Development of Graphene Aerogel Nanocomposites for Hydrogen Storage

M.Tech. Thesis

By

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DEPARTMENT OF MECHANICAL

INDIAN INSTITUTE OF TECHNOLOGY INDORE

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Development of Graphene Aerogel Nanocomposites for Hydrogen Storage

A THESIS

Submitted in partial fulfillment of the
requirements for the award of the degree
of
Master of Technology

by
ARPIT PAREKH

DEPARTMENT OF MECHANICAL
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CANDIDATE’S DECLARATION

I hereby certify that the work which is being presented in the thesis entitled **“Development of Graphene Aerogel Nanocomposites for Hydrogen Storage”** in the partial fulfillment of the requirements for the award of the degree of **MASTER OF TECHNOLOGY** and submitted in the **DEPARTMENT OF MECHANICAL, Indian Institute of Technology Indore**, is an authentic record of my own work carried out during the time period from June 2024 to May 2025 under the supervision of Prof. Shailesh Kundalwal, Professor, IIT Indore.

The matter presented in this thesis has not been submitted by me for the award of any other degree of this or any other institute.

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This is to certify that the above statement made by the candidate is correct to the best of my/our knowledge.

Signature of the Supervisor of
M.Tech. thesis (with date)
(Prof. Shailesh I Kundalwal)

ARPIT PAREKH has successfully given his/her M.Tech. Oral Examination held on **23rd May**.

Signature(s) of Supervisor(s) of M.Tech. thesis
Date: 09/06/2025

Convener, DPGC
Date: 09-06-25

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Thanks a lot, everyone…!

-ARPIT PAREKH

Dedicated to

My Beloved Family

Abstract

The transition toward clean and sustainable energy solutions has accelerated interest in advanced hydrogen storage materials. This thesis focuses on the development and characterization of reduced graphene oxide (rGO) aerogels as potential candidates for efficient hydrogen storage. The rGO aerogels were synthesized via a modified hydrothermal reduction method followed by freeze-drying, resulting in ultralight structures with highly porous 3D architectures.

A suite of characterization techniques was employed to evaluate the material's suitability for gas adsorption. BET analysis revealed a high surface area (>500 m^2/g), while BJH pore analysis confirmed a dominant mesoporous network favorable for physisorption. SEM imaging demonstrated an open, interconnected nanosheet morphology, supporting enhanced diffusion kinetics. XRD patterns indicated partial graphitization with amorphous features, and Raman spectroscopy confirmed a high defect density—both contributing to increased adsorption activity.

Overall, the structural and textural properties of the synthesized rGO aerogel suggest strong potential for application in ambient-condition hydrogen storage systems, paving the way for further optimization in clean energy technologies.

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ACRONYMS

LPE – Liquid Phase Exfoliation

CVD – Chemical Vapour Deposition

CNT - Carbon Nano tubes

GO- Graphene Oxide

rGO- Reduced Graphene Oxide

FCEV- Fuel Cell Electric Vehicles

H_2- Hydrogen

CCG- Chemically Converted Graphene

IoT- Internet of Things

SEM – Scanning Electron Microscopy

XRD- X-Ray Diffraction

BET- Brunauer-Emmett-Teller (Surface Area)

BJH- Barrett-Joyner-Halenda (Surface Area)

Chapter 1: Introduction

1.1 Carbon and its Allotropes

Carbon materials are ubiquitous in our daily lives due to their affordability and remarkable chemical and thermal stability. Carbon atoms engage in robust covalent bonds with other carbon atoms in distinct hybridization states (sp^3, sp^2, sp) as well as heteroatoms (other elements excluding carbon), giving rise to a diverse array of chemical structures including small bio-molecules, long chains and nanomaterials. This versatility results in carbon exhibiting a multitude of morphological structures at both atomic and nanoscale each endowed with unique surfaces and physicochemical properties. Generally, carbon allotropes can be classified into two groups depending on the type of covalent bond present. The first group comprise graphenic nanostructures, characterize by sp^2 hybridization, forming densely packed honeycomb like hexagonal crystal lattices, typically manifesting in 2D (two dimensional) layers with average dimension of around 500 nm. These structures may also feature sp^3 hybridized carbon atoms, indicating the presence of defect sites. Notable examples are graphene, carbon nanotubes (CNTs), carbon dots (CDs)/ fullerene. Conceptually, graphene is the fundamental "building block" of the other graphenic/graphitic nano-allotropes. A fine sectioned segment of a graphene sheet theoretically wrapped up to form the 0D fullerene, curled into 1D CNT, or stacked with additional sheet of graphene to form multilayered 2D carbon nanosheets or graphite [1]

Allotropes are different structural forms of the same element having different physical properties and chemical behaviors.

Diamond: It has tetrahedral lattice structure of sp^3-hybridized carbon atoms. Hardest natural material, electric insulator but excellent thermal conductor, transparent with high refractive index. Therefore, it is used in cutting tools, abrasives and high-performance optics

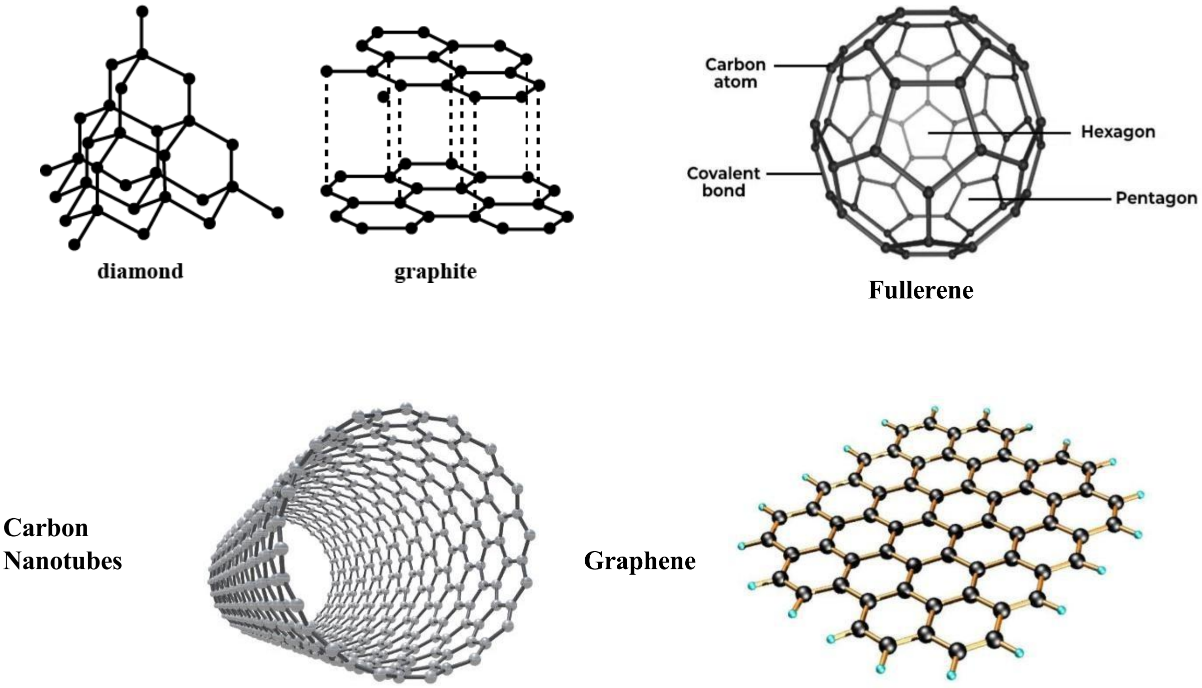


Fig 1.1: Structure of carbon allotropes

Graphite: Structure of Graphite is stacked hexagonal layers of sp^2-hybridized carbon with delocalized π-electrons. It is soft, opaque and good conductor of electricity. It is used as lubricants, pencil "lead," and electrodes in batteries.

Fullerene: It is a nanomaterial and 0D allotrope of carbon. It structure is hollow sphere arranged as 12 pentagons and 20 hexagons of C_{60} carbon atoms. It is soluble in organic solvents. It has potential application for superconductivity and drug delivery.

Carbon Nanotubes (CNT): Carbon nanotubes can be described as graphite sheets that are rolled up into cylindrical shapes. The length of CNTs is in the form of micrometres with a diameter of about 100 nm. It is called 1D structure of Carbon nanomaterial. There are two types of carbon nanotubes that are classified according to the number of carbon layers present in them. Single-walled carbon nanotubes (SWCNTs) consist of single graphene layer with diameter varying between 0.4 and 2 nm and usually occurs as hexagonal-packed bundles. Multi-walled carbon nanotubes (MWCNTs) comprises of two or several cylinder, each made up of graphene sheets. The diameter varies from 1 to 3 nm. [2]

CNTs exhibit exceptional mechanical, electrical, and thermal properties, making them one of the most promising nanomaterials for various advanced applications. Their tensile strength is among the highest of known materials (~100 times that of steel at a fraction of the weight), and they have excellent electrical conductivity and thermal stability.

Due to their extraordinary mechanical, electrical, and thermal properties, carbon nanotubes (CNTs) have found a wide range of applications across multiple scientific and engineering disciplines: Composite Materials, Energy Storage and Conversion, Electronics and Sensors, Biomedical Applications, Thermal Management, Water Purification, etc.

Graphene: Graphene is a single layer of carbon atoms arranged in a two-dimensional honeycomb lattice. It has attracted enormous research interest due to its unique electronic structure, ultrahigh carrier mobility, tensile strength (130 GPa), and thermal conductivity (5000 W/m·K). It is one atom thick i.e 0.34 nm thickness. It has properties like lightweight & flexible, good conductor of electricity. The theoretical surface area is 2630 m^2/g, which is vital for applications in adsorption, catalysis, and energy storage.

Being the building block for other carbon nanomaterials (like CNTs and fullerenes), graphene's versatility lies in its ability to be modified chemically and structurally. It can be functionalized to improve solubility, assembled into aerogels for lightweight structural frameworks, or doped to alter electronic properties.

1.2 Production of Graphene

Graphene can be synthesized through various top-down (Start with bulk graphite, then break it down into thinner layers) and bottom-up (Build graphene atom-by-atom from carbon-containing gases) techniques, each with trade-offs in terms of quality, scalability, cost, and application suitability. Below are the most widely used methods:

(i) Micromechanical Exfoliation

Micromechanical Exfoliation also known as Scotch Tape Method. It peels graphene layers off graphite with adhesive tape. It is top-down approach. This is the original method used by Geim and Novoselov (2004) to isolate graphene from graphite, earning them the Nobel Prize.

Process: A piece of graphite is repeatedly peeled using adhesive tape to thin down layers. The tape is then pressed onto a silicon wafer, leaving behind monolayer to few-layer graphene flakes.

It yields high quality defect free Graphene but is non-scalable and not suitable for bulk production.

(ii) Liquid Phase Exfoliation (LPE)

It is a top-down, scalable approach that uses sonication to exfoliate graphite in a solvent.

Process: Graphite is dispersed in a suitable solvent (e.g., NMP, water + surfactant), then subjected to ultrasonication. These breaks van der Waals forces between layers, producing single or few layer graphene. Centrifugation separates unexfoliated graphite.

It has advantages like cost-effective, scalable, and environmentally friendly but also limitations like Graphene quality is lower compared to micromechanical exfoliation or CVD.

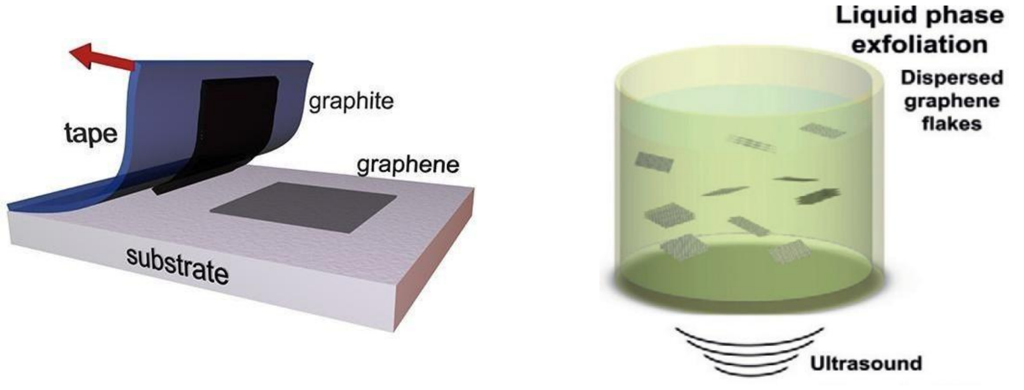


Fig 1.2: Schematic showing Micromechanical exfoliation and LPE.[3]

(iii) Chemical Vapour Deposition (CVD)

A bottom-up, high-quality method for growing large-area graphene on metal substrates.

Process: Hydrocarbon gas (e.g., methane) is decomposed at high temperature (~1000°C) on a metal substrate like copper or nickel under a reducing atmosphere (H_2/Ar). Carbon atoms deposit and self-assemble into a graphene layer.

Advantages: Produces continuous, large-area, monolayer graphene. Good for electronics, flexible displays, and sensor fabrication. Therefore, it is used in transparent electrodes, wearable electronics, and semiconductors.

Limitations: High cost and complex setup. Requires transfer process to move graphene from metal to insulating substrate, which may introduce defects.

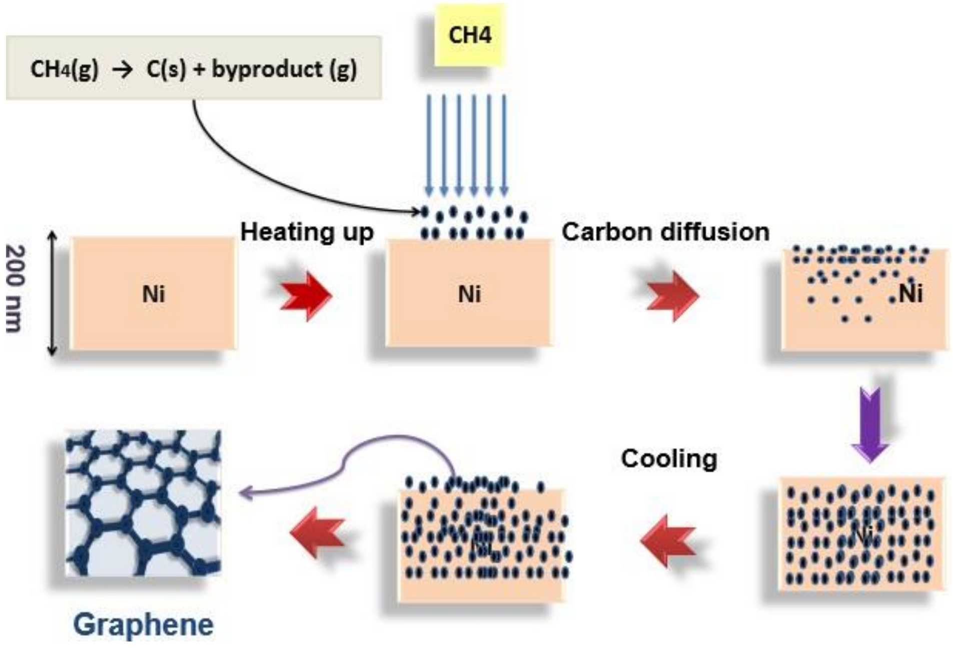


Fig 1.3: Schematic diagram showing CVD process on Nickel.[4]

1.3 Properties of Graphene

- With Tensile strength of ~130 GPa and Young's modulus ~1 TPa it is one of the strongest material ever tested.
- Flexibility: Can bend up to 25% strain without breaking.
- Thermal conductivity 5,000 W/mK which outperforms diamond.
- High electron mobility over 200,000 $cm^2/V{\cdot}s$ at room temperature (in suspended samples), much higher than silicon.
- Electrical conductivity up to 10^6 S/m due to its delocalized π-electron system.
- Graphene absorbs only ~2.3% of visible light per layer, making it nearly transparent, suitable for transparent conductive electrodes in touchscreens, OLEDs, and solar cells.

- Pristine graphene is relatively inert, but its surface can be functionalized (e.g., with oxygen, nitrogen, polymers) to enhance solubility or tailor reactivity.
- High surface area: Theoretical surface area up to 2630 m^2/g, which is excellent for adsorption, sensor design, and energy storage applications.
- Anticorrosive, Anti-bacterial
- Graphene exhibits an exceptionally low Joule heating effect

1.4 Applications of Graphene

Graphene's unique combination of electrical, mechanical, thermal, and chemical properties makes it a versatile material across a wide range of fields from next-gen electronics to clean energy and biomedicine.

- Electronics & Semiconductors: flexible displays, high-speed transistors, electronic sensors.
- Energy Storage & Generation: used as anodes in batteries, supercapacitors, Fuel cells.
- Composites & Coatings: lightweight structural composites, anti-corrosion coatings as graphene layers act as impermeable barriers to moisture, oxygen, and chemicals, transparent coatings for cultural artifacts.
- Biomedical Applications: drug delivery, biosensors.
- Filtration & Water Purification: Graphene oxide membranes, oil-water separation.
- Aerospace & Defense: EMI (electromagnetic interference) shielding, ultra-thin graphene films reduce radar visibility, useful in stealth technology, structural health monitoring.
- Sensors & IoT Devices: Gas sensors, wearable sensors, environmental monitoring.
- Graphene aerogels and composites show enhanced hydrogen adsorption capacity via physisorption and spillover mechanisms.

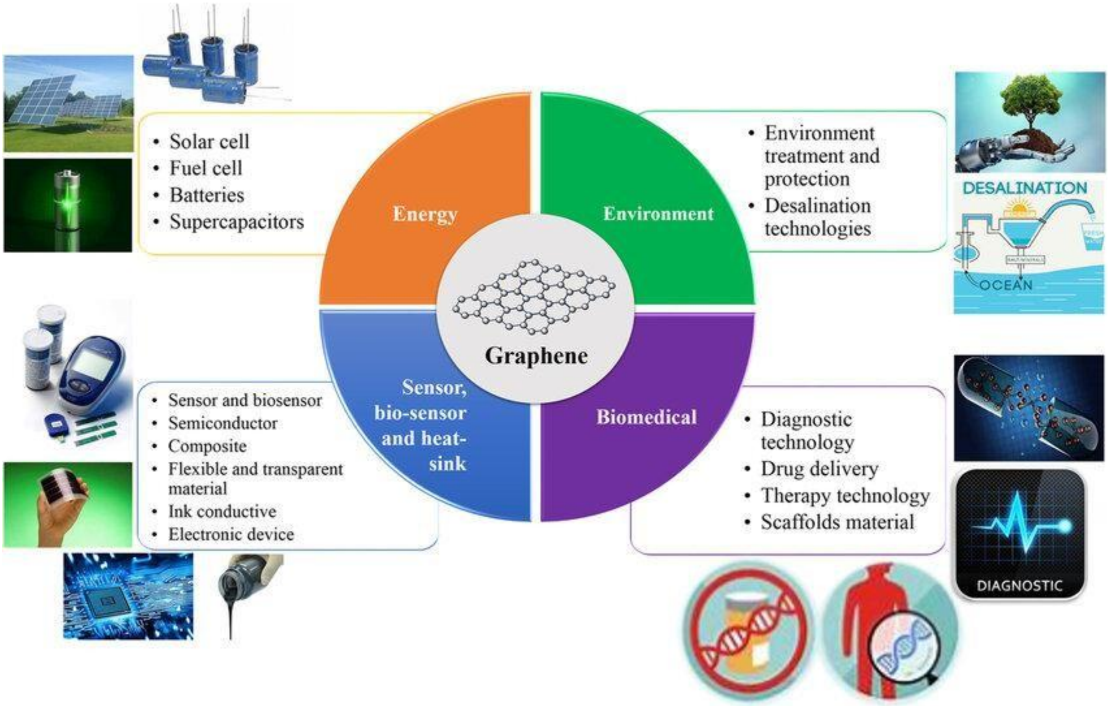


Fig 1.4: Various Applications of Graphene.[5]

1.5 Hydrogen Storage

Hydrogen is considered to be the future of green energy. But there are several challenges in it's production and storage methods as it is very light and highly inflammable gas. Some of these are:

- Safety: Because it's highly flammable and volatile.
- Energy Density: While hydrogen has a high gravimetric energy density (energy per mass), its volumetric energy density (energy per volume) is low, making it difficult to store large amounts compactly.
- Cost: Storing hydrogen is expensive due to the energy and material requirements.

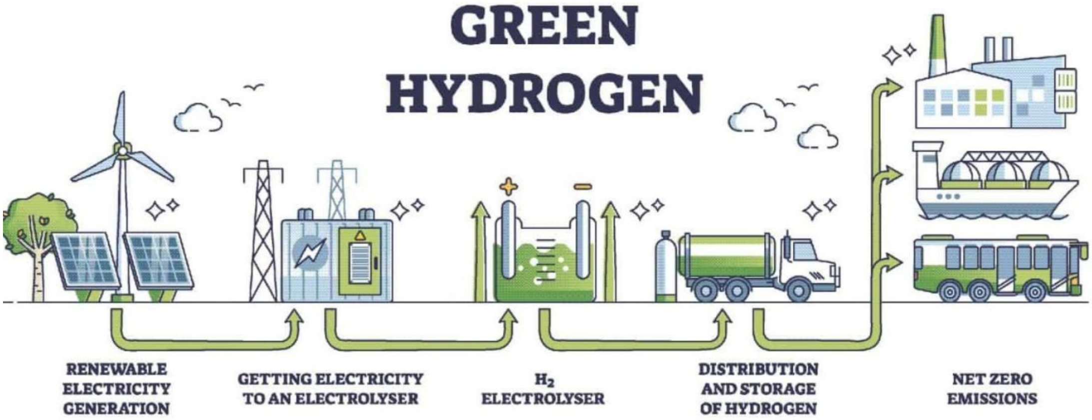


Fig 1.5: Green Hydrogen economy

1.5.1 Gas compression

Process:

Hydrogen gas is compressed and stored in high-pressure tanks typically at 350–700 bar (5,000–10,000 psi). Advanced tanks use materials like carbon fiber composites for improved strength and reduced weight.

Applications:

- Fuel cell electric vehicles (FCEVs) like Toyota Mirai and Hyundai Nexo.
- Industrial hydrogen delivery via tube trailers.
- Stationary backup power systems.

Limitations:

- Low volumetric energy density compared to fossil fuels.
- Requires heavy and expensive tanks to handle high pressure.
- Compression energy penalty (~10–15% of total energy).
- Safety concerns due to high-pressure containment.

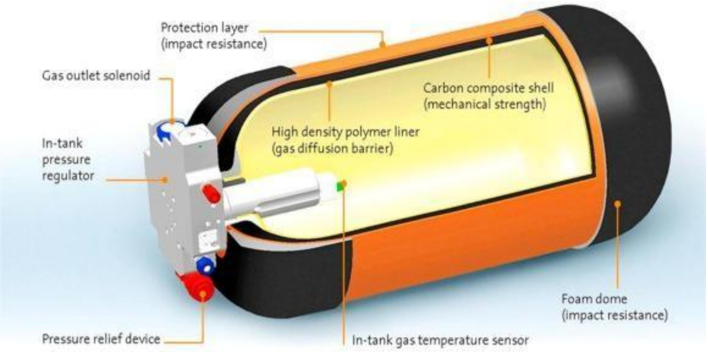


Fig 1.6: High Pressure H_2 Storage vessel

1.5.2 Liquid Hydrogen Storage

Process:

Hydrogen is liquefied by cooling it to −253°C and stored in cryogenic insulated tanks. Boil-off gas management is often included to prevent pressure build-up.

Applications:

- Used in spacecraft and launch vehicles (e.g., NASA’s Saturn V).
- Emerging use in long-distance hydrogen transport via cryogenic trucks or ships.
- Proposed for aviation fuel and large-scale energy storage.

Limitations:

- High liquefaction energy cost (~30–40% of hydrogen's energy).
- Boil-off losses due to heat ingress.
- Requires complex cryogenic infrastructure.
- Risk of embrittlement and leakage due to hydrogen diffusion.

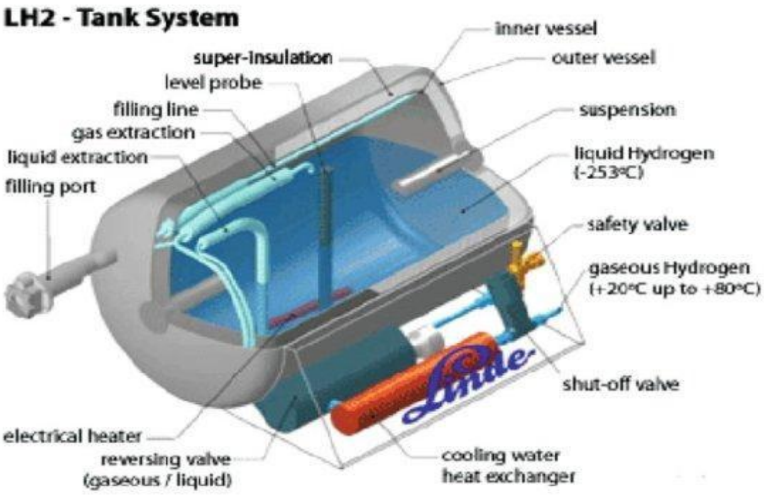


Fig 1.7: Cryogenic H_2 Storage pressure vessel

1.5.3 Metal Hydride Storage

Process:

Hydrogen is chemically absorbed by certain metals or alloys (e.g., $LaNi_5$, TiFe, MgH_2) to form metal hydrides. Hydrogen can be reversibly absorbed/desorbed under moderate temperatures and pressures.

Applications:

- Portable hydrogen storage for small fuel cells (e.g., laptops, drones).
- Backup power systems with limited space.
- Suitable for stationary hydrogen buffering.

Limitations:

- Heavy storage systems due to metal weight.
- Slow kinetics for hydrogen release (especially at low temperatures).
- Some materials require high desorption temperatures (>300°C).
- Limited gravimetric storage capacity (often <2 wt% H_2).

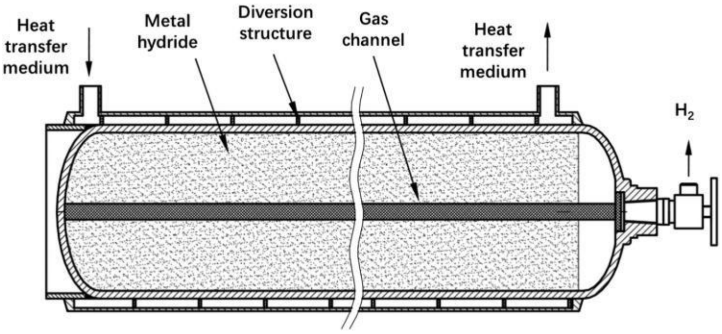


Fig 1.8: Metal hydride H_2 Storage tank

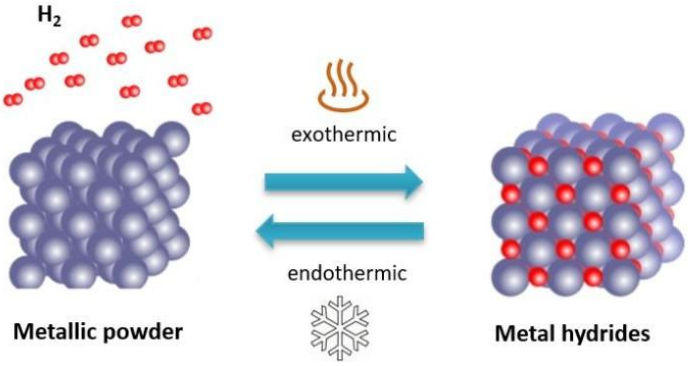


Fig 1.9: Schematic of metal hydride sorption process

1.5.4 Carbon-based Storage (Adsorption)

Process:

Hydrogen is physically adsorbed on high-surface-area materials such as:

- Activated carbon
- Carbon nanotubes (CNTs)
- Graphene and graphene aerogels

Hydrogen molecules are attracted to surface of porous materials through Vander Waals Force. Adsorption typically occurs at cryogenic temperatures (77 K) and moderate pressures (5–100 bar).

Applications:

- Being explored for vehicular and mobile energy systems.
- Promising for compact, low-weight hydrogen tanks.
- Relevant in research for future storage materials.

Limitations:

- Low hydrogen uptake at room temperature (usually <1 wt%).
- Requires cryogenic cooling to reach meaningful adsorption.
- Performance depends heavily on pore size, surface area, and structure.
- Still in experimental phase—limited commercial deployment.

1.6 Research Gap and motivation

The review of current hydrogen storage technologies namely compressed gas, liquid hydrogen, metal hydrides, and carbon-based adsorption systems; reveals a fundamental trade-off between storage capacity, safety, energy efficiency, and operational practicality. While compressed gas and liquid hydrogen storage are commercially utilized, they are hindered by issues such as high energy input, safety concerns, and infrastructure complexity. Metal hydrides offer better safety and reversibility but suffer from low gravimetric capacity and high desorption temperatures, limiting their real-world applicability.

Among the emerging alternatives, carbon-based materials especially nanostructured forms like CNTs, graphene, and their composites exhibit potential for lightweight, tunable, and reversible hydrogen storage via physisorption.

However, their performance is still restricted under ambient conditions due to weak van der Waals forces and low adsorption at room temperature.

This gap in performance, particularly under practical conditions, motivates the investigation into advanced carbon-based nanomaterials such as graphene aerogel nanocomposites, which offer synergistic advantages of high surface area, low density, hierarchical porosity, and tunable chemistry.

Thus, the goal of this research is to develop and evaluate graphene aerogel-based systems tailored for enhanced hydrogen storage, focusing on material synthesis, structural optimization, and performance benchmarking, with the aim of bridging the gap between experimental potential and commercial viability.

Chapter 2: Graphene Oxide (GO)

While pristine graphene is outstanding in terms of electrical, thermal, and mechanical properties, it's hydrophobic, chemically inert, and difficult to process in solution so making large-scale applications tricky. To overcome this, it can be chemically modified to make GO and rGO.

Graphene Oxide (GO) is made by introducing oxygen-containing functional groups into graphite. This makes it hydrophilic (water-friendly) and chemically reactive.

Reduced Graphene Oxide (rGO) is formed by partially restoring the sp^2-carbon network of GO—recovering some conductivity while retaining ease of processing.

These derivatives are more practical for bulk applications, especially in composites, energy storage, sensors, and biomedicine.

2.1 Graphene Oxide (GO): Structure & Chemistry

Graphene oxide (GO) is a chemically modified form of graphene that has garnered significant attention due to its unique structure, tunable chemistry, and extensive potential in applications such as energy storage, catalysis, environmental remediation, and biomedical engineering. Derived typically from graphite via oxidation, GO exhibits a highly functionalized, two-dimensional (2D) structure rich in oxygen-containing groups, which dramatically alters its properties compared to pristine graphene.

GO consists of a single atomic layer of carbon atoms arranged in a hexagonal honeycomb lattice, similar to graphene. However, what differentiates GO is the disruption of sp^2 hybridization due to the presence of various oxygenated functional groups. This leads to a partially sp^3 hybridized network with regions of sp^2-bonded carbon islands embedded within an sp^3 matrix.

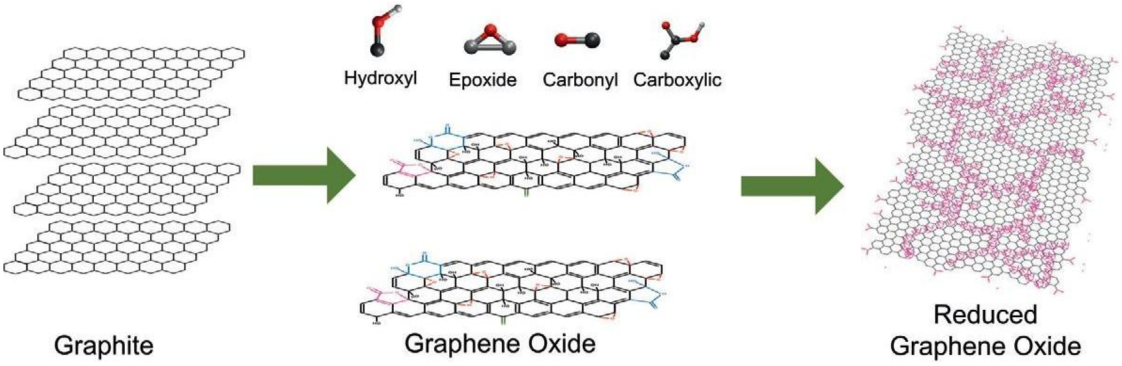


Fig 2.1: Structure of Graphite GO and rGO

2.1.1 Morphology and Layering

- GO sheets are generally wrinkled, crumpled, and corrugated, which increases their surface area.
- GO can exist in monolayer or multilayer forms, depending on the synthesis and exfoliation method.
- The interlayer spacing (typically 0.6–1.2 nm) is increased compared to graphite due to the presence of functional groups and water molecules.

2.1.2 Functional Groups and Distribution

The surface chemistry of GO is dominated by oxygen functionalities that decorate both the basal plane and edges of the sheets:

- Basal Plane: Contains epoxide (–O–) and hydroxyl (–OH) groups.
- Edges: Terminated with carboxylic (–COOH), carbonyl (C=O), and phenolic groups.
- The incorporation of oxygen atoms disrupts the delocalized π-electron network of graphene, converting sp^2-hybridized carbons to sp^3.This leads to a loss of electrical conductivity and significant changes in mechanical and optical properties.
- Reduction (chemical or thermal) can partially restore the sp^2 network, forming reduced graphene oxide (rGO), a material with intermediate properties between GO and graphene.

These groups not only make GO hydrophilic and dispersible in water but also offer sites for further chemical modification or reduction.

2.2 Graphene Oxide synthesis

Graphene oxide is typically synthesized through oxidation of graphite, employing methods such as:

- Hummers' method (and its modifications): Most commonly used, involves potassium permanganate ($KMnO_4$) and sulfuric acid (H_2SO_4).
- Brodie and Staudenmaier methods: Historical techniques involving strong oxidizers like nitric acid (HNO_3) and potassium chlorate ($KClO_3$).

These methods result in: Intercalation of oxidizing species between graphite layers, cleavage of π bonds and formation of epoxy and hydroxyl groups and subsequent exfoliation into single or few-layer GO sheets.

The abundance of oxygen groups makes GO chemically versatile and reactive. It can undergo reduction to restore electrical conductivity and tailor properties for specific applications. GO serves as an excellent precursor for nanocomposites, membranes, and sensors due to these modification capabilities. Understanding and controlling the structure–property relationships in GO are essential for tailoring it for specific applications, such as hydrogen storage, drug delivery, water purification, and composite reinforcement.

2.2.1 Hummers' Method

Hummers' method is a common method for the preparation of graphite oxide, where $KMnO_4$ solution is added to a mixture of graphite, $NaNO_3$, and H_2SO_4 as depicted in Fig 2.2.

Process Overview:

1. Oxidation: Graphite is mixed with $NaNO_3$ and concentrated H_2SO_4 under cooling (0–5 °C). $KMnO_4$ is added slowly to avoid overheating. The mixture is stirred for several hours.

2. Exfoliation: The strong oxidative conditions introduce oxygen-containing groups (epoxide, hydroxyl, carboxyl) into the graphite layers, expanding and weakening the interlayer van der Waals forces.
3. Termination: The reaction is quenched with ice and H_2O_2, turning the solution yellowish-brown and releasing Mn^{2+} ions.
4. Purification: The resulting GO is repeatedly washed with water, HCl, and centrifuged to remove residual ions and acids.

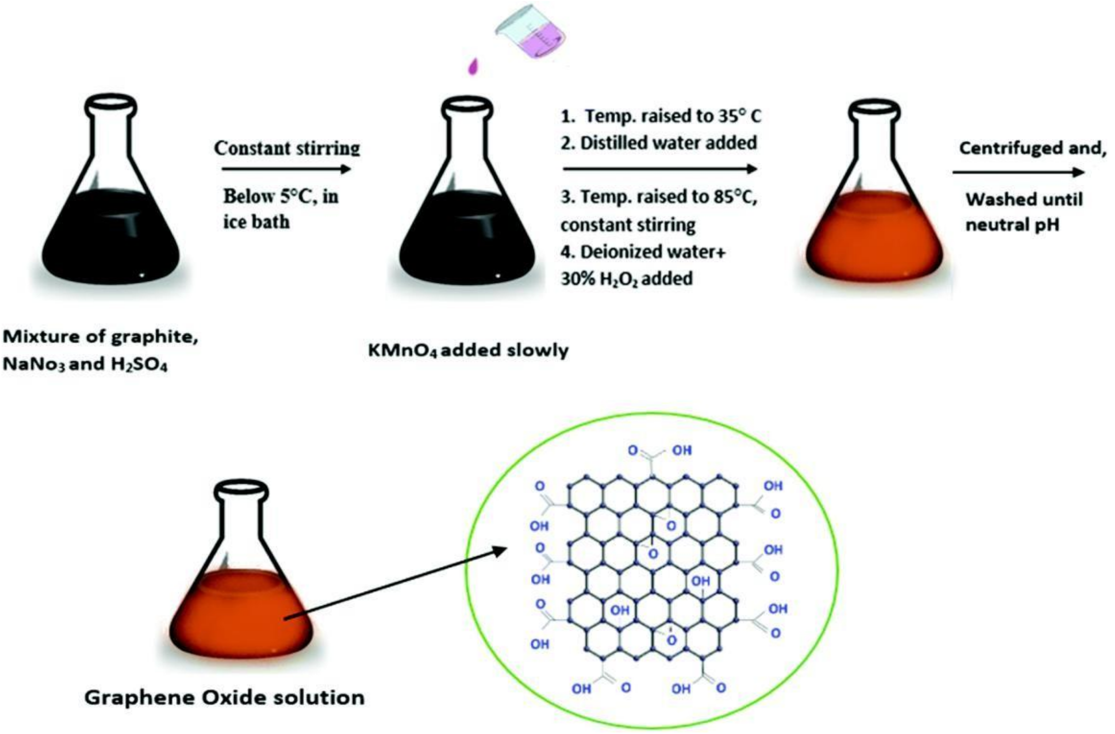


Fig 2.2: Schematic of Hummers method of GO production.[6]

2.2.2 Improved Hummers' Method

We'll use Improved Hummer's method for GO production as it reduces hazardous byproduct gases (e.g., NO_2, N_2O_4) and produces higher-quality graphene oxide.

Materials Required:

1. **Graphite powder**: Starting material.
2. **Sulfuric acid (H_2SO_4)**: Strong oxidizing medium.

3. **Phosphoric acid (H_3PO_4)**: Enhances oxidation efficiency and reduces hazardous gas formation.
4. **Potassium permanganate ($KMnO_4$)**: Primary oxidizing agent.
5. **Hydrogen peroxide (H_2O_2)**: Reduces residual manganese compounds.
6. **Deionized (DI) water**: Used for washing and dilution.
7. **Ice bath**: Controls exothermic reaction.

Experiment: GO Synthesis

Fig 2.3: Experimental procedure showing GO synthesis in lab. Hot plate (Top left) and Centrifuge (Bottom left).

Reaction of **1.5g Graphite** oxidises with $KMnO_4$ after 12h at 50°C

Quenching + 30% H_2O_2

Centrifuge to separate acidic residues

520mg -GO after drying for 24h @50°C

Improved Hummers' method (Process)

Combine concentrated H_2SO_4 and H_3PO_4 in a 9:1 volume ratio. (360:40 ml)

[Significance: H_3PO_4 reduces the generation of harmful gases (like NO_2) and enhances oxidation.]

Slowly add 3g graphite powder to the acid mixture while stirring vigorously.

[Significance: Ensures uniform dispersion and prevents agglomeration of graphite flakes.]

Gradually add 18g $KMnO_4$ to the acid-graphite mixture while maintaining the temperature below 10°C using an ice bath.

[Significance: $KMnO_4$ oxidizes graphite, forming graphite oxide. Controlled temperature prevents thermal runaway and hazardous gas generation.]

$$\mathbf{3C+KMnO_4+H_2SO_4 \rightarrow C_2O_3+MnSO_4+H_2O}$$

The reaction was then heated to 50 °C and stirred for 12 h.

[Significance: Allows complete oxidation of graphite to graphene]

Quenching : Slowly add ice-cold water to the reaction mixture. Then, add 3ml 30% H_2O_2 dropwise until bubbling ceases (indicating reduction of manganese residues).

[Significance: Quenching stops the oxidation reaction and removes residual oxidants]

$$\mathbf{MnO_2+H_2O_2 \rightarrow Mn^{2+}+O_2+H_2O}$$

Centrifuge the mixture to collect the GO and wash it repeatedly with DI water until the pH is neutral (~7). recco. x3

10% HCL x1 & then Ethanol x1 check pH(5-7)

[Significance: Removes acidic residues and unreacted chemicals.]

$$\mathbf{Mn^{2+}+2Cl^{-} \rightarrow MnCl_2}$$

Drying : Freeze-dry or air-dry the GO under vacuum overnight at 50-60 °C to obtain the final product

[Significance: Freeze-drying preserves the layered structure of GO.]

$$\mathbf{C+KMnO_4+H_2SO_4+H_3PO_4+H_2O_2 \rightarrow Graphene\ Oxide+Mn^{2+}+SO_4^{2-}+O_2+H_2O}$$

2.2.3 Comparison of different methods of synthesis

Hummers Method	**Modified Hummers' Method**	**Improved Hummers' Method**
Oxidizing Agents : $KMnO_4$, H_2SO_4, $NaNO_3$	Oxidizing Agents : $KMnO_4$, H_2SO_4, $NaNO_3$	Oxidizing Agents : $KMnO_4$, H_2SO_4, H_3PO_4
Graphite to $KMnO_4$ Ratio -1:3	Graphite to $KMnO_4$ Ratio -1:6	Graphite to $KMnO_4$ Ratio -1:3
High amounts of Toxic NO_2 and N_2O_4 gases as a byproduct	Reduced NO_2 and N_2O_4 gases	Minimal NO_2 and N_2O_4 gases due to H_3PO_4 stabilization
Quality : Moderate	Improved oxidation	Higher oxidation with fewer defects
Uncontrollable Temperature & least reaction time	Controllable Temperature with more time	Controllable Temperature with maximum reaction time

[Table 2.1: Comparison of Hummers' method, Modified hummers' and Improved Hummers' method of rGO preparation.]

An improved method for the preparation of graphene oxide (GO) is used in our experiment. Currently, Hummers' method ($KMnO_4$, $NaNO_3$, H_2SO_4) is the most common method used for preparing graphene oxide. We have found that excluding the $NaNO_3$, increasing the amount of $KMnO_4$, and performing the reaction in a 9:1 mixture of H_2SO_4/H_3PO_4 improves the efficiency of the oxidation process. This improved method provides a greater amount of hydrophilic oxidized graphene material as compared to Hummers' method or Hummers' method with additional $KMnO_4$. Moreover, even though the GO produced by our method is more oxidized than that prepared by Hummers' method, when both are reduced in the same chamber with hydrazine, chemically converted graphene (CCG) produced from this new method is equivalent in its electrical conductivity. In contrast to Hummers' method, the new method does not generate toxic gas and the temperature is easily controlled. This improved synthesis of GO may be important for large-scale production of GO as well as the construction of devices composed of the subsequent CCG.[7]

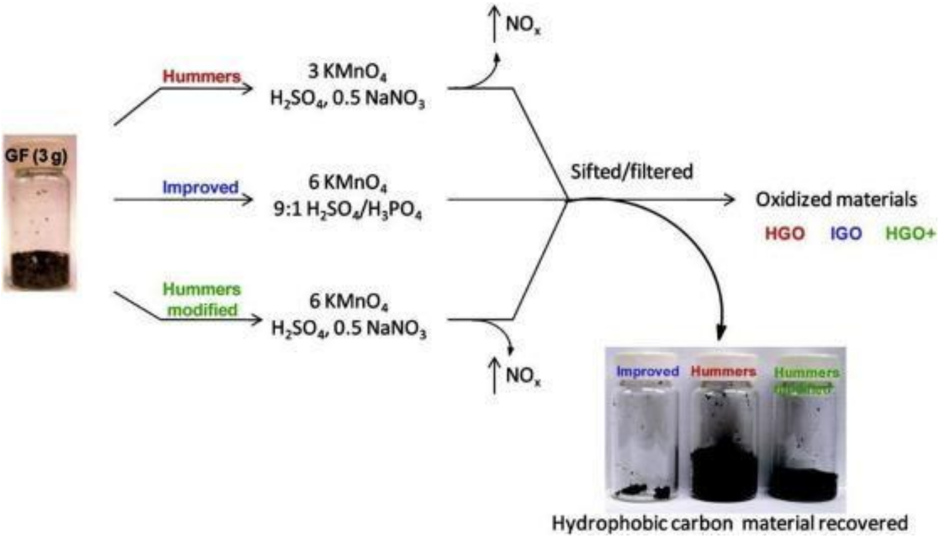
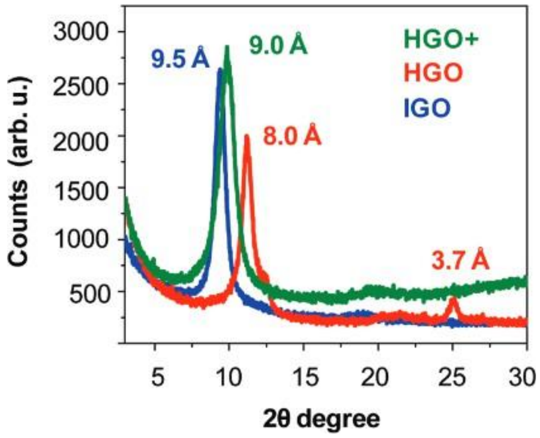


Fig 2.4: Comparison of Hummers, Modified hummers and Improved Hummers method of GO production. [7]



For XRD, the interlayer spacing of the materials is proportional to the degree of oxidation. The spacings are 9.5, 9.0, and 8.0Å for IGO, HGO+ and HGO, respectively.

Fig 2.5: XRD Spectra of Hummers (HGO), Modified Hummers (HGO+) and Improved Hummers method (IGO). [7]

2.3 GO Properties and Application

Graphene oxide exhibits a unique combination of physical, chemical, mechanical, and electronic properties that arise from its two-dimensional structure and the presence of oxygenated functional groups. These properties make GO highly versatile for a wide range of applications.

Physical and Structural Properties

- **Two-Dimensional Morphology**: GO exists as atomically thin sheets with large surface area and high aspect ratio.
- **Layered Structure**: The interlayer spacing (~0.6–1.2 nm) is larger than graphite due to the insertion of oxygen groups and water molecules.
- **Hydrophilicity**: Due to –OH, –COOH, and epoxy groups, GO readily disperses in water and other polar solvents.

Chemical Properties

- **High Reactivity**: Abundance of functional groups allows easy covalent and non-covalent functionalization.
- **Tunable Surface Chemistry**: Oxygen content and functional groups can be modified through reduction or chemical treatment.

- **Redox Activity**: GO can be reduced to reduced graphene oxide (rGO), partially restoring electrical conductivity and hydrophobicity.

Electrical Properties

- **Insulating or Semiconducting Nature**: GO is an electrical insulator due to disrupted sp^2 bonding, but rGO shows semiconducting or even conductive behavior depending on reduction extent.
- **Dielectric Behavior**: Exhibits interesting dielectric and capacitive behavior, useful in sensors and supercapacitors.

Mechanical Properties

- **High Mechanical Strength**: Though weaker than graphene, GO still retains significant tensile strength and flexibility.
- **Reinforcement Agent**: Used to improve mechanical properties of polymer nanocomposites.

Thermal Properties

- **Moderate Thermal Stability**: GO decomposes at ~200°C due to loss of oxygen groups.
- **Thermal Conductivity**: Lower than graphene but sufficient for some heat dissipation applications when partially reduced.

Applications of Graphene Oxide:

- **Supercapacitors**: GO and rGO are used as electrode materials due to their large surface area and redox activity.
- **Hydrogen Storage**: Porous GO structures and composites show promising hydrogen adsorption capacity due to surface area and functional groups.
- **Water Purification**: GO membranes can filter salts, heavy metals, dyes, and organic pollutants via size exclusion and adsorption.
- **Gas Sensors**: Sensitive to NO_2, NH_3, and other gases due to its large surface area and modifiable surface chemistry.

- **Adsorbent**: Used to remove metal ions and radioactive species from wastewater.
- **Drug Delivery**: Functionalized GO can carry anticancer or antibiotic drugs due to π–π stacking and hydrogen bonding.
- **Polymer Nanocomposites**: GO enhances strength, barrier properties, and flame retardancy in polymers like epoxy, PVA, and polystyrene.
- **Corrosion-Resistant Coatings**: Acts as a protective barrier layer in metal coatings.
- **Flexible Electronics**: GO thin films are used in transparent conductors and sensors.

Chapter 3: Reduced Graphene Oxide (rGO) Aerogel

Reduction of GO using chemical (e.g. hydrazine, ascorbic acid), thermal, or electrochemical methods removes oxygen groups and restores the conjugated graphene network However rGO is not fully equivalent to pristine graphene as it contains defects, vacancies, and residual oxygen but it regains substantial conductivity, making it suitable for practical engineering applications.

Most common Reducing agents are :Hydrazine (N_2H_4), Ascorbic Acid ($C_6H_8O_6$), Sodium Borohydride ($NaBH_4$)

3.1 Preparation of rGO

Graphene oxide (GO) is produced by oxidizing graphite, and then reduced chemically or thermally to form reduced graphene oxide (rGO), which approximates graphene. In chemical reduction process, GO is dispersed in a solvent (commonly water or ethanol) to form a uniform colloidal suspension. Then Reducing agents like Hydrazine Hydrate (N_2H_4), Sodium Borohydride ($NaBH_4$) or Ascorbic Acid (Vitamin C) is added to the suspension which reacts with the oxygen-containing groups and reduces them to produce rGO. The product is washed multiple times with deionized water and dried to obtain rGO.

Materials Required: Graphene Oxide (GO), Vitamin C, Di Water.

Procedure: Add GO in Water to make solution. Use Sonication to disperse evenly using probe sonicator for 30-60 mins or till the solution appear brownish-yellow. Add GO + L-Ascorbic Acid (C6H8O6) in ratio 1:2 and heat at 95^{0}C for 1 hr. Observe colour change from brown to black indication reduction of oxygen groups. Add cross linking agents like polymers or organic molecules like L-Ascorbic acid & heat it at 95^{0}C for 6 hr in autoclave. This process is called hydrothermal reduction method to obtain Reduced Graphene Hydrogel (RGH). Perform dialysis in water to remove soluble species. Keep in oven at 60^{0}C for 24 hr for drying to obtain → **rGO.**[8]

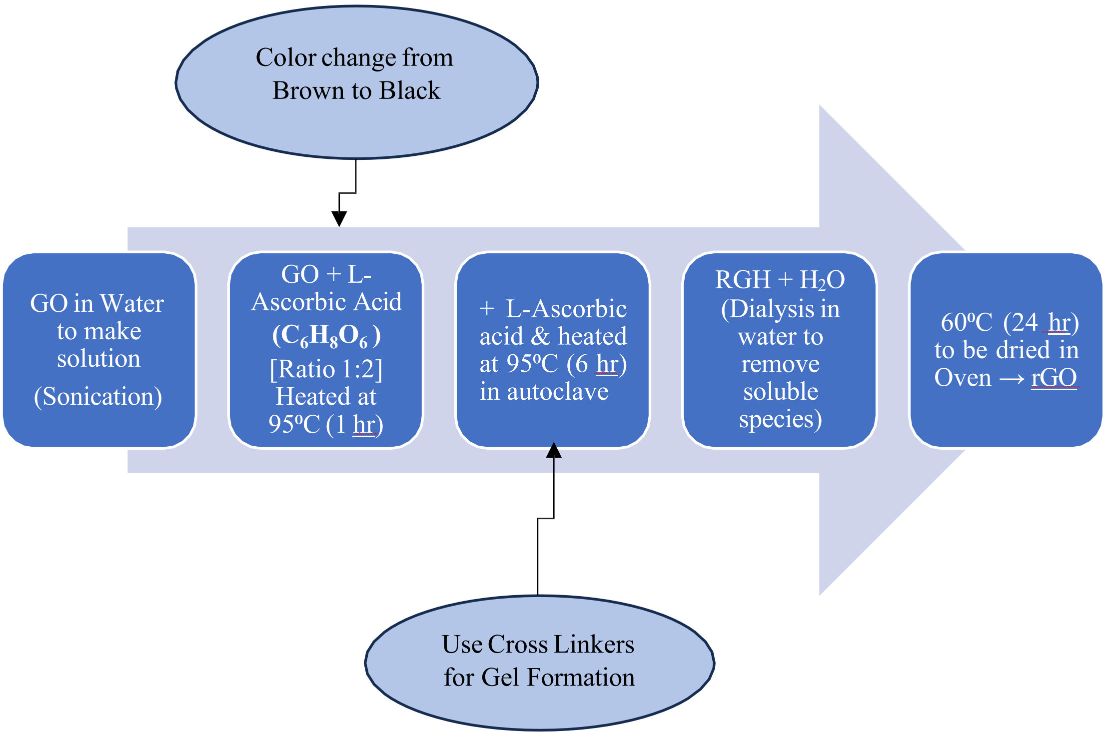


Fig 3.1: GO → rGO (Process removal of oxygen-containing functional groups)

3.2 Comparison of GO, rGO and Graphene

Property	**Graphene Oxide (GO)**	**Reduced Graphene Oxide (rGO)**	**Graphene**
Structure	2D sheet with disrupted sp^2 network, oxygenated functional groups	Partially restored sp^2 structure, some residual oxygen groups	Perfect sp^2-hybridized carbon atoms in hexagonal lattice
Functional Groups	–OH, –COOH, epoxide, carbonyl (highly functionalized)	Fewer –OH, –COOH groups, depending on reduction	No oxygen groups; chemically inert

Property	**Graphene Oxide (GO)**	**Reduced Graphene Oxide (rGO)**	**Graphene**
Hybridization	Mix of sp^2 and sp^3 hybridization	Mostly sp^2, some sp^3 due to remaining defects	Pure sp^2 hybridization
Electrical Conductivity	Insulator or poor conductor (~10^{-3} S/m)	Moderate conductor (~10^0–10^2 S/m)	Excellent conductor (~10^6 S/m)
Thermal Conductivity	Low (~5 W/mK)	Intermediate	Very high (~5000 W/mK)
Dispersibility	Easily disperses in water/polar solvents (hydrophilic)	Limited dispersibility	Poor dispersibility (hydrophobic)
Mechanical Strength	Lower than graphene due to defects	Improved, but not as strong as pristine graphene	Extremely strong (Young's modulus ~1 TPa)
Optical Appearance	Brown/yellowish	Dark brown to black	Transparent (monolayer), black in multilayer form
Bandgap	Exists (~2.2–3.5 eV, tunable)	Smaller bandgap, quasi-zero depending on reduction	Zero bandgap (semi-metallic)
Synthesis	Oxidation of graphite (e.g., Hummers method)	Reduction of GO via chemical, thermal, or electrochemical methods	Chemical vapor deposition (CVD), exfoliation, epitaxy

Property	Graphene Oxide (GO)	Reduced Graphene Oxide (rGO)	Graphene
Cost & Scalability	Cheap and scalable	Moderate, scalable	Expensive and less scalable
Key Applications	Membranes, adsorbents, sensors, drug delivery	Supercapacitors, batteries, electronics, catalysts	High-end electronics, photonics, nanoelectronics

[**Table 3.1:** Comparison of GO, rGO and Graphene.]

3.3 Synthesis of rGO Aerogel

Reduced Graphene Oxide (rGO) aerogels are ultralight, highly porous 3D structures derived from graphene oxide (GO), with improved electrical conductivity, mechanical integrity, and functional surface area due to partial removal of oxygen-containing groups.

Materials Required:

- Graphene Oxide (GO) aqueous dispersion (typically 1–5 mg/mL)
- Reducing agent:
    - Chemical: *L-ascorbic acid*, hydrazine(N_2H_4), urea, $NaBH_4$
    - Thermal: hydrothermal or annealing at 200–1000 °C
- Deionized water
- Optional: cross-linkers (e.g., ethylenediamine), templates, surfactants

The synthesis follows a three-step process:
(i) Gelation, (ii) Reduction, and (iii) Drying to form the aerogel.

Step 1: Preparation of GO Dispersion

- Disperse graphene oxide in deionized water using ultrasonication (30–60 mins) to obtain a uniform GO suspension.
- Typical GO concentration: 2–10 mg/mL depending on desired porosity and density.

Step 2: In-situ Reduction & Gelation

- Add a reducing agent like L-ascorbic acid (Vitamin C) to the GO dispersion in a 1:2 to 1:4 molar ratio (GO to reducing agent).
- Stir and transfer the mixture into a sealed Teflon-lined autoclave.
- Hydrothermal treatment: Heat the solution at 90–180 °C for 6–12 hours.
    - This step simultaneously:
        - Reduces GO to rGO by removing oxygen groups.
        - Induces self-assembly into a 3D hydrogel via π–π stacking, van der Waals interactions, and hydrogen bonding.
- After cooling, a black, monolithic rGO hydrogel is obtained.

Optional Modifications:

- Add metal nanoparticles, polymers, or doping agents (e.g., N, B) before hydrothermal step to tailor conductivity or storage capacity.

Step 3: Washing

- Wash the hydrogel with deionized water several times to remove unreacted chemicals and residual ions.

Step 4: Drying to Form Aerogel

- To preserve the porous 3D structure, dry using one of the following methods:
    - Freeze-drying (lyophilization): Freeze the hydrogel at –20 to –80 °C, then sublimate under vacuum for 24–48 hours.
        - Best for retaining high porosity and low density.

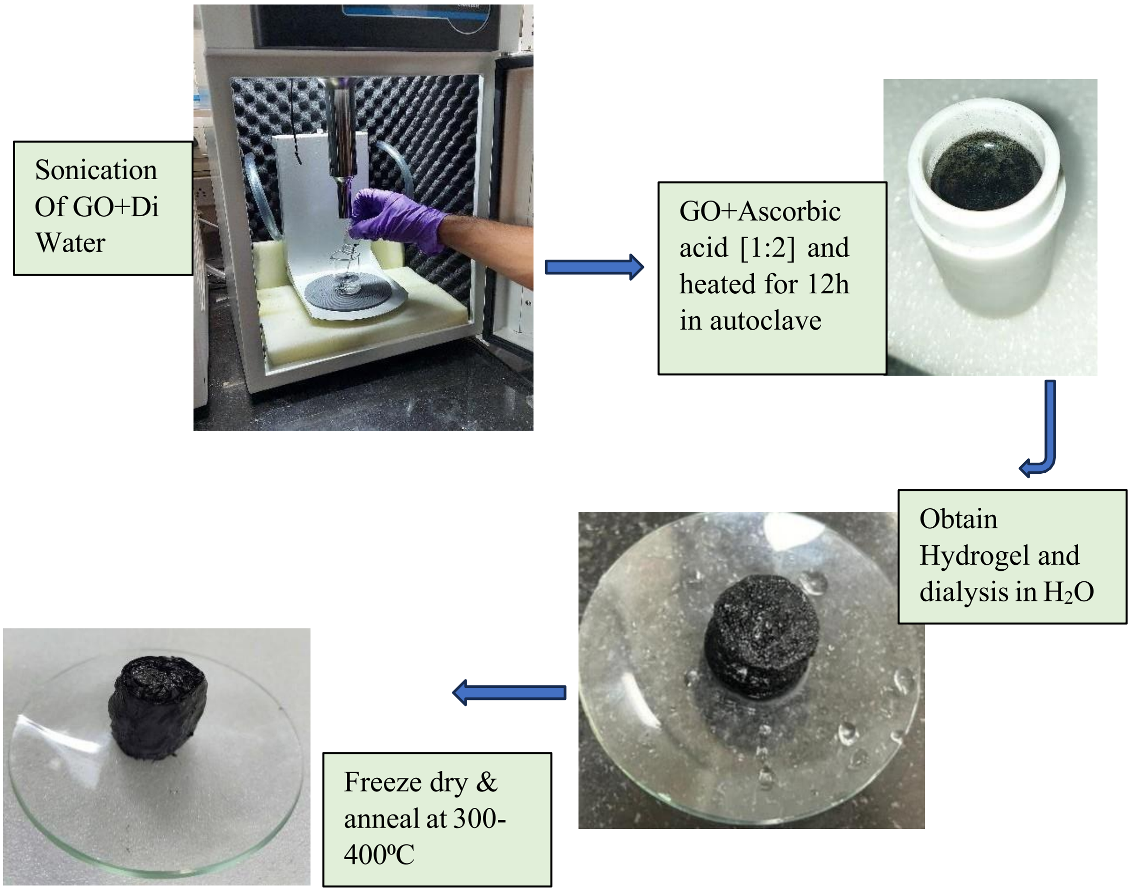
  - Supercritical CO_2 drying: Effective but expensive.
  - Ambient drying: Causes partial collapse, less porous.

Post-Treatment

- Thermal annealing at 200–800 °C under inert gas (N_2, Ar) can further reduce oxygen content and enhance conductivity.
- Structural analysis via **SEM, BET, Raman, XRD** is to be done after this.

Experiment: rGO Aerogel preparation in lab

[**Fig 3.2:** GO → rGO aerogel fabrication in lab]



3.4 Properties of rGO Aerogel

- **Ultralight Density:** rGO aerogels are among the lightest known solid materials with density around ~0.02–0.05 g/cm^3
- **High Porosity:** Aerogels are highly porous with porosity >90%
- **Electrical Conductivity:** Typical rGO aerogels exhibit conductivities in the range of 2–4 S/m, which is significantly higher than their GO counterparts
- **Thermal Insulation:** Despite their electrical conductivity, rGO aerogels demonstrate excellent thermal insulation properties, with low thermal conductivity (~0.02–0.05 W/m·K)
- **Hydrophobicity:** While pristine GO is hydrophilic, rGO aerogels can be tailored to exhibit hydrophobic or superhydrophobic surfaces through reduction and surface functionalization
- **Mechanical Flexibility & Compressibility:** Due to their interconnected networks, rGO aerogels can withstand compressive strain and exhibit recoverable deformation
- **Chemical Tunability:** Owing to the presence of residual oxygen functional groups and the graphene backbone, rGO aerogels are chemically modifiable. They can be doped (e.g., with N, B, or S) or decorated with nanoparticles, expanding their utility in electrocatalysis, gas sensing, or drug delivery

Chapter 4: Results and Discussion

To evaluate the structural, morphological, and physicochemical properties of the synthesized rGO aerogel, a comprehensive set of characterization techniques was employed with a focus on their suitability for hydrogen storage applications.

Scanning Electron Microscopy (SEM) was used to examine the surface morphology and internal architecture of the aerogel. This provided visual confirmation of the 3D porous network, essential for maximizing surface area and facilitating gas diffusion. The interconnected sheet-like structure is critical for enhancing adsorption and ensuring mechanical stability.

X-ray Diffraction (XRD) analysis helped identify the crystallinity and interlayer spacing of the material. Monitoring the shift and intensity of characteristic peaks allows us to confirm the reduction of GO and the partial restoration of the graphitic structure, which directly influences conductivity and adsorption behavior.

Brunauer–Emmett–Teller (BET) surface area and pore size analysis were crucial in determining the textural properties of the aerogel. A high surface area and optimal pore distribution are key factors in improving hydrogen uptake, as they provide more active sites for adsorption.

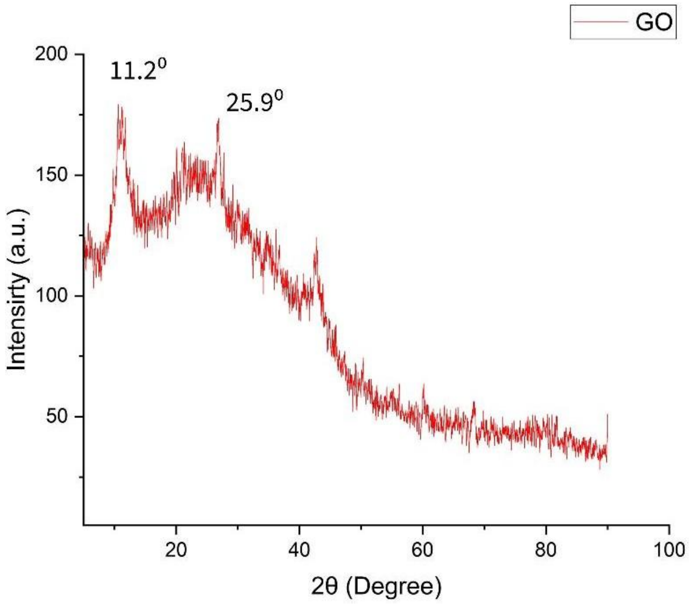
Raman Spectroscopy was employed to investigate the structural disorder and degree of reduction. The D and G bands give insight into the defect density and the sp^2/sp^3 carbon content ratio, which affect both conductivity and chemical reactivity—important for tuning hydrogen interaction with the material.

Together, these characterization techniques provided a comprehensive understanding of the material's structure-property relationships, confirming that the synthesized rGO aerogel possesses the fundamental traits required for efficient hydrogen storage.

4.1 X-Ray Diffraction (XRD) plot of rGO Aerogel

Sample 1 : Partially reduced GO

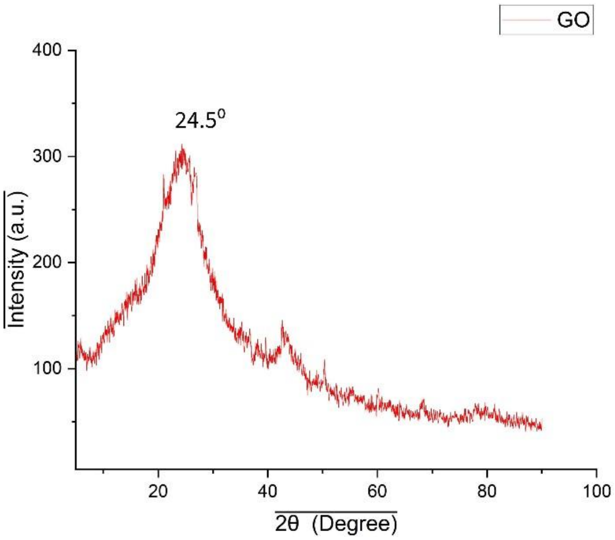
- **Reduction Temperature 80°C, Time- 24 hrs**



[Fig 4.1: XRD result- Partially reduced GO]

Sample 2 : Reduced GO

- **Reduction Temperature 150°C, Time- 72 hrs**



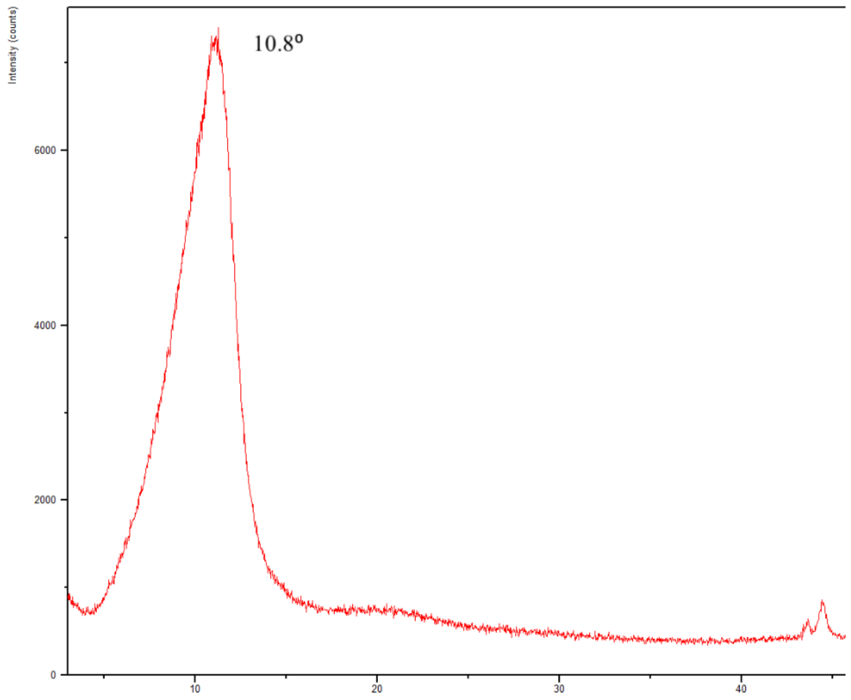
[Fig 4.2: XRD result- Reduced GO]

Check Peak Positions:

- In the case of **graphene oxide (GO)**, a distinct diffraction peak typically appears around **10–12° (2θ)**, corresponding to the (001) plane. This peak indicates the presence of oxygen-containing functional groups and increased interlayer spacing due to oxidation.
- Upon reduction to **reduced graphene oxide (rGO**), this peak diminishes or disappears, and a broader peak emerges around **24–26°**, which is attributed to the (002) plane of partially restored graphitic domains. This shift confirms the removal of oxygen functionalities and partial restacking of graphene layers.
- For comparison, **pristine graphite** shows a sharp, well-defined peak at approximately **26.5°**, indicating a highly ordered crystalline structure.

Sample 3 : Graphene Oxide (GO)

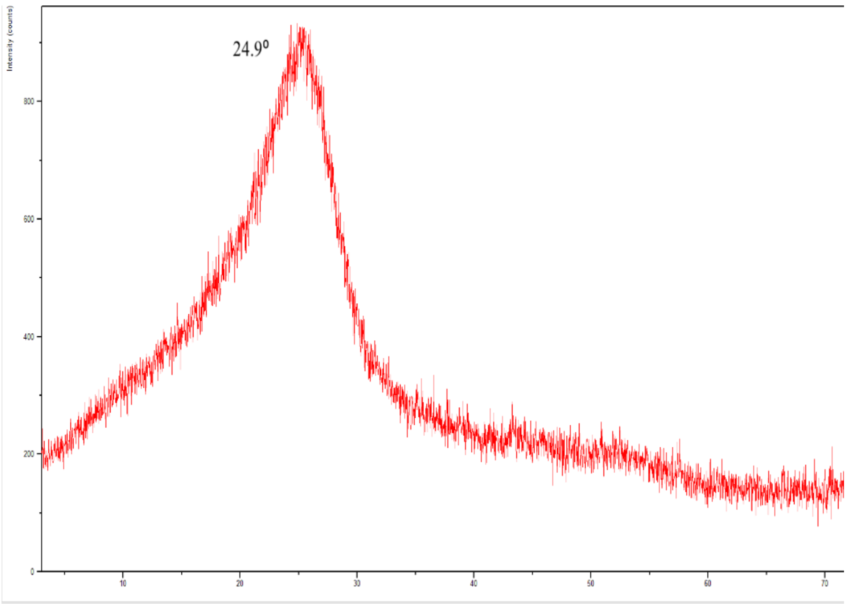
- **Reduction Temperature 50°C, Time- 24 hrs**



[Fig 4.3: XRD result- GO]

Sample 4 : rGO Aerogel

- **Reduction Temperature 50°C, Time- 24 hrs**



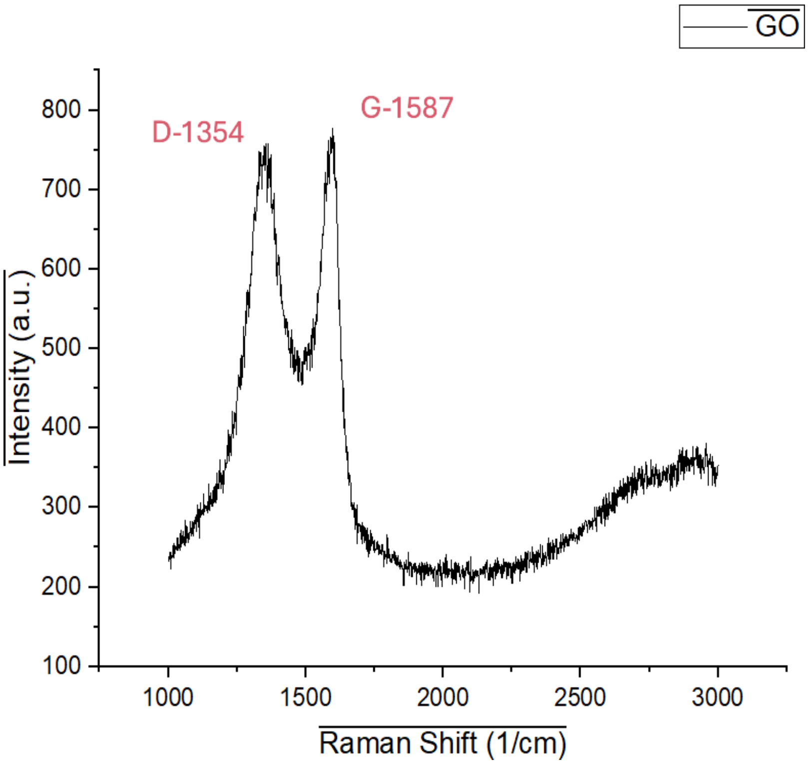
[Fig 4.4: XRD result- rGO Aerogel]

The broadness and lower intensity of the rGO peak reflect structural disorder and a turbostratic stacking of sheets, which is desirable for maintaining high surface area in aerogel form.

This transition in peak position and shape provides direct evidence of successful reduction and structural transformation from GO to rGO.

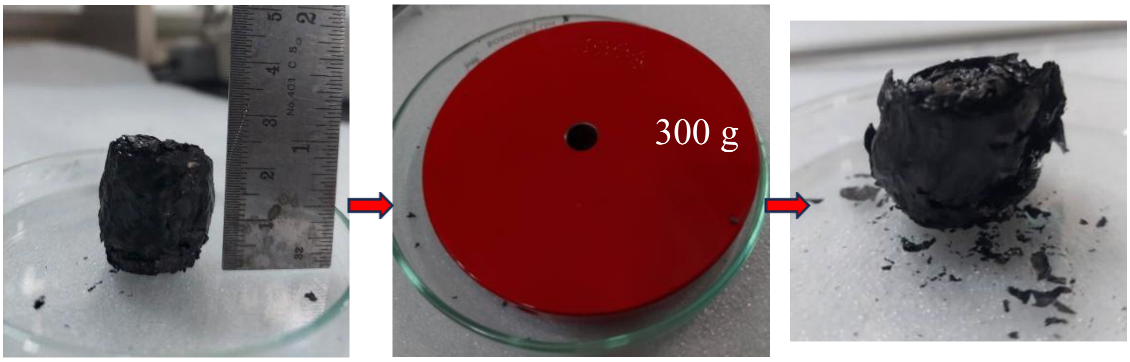
4.2 Raman Spectroscopy of GO

- D-band (~1350 cm^{-1}) shows defects, edges, and functional groups.
- G-band (~1580 cm^{-1}) represents graphitic carbon
- Higher I_D/I_G ratio means more disorder. Which suggests more adsorption sites.
- Typically, for GO, I_D/I_G ~ 0.8 to 1.1 and for rGO >1
- 2D-band (~2700 cm^{-1}) appears weak in GO, rGO typically have a stronger and sharper 2-D band



[Fig 4.5: Raman spectroscopy result- GO]

4.3 Test for Mechanical Strength & Elasticity



[Initial Graphene Aerogel (wt-62mg)] ~ [After applying 300g compressive load]

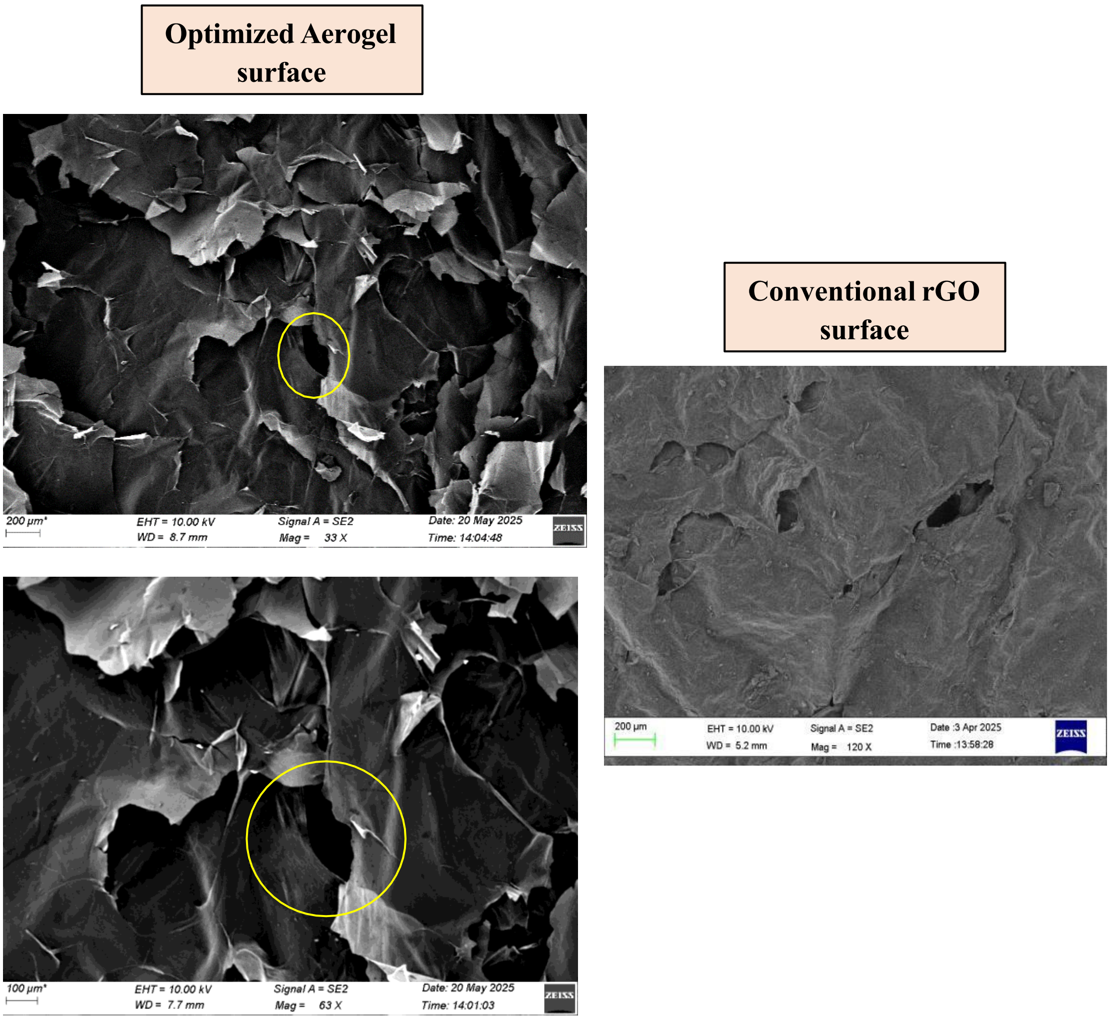
[Fig 4.6: Elastic compression test. Aerogel before (left) and after (right)**]**

Results:

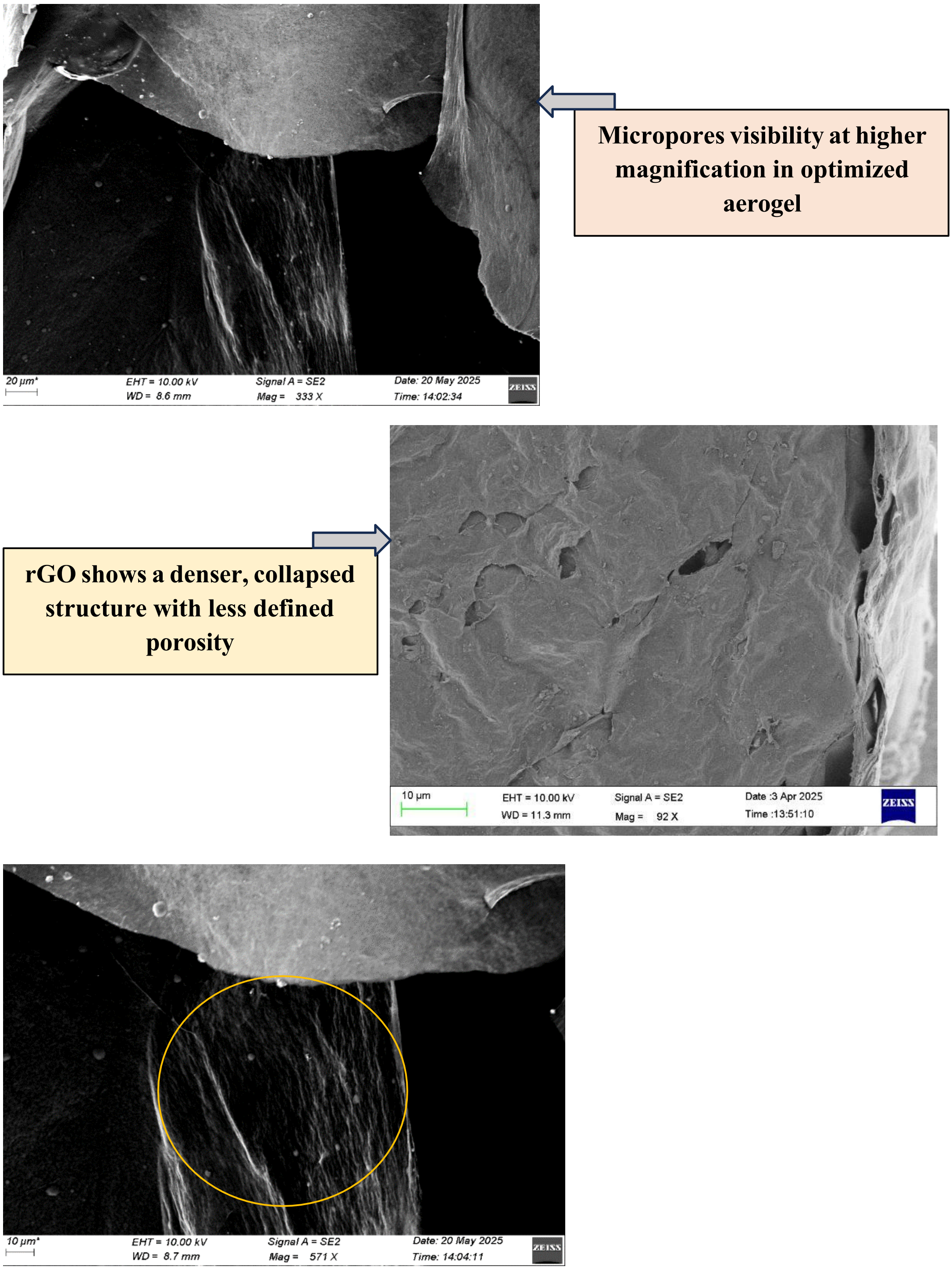
- The rGO aerogel exhibited approximately **70% elastic recovery** after repeated compression cycles (load: 300 g applied five times), indicating decent structural resilience and flexibility under cyclic mechanical stress.

- Density of aerogel is found to be **16.8 mg cm^{-3}** which is in the range of [10-30 mg cm^{-3}]. This low density supports the material's potential for lightweight applications and efficient volumetric energy storage.
- The structure showed noticeable weakening upon exposure to ambient moisture, suggesting **hydrophilic interactions** or incomplete reduction of GO. This highlights the need for surface modification or hydrophobic treatment for long-term mechanical stability in humid environments.

4.4 Test for Morphology: SEM



[Fig 4.7: SEM images of Aerogel at 200µm(Top-left), 100µm(Bottom-left) and rGO surface at 200µm (right). Pores are highlighted**]**



Micropores visibility at higher magnification in optimized aerogel

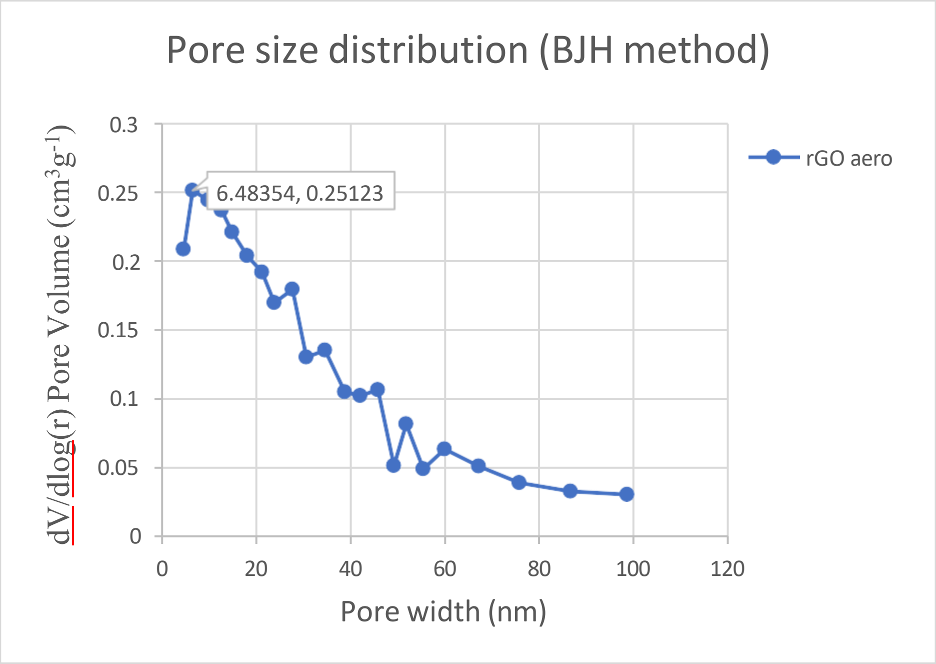
rGO shows a denser, collapsed structure with less defined porosity

[Fig 4.8: SEM images of Aerogel at 20μm(Top-left), 10μm(Bottom-left) and rGO surface at 10μm (Centre-right). Pores are highlighted**]**

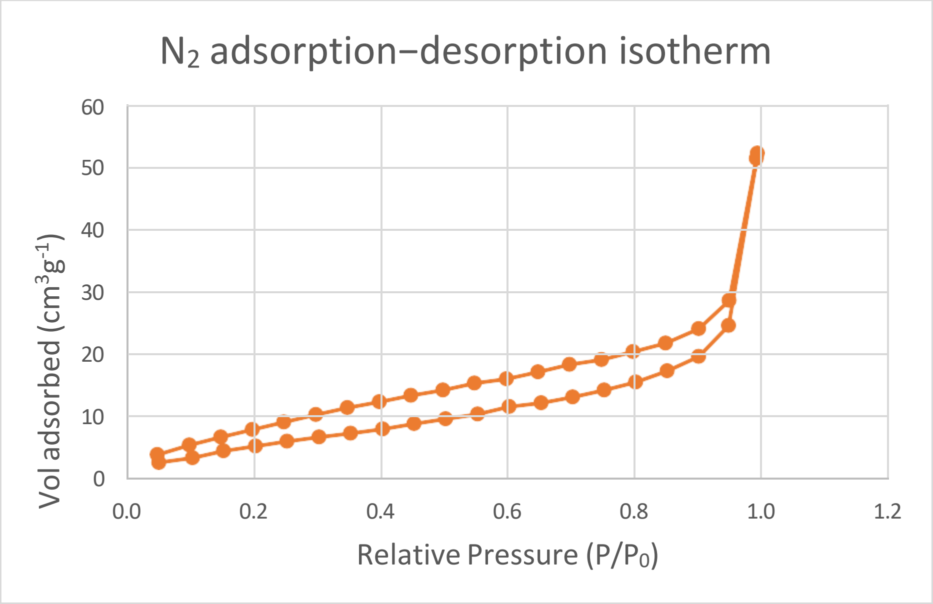
- SEM analysis clearly demonstrates that the rGO-based aerogel exhibits a highly porous, interconnected network of wrinkled nanosheets with well-defined macro- and mesopores (Fig 4.7), which are significantly more favourable for gas diffusion and storage compared to the relatively dense and less porous structure observed in conventional rGO.

- Micropores visibility at higher magnification in optimized aerogel (Fig 4.8) whereas rGO shows a denser, collapsed structure with less defined porosity.

4.5 Test for Surface Area- BET & BJH : rGO Aerogel



[Fig 4.9: BJH analysis Graph of Aerogel**]**



[Fig 4.10: BET analysis Graph of Aerogel**]**

The textural properties of the synthesized rGO aerogel were investigated using Brunauer–Emmett–Teller (BET) (Fig 4.10) surface area analysis and Barrett–Joyner–Halenda (BJH) (Fig 4.9) pore size distribution. The BET surface area was found to be **518.2 m²/g,** indicating a highly porous structure with substantial accessible surface area—an essential trait for applications involving adsorption and gas storage, such as hydrogen uptake.

The total pore volume was measured to be **0.261 cm³/g,** and the average pore radius calculated from the BJH model was approximately **3.2 nm, placing the majority of the pores in the mesoporous range (2–50 nm).** Mesopores are particularly beneficial for enhancing gas diffusion kinetics and minimizing diffusion resistance within the material matrix. Mesopores facilitate fast gas diffusion, and when coupled with high surface area, they contribute significantly to overall H_2 uptake, especially at room temperature.

The **nitrogen adsorption–desorption isotherm exhibits a Type IV behavior with a well-defined H3-type hysteresis loop**, which is characteristic of slit-like pores formed by the aggregation of plate-like particles such as graphene sheets. This further supports the SEM observation of wrinkled and layered structures in the aerogel network.

Overall, the BET and BJH results confirm that the rGO aerogel possesses a highly mesoporous architecture with a large surface area and favorable pore structure, making it a strong candidate for gas storage and energy-related applications

Chapter 5: Conclusion and scope for future work

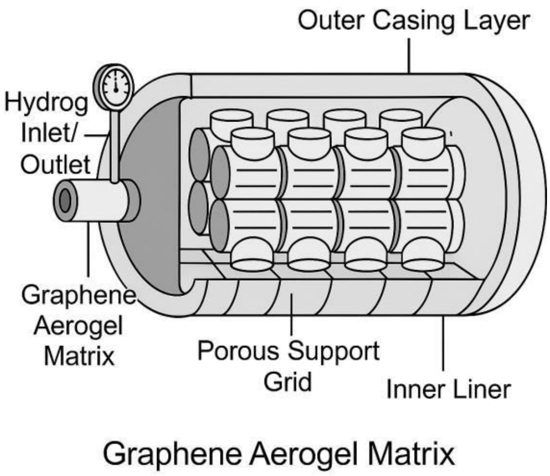
5.1 Conclusion

Test performed	Key Results	H_2 Storage Relevance
BET surface analysis	512.8 m²/g	High adsorption capacity
BJH	Pore volume - 0.261 cm^3/g Pore size – 3.2 nm	Mesoporous, - Ideal for physisorption
SEM (Surface morphology)	Porous, open structure	Fast diffusion, low density
XRD (X ray diffraction)	Broad 002 peak at 24.9°	Amorphous graphene, more pores
Raman Spectroscopy	ID/IG = 1.2	High defect density for adsorption

[Table 5.1: Test Results and conclusion.]

The comprehensive material characterization confirms the successful synthesis of a structurally optimized rGO aerogel tailored for hydrogen storage applications. The **BET surface area** of over **500 m²/g** and **mesoporous structure** (3.2 nm pore size, 0.261 cm³/g pore volume from BJH analysis) suggest a high capacity for physical adsorption, which is crucial for efficient hydrogen uptake. SEM imaging revealed a highly porous, interconnected morphology that promotes rapid gas diffusion and contributes to the material's ultralight nature.

XRD analysis exhibited a broad (002) peak around **24.9°**, indicating an amorphous graphitic structure with enhanced interlayer spacing and pore accessibility. Raman spectroscopy, with an ID/IG ratio of 1.2, further confirmed the presence of structural defects, which act as additional active sites for hydrogen adsorption. Together, these results demonstrate that the synthesized rGO aerogel offers a favorable combination of high surface area, mesoporosity, defect density, and open architecture, making it a promising candidate for hydrogen storage systems under ambient conditions.



[Fig 5.1: Schematic representation of aerogel matrix in pressure vessel.]

5.2 Future Work Scope

- Further enhancement could be achieved by nitrogen, boron doping to increase adsorption sites, or by introducing Titanium (Ti) or palladium (Pd) nanoparticles to leverage hydrogen spillover.
- Hydrogen adsorption is maximized in micropores ranging from 0.6 to 1.2 nm due to optimal van der Waals interaction; increasing microporosity via strategic doping could significantly improve storage capacity.
- The U.S. DOE specifies a hydrogen uptake target of ~5.5 wt% at operating conditions of −40 to 60 °C and 100 bar. While this project focused on optimizing structural and surface properties, future work could benchmark storage capacity against this standard.

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