using multiple analytical techniques. The aerogel samples were supplied as monolithic discs measuring 14 mm in diameter and 4 mm in thickness. The material was sourced from Aerogel Technologies, LLC (Boston, MA, USA), sold under the general product name Carbon Aerogel (CAS 7440-44-0) as listed in the manufacturer's Safety Data Sheet. Each disc was handled carefully with clean gloves to avoid surface contamination. In this chapter, the instrumentation and procedures used for FTIR spectroscopy, UV–Vis–NIR spectroscopy, infrared thermal imaging, and CO₂ adsorption testing are described in detail to ensure reproducibility and clarity for future studies.

3.1 Sample Description and Preparation

The test material consisted of black carbon aerogel discs (14 mm Ø × 4 mm thick) synthesized through a sol–gel process followed by carbonization. Each disc was a rigid, lightweight solid with a bulk density of approximately 0.2 g/cm³. No additional chemical treatment or surface modification was applied, and the samples were tested as received to represent the baseline properties of the carbon aerogel. For all characterizations, the intact discs were used directly. Measurements were performed under ambient laboratory conditions (~22 °C, 1 atm) unless otherwise stated. Because the carbon aerogel discs were tested as received with no added functional groups (e.g., amines), CO₂ capture in this study is expected to occur primarily via physisorption within the micro/ultramicroporous network. In porous carbons, physisorption is governed by van der Waals and electrostatic interactions and is therefore reversible with temperature/pressure changes. By contrast, chemisorption in aerogel-based sorbents typically

arises when reactive surface functionalities (most commonly amines) are introduced, enabling carbamate/bicarbonate formation; such chemical functionalization was not applied in the present work.

3.2 Fourier Transform Infrared (FTIR) Spectroscopy

The optical characteristics of the carbon aerogel were analyzed in the infrared region using a Bruker Vertex 70 FTIR spectrometer equipped with a gold-coated diffuse reflectance integrating sphere and dual detectors. The measurements were conducted to determine the direct transmittance (T) and diffuse reflectance (R), from which absorption (A) was calculated according to the energy balance relationship:

$$R + A + T = 1 \quad (1)$$

Both transmittance and reflectance modes were measured, and each experiment was repeated three times to ensure consistency and reproducibility. Data acquisition and processing were performed using OPUS software, which facilitated high-precision spectral evaluation across both the mid-infrared (MIR, 2000–8000 nm) and near-infrared (NIR, 1000–2500 nm) ranges. The system employed a Deuterated Lanthanum- α -Alanine doped TriGlycine Sulfate (DLaTGS) detector for both spectral regions. A quartz beamsplitter was used for NIR measurements, while a potassium bromide (KBr) beamsplitter was utilized for MIR analysis. All experimental parameters were kept constant throughout testing to ensure comparability: a spectral resolution of 8 cm⁻¹, 1024 scans for both background and sample, and an 8 mm aperture diameter to achieve optimal balance between resolution and signal-to-noise ratio.

To enhance measurement stability and ensure accurate optical alignment within the integrating sphere, a custom 3D-printed PLA sample holder was designed and fabricated. The holder was modeled using CAD software and printed using a Prusa i3 MK3S+ printer. It was dimensioned specifically to fit the integrating-sphere port of the Bruker Vertex 70 and featured a precision-cut central window (4.0×4.0 cm) to accommodate standard aerogel disc dimensions ($14 \text{ mm} \times 4 \text{ mm}$). The design included a recessed seating area and securing edges that prevented sample movement during measurements while minimizing unwanted light scattering. This custom fixture ensured consistent optical path alignment, improved measurement repeatability, and prevented physical damage to the delicate aerogel surface.

The CAD rendering of the designed holder and a photograph of the FTIR spectrometer setup are presented in Figure 3.2, showing the overall system configuration used during optical characterization.

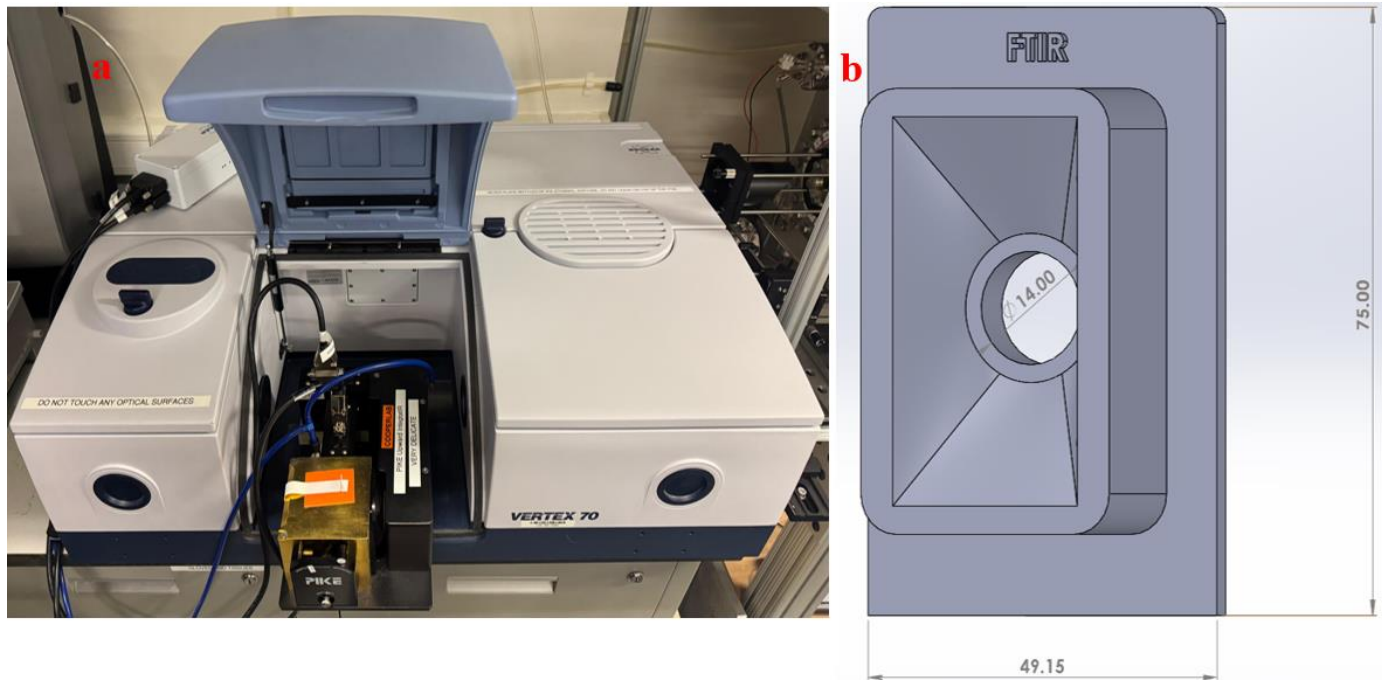


Figure 3.1.(a) photograph of the FTIR spectrometer setup used for optical characterization; (b) CAD model of the 3D-printed PLA sample holder designed for the Bruker Vertex 70 integrating sphere. Dimensions are shown in mm.

3.3 UV–Visible Spectroscopy

The optical behavior of the carbon aerogel; including its direct transmittance, hemispherical transmittance, and total relative reflectance (diffuse and specular) was characterized using the Shimadzu UV-2600 spectrophotometer. Measurements were conducted over a wavelength range of 190–1400 nm, encompassing the ultraviolet (UV), visible, and near-infrared (NIR) regions. Hemispherical transmittance and total reflectance were obtained using the ISR-2600 Plus integrating sphere attachment, which includes an external detector and enables measurements extending into the NIR region. Direct transmittance, in contrast, was measured without the integrating sphere, utilizing the internal detector of the spectrophotometer. To ensure proper

alignment and secure positioning of the aerogel throughout all measurements, a custom sample holder was designed and fabricated. A CAD representation of this holder is shown in Figure 3.

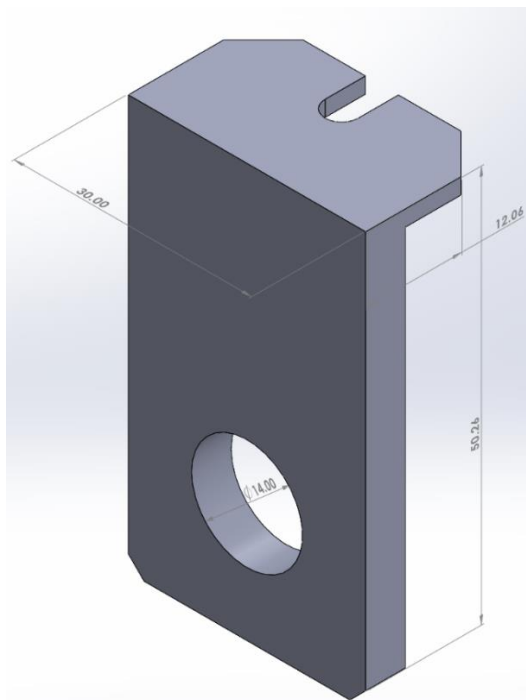


Figure 3.2. CAD model of the custom sample holder used for UV-Vis measurements. Dimensions are shown in mm.

To maintain consistency and reproducibility, the measurement procedure was standardized for all experiments. The sample was positioned parallel to the incident light beam, with the detector located directly opposite to collect transmitted light. Key optical settings including the 5 nm slit width, slow scan speed, and 2 nm sampling interval were kept constant to balance noise reduction with high-quality spectral resolution.

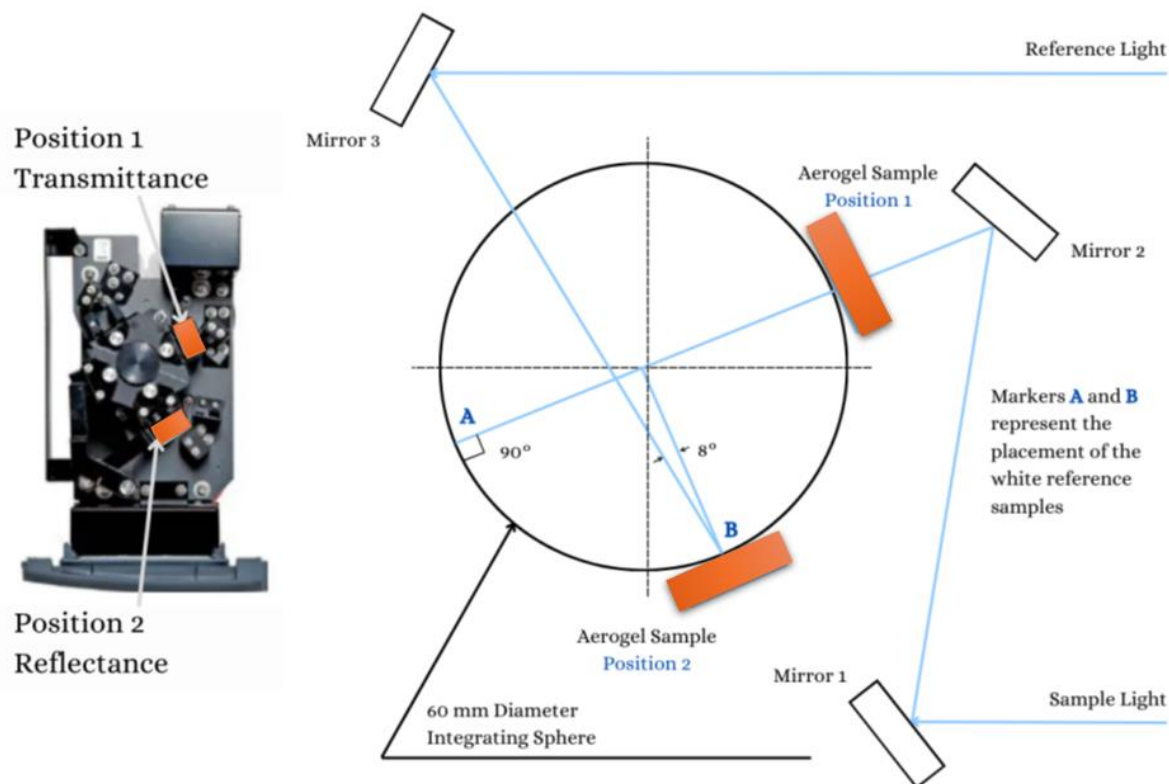


Figure 3.3. Schematic of the Shimadzu UV-2600 spectrophotometer with the ISR-2600 Plus integrating sphere attachment. The diagram shows the light path and the sample positions used during data acquisition

The sample/reference (S/R) exchange mode was adjusted depending on the measurement type: normal S/R for transmittance and inverse S/R for reflectance. Prior to each measurement, a baseline correction scan was performed over the full wavelength range. As indicated in Figure 4, two BaSO₄ white reference plates were required during baseline correction. For direct transmittance (DT), the baseline scan was first recorded without the sample, followed by the measurement with the carbon aerogel inserted. The same procedure was applied to hemispherical transmittance (HT). For total reflectance (TR), the baseline scan was performed with both reference plates in place; subsequently, the plate at Position 2 was replaced with the aerogel sample to record the reflectance spectrum. After each baseline correction scan, spectral data were collected

using UVProbe, the software integrated with the Shimadzu UV-2600 system, allowing real-time processing and organized storage of all acquired measurements.

3.4 Thermal Imaging Characterization (Infrared Thermography)

The photothermal effect refers to the process in which absorbed light is converted into heat within a material. When a material interacts with incident photons, their energy is dissipated as thermal energy, resulting in a measurable temperature increase. This behaviour is characteristic of light-absorbing materials and forms the basis of many light-driven heating applications.

In this work, photothermal heating was investigated using a 550 W xenon arc-lamp solar simulator (Sciencetech SF300-C). The lamp was mounted above the sample on a height-adjustable scissor lift, allowing precise control of the lamp–sample distance, as shown in Figure 5. The simulator generates a circular illumination spot approximately 5.1 cm (2 inches) in diameter and is powered by an adjustable 300 W supply.

The thermal response of the carbon aerogel was investigated using a FLIR A6700 MWIR thermal camera, sensitive to 3–5 μm radiation with < 0.1 K temperature sensitivity. The objective was to visualize the heating–cooling behaviour of the aerogel and obtain a reliable emissivity estimate. The aerogel disc was placed on a ceramic insulating plate. Thermal images were captured from two perspectives (top-down and lateral) with the camera positioned ~ 24 cm from the sample. The lateral imaging configuration and the laboratory setup of the xenon arc-lamp system are shown in Figure 6. To estimate emissivity, a piece of electrical tape ($\epsilon \approx 0.95$) was attached to the surface; after uniform heating to ~ 50 °C on a hot plate, the apparent temperatures of the tape and aerogel were nearly identical, indicating that the aerogel also exhibits high emissivity (~ 0.9 – 0.95). Thus, $\epsilon \approx 0.95$ was used in all subsequent thermal calculations.

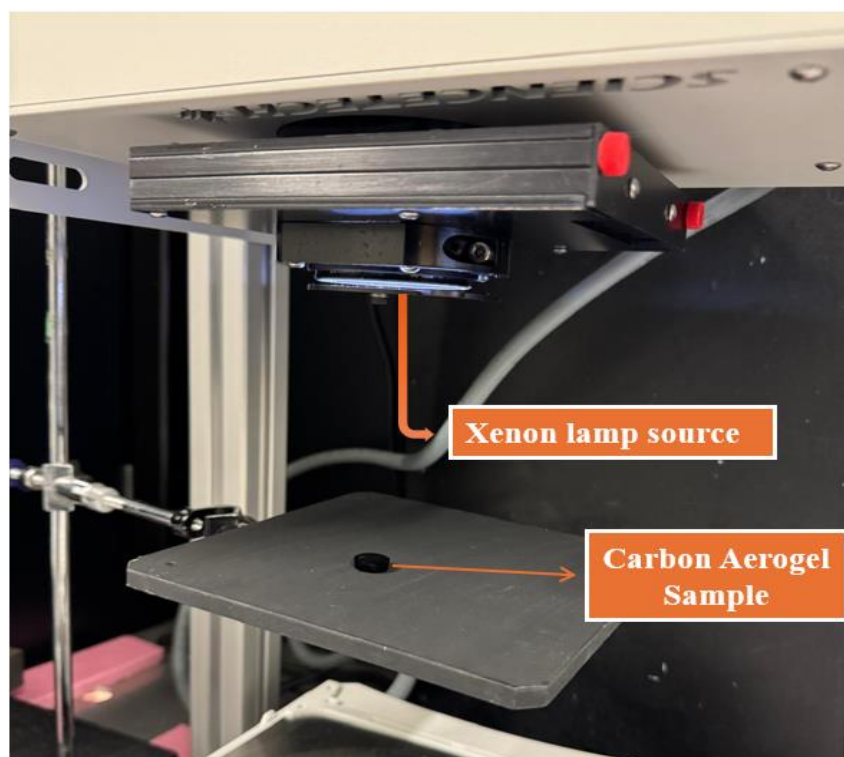


Figure 3.4. Adjustable xenon arc-lamp setup for photothermal heating of the carbon aerogel.

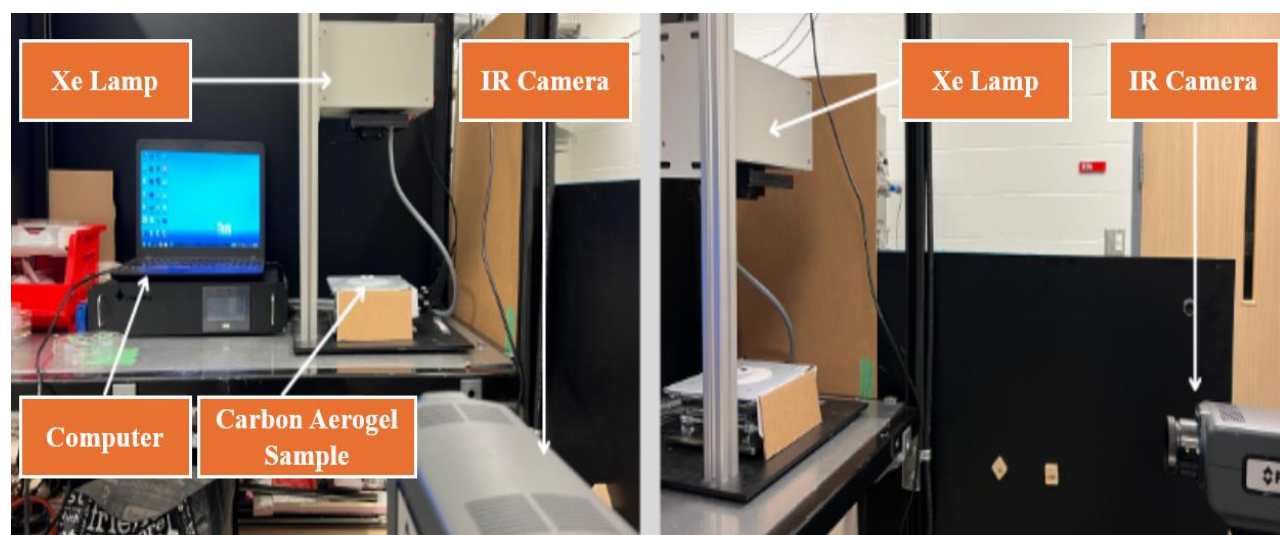


Figure 3.5. Lateral arrangement of the thermal camera and the laboratory setup of the xenon arc-lamp system.

Transient heating–cooling experiment: The aerogel, initially at room temperature ($\sim 25\text{ }^{\circ}\text{C}$), was exposed to continuous illumination from the incident solar-simulated light, and its thermal response was recorded at specific time intervals (1 s, 15 s, 30 s, 1, 2, 3, 4, 5, and 10 min). Under illumination, the material exhibited rapid photothermal heating and developed a uniform temperature distribution across the disc. After the light source was turned off, the surface temperature decreased gradually and uniformly, with no observable localized hotspots, indicating homogeneous thermal behavior throughout the structure.

Overall, thermal imaging confirmed that the carbon aerogel possesses high infrared absorptivity/emissivity and extremely low thermal conductivity, enabling rapid energy uptake while retaining heat for extended durations. These characteristics are highly desirable for temperature-swing processes that require fast heating and controlled cooling cycles. All measurements were recorded as thermal image sequences (.seq files) to extract spatially resolved temperature–time profiles.

3.5 CO₂ Adsorption Testing

The CO₂ adsorption performance of the carbon aerogel was evaluated using a custom-built continuous-flow adsorption/desorption breakthrough measurement apparatus designed to provide precise control of gas composition, flow rate, and temperature. A premixed stream of 11% CO₂ and 89% N₂ was delivered using a Masterflex mass flow controller (MFC), Model 32907-59 (Cole-Parmer, USA), operating at a total flow rate of 111 SCCM. This flow rate was selected to ensure stable residence time within the reactor while preventing internal mass-transfer limitations.

Post-combustion flue gas is the combustion exhaust stream and is typically at ~ 1 atm. Its dry CO₂ level is fuel-dependent (commonly ~ 10 – 15 vol% for coal and ~ 3 – 5 vol% for natural gas). Real flue gas also contains H₂O (humidity), residual O₂, and trace impurities (e.g., NO_x/SO_x), which can

affect adsorption kinetics/capacity and long-term stability. In this thesis, breakthrough tests used a dry CO₂/N₂ surrogate (11% CO₂, balance N₂) at near-ambient conditions. Here, “flue-gas-like” refers only to matching the CO₂ partial pressure range, not the full flue-gas composition.

3.5.1 Sample Mounting and Holder Design

The carbon aerogel discs (14 mm diameter, 4 mm thickness) were placed inside a custom-fabricated stainless-steel sample holder designed specifically for this work. The holder was sealed using Teflon O-rings to prevent leakage and ensure accurate measurement of breakthrough and regeneration behavior. A photograph of the stainless-steel sample holder used for CO₂ adsorption testing is shown in Fig. 7a. A CAD rendering of the stainless-steel holder is provided in Fig. 7b, illustrating the flow channels, sealing configuration, and overall geometry. Fig. 7c presents the carbon aerogel discs positioned inside the assembled holder, demonstrating the actual experimental configuration.

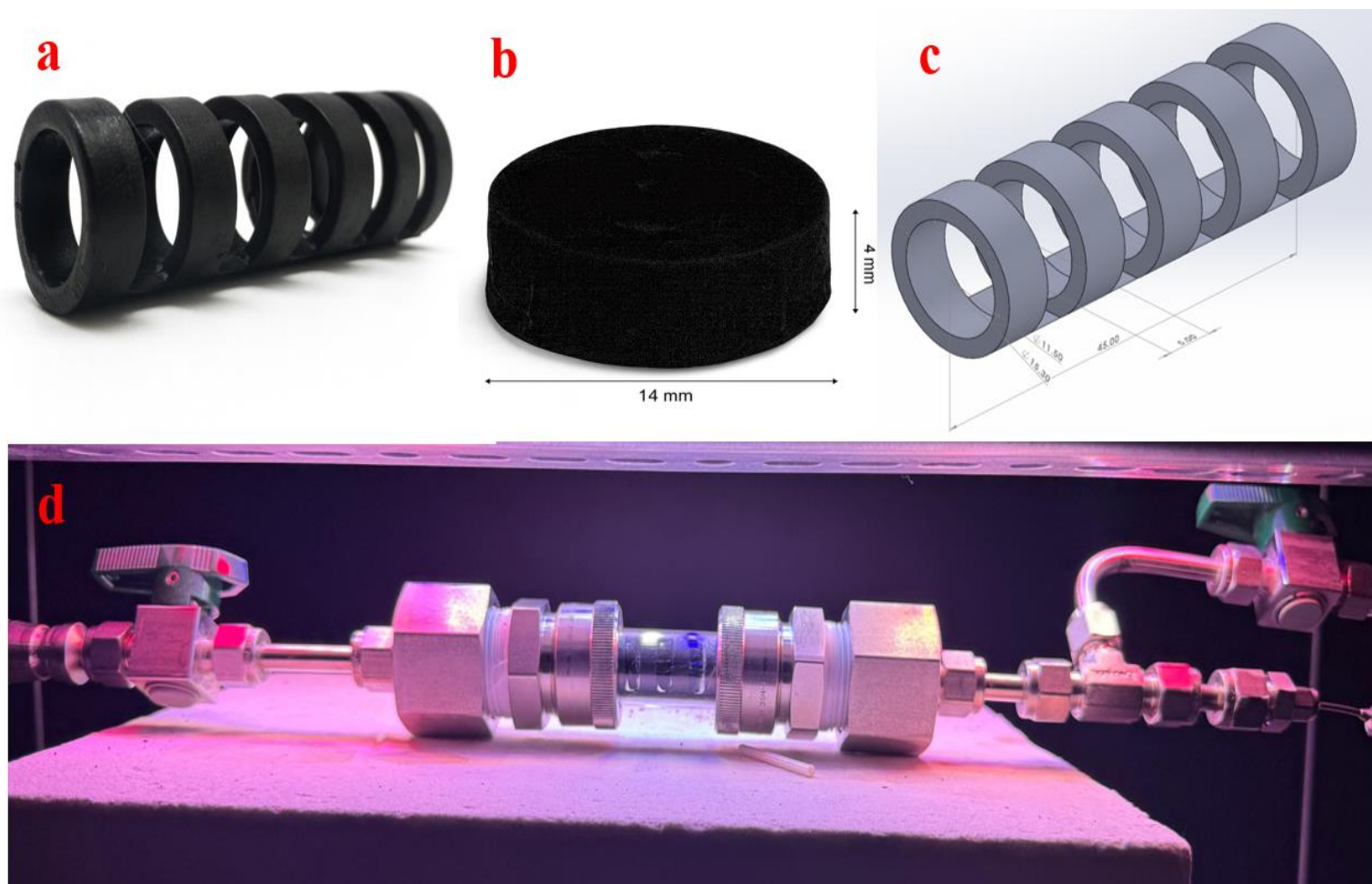


Figure 3.6. (a) Photograph of the stainless-steel sample holder. (b) Photograph of the carbon aerogel disc sample. (c) CAD model of the holder. (d) Photograph of the carbon aerogel placed inside the holder in the chamber. Dimensions are shown in mm.

3.5.2 Gas Monitoring and System Validation

The outlet gas composition was monitored in real time using an online CO₂ analyzer (Model 906; Quantek Instruments Inc., MA, USA), capable of detecting low concentrations with high stability. To verify system reproducibility and identify any instrument- or tubing-related contributions, each complete test sequence was repeated three times with the aerogel sample, followed by three blank tests using the empty sample holder.

3.5.3 Adsorption–Desorption Procedure

A full adsorption–desorption cycle consisted of four controlled stages, described below.

1. Cleaning and Pre-Regeneration

Before each adsorption test, the aerogel was regenerated by heating to approximately 110 °C using a high-intensity external light source. During this heating step, the system was purged with N₂ (111 SCCM) for 30 minutes to remove moisture and any pre-adsorbed CO₂. After heating, the light source was switched off, and the system was allowed to cool back to ~25 °C with the aid of a cooling fan to restore baseline temperature conditions.

2. Gas Stabilization

The CO₂/N₂ mixture was then introduced into the system, bypassing the sample holder. The gas was allowed to flow until the outlet concentration reached a stable value of **11% CO₂**, which was held for an additional 2 minutes. This ensured that the inlet conditions entering the sample remained constant throughout the experiment.

3. Adsorption Step

The valves were reconfigured to direct the gas mixture through the aerogel-filled sample holder. Upon contacting the carbon aerogel, the outlet CO₂ concentration sharply decreased from ~11% to ~0%, indicating rapid adsorption. As the material approached saturation, the concentration gradually returned to ~11%. To guarantee complete uptake, the flow was maintained for an additional 5 minutes after the return to baseline.

4. Photothermal Desorption (Regeneration)

The desorption step utilized photothermal heating, leveraging the strong light-absorbing properties of the carbon aerogel. The system was switched back to pure N₂ flow (111 SCCM) while a solar-simulated LED light source (SolarSystem 550; California Lightworks, USA) irradiated the aerogel surface. The illumination intensity was 2.3 kW/m², which is more than twice the AM1.5 solar spectrum (which has an intensity of 2.3 kW/m²) and is readily achievable with the LED system. The fixed-bed reactor was equipped with a transparent glass lid, allowing direct irradiation of the aerogel. Photothermal heating rapidly increased the sample temperature to ~110 °C, initiating CO₂ desorption.

During regeneration, the outlet CO₂ concentration initially increased as CO₂ was released from the aerogel, then decreased back to 0%, confirming complete desorption and full regeneration. The flow rate during desorption was kept identical to the adsorption step to ensure comparable flow dynamics. A photograph of the photothermal desorption setup with the solar-simulated LED light source is shown in Fig. 9, illustrating the illumination geometry and reactor configuration used for thermal regeneration.

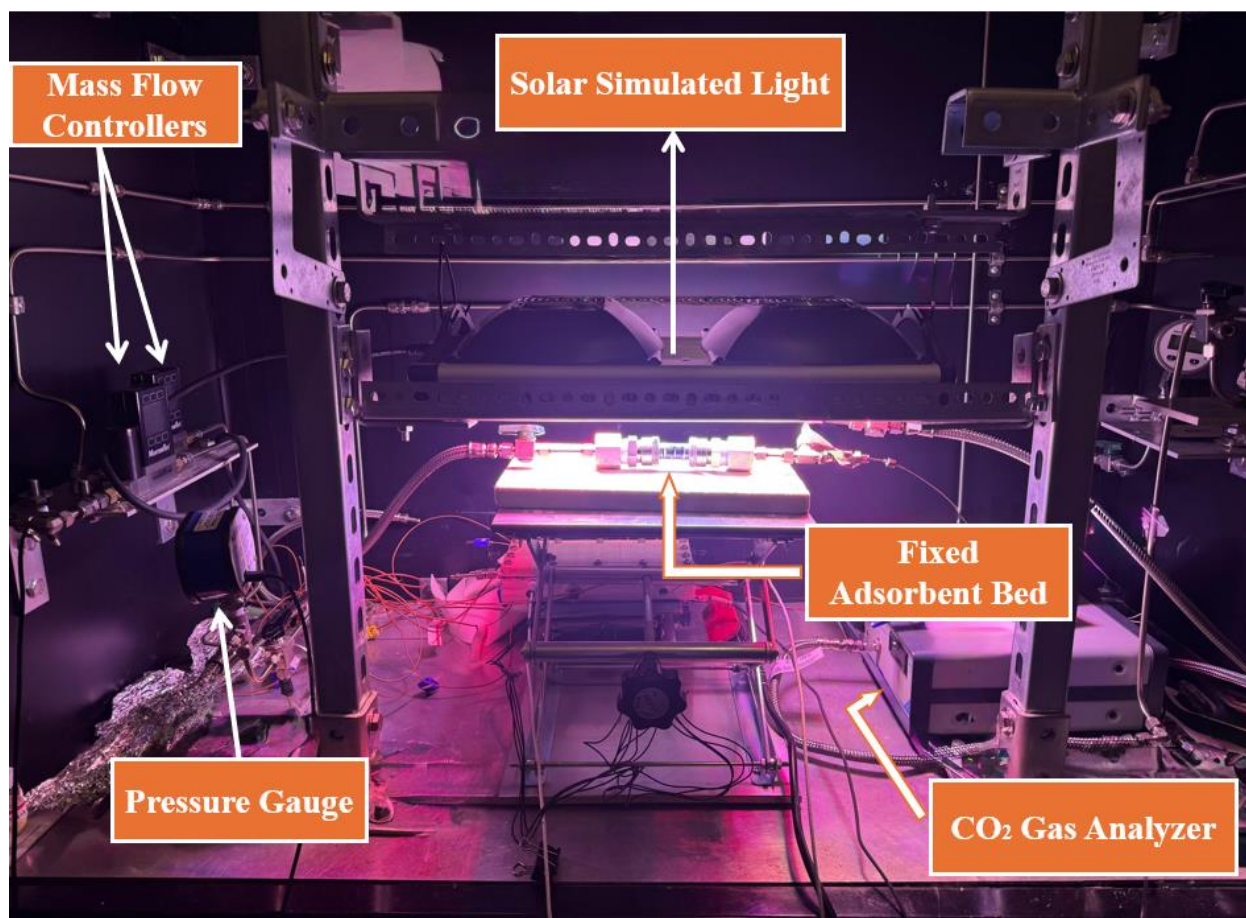


Figure 3.7. Photothermal CO₂ desorption setup using a solar-simulated light source.

4 Results and Discussion

4.1 UV-Vis Spectroscopy

The UV–Vis–NIR characterization provided clear evidence of the strong optical attenuation exhibited by the carbon aerogel. As shown in Figure 9, the transmittance remained effectively zero throughout the entire wavelength range investigated (220–1400 nm), demonstrating that incident radiation does not penetrate the 4 mm thick monolith. This behaviour reflects the combined effects of multiple internal scattering events and the intrinsic absorptive nature of carbon-based porous networks. Reflectance values were likewise low, remaining below approximately 5% across all wavelengths. Accordingly, the calculated absorptance consistently exceeded 95%, confirming that the aerogel functions as a broadband, near-ideal black absorber.

The recorded spectra did not display discrete absorption bands or features typically associated with well-defined molecular or crystalline electronic transitions. Instead, the spectral profile was smooth and featureless—an outcome characteristic of disordered carbon architectures composed of amorphous carbon regions interspersed with locally graphitic domains. In such materials, optical attenuation arises from a combination of π – π^* transitions, defect-state absorption, and free-carrier contributions, all of which span wide spectral ranges rather than producing narrow resonances. Comparable broadband absorption behaviour has been reported for various carbon-based aerogels, foams, and activated carbons, supporting the structural interpretation of the material used in this study.

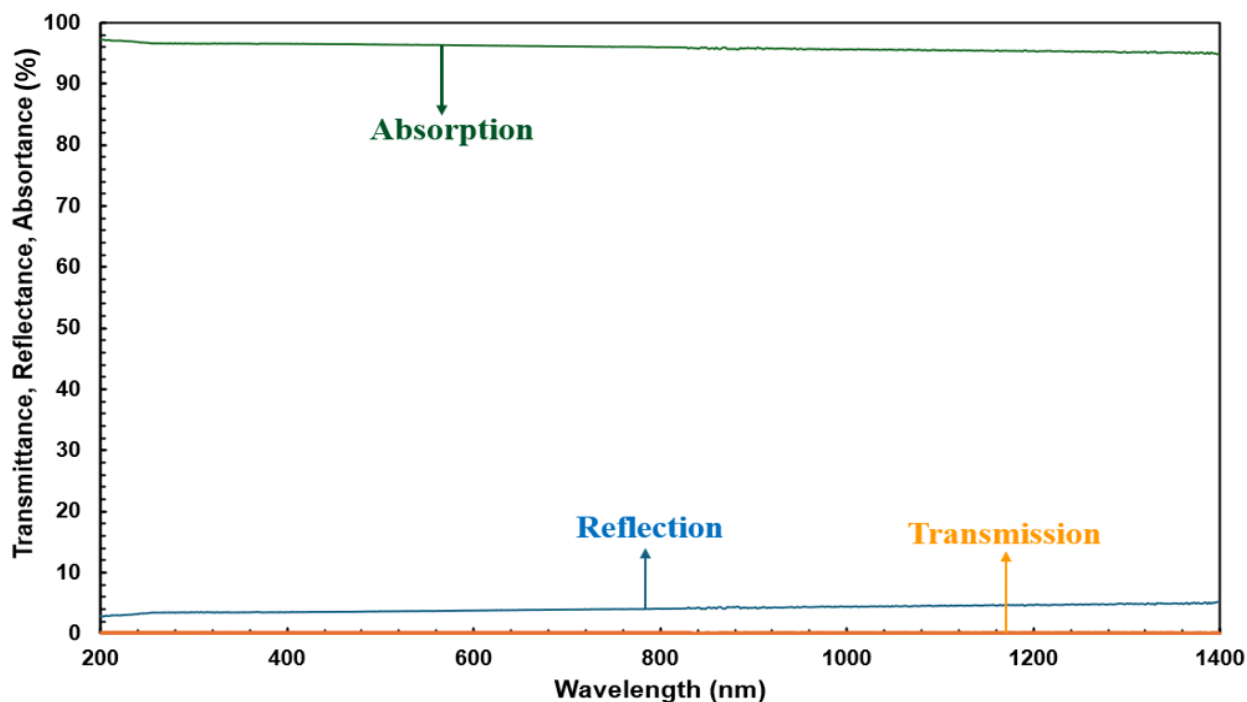


Figure 4.1. UV-Vis-NIR spectra of the carbon aerogel disc. The material shows negligible transmittance and very low reflectance (<5%) across 220–1400 nm, resulting in broadband absorptance above 95%.

This uniformly high absorptance has direct relevance for CO₂ capture applications. First, it indicates that the aerogel can efficiently convert incident ultraviolet, visible, and near-infrared radiation into thermal energy, enabling rapid photothermal heating even in relatively thin samples. Second, the absence of transmission and the low reflectance suggest that the absorbed energy is dissipated within the bulk of the porous network, promoting spatially uniform temperature elevation. Such characteristics are advantageous for temperature-swing adsorption (TSA) and photo-assisted regeneration processes, where rapid and homogeneous heating accelerates CO₂ desorption while minimizing thermal gradients that could otherwise impact structural integrity or reduce sorbent performance.

Overall, the UV-Vis-NIR results demonstrate that the carbon aerogel possesses a broadband, highly absorbing optical response that complements its high surface area and interconnected pore

network. These features collectively enhance its potential as an efficient and robust photothermal sorbent for next-generation CO₂ capture and regeneration technologies.

4.2 Fourier Transform Infrared (FTIR) Spectroscopy

FTIR measurements were performed on the disc-shaped carbon aerogel sample to quantify its wavelength-dependent reflectance, transmittance, and absorptance, with a focus on optical properties relevant to radiative heat transfer and infrared thermography. The investigated spectral window from 1 to 21 μm spans the upper near-infrared (NIR, $\sim 0.7\text{--}2.5\ \mu\text{m}$) and most of the mid-infrared (MIR, $\sim 2.5\text{--}25\ \mu\text{m}$) regions, thereby covering both solar-relevant wavelengths and those associated with thermal radiation near ambient temperatures. Figure 10 shows the measured spectral transmittance $T(\lambda)$ and reflectance $R(\lambda)$, together with the absorptance $A(\lambda)$. The absorptance was obtained from the measured quantities using:

$$A(\lambda) = 1 - R(\lambda) - T(\lambda) \quad (2)$$

The spectra are assumed to represent total (specular + diffuse) hemispherical values at near-normal incidence obtained with an integrating-sphere configuration. It is important to note that these optical spectra were obtained under ambient laboratory air conditions, so they primarily represent the effective response of the solid + air-filled pore network. In real post-combustion environments, the surrounding gas phase (and potentially the pore gas) differs substantially and includes CO₂ and H₂O, both of which are infrared-active and can absorb and emit radiation—most notably CO₂ near $\sim 4.3\ \mu\text{m}$ and $\sim 15\ \mu\text{m}$, and water vapor with strong bands including $\sim 2.7\ \mu\text{m}$ and $\sim 6.3\ \mu\text{m}$. Consequently, while the measured spectra remain appropriate for describing the intrinsic material optical behavior, radiative heat transfer in an operating adsorber can also depend on gas

composition, temperature, and optical path length through gas-phase participation. In addition, for FTIR measurements conducted without strict purge control, changes in ambient CO₂/H₂O between background and sample scans can introduce small atmospheric features, which is why purge/vacuum stabilization is commonly recommended in FTIR practice.

A second implication is that the optical characterization was not performed at controlled CO₂ loading states (e.g., after CO₂ saturation). In principle, replacing air in the pore space with CO₂-rich gas and/or accumulating adsorbed CO₂ could slightly alter the effective optical response by changing the effective refractive index of the pore medium and modifying scattering conditions; however, for the present 4-mm disc the material is already effectively opaque over most of the MIR ($T(\lambda)$ drops below $\sim 0.2\%$ for $\lambda \gtrsim 3.5 \mu\text{m}$), so any loading-dependent changes are expected to be second-order except potentially near strong CO₂/H₂O bands. Future work should therefore quantify any loading- or gas-composition dependence using in-situ optical methods under controlled atmospheres (e.g., FTIR diffuse reflectance/DRIFTS or emissivity measurements in an environmental cell) at defined CO₂ partial pressure and humidity.

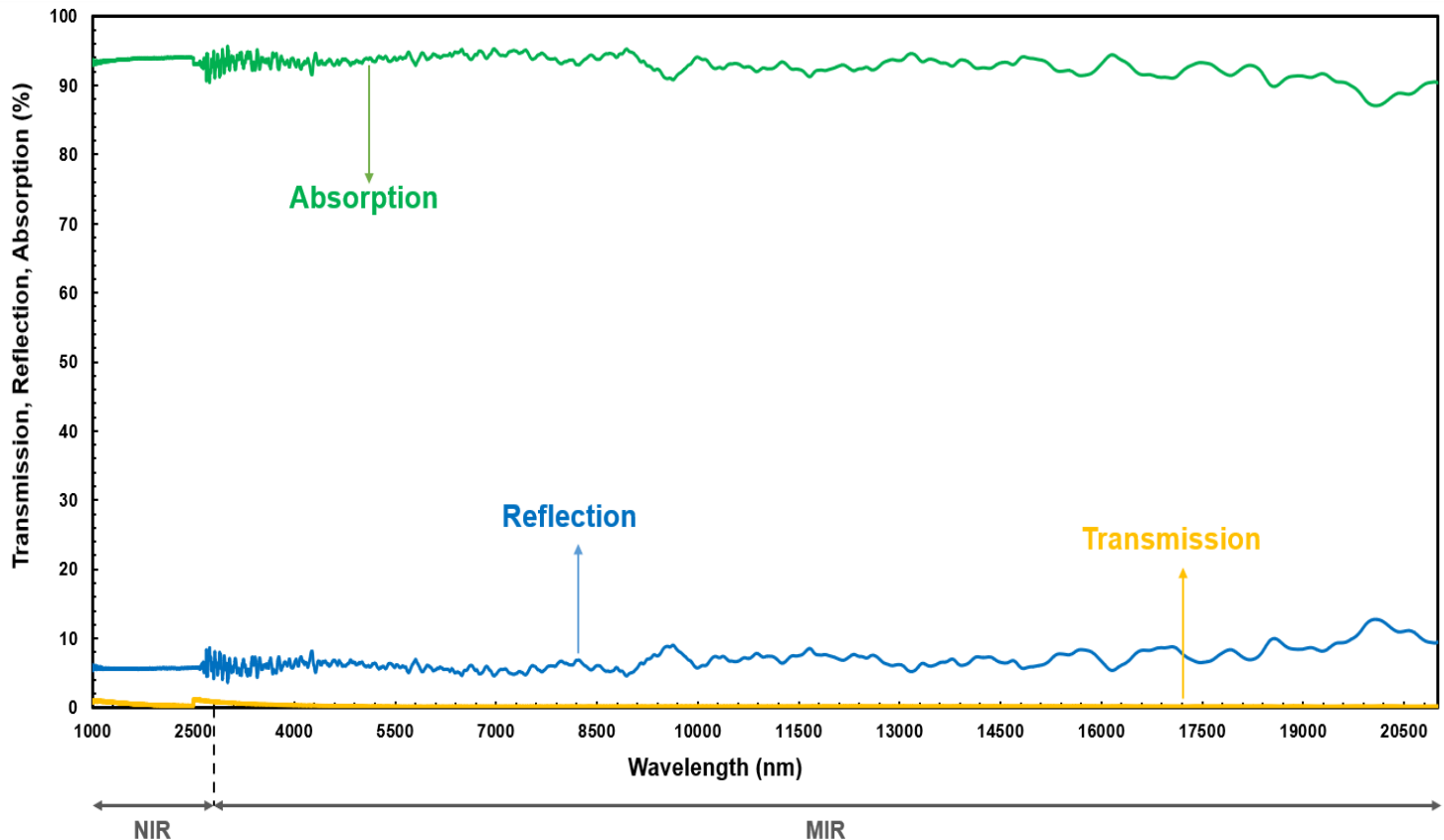


Figure 4.2. FTIR spectra of the carbon aerogel showing reflectance, transmittance, and absorbance across the 1–20.5 μm wavelength range.

Across the entire NIR–MIR range, the transmittance remains very small. In the short-wavelength region (1–2.5 μm), $T(\lambda)$ is approximately 0.8–1.3 %, and it drops below ~ 0.2 % for $\lambda \gtrsim 3.5$ μm . The 4-mm carbon aerogel disc can therefore be treated as effectively opaque over most of the measured range. The reflectance is also relatively low but exhibits some spectral structure. In the NIR and short-wavelength MIR regions (1–7 μm), $R(\lambda)$ is typically 5–6 %, apart from a narrow region around 2.5–3.2 μm where interference fringes produce oscillations between roughly 4 and 9 %. In the central MIR region, a clear local maximum of about 8–9 % occurs near $\lambda \approx 9$ μm , followed by an undulating increase from ~ 6 –7 % to ~ 11 –12 % between 10 and 21 μm . Because $T(\lambda)$ is negligible beyond ~ 3.5 μm , the absorbance closely follows $A(\lambda) \approx 1 - R(\lambda)$. In

the 1–7 μm region and again from ~ 10 –21 μm , $A(\lambda)$ therefore remains high, generally in the range 0.88–0.95, with only a modest reduction to ~ 0.91 –0.92 in the vicinity of the reflectance maximum.

The measured response can be understood by considering both the porous microstructure of the aerogel and the intrinsic optical properties of carbon. Carbon aerogels consist of an open three-dimensional network of nanometre-scale carbon particles and ligaments with very high porosity (often $> 90\%$) [4, 12]. The pores are filled with air, so the effective refractive index of the bulk is only slightly higher than that of air, and the Fresnel reflection at the air–aerogel interface is modest. This explains the relatively low reflectance (generally below 10%) despite the visually “black” appearance of the material. Once incident radiation enters the aerogel, however, it undergoes intense multiple scattering within the tortuous pore network. The greatly increased optical path length through the strongly absorbing carbon skeleton drives the overall absorptance toward unity and suppresses transmission, producing the near opacity observed throughout the NIR and MIR. Similar light-trapping and ultra-black behaviour have been reported for carbon-aerogel-based absorbers and coatings and are regarded as key advantages of these materials for photothermal and thermal-management applications.

Because the carbon aerogel disc is effectively opaque ($T(\lambda) \approx 0$) for $\lambda \gtrsim 3.5\ \mu\text{m}$, Kirchhoff’s law of thermal radiation can be applied in its opaque form. The spectral emissivity $\varepsilon(\lambda)$ is then approximately equal to the spectral absorptance $\alpha(\lambda)$, which is in turn given by $A(\lambda)$:

$$\varepsilon(\lambda) \approx \alpha(\lambda) = A(\lambda) \approx 1 - R(\lambda) \quad (3)$$

Only in the short-wavelength NIR region (1–2.5 μm) does the difference between $1 - R(\lambda)$ and $1 - R(\lambda) - T(\lambda)$ reach ~ 0.01 –0.02. In the NIR, the measured reflectance implies $\varepsilon(\lambda) \approx 0.93$ –0.95. In the MIR, emissivity remains high overall: approximately 0.90–0.95 for 3–7 μm and 10–

21 μm , with a local minimum of $\sim 0.88\text{--}0.92$ around 9 μm corresponding to the reflectance peak. When averaged over the 7.5–13 μm band used by the thermal camera, the effective emissivity is therefore in the range $\varepsilon \approx 0.92 - 0.94$. These values are consistent with reported emissivities of carbon-based aerogels and related carbon materials [7, 34, 35], which typically fall in the range 0.7–1.0 in the 3–5 μm and 8–14 μm bands depending on density and composition, and can approach ~ 0.98 for optimized black coatings. For the present samples, an emissivity setting $\varepsilon_{\text{cam}} \approx 0.93 - 0.95$ in the IR camera is therefore well supported by the FTIR data and is used consistently in the thermal imaging experiments.

Overall, the FTIR analysis across the NIR and MIR ranges shows that the disc-shaped carbon aerogel behaves as a broadband, nearly opaque, high-emissivity absorber. This behaviour is a direct consequence of its porous carbon microstructure and the strong intrinsic absorption of carbon, and it underpins the rapid surface heating and efficient radiative exchange observed in the photothermal and CO_2 -capture experiments discussed in subsequent sections of this thesis.

4.3 Thermal Imaging Characterization

The photothermal behaviour of the carbon aerogel was investigated using a mid-wave infrared (MWIR) thermal imaging system while the sample was subjected to controlled radiative heating. A xenon arc lamp calibrated to 1 Sun (100 mW cm^{-2}) was employed as the illumination source, providing a continuous and spectrally broad irradiance profile comparable to the AM1.5 terrestrial solar spectrum. Thermal images were recorded from both top and lateral orientations to capture the surface response and the through-thickness temperature distribution within the aerogel disc. Representative temperature maps are shown in Figure 10, and the corresponding temperature–time profiles extracted from selected regions of interest are presented in Figure 11. These measurements

provide insight into the photothermal conversion efficiency, heat transport mechanisms, and transient thermal response of the material.

4.3.1 Heating Cycle

Upon exposure to incident light from the xenon-lamp, the carbon aerogel exhibited a rapid temperature rise consistent with strong radiative absorption and efficient photothermal conversion. From the top-view images, the surface temperature increased sharply from ambient ($\sim 28\text{ }^{\circ}\text{C}$) to approximately $58\text{ }^{\circ}\text{C}$ within the first 15 s. This fast response reflects the high broadband absorptance of the aerogel, as evidenced by the UV–Vis–NIR results in Section 4.1, in combination with its low thermal conductivity, which limits heat dissipation away from the illuminated region.

Following this initial transient, the surface temperature rose more gradually and reached a steady-state temperature of approximately $64\text{ }^{\circ}\text{C}$ within 1–2 min. Beyond this point, the temperature remained effectively constant over the 10 min heating period because the absorbed radiative power became balanced by the heat lost to the surroundings.

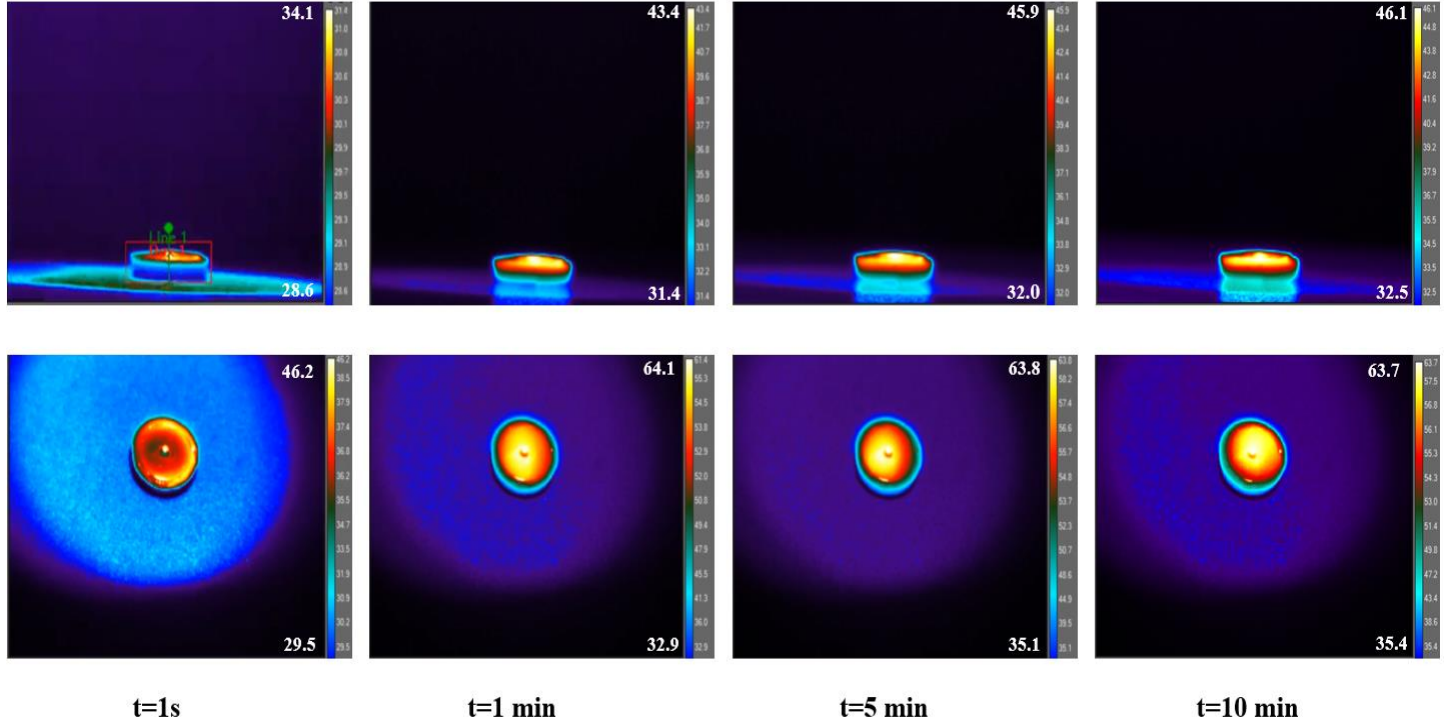


Figure 4.3. Thermal imaging results of the carbon aerogel during heating at 1 Sun irradiance, recorded at selected time intervals ($t = 1$ s, 1 min, 5 min, 10 min) from the top and lateral views.

In contrast, the lateral-view images revealed a lower temperature rise and a more moderate time response. The edge temperature increased from ~ 28 °C to ~ 40 °C within 15 s and subsequently approached a steady value of ~ 46 °C after roughly 2–3 min, which is about 18 °C lower than the illuminated surface. This pronounced surface–edge temperature gradient arises from the inherently directional nature of the radiative input: the top surface absorbs most of the incident radiation directly, whereas the edges receive only indirect heating through in-plane thermal conduction and diffuse scattering from the illuminated region. Additionally, the exposed edges lose heat more effectively to the surrounding air, further contributing to their lower temperature relative to the surface.

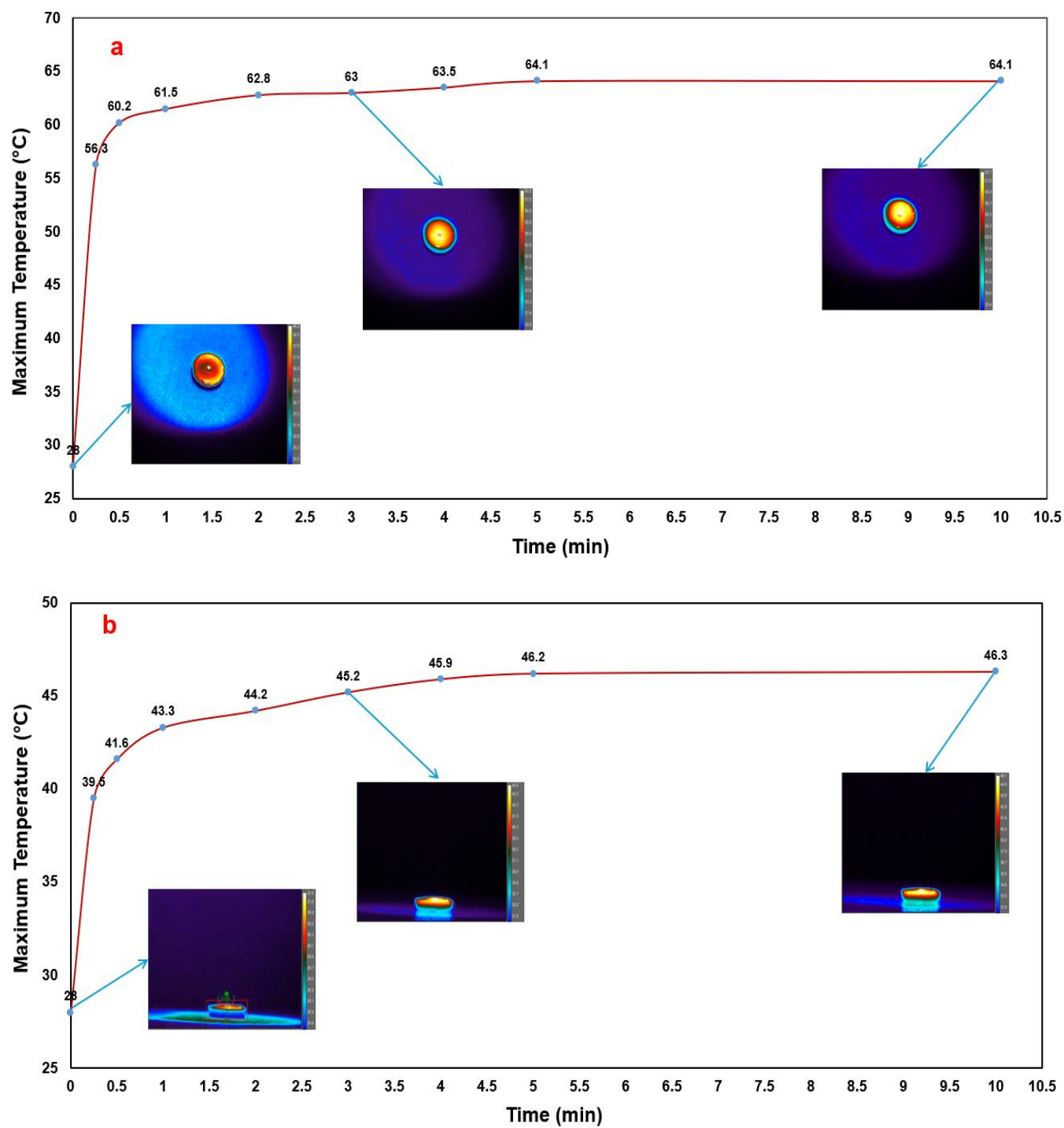


Figure 4.4. Temperature profile of the carbon aerogel during heating, showing maximum recorded values from the top (a) and lateral (b) surfaces.

The spatial distribution of temperature in the top-view images also revealed a slightly hotter central region, suggesting mild non-uniformities in the irradiance field or reflective focusing effects from nearby optical/mechanical components. In the lateral view, localized bright regions at the disc–holder interface confirmed that the holder acts as a secondary heat sink, promoting conductive heat transfer away from the aerogel. Overall, the heating cycle demonstrates that the carbon aerogel can be heated rapidly and efficiently under moderate radiative fluxes, while the maximum attainable temperature and its spatial distribution are strongly influenced by geometry, illumination configuration, and boundary conditions.

4.3.2 Cooling Cycle

The cooling behaviour of the aerogel was examined immediately after the xenon lamp was switched off. Representative thermal images at selected time intervals are shown in Figure 12, and the corresponding temperature–time curves are presented in Figure 13. From the top view, the surface temperature decreased from $\sim 64\text{ }^{\circ}\text{C}$ to $\sim 38\text{ }^{\circ}\text{C}$ within the first 15 s, followed by a gradual decline to $\sim 31\text{ }^{\circ}\text{C}$ over a 10 min interval, approaching the ambient laboratory temperature. The lateral surface cooled from $\sim 46\text{ }^{\circ}\text{C}$ to $\sim 33.5\text{ }^{\circ}\text{C}$ in 15 s and then to $\sim 30\text{ }^{\circ}\text{C}$ by the end of the same period.

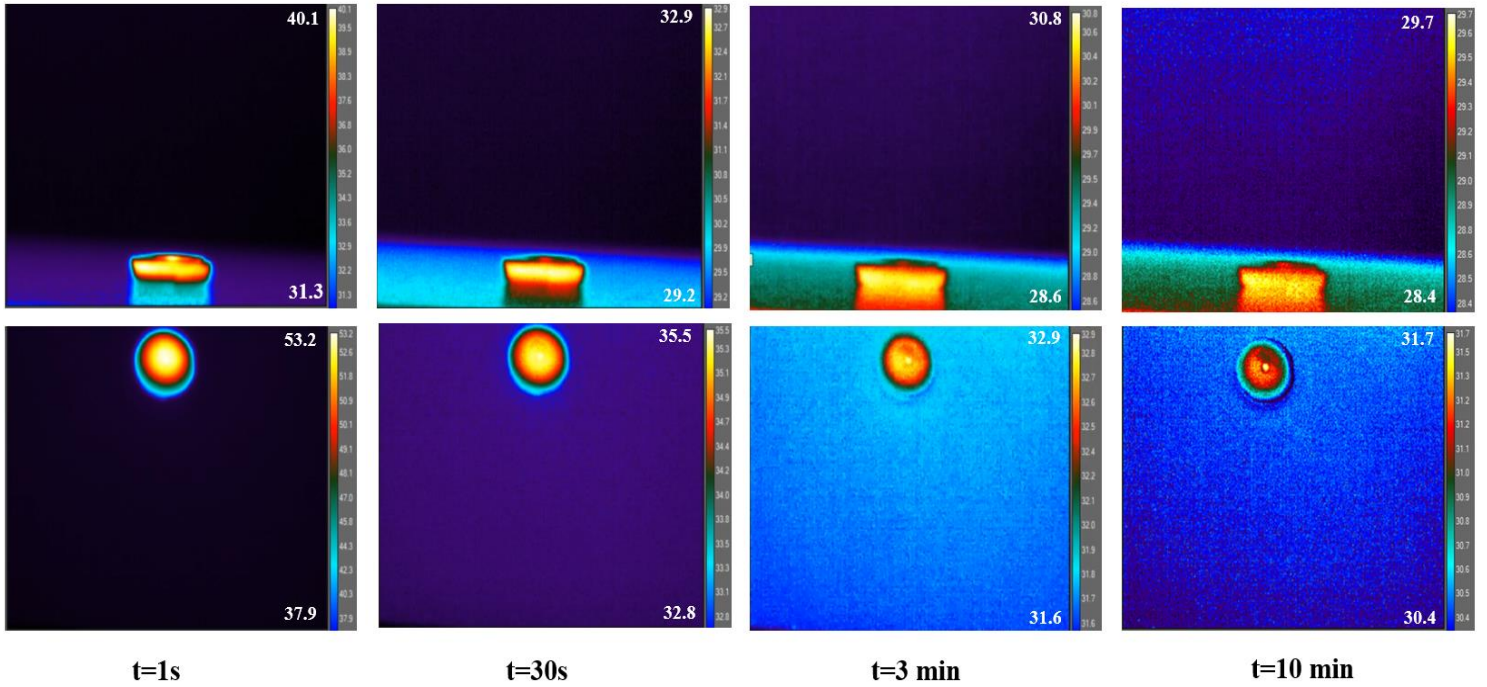


Figure 4.5. Thermal imaging results of the carbon aerogel during cooling following lamp switch-off, recorded at selected time intervals ($t = 1 \text{ s}$, 30 s , 3 min , 10 min) from the top and lateral views.

Both orientations exhibited a characteristic two-stage cooling response. The initial stage was dominated by relatively large temperature gradients between the aerogel and its surroundings, leading to rapid heat loss driven mainly by natural convection to the surrounding air and thermal radiation to the environment. In the subsequent stage, as the temperature difference decreased, the cooling rate slowed, and the system approached thermal equilibrium asymptotically. Conduction through the sample holder contributed to heat dissipation throughout the entire cooling process but became more comparable to convective and radiative losses at later times.

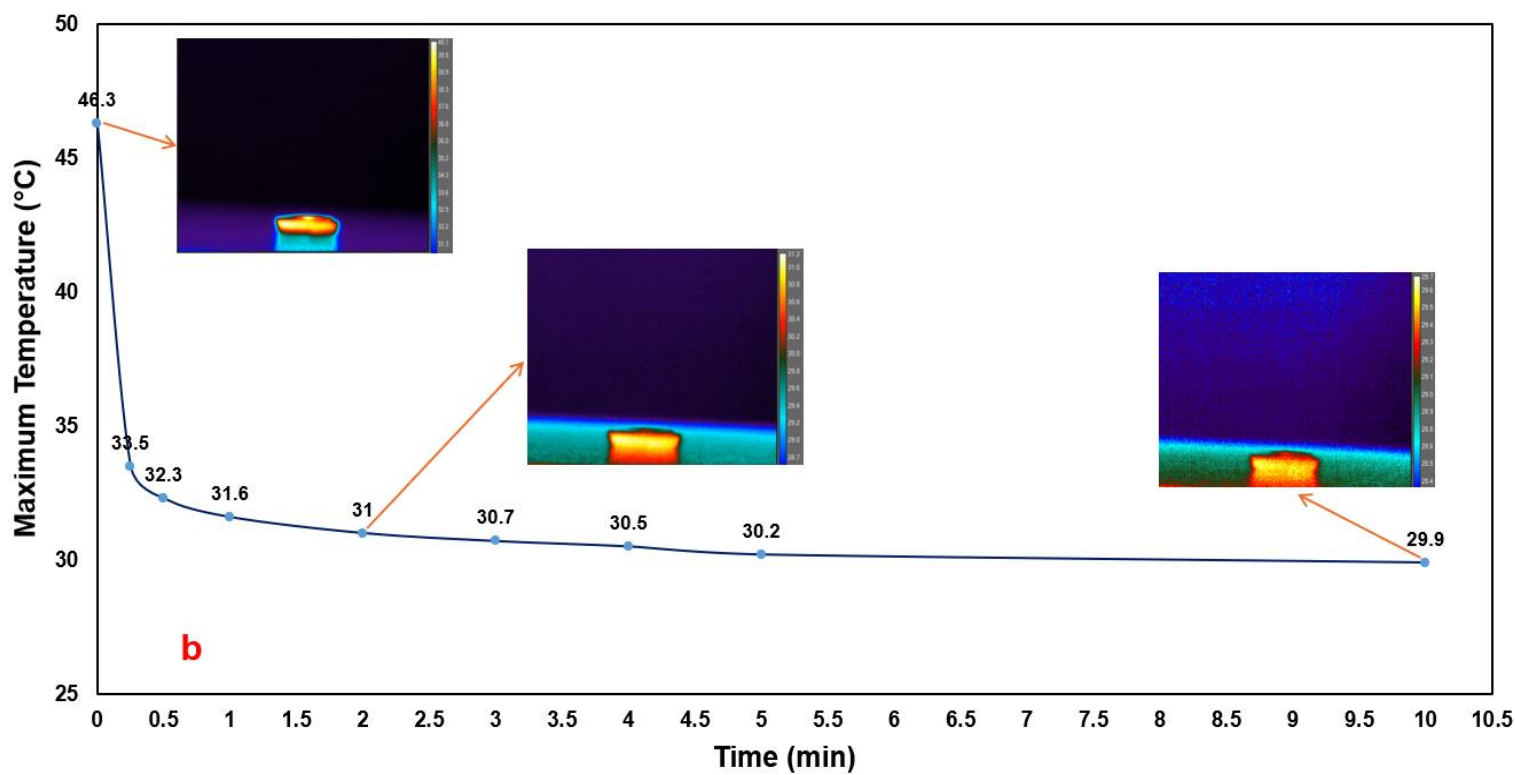
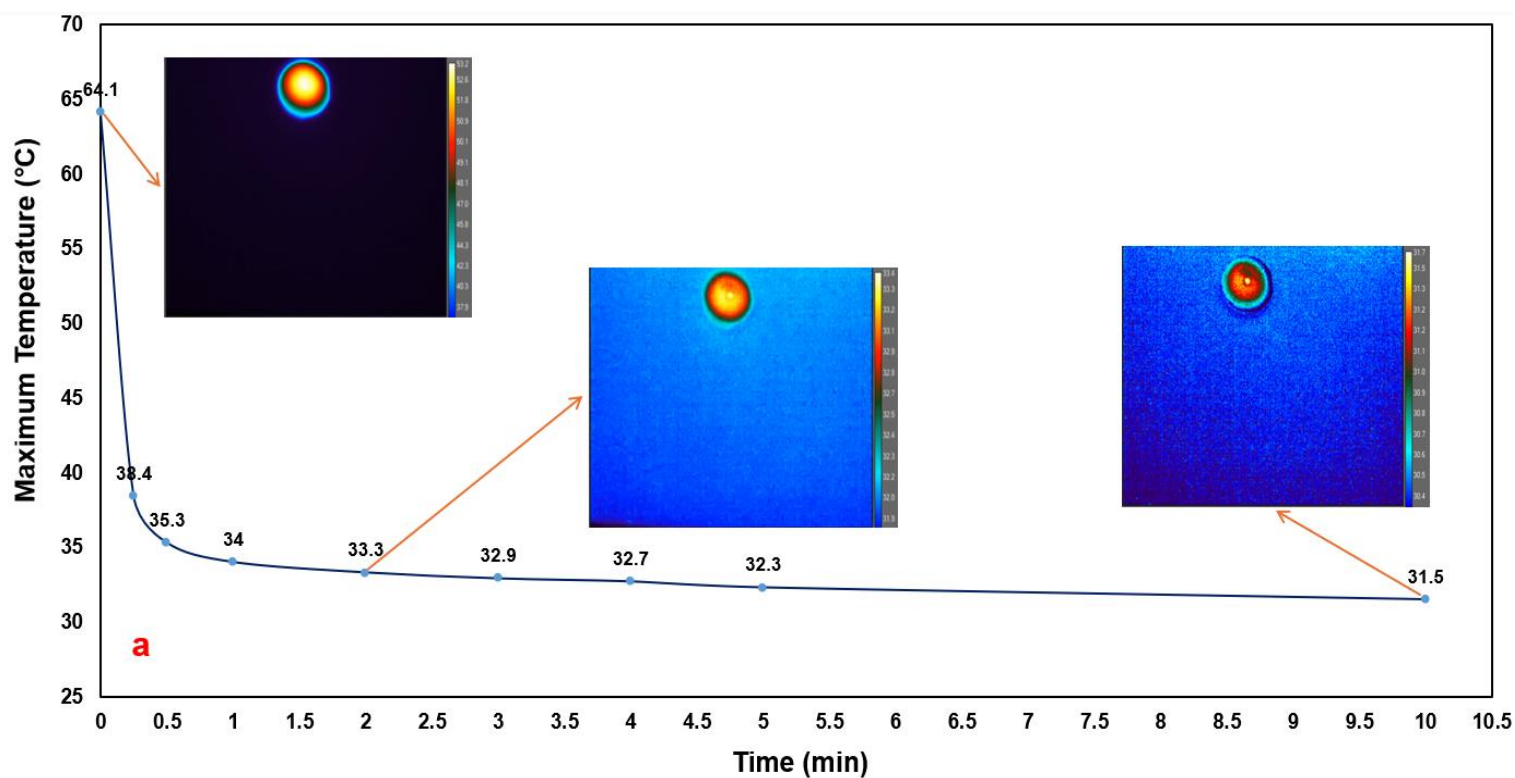


Figure 4.6. Temperature profile of the carbon aerogel during heating, showing maximum recorded values from the top (a) and lateral (b) surfaces

The close similarity between the top- and side-view cooling curves during the later portion of the transient suggests that, once the external illumination is removed, heat dissipation becomes more uniform and is governed primarily by the intrinsic thermal properties of the aerogel and the ambient conditions, rather than by the directional nature of the earlier radiative input. The relatively short characteristic cooling time is consistent with the ultralow density, high porosity, and low heat capacity of carbon aerogels, which limit thermal energy storage and favour rapid equilibration with the environment.

4.3.3 Implications for CO₂ Capture and Regeneration

The thermal imaging results have direct implications for the use of carbon aerogels in thermally driven CO₂ capture processes. The rapid temperature rise during illumination indicates that the material can efficiently convert incident solar-like or lamp-based radiation into heat, enabling fast attainment of desorption temperatures in temperature-swing or photothermal-swing adsorption cycles. The relatively low thermal mass and rapid cooling behaviour are likewise advantageous, as they allow quick transitions between adsorption (low-temperature) and desorption (high-temperature) steps, potentially reducing cycle times and improving process productivity.

The observed temperature gradients also highlight important design considerations for reactor and system integration. In practical configurations, optimization of sample geometry, illumination uniformity, thermal insulation, and contact with supporting structures will be necessary to minimize edge losses and to achieve more homogeneous temperature distributions during regeneration. Nonetheless, the combination of efficient photothermal response, rapid thermal cycling, and structural stability demonstrated here confirms that carbon aerogels possess favourable thermal characteristics for integration into solar-assisted or radiatively driven CO₂ desorption systems.

4.3.4 Comparison with photothermal CO₂ capture literature

The photothermal response measured here provides a direct basis for comparison with emerging photothermal and solar-assisted CO₂ capture studies. Under 1 Sun illumination, the carbon aerogel surface heats rapidly from ~28 °C to ~58 °C within ~15 s and reaches a steady-state near ~64 °C within 1–2 min, while lateral imaging shows the disc edge stabilizing near ~46 °C, producing a surface–edge gradient of ~18 °C. These observations are consistent with the conceptual foundation of solar-assisted swing capture: strong radiative absorption enables fast heating, while the thermal boundary conditions and geometry govern the attainable temperature and spatial uniformity.

Compared with carbon monolith STSA, the present work differs in both architecture and measurement emphasis. Wu et al. [150] demonstrate intermittent illumination applied to porous carbon monoliths to enable adsorption/desorption cycling and report a high capture rate under 1,000 W m⁻² illumination [151]. In contrast, the present study focuses on a commercial carbon aerogel disc and quantitatively links (i) broadband optical attenuation, (ii) transient thermal maps and gradients, and (iii) dynamic breakthrough-derived CO₂ uptake and photothermal regeneration. This provides complementary information that is not always available in STSA-focused studies: specifically, the measured temperature gradients identify a practical limitation for monolithic photothermal contactors—illumination can create non-uniform internal thermal driving forces that may limit regeneration completeness unless thickness, illumination geometry, or conductive pathways are optimized.

Relative to DAC-focused photothermal regeneration in aerogels and polymer sorbents, the present work also occupies a distinct operating regime. Kataoka et al. use an amine-modified silica/carbon aerogel with carbon black as a photothermal component and report CO₂ desorption after LED irradiation (2.0 kW m⁻²) to ~70 °C, achieving high desorption efficiency. Leick et al. demonstrate

photo-swing DAC using PEI with TiN photothermal absorbers and report strong photo-driven desorption relative to thermal desorption and stable cycling. In contrast, the present work investigates non-functionalized carbon aerogel under flue-gas-like CO₂ concentrations (12% CO₂/88% N₂) and uses photothermal heating to reach ~110 °C during regeneration [152].

This comparison highlights a key difference in mechanism and design: while many DAC photothermal systems rely on amine chemistry and added photothermal fillers, the carbon aerogel itself acts as an intrinsically strong absorber and thermally stable monolith. The tradeoff observed here is that partial regeneration can occur, consistent with diffusion limitations and strongly retained CO₂ in ultramicropores, implying that photothermal contactor design (thickness, illumination uniformity, and optional vacuum assistance) becomes critical to maximize cyclic working capacity.

4.4 CO₂ Adsorption Capacity

The CO₂ adsorption performance of the carbon aerogel was evaluated through dynamic breakthrough experiments using a gas mixture of 11% CO₂ and 88% N₂. Adsorption was conducted at 25 °C, followed by photothermal desorption at 110 °C under solar-simulated illumination. Representative breakthrough and desorption curves are shown in Figure 15, illustrating the characteristic behaviour of the inlet and outlet CO₂ concentrations during each stage. To ensure experimental reproducibility and statistical reliability, each test was repeated three times for both the blank holder and the aerogel-loaded holder. The integrated breakthrough volumes for adsorption and desorption from all six experiments are summarized in Table 1, and the corresponding standard deviations are reported in Table 2.

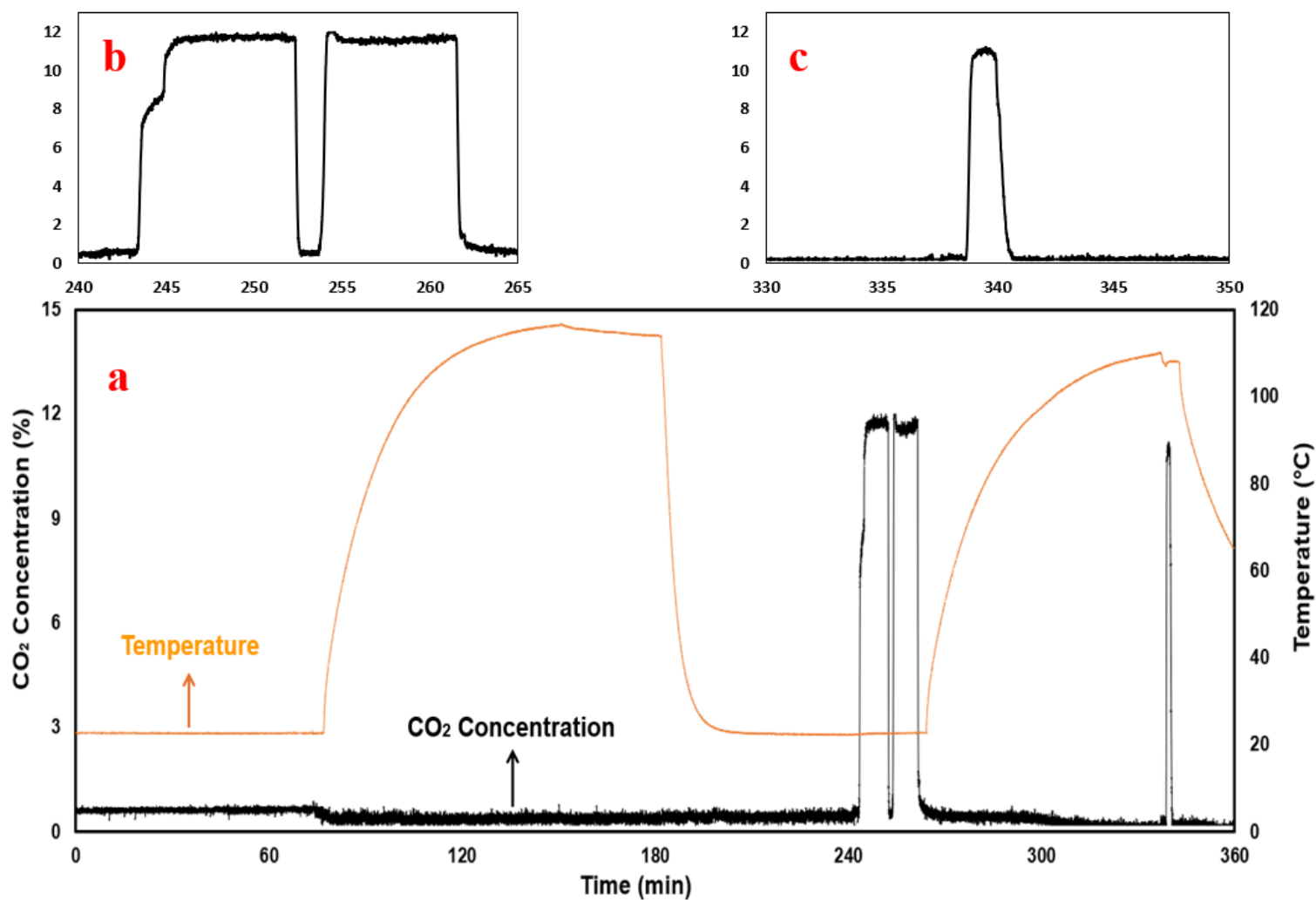


Figure 4.7 Breakthrough curves and temperature profiles for the carbon aerogel samples during CO₂ adsorption at 25 °C and photothermal desorption under solar-simulated illumination ($T \approx 110$ °C): (a) full profile, (b) adsorption region, and (c) desorption region.

Table 1. Integrated CO₂ adsorption and desorption volumes for blank runs and carbon aerogel samples, obtained from three repeated breakthrough experiments

Sample	Adsorption (cm³)	Desorption (cm³)
Blank run 1	18.13	15.82
Blank run 2	19.01	15.50
Blank run 3	19.06	16.12
Carbon Aerogel run 1	23.60	18.33
Carbon Aerogel run 2	23.65	19.14
Carbon Aerogel run 3	23.6	18.34

Across the three blank runs, the average integrated adsorption and desorption volumes were 18.73 cm³ and 15.81 cm³, respectively. The standard deviations, 0.523 cm³ for adsorption and 0.310 cm³ for desorption, indicate modest variability, attributable to minor fluctuations in system flow stability, detector sensitivity, and unavoidable noise within the gas-handling setup. Importantly, the magnitude of these deviations remains small relative to the absolute breakthrough volumes, confirming that the baseline system behaviour is stable and that the dead-volume correction is reliable.

Table 2. Average $\pm$ standard deviation of integrated CO₂ adsorption and desorption volumes for blank runs and carbon aerogel samples, based on three repeated breakthrough experiments for each condition.

Sample	Adsorption (cm³)	Desorption (cm³)
Blank runs	18.73 $\pm$ 0.523	15.81 $\pm$ 0.310
Carbon Aerogel runs	23.62 $\pm$ 0.0289	18.60 $\pm$ 0.4648

The carbon aerogel runs showed higher integrated volumes, with mean values of 23.62 cm³ during adsorption and 18.60 cm³ during desorption. The standard deviations for these runs were 0.0289 cm³ for adsorption and 0.4648 cm³ for desorption. The remarkably small adsorption standard deviation (0.0289 cm³) demonstrates excellent reproducibility between aerogel runs and highlights the stability of the aerogel's CO₂ uptake characteristics across repeated cycles. In contrast, the slightly higher desorption standard deviation (0.4648 cm³) suggests greater variability during the regeneration step. This behaviour is consistent with the sensitivity of desorption to photothermal heating uniformity, gas flow redistribution, and the release kinetics of strongly bound CO₂ species within ultramicropores (<0.7 nm). Even so, the magnitude of this variation remains moderate and does not detract from the overall reproducibility of the aerogel's performance. Subtracting the blank contributions yields net CO₂ adsorption and desorption volumes of 5.06 ± 0.52 cm³ and 3.08 ± 0.56 cm³, respectively. Converting these values to molar uptake using the ideal molar volume at STP (22,414 cm³ mol⁻¹) results in absolute uptakes of 0.226 ± 0.023 mmol (adsorption) and 0.137 ± 0.025 mmol (desorption). Normalizing by the total aerogel mass of 0.12 g produces adsorption and desorption capacities of 1.88 ± 0.19 mmol g⁻¹ (82.6 ± 8.6 mg mg g⁻¹) and 1.14 ± 0.21 mmol g⁻¹ (50.2 ± 9.1 mg g⁻¹), respectively.

The difference between adsorption and desorption capacities indicates that a fraction of CO₂ remains trapped in the aerogel after the photothermal regeneration step. This residual loading likely arises from slow CO₂ release from ultramicropores, diffusion limitations within the interconnected pore network, and strongly physisorbed species that require stronger driving forces (e.g., vacuum, higher temperatures) for complete desorption. Such adsorption–desorption hysteresis is well-recognized in carbon-based sorbents and reflects the influence of pore size distribution and adsorption potential wells.

The triplicate dataset and standard deviations provide insight into the robustness of both the measurement system and the aerogel material. The greater variability in blank adsorption values relative to aerogel adsorption values underscores the stable intrinsic performance of the sorbent compared to minor system fluctuations. Meanwhile, the elevated desorption variability for the aerogel highlights the importance of photothermal uniformity and thermal management in future optimization.

The measured adsorption capacity of 1.88 mmol g^{-1} falls squarely within the range typically reported for carbon aerogels and activated carbons under similar dilute CO_2 conditions ($1\text{--}3 \text{ mmol g}^{-1}$ at near-ambient temperatures and $10\text{--}15\% \text{ CO}_2$ [4]). These results confirm that the high porosity, large surface area, and accessible microporosity of the carbon aerogel promote effective CO_2 capture. The photothermal desorption behaviour shown in Figure 14 demonstrates the feasibility of solar-assisted regeneration, although further improvements in thermal uniformity or pore-engineering strategies may enhance the working capacity.

Overall, the carbon aerogel demonstrates consistent, reproducible, and meaningful CO_2 adsorption under dilute conditions, along with effective, though not complete, photothermal regeneration. Future developments such as micropore tuning, nitrogen or amine functionalization, or hybrid thermal–vacuum regeneration could further enhance performance for cyclic CO_2 capture systems.

5 Conclusions and Future Work

5.1 Overview of the Study

This thesis investigated the suitability of a commercial carbon aerogel as a photothermal sorbent for CO₂ capture under dilute flue-gas-like conditions. The work was motivated by the urgent need to develop robust, thermally stable, and regenerable solid sorbents that can operate efficiently in temperature-swing or photo-assisted adsorption processes. While carbon aerogels have been widely discussed as promising candidates owing to their low density, high porosity, and favourable thermal properties, comprehensive studies that link their optical response, photothermal behaviour, and CO₂ uptake performance remain comparatively limited in the literature.

Building on these gaps, the thesis was structured around three main objectives. First, the optical properties of a disc-shaped carbon aerogel (14 mm diameter, 4 mm thickness, bulk density $\approx 0.2 \text{ g cm}^{-3}$) were characterised using FTIR and UV–Vis–NIR spectroscopy, with the aim of quantifying its reflectance, transmittance and absorptance across a broad spectral range and testing the hypothesis that the aerogel behaves as an exceptionally strong absorber. Second, the photothermal response of the same aerogel under simulated solar illumination was examined via infrared thermography to assess how efficiently the material converts incident radiation into heat, how temperature gradients develop, and how quickly the sample can be heated and cooled. Third, the CO₂ adsorption capacity and photothermal regeneration behaviour were measured using dynamic breakthrough experiments with a 12 % CO₂ / 88 % N₂ mixture at near-ambient temperature, to determine whether the high surface area and microporosity of the aerogel translate into a meaningful CO₂ uptake on the order of a few mmol g⁻¹, even without chemical functionalization.

The experimental methodology described in Chapter 3 integrated these elements using custom sample holders for optical and adsorption measurements and a combination of FTIR, UV–Vis–NIR spectroscopy, thermal imaging and breakthrough analysis. Chapter 4 then presented and discussed the resulting optical spectra, thermal maps and adsorption–desorption curves in a unified framework, providing the basis for the conclusions and recommendations summarised in this chapter.

5.2 Key Conclusions

5.2.1 Optical properties of the carbon aerogel

The first objective of this thesis was to determine whether the carbon aerogel exhibits the broadband, near-black optical behaviour that is desirable for photothermal CO₂ capture systems. UV–Vis–NIR measurements over 220–1400 nm showed that the 4 mm thick aerogel disc is effectively opaque in this range: transmittance was essentially zero at all wavelengths, and reflectance remained below approximately 5 %. Consequently, the calculated absorptance exceeded 95 % throughout the entire UV–Vis–NIR spectrum, confirming that the material behaves as a strong broadband absorber in this region.

Complementary FTIR spectroscopy extended this analysis into the near- and mid-infrared (1–21 μm), a spectral window that is highly relevant for radiative heat transfer and for thermal imaging. Across the NIR–MIR range the transmittance remained very small: in the short-wavelength region (1–2.5 μm) it was on the order of 0.8–1.3 %, and it dropped below about 0.2 % at longer wavelengths, indicating that the disc is effectively opaque for thermal radiation. The reflectance was also relatively low, typically around 5–6 % in the 1–7 μm range, with modest spectral features

including a local maximum of approximately 8–9 % and an increase toward roughly 11–12 % between 10 and 21 μm . Because transmission is negligible beyond $\sim 3.5 \mu\text{m}$, the absorptance closely follows $1-R$ and generally falls in the range 0.88–0.95 over most of the NIR–MIR spectrum.

Taken together, these measurements demonstrate that the investigated carbon aerogel behaves as a broadband, nearly opaque, high-emissivity absorber across both solar and thermal radiation bands. This behaviour is consistent with its porous carbon microstructure and the strong intrinsic absorption of disordered carbon networks and underpins the subsequent photothermal experiments and emissivity assumptions used in the thermal imaging analysis. The initial hypothesis that the aerogel's black carbon framework and porous structure would lead to exceptionally high absorption is therefore strongly supported by the optical data.

5.2.2 Photothermal response and heat-transfer behaviour

The second objective was to quantify how efficiently the carbon aerogel converts incident radiation into heat and how the resulting temperature field evolves in space and time. Infrared thermography under xenon-lamp illumination at 1 Sun (100 mW cm^{-2}) showed a rapid temperature rise at the illuminated surface: the top-view temperature increased from ambient ($\sim 28^\circ\text{C}$) to approximately 58°C within the first 15 s, then rose more gradually to a steady-state value of about 64°C within 1–2 min. Thereafter, the temperature remained essentially constant over a 10 min illumination period, indicating that the absorbed radiative power was balanced by convective and radiative heat losses to the surroundings.

In contrast, the lateral view revealed lower temperatures and a more moderate time response. The edge temperature increased from $\sim 28^\circ\text{C}$ to $\sim 40^\circ\text{C}$ within 15 s, approaching a steady value near

46 °C after roughly 2–3 min—around 18 °C lower than the illuminated surface. This pronounced surface–edge temperature gradient reflects the directional nature of the radiative input: the top surface directly absorbs most of the incident flux, while the edges receive heat only through in-plane conduction and diffuse scattering, and experience enhanced convective cooling due to their exposure. Spatial temperature maps further indicated a hotter central region and localised heating at the disc–holder interface, confirming that both illumination non-uniformities and heat conduction into the holder influence the steady-state distribution.

The cooling cycle, recorded immediately after switching off the lamp, demonstrated that the aerogel also cools rapidly. The top surface temperature decreased from ~64 °C to ~38 °C within 15 s and approached ~31 °C (close to ambient) over a 10 min interval, while the lateral surface cooled from ~46 °C to ~33.5 °C in 15 s and to ~30 °C by the end of the same period. This two-stage cooling response—initial rapid decay followed by a slower approach to equilibrium—is consistent with the very low density, high porosity, and low heat capacity of carbon aerogels, which limit thermal energy storage and favour quick equilibration with the environment.

Overall, the thermal imaging experiments confirm that the carbon aerogel can be heated quickly and efficiently under moderate radiative fluxes and can likewise be cooled rapidly when illumination is removed. These characteristics are beneficial for temperature-swing or photothermal-swing adsorption cycles, because they enable fast transitions between adsorption and desorption conditions and thereby have the potential to shorten cycle times and improve process productivity. At the same time, the observed temperature gradients highlight the importance of optimising sample geometry, insulation and illumination uniformity to achieve more homogeneous temperature distributions in practical devices.

5.2.3 CO₂ adsorption capacity and photothermal regeneration

The third objective was to evaluate the CO₂ adsorption performance of the carbon aerogel under dilute conditions and to assess its regenerability via photothermal heating. Dynamic breakthrough experiments were conducted in a fixed-bed configuration

using a 12 % CO₂ / 88 % N₂ mixture at approximately 25 °C for adsorption, followed by photothermal desorption at ~110 °C under irradiation from a solar-simulated source. The outlet CO₂ signal showed a sharp drop from ~12 % to ~0 % upon switching the flow through the aerogel, followed by a gradual return to the baseline concentration as the material approached saturation, indicating rapid initial uptake and a well-defined breakthrough front.

Integrated breakthrough volumes from three replicate runs with the aerogel and three blank runs with the empty holder were used to compute net CO₂ uptake. The mean integrated adsorption and desorption volumes were 23.62 cm³ and 18.60 cm³ for the aerogel, compared to 18.73 cm³ and 15.81 cm³ for the blank, with modest standard deviations that confirm good reproducibility of both the system and the material. After subtracting the blank contributions, the net adsorption and desorption volumes were 5.06 cm³ and 3.08 cm³, respectively. Converting these values to molar uptake using the ideal molar volume at STP and normalising by the 0.12 g aerogel mass yielded adsorption and desorption capacities of 1.88 mmol g⁻¹ (82.6 mg g⁻¹) and 1.14 mmol g⁻¹ (50.2 mg g⁻¹).

These capacities fall squarely within the range typically reported for carbon aerogels and activated carbons under comparable conditions (1–3 mmol g⁻¹ at near-ambient temperatures and CO₂ volume fractions of around 10–15 %), confirming that the high porosity and accessible microporosity of the present aerogel translate into effective CO₂ capture. However, the difference

between adsorption and desorption capacities indicates that a fraction of CO₂ remains trapped after the photothermal regeneration step. This residual loading is attributed to slow release from ultramicropores, diffusion limitations within the pore network, and strongly physisorbed species that would require stronger driving forces (e.g., higher temperatures or vacuum) to be fully desorbed behaviour that is well-known for many carbonaceous sorbents.

Mechanistically, CO₂ capture in this study is expected to be predominantly physisorption, since the carbon aerogel discs were tested as received with no added reactive functionalities (e.g., amines). In porous carbons, physisorption proceeds mainly via pore filling in micro/ultramicropores and weak dispersive/dipole–quadrupole interactions and is therefore inherently reversible with temperature and/or pressure changes. By contrast, chemisorption typically requires specific reactive surface groups that form stronger chemical species (e.g., carbamates/bicarbonates in amine-based sorbents), which are not intentionally introduced here. The observed adsorption–desorption gap is therefore attributed to strong physisorption in ultramicropores and diffusion/kinetic limitations, rather than irreversible chemical bonding.

Overall, the breakthrough experiments demonstrate that the studied carbon aerogel provides consistent, reproducible CO₂ uptake under dilute conditions and can be regenerated photothermally to a substantial extent, though not fully. The initial expectation that the aerogel would exhibit a CO₂ capacity on the order of a few mmol g⁻¹ is therefore validated, while the observed partial hysteresis underscores the need to further optimise regeneration conditions and pore architecture to maximise the working capacity.

5.2.4 Overall assessment and contributions

Taken together, the results from Chapters 3 and 4 provide an integrated picture of how the optical, thermal and adsorption properties of a carbon aerogel interact in the context of CO₂ capture. The

material exhibits near-black, high-emissivity behaviour across solar and thermal wavelengths; it heats and cools rapidly under moderate radiative fluxes; and it delivers a CO₂ adsorption capacity that is competitive with other non-functionalised carbon sorbents under dilute conditions, with reasonably effective photothermal regeneration.

In conventional temperature-swing adsorption (TSA) for post-combustion CO₂ capture, practical deployment is often limited by (i) high regeneration energy, because a significant fraction of supplied heat is consumed as sensible heat to warm the sorbent and reactor internals rather than to desorb CO₂, (ii) long cycle times, driven by heat-transfer resistances and slow bed heating/cooling, and (iii) non-uniform regeneration, where temperature gradients produce spatially varying working capacity and incomplete desorption. Recent TSA analyses and reactor reviews highlight that the temperature-swing energy requirement scales with the heat capacities and temperature change of the sorbent/reactor system and is strongly coupled to the achievable CO₂ working capacity, while poor heat transfer can translate directly into long heating and cooling steps [148].

The present results address these TSA limitations in three ways. First, the aerogel's near-black optical response enables efficient coupling of radiative energy into the sorbent, supporting photothermal/solar-assisted TSA concepts that aim to reduce parasitic heating of bulk reactor hardware by depositing energy where it is needed for desorption. Second, the measured rapid heating and cooling under solar-like irradiation indicate that meaningful thermal swings can be achieved on short time scales, which is consistent with strategies for reducing TSA cycle time and improving productivity through faster thermal transitions. Third, the thermal imaging quantitatively reveals surface–edge temperature gradients, providing a clear design target for scale-up: practical photothermal TSA will require improved illumination uniformity, insulation, and thermal-contact design to avoid cold zones that limit desorption and reduce working capacity.

These findings align with recent reports on solar/photothermal swing adsorption using carbonaceous monolith-type sorbents, where localized radiative heating can accelerate regeneration, but thermal uniformity remains a key reactor-level constraint [149].

Finally, the breakthrough results demonstrate reproducible uptake under dilute flue-gas-like CO₂ levels, while the adsorption–desorption gap highlights a remaining TSA challenge: incomplete regeneration can occur when a fraction of CO₂ is retained in ultramicropores or released slowly due to diffusion limitations. This points to specific process routes, e.g., higher regeneration temperature, longer purge, reduced pressure (TVSA/VSA), and optimized monolith geometry/thermal management—that can increase the recoverable working capacity in future reactor implementations [149, 150].

Considering recent photothermal CO₂ capture literature, the main contribution of this thesis is to provide a single-material, experimentally integrated linkage between optical absorption/emissivity, transient photothermal temperature fields, and dynamic breakthrough-based CO₂ uptake/regeneration for a monolithic carbon aerogel. While prior studies have demonstrated solar thermal swing adsorption in porous carbon monoliths under intermittent illumination and photothermal regeneration of chemisorptive DAC sorbents using aerogel or polymer architectures [151], comparatively fewer works quantify how broadband optical behavior and spatially non-uniform heating translate into measurable regeneration effectiveness under flue-gas-like concentrations. Here, the carbon aerogel is shown to function as an intrinsically strong photothermal absorber and a competitive physical sorbent under dilute flue-gas-like conditions, achieving net adsorption of 1.88 mmol g⁻¹ and photothermal desorption of 1.14 mmol g⁻¹ under the tested conditions. The observed incomplete regeneration further identifies a practical design constraint for photothermal/TSA hybrid systems, that ultramicropore diffusion and temperature

gradients can limit working capacity thereby directly motivating future improvements such as pore-structure tuning, reduced thickness or improved illumination uniformity, and hybrid thermal–vacuum regeneration strategies.

Beyond these performance metrics, the thesis contributes methodological advances by developing and demonstrating custom sample holders for optical and breakthrough measurements and by combining FTIR/UV–Vis spectroscopy, thermal imaging and dynamic adsorption experiments within a single, coherent experimental framework. This integrated approach helps to bridge the gap between fundamental characterisation of carbon aerogels and their practical evaluation as CO₂ sorbents and provides a template for future studies aiming to link structure, optical behaviour, photothermal response and adsorption performance more systematically.

5.3 Limitations

While the findings of this thesis provide valuable insights into the behaviour of carbon aerogels for CO₂ capture, several limitations should be acknowledged when interpreting the results and extrapolating them to other systems.

5.3.1 Material and characterization limitations

The study focused on a single commercial carbon aerogel formulation supplied as monolithic discs with fixed dimensions (14 mm × 4 mm) and a nominal bulk density of ~0.2 g cm⁻³. Consequently, the effects of key structural parameters such as density, pore size distribution, or surface chemistry on optical, thermal and adsorption behaviour could not be systematically explored. In addition, while the literature provides extensive information on typical surface areas and pore structures for carbon aerogels, detailed textural characterisation (e.g., N₂ physisorption to determine BET surface area and micropore volume) and surface-chemistry analysis (e.g., XPS or Raman spectroscopy)

were not performed directly on the specific sample examined here. This limits the ability to quantitatively correlate the measured performance with microstructural descriptors for this material.

5.3.2 Experimental and operating-condition limitations

The optical measurements were conducted at near-normal incidence in an integrating-sphere configuration and used a single disc thickness, which means that the relative contributions of intrinsic absorption versus multiple scattering and sample thickness were not independently resolved. The thermal imaging experiments were performed under one illumination level (1 Sun) and in a specific laboratory geometry; although they clearly revealed fast heating and distinct temperature gradients, they do not directly cover the range of fluxes and configurations that might be encountered in large-scale solar collectors or industrial reactors.

This solar-collector dependence is important because the achievable regeneration temperature window constrains the CO₂–sorbent interaction strength that can be targeted through functionalization: binding that is too strong may not be fully regenerated under realistic solar thermal conditions, whereas binding that is too weak may reduce working capacity under dilute CO₂.

Similarly, the CO₂ adsorption tests were carried out at a single inlet composition (12 % CO₂ in N₂), near-ambient pressure, and one desorption temperature (~110 °C) with a fixed flow rate. These conditions represent an important but limited subset of possible operating regimes. The study did not explicitly resolve CO₂/N₂ selectivity through multi-component isotherms or separate single-component adsorption measurements, nor did it explore the influence of moisture, higher pressures, or very low CO₂ concentrations (e.g., for direct air capture scenarios). Furthermore,

although the breakthrough protocol included three repeated adsorption–desorption cycles, longer-term cycling over tens or hundreds of cycles was not investigated, so conclusions about long-term structural stability and performance degradation remain tentative.

5.3.3 Modelling and scale-up limitations

The present work is primarily experimental and does not include detailed modelling of coupled heat and mass transfer within the aerogel or within a packed-bed reactor. As a result, while qualitative interpretations of the observed temperature gradients and breakthrough shapes were provided, a rigorous quantitative description of transport processes and a complete process-level assessment (e.g., energy efficiency, productivity, cost) were beyond the scope of the thesis. The implications of the measured properties for full-scale CO₂ capture systems therefore need to be evaluated with dedicated reactor and process models in future work.

Beyond the lack of coupled heat–mass transfer modelling, the present study also does not resolve several practical feasibility constraints that can govern scale-up of carbon-aerogel-based contactors. Many carbon aerogels are known to be brittle/fragile, so mechanical integrity under handling, sealing, vibration, and repeated thermal cycling must be established using compressive testing and attrition/dusting assessments for the target reactor geometry. In addition, the manufacturing route can be a dominant scalability and cost driver: conventional carbon aerogel production may require extensive solvent processing and supercritical or freeze-drying steps, and the use of costly precursors (e.g., resorcinol) and long cycle times has been identified as a barrier to industrial-scale production. These considerations reinforce the need for future work that combines reactor-level modelling with mechanical qualification and an explicit techno-economic comparison against lower-cost carbon sorbents (e.g., activated carbons) and other leading adsorbents under realistic flue-gas conditions [153].

Finally, the optical characterization (UV–Vis–NIR and FTIR) was performed in ambient air and was not repeated under representative flue-gas compositions or at controlled CO₂ loading states of the sorbent. Because CO₂ and H₂O radiatively participate in the infrared, gas composition can influence reactor-scale radiative exchange and may also affect apparent FTIR data if atmospheric content differs between background and sample scans. Future work should therefore perform in-situ optical measurements under controlled CO₂/N₂/H₂O atmospheres at defined sorbent loadings to separate intrinsic solid response from gas-phase contributions and to support coupled radiative/heat–mass transfer modelling for regeneration.

5.4 Recommendations for Future Work

The results and limitations discussed above suggest several technically grounded directions for further research. These recommendations are grouped into material development, advanced characterisation, extended adsorption testing, and system-level design and modelling.

5.4.1 Material design and surface functionalization

A natural next step is to move beyond a single commercial aerogel and investigate a family of carbon aerogels with systematically varied properties. Building on the synthesis and activation strategies reviewed in the background chapter, future studies could:

- Vary precursor chemistry, gelation conditions, and carbonisation/activation protocols (e.g., CO₂, steam, or KOH activation) to tailor microporosity, mesoporosity, and bulk density, thereby elucidating how these parameters affect optical absorptance, emissivity and CO₂ uptake.
- Introduce heteroatom doping (e.g., nitrogen, oxygen, or phosphorus) during carbonisation or via post-treatment to modify surface polarity and enhance specific interactions with CO₂.

- Reduce mid-IR emissivity while preserving solar absorptance: The present aerogel behaves as a near-black absorber, which is excellent for solar absorption but can also imply high mid-IR emissivity and therefore higher radiative heat losses at elevated temperature. Future work could target spectral selectivity using approaches adapted from solar-selective absorber technology (e.g., thin low-emittance coatings or layered reflector designs) while ensuring the coating does not block pores, degrade mass transfer, or fail under thermal/humidity cycling.
- Incorporate amine-based functionalities (such as polyethyleneimine or other polyamines) into the aerogel network or onto its surface, drawing on approaches that have shown strong enhancements in CO₂ capacity and selectivity in other porous sorbents.

In selecting among these functionalization pathways, it is important to consider the temperature window that can practically be reached using solar energy, because this window dictates the optimal CO₂ bonding strength required to achieve high cyclic working capacity with solar-driven regeneration. Under standard terrestrial conditions, ‘1 Sun’ corresponds to an incident irradiance on the order of $\sim 1000 \text{ W m}^{-2}$. However, the useful thermal power and attainable operating temperature depend strongly on collector type, optical concentration, insulation, and ambient losses. Flat-plate collectors are typically designed for relatively low-to-moderate working temperatures, while medium-temperature solar process heat commonly targets the $\sim 80\text{--}160 \text{ }^{\circ}\text{C}$ regime using configurations such as CPC-evacuated tube collector systems. Consequently, if the regeneration step is intended to rely predominantly on solar thermal input, the adsorbent should be engineered so that desorption can proceed efficiently within this practical temperature window. This has direct implications for the CO₂ bonding strength that should be targeted. If the interaction is too weak, regeneration is easy at modest temperature, but CO₂ uptake and selectivity can drop—

especially as the bed warms—leading to low working capacity under dilute conditions. If the interaction is too strong, uptake (particularly at low CO₂ partial pressure) may improve, but regeneration becomes more demanding: higher temperature, longer purge times, and/or reduced pressure may be required to restore capacity, and the per-cycle energy requirement increases because both the desorption enthalpy and the sensible heat of the sorbent/contacter contribute to TSA energy consumption. For many porous carbons, physisorption-dominated CO₂ capture typically corresponds to heats of adsorption on the order of ~25–40 kJ mol⁻¹, consistent with reversible pore filling in micro/ultramicropores. In contrast, amine-based chemisorbents often exhibit substantially larger heats of adsorption (commonly reported on the order of ~60–100 kJ mol⁻¹ depending on the amine chemistry and mechanism), reflecting stronger chemical interactions such as carbamate/bicarbonate formation. These ranges suggest that, for solar-assisted regeneration constrained to roughly ~80–160 °C, moderate interaction strengths are generally desirable for post-combustion-like CO₂ concentrations to preserve regenerability and cycle efficiency, whereas stronger chemisorption strategies are most justified for very low CO₂ partial pressures (e.g., DAC) if the regeneration strategy (temperature, pressure, and time) remains compatible with the available solar thermal input [9].

Accordingly, future functionalization of carbon aerogels can be pursued along two complementary routes, chosen based on the intended capture scenario and solar thermal constraints. (i) ‘Moderate-binding’ optimisation for flue-gas capture can focus on tailoring ultramicroporosity and introducing controlled heteroatom doping or mild surface oxidation (e.g., N/O/P incorporation or post-treatments) to enhance CO₂ affinity and selectivity without pushing the binding strength beyond what can be reversed within the solar temperature window. This route aims to increase working capacity while retaining rapid photothermal swing behaviour and avoiding excessive

regeneration temperature. (ii) ‘Strong-binding’ chemisorption for very dilute CO₂ can incorporate amine functionalities to increase uptake at low partial pressure; however, amine loading and dispersion must be optimised because excessive loading can block pores, increase heat capacity, and introduce strong water co-adsorption, each of which can slow solar-driven regeneration and increase the sensible-heat penalty [9, 154].

Finally, to ensure that functionalization targets are aligned with solar-driven regeneration, future studies should quantify adsorption energetics and cyclic performance in a process-relevant manner. This includes (a) acquiring multi-temperature CO₂ isotherms and extracting heats of adsorption, (b) performing cyclic breakthrough tests under representative feeds (including humidity and impurities), and (c) evaluating regeneration under illumination/collector-relevant boundary conditions to determine whether the selected bonding strength yields the desired trade-off between uptake, selectivity, and solar-achievable regeneration temperature [9].

5.4.2 Extended structural and spectroscopic characterisation

To better link structure and performance, future work should complement the existing optical and adsorption measurements with more comprehensive structural and surface-chemistry characterisation of the specific aerogel samples under study. Recommended techniques include:

- **Gas sorption analysis** (e.g., N₂ physisorption) to quantify BET surface area, pore-size distributions and micropore volumes, enabling direct correlation between microstructure and CO₂ uptake.
- **Electron microscopy** (SEM/TEM) to visualise the pore architecture, particle connectivity and potential structural changes after multiple adsorption–desorption cycles.

- **Spectroscopic probes** such as Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) to characterise graphitic versus amorphous carbon domains and to identify surface functional groups relevant to CO₂ adsorption.

On the optical side, it would be beneficial to perform angle-dependent and polarisation-resolved measurements, as well as temperature-dependent FTIR studies, to more fully characterise the radiative properties of the aerogels under conditions representative of operating temperatures.

5.4.3 Advanced adsorption testing and process optimisation

The breakthrough experiments in this thesis provide a valuable baseline for the adsorption capacity and photothermal regenerability of the carbon aerogel, but further work is needed to map out its performance under a broader range of conditions and to quantify key process metrics such as selectivity and working capacity. Specific recommendations include:

- **Varying feed composition and pressure.** Conducting adsorption tests at different CO₂ volume fractions (e.g., 5–20 %) and pressures, including sub-ambient and slightly elevated pressures, would help to construct multi-point isotherms and assess performance across various industrial scenarios.
- **Direct determination of CO₂/N₂ selectivity.** Combining single-component adsorption measurements with multi-component (CO₂/N₂) breakthrough experiments would allow more rigorous determination of selectivity and competitive adsorption effects, addressing one of the original objectives that could not be fully realised in the present work.
- **Influence of humidity and contaminants.** Introducing controlled amounts of water vapour or other flue-gas impurities (e.g., O₂, SO_x, NO_x) into the feed stream would clarify

the robustness of the aerogel under more realistic gas compositions, especially given the hydrophobic nature of many carbon aerogels.

- **Extended cycling and regeneration strategies.** Performing long-term cycling tests (tens to hundreds of adsorption–desorption cycles) under photothermal regeneration, and comparing them with alternative schemes (e.g., combined thermal–vacuum swing or stepwise temperature ramps), would help to quantify working capacity, regeneration efficiency and potential degradation mechanisms.

These studies would provide a more complete picture of the aerogel’s practical viability as a CO₂ sorbent and help to identify operating windows in which its advantages are maximised.

5.4.4 System-level design, modelling and integration

Finally, there is considerable scope for extending the present work toward system-level design and modelling of photothermal CO₂ capture processes using carbon aerogels. Future efforts could focus on:

- **Coupled heat–mass transfer modelling.** Developing 1D or 2D models of radiatively heated aerogel beds that incorporate realistic optical properties, thermal conductivity, heat capacity, and adsorption isotherms would enable quantitative interpretation of the temperature gradients and breakthrough profiles observed in this thesis and guide the optimisation of disc thickness, bed packing, and illumination geometry.
- **Reactor and holder optimisation.** Building on the custom holder designs introduced here, numerical simulations could be used to minimise edge heat losses, improve flow distribution, and reduce dead volume, with subsequent experimental validation using updated hardware.

- **Energy and process-level assessment.** Integrating the measured and model-predicted performance into process simulations (e.g., cycle-averaged energy consumption, throughput, and sorbent inventory) would help to benchmark photothermal carbon-aerogel systems against conventional temperature-swing adsorption and alternative CO₂ capture technologies.

Such multi-scale analysis, from material to device to process, would be an important step toward evaluating the true potential of carbon aerogels in industrial CO₂ capture and could inform the design of next-generation solar-assisted capture systems.

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