

Review

Cellulose, graphene and graphene-cellulose composite aerogels and their application in water treatment: a review

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Abstract

Due to their unique properties such as outstanding specific surface areas, aerogels can potentially offer a cost-effective and efficient water and wastewater treatment solution thereby solving the worldwide ever-growing water pollution conundrum. This review explores the developments and progress in the application of cellulose, graphene, and cellulose-graphene composite aerogels in addressing water treatment challenges. The objective of this study was to critically review the state-of-the-art with respect to synthesis methods and properties of cellulose, graphene, and cellulose-graphene composite aerogels and their application, effectiveness and performance in water and wastewater treatment. It is evident from the review that cost-effective aerogel production methods that can be scaled up easily still need to be developed to enable commercial viability of aerogels in water treatment. Furthermore, the mechanical properties of aerogels, particularly stiffness and strength, are a hinderance to expanding their utility. The potential for functionalization of the aerogels through fabrication of aerogel nanocomposites or incorporation of functional groups was recognized as a viable method to enhance their properties and effectiveness in water treatment and other advanced industrial-scale applications. Graphene-cellulose composite aerogels have enhanced properties, removal capacities and efficiencies for pollutants from water compared to aerogels prepared from the individual materials. The regeneration ability and reusability of the composite aerogels make them an attractive option for practical applications. Although the potential of cellulose, graphene, and cellulose-graphene composite aerogels in water treatment applications is apparent, the impact of their production, utilization and subsequent disposal on sustainability and the environment is yet to be fully studied.

Keywords Cellulose aerogels · Graphene oxide aerogels · Composite aerogels · Water treatment · Waste water · Removal of heavy metals · Super critical drying

1 Introduction

Aerogels are highly porous, solid materials in which the inner structure is filled with or composed of air [1]. Aerogels possess a diverse range of exceptional characteristics, such as high porosity, low density, large surface area, excellent thermal insulation, high mechanical strength, remarkable optical properties, and chemical stability, which makes them an attractive material for a broad range of applications such as energy storage, thermal insulation, catalysts, sensor, oil spill clean-up, personal care and water treatment [2–5].

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Since, the invention of aerogels by Kistler in the 1930s and further developments in the 1980s, aerogel synthesis techniques have been largely refined and the development of aerogels included many more materials with vast properties. These include superhydrophobic aerogels with self-cleaning properties, sulfonated zirconia aerogels, titanium dioxide aerogels, aerogel composites, organic cellulose based aerogels, carbon aerogels, hybrid organic–inorganic aerogels, allophanes and many more [6]. The introduction and development of varying types of aerogels was accompanied by research based on comprehending the properties of aerogels which include mechanical, luminescent, conducting, magnetic and thermal properties.

Due to better understanding of aerogel properties, current research has been mainly focused on their industrial applications. Some of the potential applications include wastewater treatment, retainment of nuclear waste, carbon dioxide capture, chemical sensors, water repellent coatings, heterogenous catalysts, metal casting moulds, energy storage devices, acoustic transducers, pharmaceutical drug carriers, and non-flammable cryogenic insulators [4, 7].

The aim of this study was to bring to the forefront the headways made in the development of high-performance and functionalized cellulose and graphene-based aerogels and their applications in advanced industrial-scale applications, particularly water and wastewater treatment. The study also sought to highlight the existing challenges in the development and utilization of these aerogels in water treatment, and to explore the proposed solutions to overcome these. Therefore, the objective of the study was to critically review the state-of-the-art with respect to the aerogel synthesis methods, properties, their application, effectiveness and performance in water and wastewater treatment.

2 Aerogel fabrication methods

Depending on the application and user requirements aerogels can typically be produced in distinct forms, such as monoliths (can be complex or customized), granules or powder and aerogel fibres using different methods [1, 8]. Aerogels are further classified as organic or inorganic.

Initially metallic salts were used as precursors for the formation of oxide gels which were then synthesized into aerogels. Kistler used sodium metasilicate (Na_2SiO_3) as the precursor because of its affordability. However, this process required the removal of the sodium chloride (NaCl) which forms during the synthesis. This made it challenging to use aerogels in industrial applications as the NaCl required a lengthy dissolution process [8]. Peri [9] was the first to use alkoxides as precursors, which eliminated the lengthy removal of NaCl and accelerated the synthesis of silica aerogels. This transition made it possible to directly dry the gels by a supercritical process, eliminating the water dissolution step [9].

The variety of precursors which can be used for fabricating aerogels has also increased, to include oxyalkoxides and an extensive list of $\text{XSi}(\text{OR})_3$ precursor molecules, in which the ligand X is an organic group, which will impart properties such as hydrophobicity and is also capable of undergoing its own polymerization. R is an organic group or chain (with a methyl, propyl, phenyl, amine end etc.) [10].

Several aerogel fabrication methods have been developed and improved. For instance, Sandia National Laboratories developed a facile kinetic growth model for controlling the texture of the silica aerogels [11]. This process was later improved by Tillotson and Hrubesh [12], by substituting the alcohol with an aprotic solvent, in order to prohibit reverse hydrolysis reactions. This improved method increased the porosity of the silica aerogel monoliths by 99.8% (by volume) and reduced the density to 3 kg m^{-3} , producing aerogels of the lowest density [12].

Aerogels can be synthesized by the sol–gel technique, in which micro-particles or molecules with a diameter ranging from 1 to 1000 nm are dispersed in a solution in which they agglomerate and coalesce to form a consistent network (gel) under controlled conditions [1, 6, 10]. The sol–gel process consists of three steps, synthesis, aging and drying.

2.1 Synthesis

The synthesis step includes hydrolysis and condensation reactions of identified/selected precursors, which includes metallic salts, metal alkoxides or other organic or inorganic compounds which can be hydrolysed, using water or alcohols as a liquid/reactant, in the presence of a catalyst and surfactant [6, 7]. These reactions will result in the formation of wet gels, characterized by a viscoelastic body which consists of a porous solid phase with liquid trapped in its pores [13]. The gels formed are referred to as alcogels, aqua-gels or hydrogels [1]. During the formation of the gel, the final shape of the aerogel, which includes monoliths, granules or powder can be formed by moulding [7]. Figure 1 shows aerogel synthesis methods.

2.2 Aging

Aging occurs during condensation reactions which take place to form and strengthen the network structure of the gels, the process can also be referred to as crosslinking [7]. During this step the gel network grows and improves in mechanical properties, textural properties porosity, surface area and pore size [14, 15]. The aging step requires hours or days to complete in a suitable solvent [16]. The kinetics of the aging step is influenced by factors such as temperature, pH of the medium and time [17, 18].

2.3 Drying

The drying step is considered to be the most critical step in aerogel formation. Its main purpose is to extract the solvent from the gel (by evaporation), leaving the solid network structure behind. Drying also assists in preserving the microstructure formed [7]. Different approaches can be used for drying, each approach has advantages and disadvantages. These include supercritical drying, ambient—pressure or subcritical drying, freeze drying and microwave—assisted drying [1, 8, 10, 13, 19–27].

3 Properties of aerogels

Aerogel structures are distinguished by well-defined and interconnected tube-shaped pores with diameters ranging from 2 to 50 nm [1, 28]. Because of their ultralow density ($0\text{--}0.5\text{ g cm}^{-3}$), aerogels are extremely light weight, which makes them the lightest solid materials to be produced industrially and ideal to be used in applications requiring weight reduction [1]. The ultralow density is attributed to the high porosity of the aerogels (50–99%), which also contributes to their low thermal conductivities ($0.01\text{--}0.03\text{ W m}^{-1}\text{ K}^{-1}$) and large surface areas ($200\text{--}1000\text{ m}^2\text{ g}^{-1}$) [13, 29].

The ability to modify the chemistry of aerogels makes them unique and compelling. The surface area, pore size and processing technique of aerogels can be customized, different functional groups can be incorporated in order to provide desired chemical, physical and mechanical properties [19]. The molecules which form the structure of some aerogels are bonded by strong chemical bonds which are able to resist deformation under pressure. The presence of strong chemical bonds in the structure of the aerogels permits them to have relatively high mechanical strengths [6]. The preparation

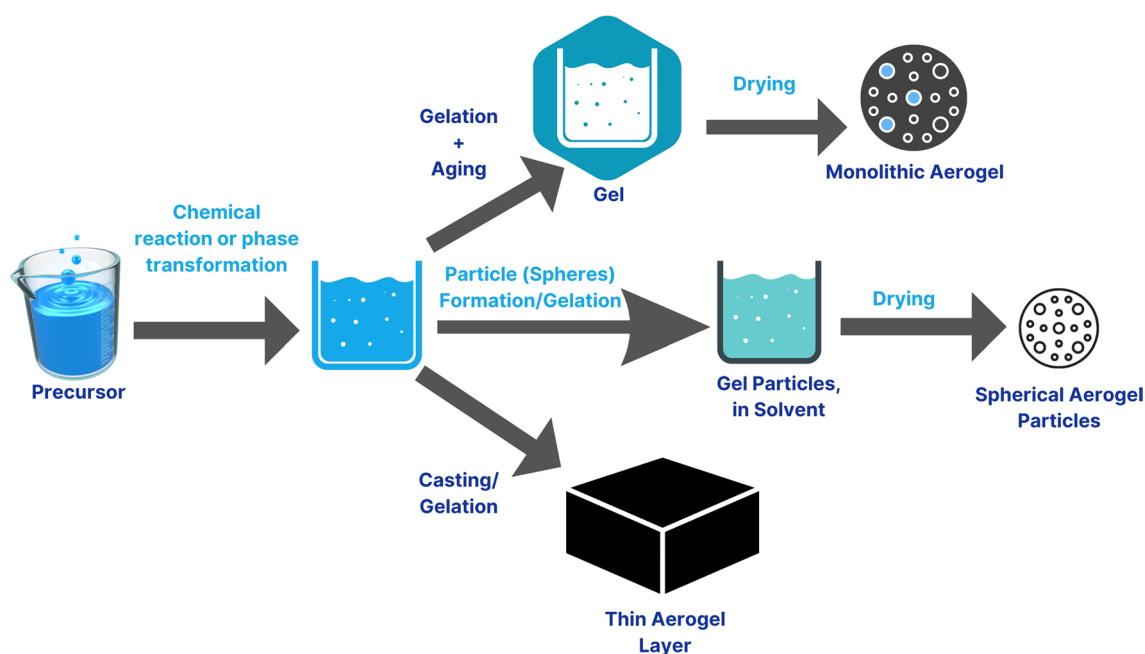


Fig. 1 Synthesis of aerogels [7]

conditions have a significant influence on the network structural connection, which ultimately influences the mechanical properties [1].

Aerogels are capable of displaying distinctive optical properties which include translucence, depending on their configuration and structure [1]. Because of their strong chemical bonds, aerogels typically have great chemical stability and resistance to degradation, which makes them ideal for use in harsh environments or in applications where long-term stability is important [6].

The combination of ultra-low density, high porosity, large surface area, excellent thermal insulation, high mechanical strength, unique optical properties, and chemical stability make aerogels a highly versatile class of materials with great potential for innovation and advancement in many fields. Zhu et al. [3] demonstrated the potential of using aerogel materials as sensors in wearable electronic devices, by fabricating a highly robust and conductive carbonized bamboo aerogel enclosed with silicon resin. The composite aerogel indicated excellent features which made it suitable to be used as a wearable sensor in personal health care and artificial intelligence systems. The features included high flexibility, very low Young's Modulus (0.09 MPa), a large failure strain (275%), strain sensing performance, a short responsive time and a stable response to cyclic loading over 1000 cycles [3]. The National Aeronautics and Space Administration (NASA) employed aerogels in their Stardust spacecraft to successfully capture cosmic dust particles in space in their mission. The super lightweight silica aerogels also functioned to protect the space mirrors from collision with the dust particles travelling at speeds exceeding 18,000 km [30]. Gu and Yushin [31] conducted a detailed review of nanostructured carbon materials for electrochemical capacitor applications, which included studies conducted on carbon aerogels. In their study, it was indicated that carbon aerogels have the potential to be used as energy storage materials in supercapacitors for commercial large scale applications [31]. Other potential applications of composite aerogels which have been investigated include: thermal insulation, gas storage and catalysis [28, 32]. Table 1 summarizes the most significant properties of aerogels.

4 Applications of aerogels in water and wastewater treatment

Aerogels have garnered increasing interest for their potential applications in water and wastewater treatment. The high surface area and porosity of aerogels make them ideal materials for adsorbing pollutants and contaminants from water and wastewater. Adsorption using aerogels is an effective process for the removal of various pollutants, including heavy metals, dyes, organic compounds, and other contaminants from water and wastewater. The highly porous structure of aerogels allows them to provide a large surface area for adsorption, thus enabling the removal of pollutants with high efficiency. Additionally, aerogels can be engineered to possess specific surface functionalities, such as surface charges, that can enable their selectivity for particular contaminants. The unique mechanical properties of aerogels make them highly durable and resistant to the harsh chemical and physical conditions encountered in wastewater treatment processes, enabling them to maintain their adsorption capacity over prolonged periods [39–41]. Jiang et al. [2] fabricated a multifunctional gelatine-based composite aerogel, by dispersing TiO₂ nanoparticles in the three-dimensional structure of an aerogel which was crosslinked with branched polyethyleneimine (PEI). The composite aerogel exhibited a highly porous structure and excellent amphiphilic surface which enabled it to separate oil/water mixtures. The composite aerogel was also successful in adsorbing cationic and anionic organic dyes with good adsorption capacity as well as removing copper ions from aqueous solutions with good reusability [2].

The use of aerogels in water and wastewater treatment has the potential to offer a cost-effective and efficient solution for the growing problem of water pollution. By using aerogel-based adsorbents, contaminants can be effectively removed from wastewater, thus making it safe for discharge or reuse. Furthermore, the reusability and regeneration ability of aerogel-based adsorbents can make the process more sustainable and economical. Aerogel-based adsorbents can be regenerated by desorption of the adsorbed pollutants, which allows for the adsorbents to be reused multiple times, thus reducing the amount of waste generated in the treatment process. Therefore, the use of aerogels in wastewater treatment is a promising avenue for the development of sustainable and effective technologies for water purification [19, 42].

This review explores the applications of cellulose, graphene, and cellulose-graphene composite aerogels in water treatment. The next sections of the review are focused on the work conducted on these three types of aerogels and their use in water treatment applications.

Table 1 Significant properties of aerogels

Property	Description	Illustrative values
Surface area	Exceptionally high surface areas	200–100 m ² g ⁻¹ [13, 29]
Porosity	High porosity	50–99% [13, 29]
Density	Extremely low density	0–0.5 g cm ⁻³ [1]
Strength	Relatively high mechanical strength and stiffness	Compressive strength: silica aerogels: 1–100 MPa. Graphene aerogels: 1–200 MPa [33, 34]
Thermal insulation	Highly thermally insulative, low thermal conductivity	0.01–0.03 W m ⁻¹ K ⁻¹ [13, 29]
Transparency	Some aerogels, e.g. silica-based aerogels, are transparent to visible light	90 to 99.8% in the visible spectrum (400–700 nm) [10]
Hydrophobicity/hydrophilicity	Can be hydrophobic or hydrophilic	Contact angles ranging from as low as 100° for hydrophilic aerogels to over 150° for highly hydrophobic aerogels [10, 35]
Chemical stability	Chemically stable, resistant to degradation and corrosion in various environments	Environments, including acidic, alkaline, and aqueous solutions [10]
Electrical conductivity	Carbon-based aerogels are high electrically conductive	100 S/cm to over 1000 S/cm [34, 36]
Flexibility	Can be flexible or rigid depending on the composition and processing methods	Polymer-based aerogels can be highly flexible, allowing for bending and deformation, while inorganic aerogels like silica aerogels tend to be more rigid [37, 38]

4.1 Cellulose based aerogels

In the current era, scientific and technological movement is oriented towards materials that are ecologically sound, sustainable techniques and procedures, and renewable resources and energy. The prospect of exhaustion of non-renewable resources such as coal, petroleum, and gas in the future, and the negative impact on the environment due to petroleum-based polymers, has resulted in a growing need for biodegradable resources that are not based on petroleum, are carbon neutral, and present minimal risks to the environment, animals, and human health and safety [43].

Cellulose, which is a natural linear macromolecule obtained abundantly from plants such as cotton, bast fibres, flax, hemp, sisal, jute or wood perfectly fits the description for the biodegradable resources which are sought after. The production of cellulose fibres requires a chemical process for separating the cellulose from lignin, hemicellulose and other components of the plants cells [44, 45].

The arrangement of grown cellulose occurs in fringes and fibrils, where regions of crystalline organization are interspersed with areas of disorder, as depicted in Fig. 2a. A schematic representation of the dissection of a cellulose fibre, from its macroscopic form as found in plant cells to its polymeric unit, is illustrated in Fig. 2b [46].

The structure of cellulose comprises repetitive anhydrous glucose units covalently linked by acetal functionalities that encompass cellulose hydroxyl (OH) groups. The principal OH groups along the chains of macromolecules can be readily altered through interaction with functional groups, resulting in a diverse array of cellulose derivatives. The reactivity of these anhydrous glucose units varies widely, influenced by both steric effects of reagents and supra-molecular structures [47, 48]. Derivatives of cellulose can be produced in solvent systems by using techniques such as esterification, acylation, grafting, and etherification [49].

Due to its abundant availability, renewability, and biodegradability, cellulose represents an excellent material option for sustainable fabrication of aerogels. However, the efficient chemical modification of cellulose through grafting often necessitates the use of aprotic solvents, toxic reactants, and harsh conditions, which adversely impact dispersibility and necessitate further purification steps [49].

Despite being hydrophilic and capable of significant swelling in water, cellulose does not dissolve in water or most organic solvents. However, it is biodegradable, and the use of cellulose aerogels aligns with the principles of green materials. The cellulose polymer can be broken down chemically into glucose units using concentrated acids at high temperatures [50].

Since cellulose is highly stable chemically, the production of aerogels from cellulose requires a processing method or technology that can disintegrate the cellulose into its fundamental fibrils or even down to its polymeric chain without any degradation or chemical modification. This must be followed by rebuilding the fibrils into a low-density, open-porous gel that can be dried to form a three-dimensional aerogel structure.

The key advantages of cellulose-based aerogels are that they are made from an abundant, renewable, non-toxic, low cost, and biodegradable resource, making them more environmentally friendly than many other types of materials. A comparative analysis between cellulose aerogels and bulk cellulose materials is given in Table 2.

Fig. 2 **a** Fringe-fibril model of cellulose and **b** Macro-fibril as existing in plant cells begin a composite of micro-fibrils [46]. Reproduced with permission from Springer Nature

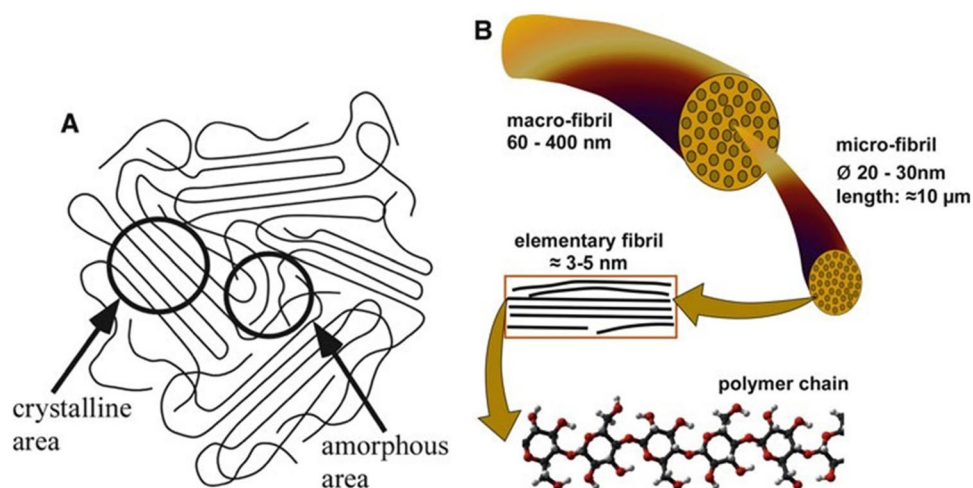


Table 2 Comparative analysis between cellulose aerogels and bulk cellulose materials

Property	Cellulose aerogels	Bulk cellulose materials
Porosity	Highly porous structure with interconnected pores, resulting in high surface area and low density [1]	Lower porosity compared to aerogels, resulting in lower surface area and higher density
Mechanical strength	Exhibits moderate to high mechanical strength and stiffness, suitable for structural applications [51]	Lower mechanical strength compared to aerogels, making them less suitable for load-bearing applications
Surface area	High surface area due to the porous structure, enabling efficient adsorption and catalysis [13, 29]	Lower surface area compared to aerogels, limiting adsorption capacity and catalytic activity
Density	Low density, often among the lowest of all solid materials, contributing to lightweight properties [1]	Higher density compared to aerogels, resulting in heavier and denser materials
Thermal insulation	Good thermal insulation properties, with low thermal conductivity and high temperature stability [13, 29]	Limited thermal insulation compared to aerogels, with higher thermal conductivity and lower temperature stability
Transparency	Some cellulose aerogels can be transparent to visible light, allowing for optical applications [1]	Bulk cellulose materials are generally not transparent to visible light
Chemical modification potential	Cellulose aerogels offer versatility for chemical modification, enabling tailored properties for specific applications [51]	Bulk cellulose materials can be chemically modified but may have limitations due to their dense structure
Processing complexity	Aerogel production involves more complex processing methods, such as supercritical drying, compared to bulk cellulose materials [1, 8, 10, 13, 19–27]	Bulk cellulose materials can be processed using simpler methods such as mechanical or chemical treatments

4.1.1 Production of cellulose based aerogels

Various techniques for the production of cellulose aerogels have been documented in the literature, and new methods continue to be developed. The approach employed is often dependent on the type of raw material used. For instance, the preparation of aerogels from hemicellulose necessitates a different method compared to that used for raw cellulose or lignocellulosic polymer mixtures [6].

Tan et al. [52] developed the initial cellulose aerogels which became popular, by using cellulose acetate as precursor and de-esterifying it. Crosslinking of the cellulose ester was achieved by using an acetone solution with toluene-2,4 diisocyanate. When the concentration of cellulose concentration was more than 5 and less than 30 wt.% gels were formed [52]. Supercritical drying was used to form aerogels with specific areas of less than $400 \text{ m}^2 \text{ g}^{-1}$ and densities ranging from 100 to 350 kg m^{-3} [52]. In the study, Tan et al. observed that the ratio of cellulose to volume of acetone, and toluene crosslinking agent had an influence on the density and shrinkage of the aerogel; small cellulose ester concentrations and larger toluene concentration led to higher shrinkage [52].

Jin et al. [26] developed a different technique for producing high quality cellulose aerogels, which eliminates the use of toxic isocyanates and uses smaller amounts of cellulose (0.5–3 wt.%), by using semi-crystalline raw cellulose. In their study, a combination of water and $\text{Ca}(\text{SCN})_2$ was used as a solvent for breaking down the extensive system of hydrogen bonds along the cellulose chain, into a colloidal system. The mixture formed was moulded into layers of 1.0 mm thickness and submerged in a methanol bath, for extraction of the salt and regeneration of cellulose as a gel, followed by freeze drying to form aerogels [26]. Figure 3 shows the process developed by Jin et al. [26].

Ishida et al. [53] produced carbon aerogels by dissolving microcrystalline cellulose in H_2SO_4 or NaOH water solutions, resulting in an aqueous suspension with 0.2–1 wt.% cellulose solid content. Fine powder aerogels with surface areas ranging from 32 to $178 \text{ m}^2 \text{ g}^{-1}$ were obtained after solidification of the gel and drying by different drying techniques including: freeze drying, supercritical drying in CO_2 (after solvent exchange) and solvent exchange drying [53]. To obtain carbon aerogels, the cellulose aerogels were pyrolyzed in an infrared furnace under a flow of nitrogen up to $600 \text{ }^\circ\text{C}$ [53].

Fischer et al. [54] used a different method to produce cellulose based monolith aerogels with specific surface areas ranging from 140 to $250 \text{ m}^2 \text{ g}^{-1}$, densities from 0.25 to 0.85 g cm^{-3} and pore sizes between 13 and 25 nm , after supercritical drying under CO_2 . In their study, the sol–gel technique was used to crosslink cellulose acetate with a non-toxic isocyanate, under a tin-based catalyst [54]. Gelation times ranging from 15 to 150 min were observed, which were highly influenced by the nature and concentration of the reagents [54]. Figure 4 shows the process used by Fischer et al. [54].

Novel highly porous aerogel-like materials were produced from gelation of different types of cellulose in sodium hydroxide solution and *N*-methyl-morpholine-*N*-oxide (NMMO) monohydrate solution, followed by supercritical drying with CO_2 by Gavillon and Budtova [55]. The types of cellulose used differed in the degree of polymerization (DP), which included microcrystalline cellulose (DP = 180), steam exploded Borregaard cellulose (DP = 500), fibrous Solucell cellulose (DP = 950 and 310) with concentrations between 3 and 8 wt.%. Distilled water was used to regenerate the samples, which resulted in a volume shrinkage of 10%. The results from their study indicated that gelation time varied exponentially with an increase in temperature. A decrease in cellulose concentration delayed gelation, by increasing gelation temperature and gelation time [55]. The aerogels obtained in this study displayed porosities above 95%, specific surface areas between 200 and $300 \text{ m}^2 \text{ g}^{-1}$ and densities from 0.1 to 0.15 g cm^{-3} . The conditions of regeneration; bath temperature and acidity, had an influence

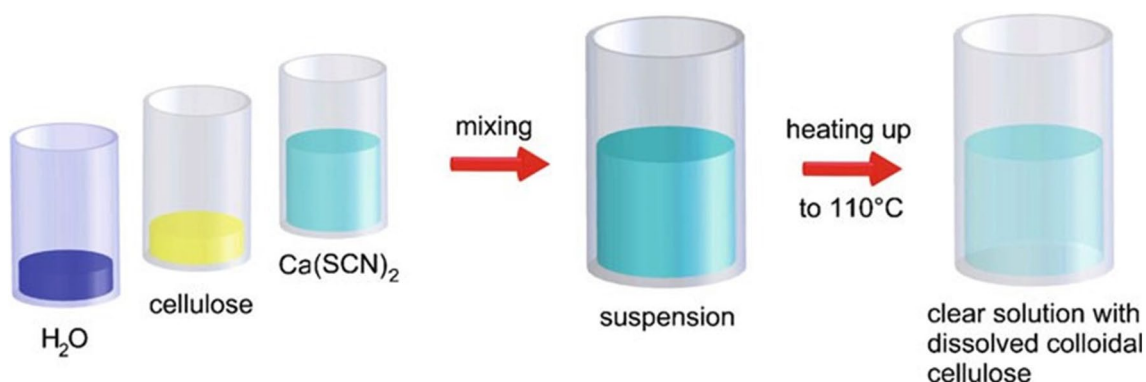


Fig. 3 Schematic [46] of the cellulose aerogel production process developed by Jin et al. [26]. Reproduced with permission from Springer Nature

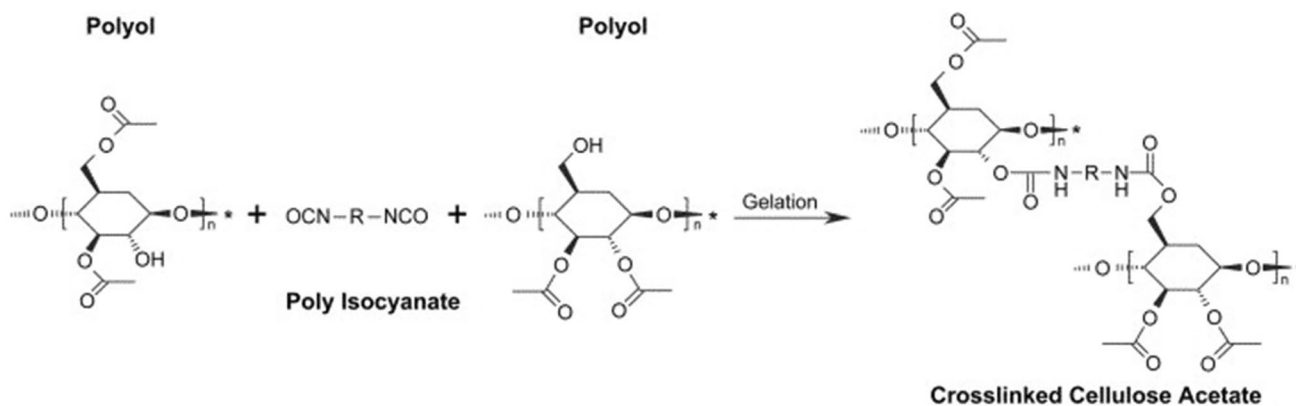


Fig. 4 Schematic of crosslinking by urethane bonding [54]. Reproduced with permission from Elsevier

on the porosity, which can also be controlled by addition of a surfactants such as Simusol and alkylpolyglycoside (APG) [55]. Figure 5 shows the process used by Gavillon and Budtova [55].

Pinnow et al. [56] successfully produced highly porous cellulose aerogels with densities below 0.06 g cm^{-3} and pores ranging from 0.5 to 1 nm, by gelation of sodium carbamate in solutions of sodium hydroxide or dilute sulphuric acid at higher temperatures. This study indicated that sodium carbamate can be used to produce cellulose aerogels with highly porous structure and large specific surface areas, instead of thiocyanate which is toxic [56].

Deng et al. [57] utilized an ionic liquid to dissolve cellulose and subsequently crafted aerogels through freeze-drying, instead of utilizing hydroxide or hydrate melts. By employing scanning electron microscopy and nitrogen adsorption techniques, it was discovered that a foam with a 3D open fibrillar network structure can be created through rapid freezing and freeze-drying processes. This foam had a specific surface area of $186 \text{ m}^2 \text{ g}^{-1}$ and a porosity of 99% [57]. Buchtova and Budtova [58] synthesized porous and lightweight cellulose aerogels with excellent versatility by using a dissolution-coagulation process followed by diverse drying methods. Cellulose was dissolved in a mixture of ionic liquid and dimethyl sulfoxide (DMSO), and then coagulation was achieved using ethanol [58].

Cellulose filament aerogels are a type of aerogel made from cellulose nanofibrils or filaments. These filaments are extracted from natural sources, such as wood pulp, and have diameters of between 20 and 100 nm and lengths of several μm [6, 59]. The production process of cellulose filament aerogels is similar to monolith aerogels production, which involves suspending the filaments in solvent and then using a freeze-drying process to remove the water and leave behind a three-dimensional, highly porous structure. This process can also involve the addition of a crosslinking agent, which increases the strength and stability of the aerogel [59].

A cellulose aerogel based on nanostructured filament of an open porous structure was produced using a sol-gel method and fibres obtained from a wet spinning technique by Schmenk et al. [60] and Hacker et al. [61]. Although, the aerogel fibres obtained had sponge-like mesopores, with a specific surface area of approximately $180 \text{ m}^2 \text{ g}^{-1}$, they were also rather dense, with a density of 124 kg m^{-3} , which suggests that aerogel filament spinning can be further improved [60, 61].

Alakalunmaa et al. [59] used softwood polysaccharides to prepare hydrogels through crosslinking-aided gelation, with spruce galactoglucomannans (GGM) as the primary component. Cellulose nanofibrils were utilized to form a reinforcing network, which was further crosslinked with a GGM matrix by an inorganic salt, ammonium zirconium carbonate, known to be safe for use in food packaging. The hydrogels were freeze-dried to form low-density, stiff aerogels with almost all of their volume comprising of air-filled pores. These aerogels demonstrated high water absorption capacity, up to 37 times their initial weight, and showed elasticity, and repeatable and reversible sponge capacity when immersed in water. The approach presents a new way of assembling wood polysaccharides, offering various possibilities for using the underutilized GGM. The

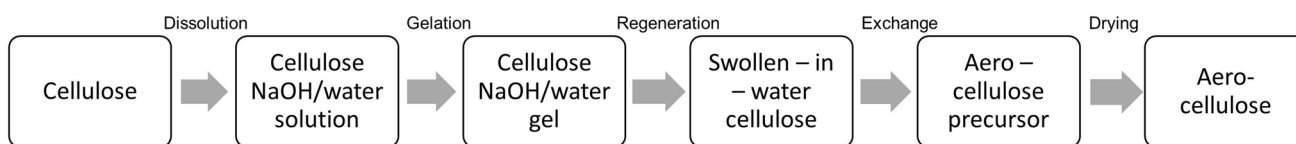


Fig. 5 Schematic of the main steps in the preparation of aero-cellulose from cellulose/NaOH/water solutions [55]

resulting bio-based materials could have potential applications in food packaging and contribute to reducing the usage of petroleum-based plastics in the future [59].

An interesting prospect would be to investigate the possibility of producing transparent cellulose filament aerogels, using techniques for synthesising transparent cellulose monoliths aerogels as described by Cai et al. [62]. Some of the potential applications of cellulose filament aerogels include thermal insulation, sound absorption, and biomedical engineering. They are also being investigated for use in packaging materials and as a sustainable alternative to synthetic materials [59, 62]. Cellulose filament aerogels represent an exciting area of research with many potential applications. However, further research is needed to optimize their properties and develop new applications. Table 3 illustrates precursor materials for cellulose-based aerogels and their sustainability aspects across past, present, and future trends.

4.1.2 Application of cellulose based aerogels in water treatment

Although cellulosic materials contain hydroxyl groups that are capable of interacting effectively with water pollutants, their relatively low surface areas inhibit and limit their effective performance in water treatment [70, 71]. Due to their high porosity, low density, and biocompatibility, cellulose aerogels have attracted interest for their potential use in water treatment applications. They have been studied as an adsorbent material for the removal of pollutants from water, such as heavy metals, dyes, and organic compounds. The surface of cellulose can be modified by introducing cationic, anionic or hydrophobic groups, in order to improve the adsorption capacity or selectivity of certain ions/groups [72].

Daribatabar et al. [45] produced a cellulose aerogel with a specific surface area of $300 \text{ m}^2 \text{ g}^{-1}$, a density of 0.004 g cm^{-3} and porosity above 99%, using carboxy methyl cellulose and citric acid as safe and green crosslinking agents. The aerogel was used to purify aquaculture wastewater by removing nitrate (NO_3^-), nitrite (NO_2^-) and phosphate (PO_4^{3-}) compounds, which indicated removal efficiencies of 79.65, 73.04 and 98.18% respectively. The effect of varying different parameters on the removal efficiencies obtained from their study is illustrated in Fig. 6. These results indicate the ability to use safer, green and non-additionally coated cellulose nanofiber aerogels to effectively remove pollutants in wastewater treatment. In their study, adsorption and desorption experiments using NaOH were conducted to investigate the rate of desorption and adsorption efficiency after 3 cycles. The regenerated aerogel achieved adsorption capacities of 65% NO_3^- , 50% NO_2^- , and 58% PO_4^{3-} after 3 cycles [45].

Mo et al. [73] designed a nanocellulose aerogel adsorbent that imitates the honeycomb structure of wood, featuring specific covalent bonding networks. Their design provided the adsorbent with both mechanical integrity and the ability to remove contaminants rapidly, which demonstrated outstanding selectivity, superfast adsorption, and easy regeneration. Specifically, it exhibited maximum adsorption capacities for Pb (II), Cu (II), Zn (II), Cd (II), and Mn (II) of 571 mg g^{-1} , 462 mg g^{-1} , 361 mg g^{-1} , 263 mg g^{-1} , and 208 mg g^{-1} , respectively. Moreover, it showed high efficiency in removing Pb (II) species with rapid speed, reaching 87% and 100% of its equilibrium uptake in 2 min and 10 min, respectively. The adsorption isotherm and kinetics models supported the Langmuir and pseudo-second-order models, respectively, indicating that the adsorption process was primarily dominated by monolayer chemisorption. The adsorbent demonstrated superior affinity for Pb (II) over other ions in wastewater and could be regenerated at least five times [73]. This wood-inspired aerogel adsorbent holds tremendous potential for cleaning contaminated substances [73].

Li et al. [74] developed amine functionalized cellulose—based aerogel beads (CGP), from cellulose nanofibrils (CNF), for the simultaneous and efficient adsorption-reduction-sequestration of Cr(VI), in both a batch and fixed bed column studies. The aerogel beads were obtained by grafting hyperbranched polyethyleneimine (PEI) on CNF using (3-glycidyloxypropyl) trimethoxysilane (GPTMS) as the crosslinker, followed by shaping the mixture into 3 mm beads by injecting into liquid nitrogen and freeze drying at $-50 \text{ }^\circ\text{C}$ [74]. In their study, it was observed that the pH of the solution had an effect on the adsorption performance of the CGP. As the pH increased from 2 to 9, the adsorption capacity gradually decreased from 51.3 to 6.18 mg g^{-1} [74]. The adsorption isotherm correlated more to the Langmuir model, indicating a maximum adsorption capacity of 386.40 mg g^{-1} for Cr(VI). Their study also investigated the effects of interference substances such as normal organic matters, electrolyte ions and other metal ions. To study the effect of normal organic matters on the adsorption capacity of the CGP, humic acid was added to the Cr(VI) solution at a concentration of 0–100 ppm, which indicated only a slight decrease in adsorption capacity as the concentration increased (95% adsorption capacity could be maintained) [74]. The addition of anions such as NaCl, NaSO_4 , NaHCO_3 , NaNO_3 and Na_2CO_3 , reduced the adsorption capacity of the CGP for Cr(VI) as the concentration of the anions increased from 0 to 100 ppm. The CGP indicated excellent adsorption selectivity for Cr(VI) in the presence of other metal ions such as Cd(II), Cu(II), Zn (II), and Cr(III) in their chloride forms at 500 ppm [74]. A home-made fixed bed column setup was made to simulate a typical wastewater treatment process, by continuously pumping an effluent flow which

Table 3 Cellulose aerogels precursors and their sustainability

Precursor material	Past trends	Present trends	Future trends
Wood pulp	Widely used, sourced from natural forests with variable sustainability practices	Continued use, increasing focus on sustainable forestry management and certification (e.g., the Forest Stewardship Council, FSC). Research on alternative lignocellulosic sources (e.g., agricultural waste, algae) to minimize environmental impact [63]	Advanced processing techniques to improve resource efficiency and reduce waste. Implementation of circular economy principles for wood pulp utilization and waste valorisation [63]
Cotton	Traditional source with significant environmental footprint due to intensive water and pesticide usage in conventional cotton farming [64, 65]	Shift towards organic and sustainable cotton farming practices to reduce environmental impact. Research on alternative cellulose sources (e.g., cotton textile waste, agricultural residues) to promote circularity [65]	Adoption of closed-loop systems for cotton cultivation and processing, minimizing water and chemical usage. Integration of precision agriculture and biotechnological advancements for sustainable cotton production [65]
Bagasse	By-product of sugarcane processing, often underutilized or disposed of as waste, contributing to environmental pollution	Increasing utilization of bagasse for bioenergy and biomaterial production, reducing waste and promoting resource efficiency. Research on valorisation techniques (e.g., enzymatic hydrolysis, chemical modification) to enhance bagasse-derived cellulose quality and yield [66]	Development of integrated biorefinery systems for sugarcane processing, maximizing value recovery from bagasse while minimizing environmental impact. Exploration of advanced conversion technologies (e.g., biorefinery platforms, biocomposite production) for diversified applications [67]
Bamboo	Considered a sustainable and rapidly renewable resource, but traditional harvesting methods may lead to habitat destruction and soil degradation	Adoption of sustainable bamboo cultivation practices, including agroforestry and forest management certification, to minimize ecological footprint. Research on engineered bamboo materials and processing techniques for improved performance and resource efficiency [68]	Implementation of innovative harvesting and processing technologies (e.g., precision forestry, biorefinery integration) to optimize bamboo resource utilization and reduce environmental impact. Integration of bamboo cultivation with ecosystem restoration and biodiversity conservation initiatives [69]

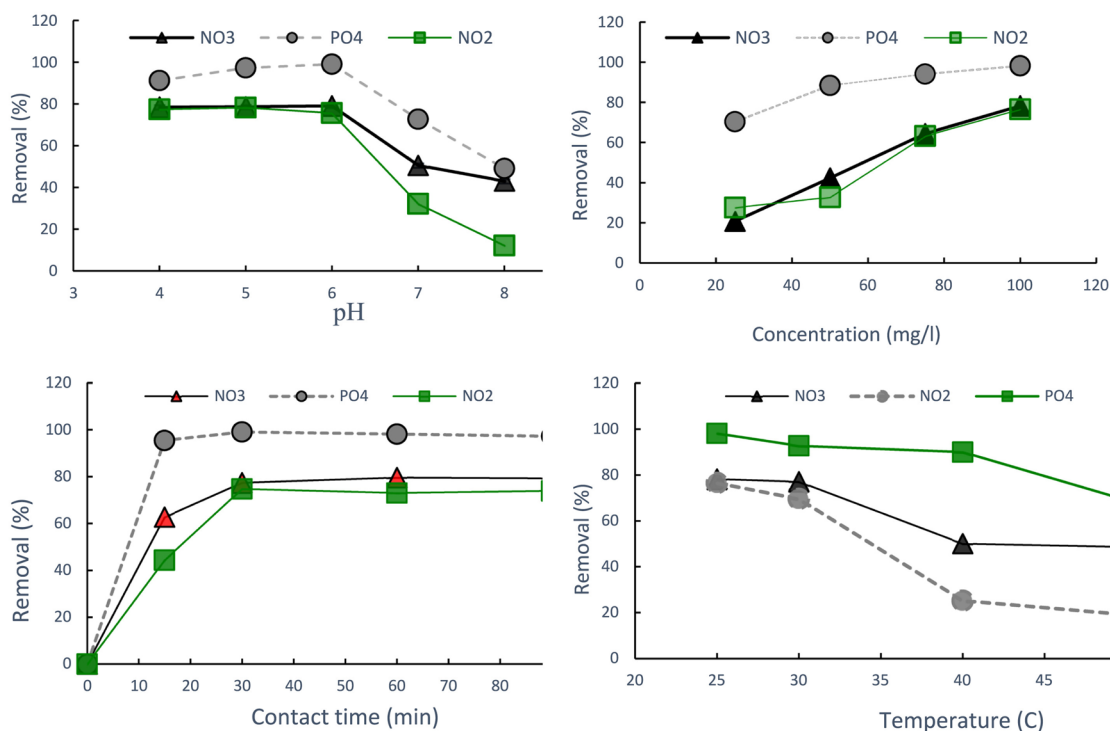
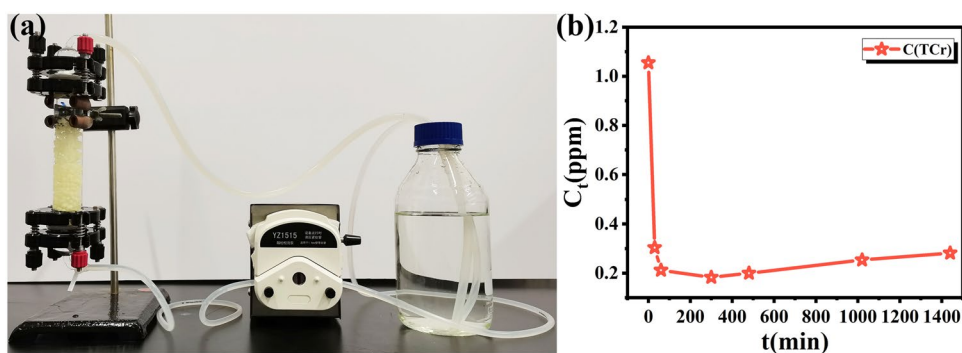


Fig. 6 The effect of different physiochemical parameters on the adsorption of nitrate, nitrite and phosphate compounds by CNF aerogel. **a** pH (time=30 min, concentration=100 mg L⁻¹, weight=0.1 g); **b** concentration (time=30 min, pH=6, weight=0.1 g); **c** contact time (pH=6, concentration=100 mg L⁻¹, weight=0.1 g); **d** temperature (time=60 min, concentration=100 mg L⁻¹, weight=0.1 g, pH=6) [45]. Reproduced with permission from Elsevier

Fig. 7 The digital image of the home-made fixed-bed column **(a)**; Cr(VI) concentration as a function of contact time in the fixed-bed column adsorption study (250 mg CGP, Cr(VI) solution(1 L, 1 ppm), T: 298 K, pH: 2.0) **(b)** [74]. Reproduced with permission from Elsevier



contained 1 ppm Cr(VI) as indicated in Fig. 7. Their study indicated a decrease of the Cr(VI) concentration to around 200 ppb, which is lower than the discharge limit of 500 ppb permitted in China [74].

Li et al. [75] produced a chemically crosslinked CNF aerogel. The CNF aerogel was crosslinked with 3,3-dithiodipropionic acid dihydrazide (DAD) molecules for improved mechanical properties and loaded with TiO₂ nanoparticles, for use in adsorption and degradation of Rhodamine B dye which can be found in waste water. In their study it was observed that the adsorption increased in the initial 30 min and reached saturation after 60 min. The adsorption efficiency decreased with an increase in the concentration of the dye, the optimum adsorption efficiency was found to be 58%, when the concentration of the dye was 10 mg L⁻¹ [75].

Apart from modifying the surface of cellulose aerogels to increase the adsorption capacity, the pH of the solution has an influence on the selectivity and performance of the adsorbent towards metal ions [76]. In their study, Sharma et al. [77] reported the effect of pH on the adsorption capacity of nanocellulose prepared using the nitro-oxidation method, for removal of cadmium (II) (Cd²⁺) ions from water. At pH 7 adsorption efficiency was the highest, compared to lowest adsorption efficiencies at lower and higher pH [77].

Other supporting materials such as methacrylic acid and maleic acid can be grafted onto the nanocellulose surface to improve the adsorption capacity towards metal ions, by increasing the sites of adsorption available [78].

Phanthong et al. [79] fabricated a nanocellulose sponge which was treated with stearoyl chloride to obtain super-hydrophobicity and super-oleophilicity. The nanocellulose sponge showed a superior affinity to several compounds including, dichloromethane, silicon oil, toluene, vacuum pump oil, ethanol, acetone, n-octane, and n-hexane, even after ten cycles, which was recovered by simply squeezing the liquid [79].

Aerogel recovery and absorption of oils and organic solvents remains problematic. This issue has prompted some researchers to incorporate a magnetic feature into the aerogels, making it simpler to recover the aerogels after absorption of oils by simply using a magnet [80].

4.2 Graphene based aerogels

Graphene-based aerogels are a relatively new type of material that has generated a lot of interest among scientists due to their exceptional properties and potential applications. These materials are composed of three-dimensional structures made up of sheets of graphene, which are two-dimensional layers of carbon atoms arranged in a hexagonal pattern [81]. The resulting aerogels have large surface areas, are highly porous, have excellent compressibility, recoverability, and thermal insulation properties [82, 83].

Graphene-based aerogels are incredibly lightweight and strong, making them ideal for use in aerospace and structural engineering. Furthermore, these materials exhibit excellent thermal and electrical conductivity, which makes them well-suited for use in energy storage and conversion devices [5, 84, 85]. The high surface area of graphene-based aerogels is a notable property, which provides a large number of active sites for chemical reactions. This makes them valuable in areas such as catalysis and gas sensing [34, 81, 86].

However, the production of graphene-based aerogels is not without its challenges. One of the primary issues is the difficulty in producing graphene sheets with high purity and uniformity, which can affect the mechanical and electrical properties of the aerogel. Additionally, the production process can be complex and costly, which can limit the scalability of the technology [87].

The most appropriate precursor for manufacturing 3D gel networks based on graphene is graphene oxide (GO). GO has the advantage of being easily dispersed in many solvents and its physical properties, including dielectric, electronic, and mechanical properties can be easily manipulated and adjusted within a broad range [82]. To obtain the desired sp^2 bonded carbon properties in graphene, a reduction process is necessary, given the high level of oxidation and lattice defects in GO. Reduction of GO to graphene can be achieved through various methods such as self-assembly, hydrothermal, chemical, thermal, photoreduction, microorganisms, microwave, electrochemical, and cross-linking. The resulting graphene can be used to form a 3D gel network with pores ranging from sub-micron to micron scale [88].

High volume drying methods such as freeze-drying or supercritical CO_2 drying are utilized to prevent network collapse of the alcogel or hydrogel, caused by capillary forces and to achieve high porosity in graphene-based aerogels [81]. The drying methods have an influence on the pore sizes, for example; CO_2 drying generates numerous macro-pores along with a small number of mesopores, while freeze-drying produces a high number of mesopores that merge into larger macro-pores due to gradual ice crystal growth [83].

Overall, graphene-based aerogels hold great promise as a new class of materials with unique properties and potential applications in a range of fields including water treatment. Ongoing research in this area is likely to lead to the development of more efficient and cost-effective production methods, further enhancing the potential of these materials.

4.2.1 Production methods for graphene-based aerogels

Graphene based aerogels are synthesized by using GO as a precursor. To obtain GO, graphite powder is typically exfoliated via a chemical procedure by treating it in a blend of potassium permanganate, sodium nitrate and concentrated sulfuric acid [81]. However, the use of these chemicals poses toxicity threats and leads to environmental issues. Researchers have been working on developing alternative methods which are milder and less harmful to the environment. These methods include the use of reducing agents such as vitamin C, organic acids, amino acids, microorganisms, plant extracts, sugars, proteins and other antioxidants [89]. Another method of exfoliating graphite which has been recently developed, is through the use of electrochemical methods [90].

As a result of graphite oxidation, diverse oxygen groups, particularly epoxy and hydroxy groups, attach to the edges and basal planes of the GO surface. These functional groups contribute to the high hydrophilicity of the oxide sheets,

enabling the dispersion of graphite oxide in solvents and the creation of stable aqueous colloids. This low-cost and straightforward solution process facilitates the construction of 3D GO structures [91–93].

Besides the reduction of GO, hydrothermal reduction is typically used for synthesis of other materials such as nanoparticles and metal oxides. In this process a material is reduced using hydrogen gas in a high pressure, high temperature (above 100 °C and below 300 °C) and water-based environment. These conditions allow the water to reach a supercritical state and are advantageous in controlling the size, shape, and properties of the final product.

To alter the structure and chemical composition and achieve desired physical properties of aerogels in graphene systems, a variety of reagents are employed. However, caution must be taken to avoid undesired reduction of the graphene oxide, which can be prevented by keeping the temperature below 200 °C [94]. The final porous aerogel structure is achieved by drying of the hydrothermally reduced GO or composite. Hydrothermal synthesis is a simple procedure and offers the capability for large scale production, however, the high energy needs, increases the cost of manufacturing [81].

Different crosslinking agents can be used to form the 3D network structure of graphene-based aerogels. For instance, when exposed to water, GO sheets form a colloidal dispersion which is fairly stable [95]. However, due to a lack of reciprocal repulsion, GO sheets are not stable in the presence of strong acids. A reduction in pH weakens the electrostatic repulsions, and strengthens hydrogen bonding, as a result of the protonation of the carboxyl groups. Hence, a stable GO gelation can be achieved by weakening the repulsion force or by increasing the bonding force. The utilization of crosslinking agents is recognized to aid in the solidification of GO sheets by substantially increasing the adhesive properties between them. Commonly used crosslinking agents include: hydroxyl (PVA), oxygen-containing hydroxypropyl cellulose (HPC), Polyethylene Oxide (PEO) or nitrogen functional groups (polyamines), and ethylenediamine [82, 95–97].

By combining with nearby GO sheets, these polymeric substances can create hydrogen bonds and enhance the binding forces involved in the gelation process. It is noteworthy that several GO/polymer composite hydrogels are capable of responding to changes in pH. Additionally, certain quaternary ammonium salts, including cetyltrimethylammonium bromide (CTAB), tetramethylammonium chloride (TMAC), and melamine, which are relatively small in size, can be employed to help generate GO hydrogels [95].

Divalent and trivalent ions such as: Ca^{2+} , Mg^{2+} , Cu^{2+} , Pb^{2+} , Cr^{3+} , Fe^{3+} etc. have the capability to stimulate self-assembly of GO sheets, due to the interaction between the metallic ions and the GO by a bonding force [98]. The flask volume can be adjusted to obtain hydrogels of different macroscopic sizes. Moreover, the density and size of pores can be influenced by the concentration of GO. In addition, the presence of metal ions can impact the formation of chemical and hydrogen bonds between water, divalent metals, and oxygenated groups [98]. Tang et al. [99] showed that noble metals such as Au, Ag, Pd, Ir, Rh, or Pt, etc. promoted the formation of three-dimensional macro-assembly of single layered graphene oxide [99].

Biomacromolecules possess the ability to interact with GO sheets under ideal conditions. Both synthetic and natural biopolymers, including polysaccharides [100], polyvinyl alcohol [101], proteins and DNA [102], can act as crucial crosslinking agents to create porous interconnected structures by means of hydrogen bonding as well as covalent or non-covalent interactions. Wang et al. [103] fabricated and functionalized a biological graphene aerogel, by reusing microorganisms in activated sludge and ionic dyes. Employing bacteria as a cross-linking agent to produce a biological graphene aerogel offers a promising substitute for chemical techniques that can be expensive, toxic, and demand specific conditions. Xu et al. [104] reported on environmentally friendly 3D aerogels made from self-assembled GO sheets and DNA. These aerogels possessed good mechanical properties, high dye loading capacity and self-healing properties [104].

To produce an aerogel with a polymer support using a cross-linking technique, it is necessary to utilize polymer materials that are capable of bonding with graphene oxide layers. This process involves creating steric hindrances to prevent the stacking of layers, as well as reducing the exposed area [105]. For instance, Chen et al. [106] developed an aerogel by cross-linking molecularly modified poly(*m*-phenylenediamine) (mPmPD) with polyvinyl alcohol (PVA) and graphene oxide (GO). The incorporation of 3-aminophenylboronic acid ($\text{C}_6\text{H}_8\text{BNO}_2$) in the aerogel structure facilitated the confinement of poly(*m*-phenylenediamine). The resultant aerogel possessed sturdy mechanical properties and exhibited exceptional adsorption capacity for silver, with a maximum limit of 917.41 mg g^{-1} [106]. This study showed that by chemically crosslinking graphene oxide (GO) with a conducting polymer material that has adsorption capabilities, it is possible to produce low-cost aerogels with exceptional adsorption properties that can perform well in practical settings.

The sol–gel process is a chemical method that transforms small molecules or ions into solid materials through hydrolysis and condensation reactions to form a three-dimensional network of interconnected particles. This process is particularly useful in synthesizing aerogels because it allows for the creation of porous structures with high surface area and low density. By controlling the reaction conditions and precursor materials, the sol–gel process can be used to produce aerogels with varying properties. The sol–gel process is dependent upon various factors, including the type

of components, precursors, and solvents used, as well as the solution's temperature and pH, composition, stabilizing agents, and drying control chemical additives. These variables collectively determine the duration of gelation and the structure of the resulting solution [6, 107]. Elaloui et al. [108] fabricated alumina xerogel and aerogels using the sol–gel process and investigated the influence of the sol–gel process on the structure and porous texture of the aerogels. The study showed that low-temperature wet chemistry conditions have an important effect on the residual pore texture and specific area of non-doped alumina aerogels at high temperature (1200 °C) [108].

Methods that are guided by templates are effective in creating porous graphene-based aerogels that possess well-defined and shaped structures. Templates serve as agents that determine the morphology of the graphene-based aerogels [109]. This approach facilitates the production of macro-pores that are uniform and controllable, and also enables the microstructure to be adjusted according to requirements [110]. Template-directed reduction can be done in two different ways, chemical vapour deposition and template-directed self-assembly (ice template or sol-cryo method).

Chemical Vapour Deposition (CVD) produces autonomously and immaculate graphene 3D porous structures. Altering the CVD temperature permits an accurate way to manage the pore size, structure and volume [111–113]. However, the utilization of expensive reagents and intricate equipment is necessary for CVD, which in turn restricts its suitability for large-scale production. Additionally, an extra step of transferring the graphene layer onto the intended substrate is required.

The ice-template or sol-cryo directional freezing technique is a prevalent and cutting-edge method for creating 3D monolithic structures without using chemicals. This approach involves using ice grain growth to customize the morphology of the graphene-based aerogels. Different freezing methods such as unidirectional freeze-drying, non-directional freeze-drying, and air freeze-drying can be employed to determine the pore sizes and volumes. Moreover, the shape and size of the resulting 3D foams are influenced by the type of mould used during the freezing process [114].

Additive manufacturing popularly known as 3D printing, is a process of creating three-dimensional objects from a digital model by building them layer by layer. It typically involves the use of a 3D printer, which reads the digital design file and then deposits material in successive layers until the object is complete. The material used can be various types, including plastics, metals, ceramics, and even food. The 3D printer typically applies the material in a controlled manner, guided by the design specifications, and then solidifies or fuses the material to create the desired shape [115].

3D printing technology has been advancing rapidly in recent years and has become a valuable tool for prototyping and manufacturing in a range of industries, from aerospace and automotive to medicine and consumer products. It enables the production of complex shapes and geometries that may not be possible with traditional manufacturing methods, and can also reduce waste and cost by allowing for on-demand production of small quantities or custom-made parts [115]. Two 3D printing techniques have been used for manufacturing graphene aerogels, direct ink writing and stereolithography.

In principle, direct ink writing (DIW) involves extruding materials by depositing a liquid material ink that rapidly solidifies upon extrusion. To achieve a smooth surface, the ink must possess the appropriate modulus to maintain its shape after extrusion. In general, high viscosity inks have better shaping abilities, but they also increase the risk of nozzle clogging. Therefore, the development of high viscosity inks with shear-thinning behaviour that enables flow under pressure and pseudoplastic recovery after deposition is critical for the success of DIW [116, 117]. In their study, Jiang et al. [118] demonstrated that an ultralight micro-lattice of graphene aerogel can be directly printed using GO ink and an ion-induced gelation method. By introducing small quantities of Ca^{2+} ions into the GO ink, the researchers were able to transform the aqueous GO sol into a printable gel ink [118].

Superior results have been obtained by researchers using the DIW method, however, the method has not yet been implemented for practical use because of processing challenges. When printing intricate patterns for applications that demand precise size and resolution, the challenge of achieving dimensional accuracy arises. This is particularly difficult when there is a requirement to use a sufficiently small and simple structural unit without compromising the printing duration or reducing the geometry of the printed parts. In addition, the complexity of structures, such as extrusion nozzle sizes, may pose an obstacle in generating nano-inks [119, 120].

Stereolithography (SLA) is a type of 3D printing technology that uses a laser to create a three-dimensional object from a liquid resin material. The SLA process involves the use of a vat of liquid resin, a UV laser, and a computer-controlled mirror or galvanometer to selectively cure the resin to form the desired 3D shape. The liquid resin is poured into a vat and a platform is lowered into the resin to a depth corresponding to the thickness of the first layer. The UV laser is then used to selectively cure the resin in the shape of the first layer by tracing the pattern of the first slice [115, 119].

Once the first layer is complete, the platform is raised and the process is repeated for the subsequent layers, with the platform being lowered incrementally until the entire object is complete. After the object has been fully printed, it is

removed from the vat and undergoes a post-processing stage where it is cleaned and cured under UV light to finalize the solidification of the resin [115, 119].

In order for a composite to be printed using this technique: the resin must cure when exposed to the laser, it must also have a low viscosity to allow for the recoating process. A complex micro-built graphene aerogel was produced by Hensleigh et al. [121] using the projection micro stereolithography (PμSL) technique.

SLA is capable of producing highly detailed and complex parts with smooth surfaces and high dimensional accuracy, which enables fabrication of complex 3D graphene structures. The limitation with this method is the speed at which objects are printed, however, this leaves room for improvement by researchers and engineers. Figure 8 indicates some of the production methods for GO aerogels, while Table 4 illustrates precursor materials for graphene production and their sustainability aspects across past, present, and future trends.

4.2.2 Application of graphene-based aerogels in water treatment

In several studies by our group, graphene-based materials applied in adsorption-based water treatment processes to remove pollutants such as phenol and Cr(VI) performed relatively poorly compared to commercial adsorbents such as activated carbon [127–129]. This was mainly attributed to the relatively low surface areas of the graphene-based materials utilised, compared to those of commercial adsorbents such as activated carbon. The unique properties of graphene aerogels, such as their high surface area, porosity, and mechanical strength, make them a promising material for water treatment. There are several potential applications of graphene aerogels in water treatment, including the remediation of heavy metals and organic pollutants.

The high surface area and porosity of graphene aerogels allow for efficient adsorption of heavy metal ions, such as lead, mercury, and arsenic, from water. The adsorption process occurs when heavy metal ions in the water are

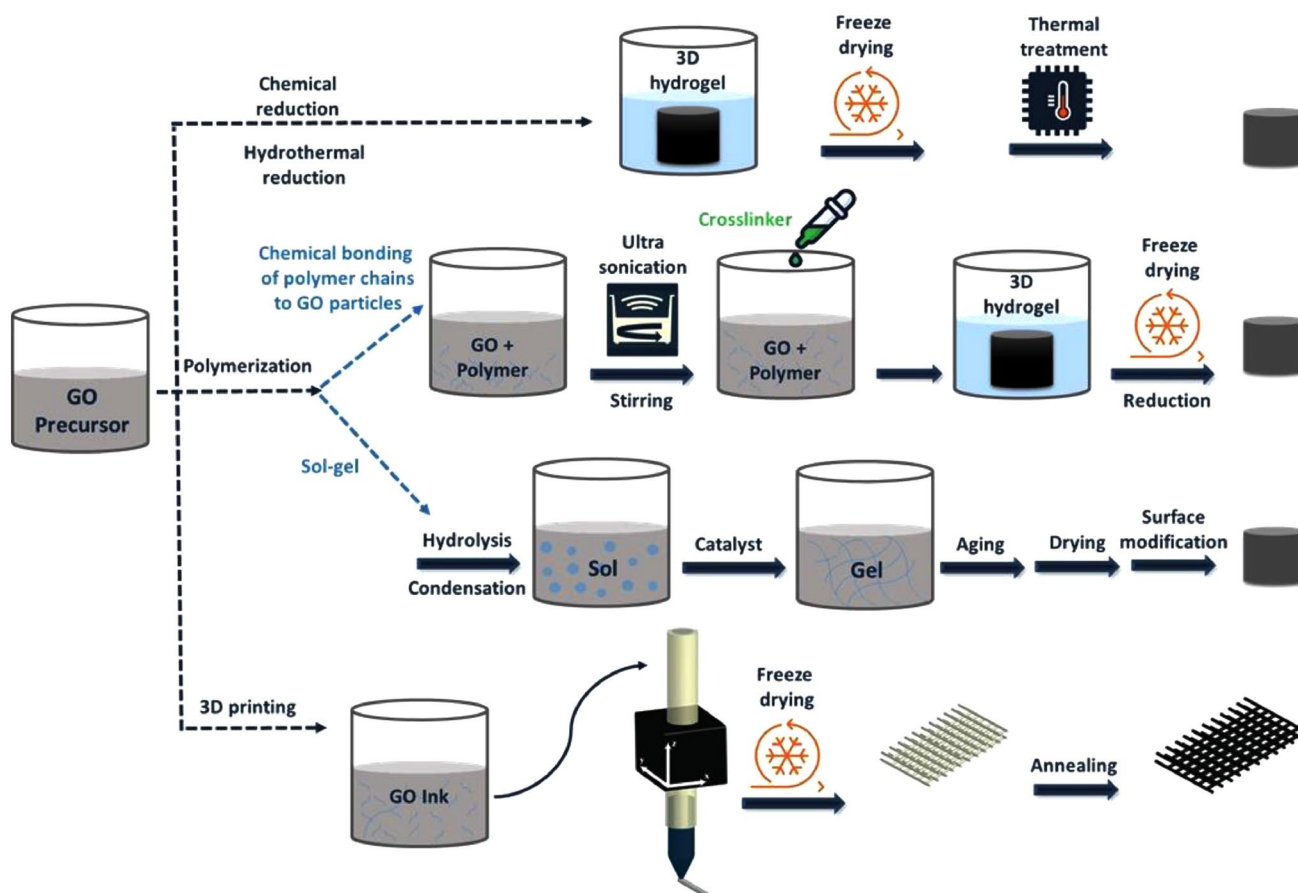


Fig. 8 A schematic representation of the most common methods for the production of GO aerogels [81]. Reproduced with permission from Elsevier

Table 4 Precursor materials for graphene oxide-based aerogels and their sustainability aspects

Precursor material	Past trends	Present trends	Future trends
Graphite	Traditional precursor for graphene production [122]	Still widely used due to its availability and reliability	Exploration of sustainable graphite sources and recycling
Graphene oxide	Emerged as a precursor for scalable graphene production [95, 123]	Extensively researched for its ease of production and versatility	Further optimization for large-scale, sustainable synthesis
Biomass-derived carbon	Increasing interest in using biomass-derived precursors	Research into sustainable biomass sources and conversion processes	Development of efficient, eco-friendly biomass-to-graphene methods [124]
Carbon dioxide	Research into direct conversion of CO ₂ to graphene	Emerging as a sustainable precursor for graphene production	Advances in CO ₂ capture and conversion technologies for graphene synthesis [125]
Sustainable polymers	Growing interest in using renewable polymers as graphene precursors	Exploration of bio-based and biodegradable polymers for graphene production [126]	Integration of sustainable polymer feedstocks and green chemistry approaches

attracted to the surface of the graphene aerogel and held there by chemical forces. The adsorption efficiency of graphene aerogels can be increased by modifying their surface properties through chemical functionalization or by doping them with other materials [85].

Studies have shown that graphene aerogels can effectively remove heavy metals from water, with removal efficiencies of up to 99% for lead, mercury, arsenic ions and other heavy metal ions. Additionally, graphene aerogels have been found to be more effective in removing heavy metals from water compared to traditional adsorbents, such as activated carbon, and compared to other water treatment methods such as ion exchange, coagulation and filtration, due to their higher surface area, adsorption capacity, long lifespan and regeneration [130].

Luo et al. [131] synthesized novel graphene oxide/carboxy methyl chitosan (GO/CMC) composite aerogels through a vacuum assisted self-assembly and freeze-drying technique, for the adsorption of heavy metals from aqueous solution. In their study, the maximum adsorption capacities of the GO/CMC were 151.30 mg g⁻¹ for Ag⁺, 95.37 mg g⁻¹ for Cu²⁺ and 249.38 mg g⁻¹ for Pb²⁺ [131]. In another study, Yu et al. [132] investigated the adsorption capacity of a lyophilized graphene oxide chitosan composite aerogel, for Cu²⁺ metal ions. Simulation of the adsorption kinetics using the pseudo-second order and Langmuir isotherm models indicated a good adsorption capacity of 2.54 × 10¹ mg g⁻¹ for Cu²⁺ ions [132]. The adsorption capacity was positively influenced by an increase in pH and temperature as well as a lower ionic strength.

Liu et al. [133] fabricated unique 3D graphene/δ-MnO₂ aerogels via self-assembly and reduction of graphene oxide, followed by in situ solution-phase deposition of ultrathin δ-MnO₂ nanosheets. The unique structural characteristic of the aerogels resulted in a fast adsorption kinetic rate and higher adsorption capacity towards heavy metal ions. The Langmuir isotherm model for the graphene/δ-MnO₂ aerogels indicated adsorption capacities of 643.62 mg g⁻¹ for Pb²⁺, 250.31 mg g⁻¹ for Cd²⁺ and 228.46 mg g⁻¹ for Cu²⁺ [133]. It was noted in the study that the aerogels were regenerated by treatment in HCl and KOH, and they maintained their original shape and could be repeatedly used for over eight cycles with no clear indication of degradation in performance, which indicated a high potential for sustainability [133].

Chen et al. [134] used graphene oxide as a precursor and oxidant to synthesize aerogels, by a facile hydrothermal technique. The surface of the aerogel was functionalized by incorporating ethylenediaminetetraacetic acid disodium salt (EDTA-2Na). Pyrrole monomer was used as a reducing agent, resulting in an EDTA-2Na/polypyrrole (Ppy)/rGO aerogel (EPGA) which had a high adsorption capacity for Cr(VI) anions, with a maximum adsorption capacity reaching 361 mg g⁻¹ at 298 K at pH of 2. The study also investigated the ability of EPGA to selectively adsorb Cr(VI) ions in the presence of coexisting ions including Cl⁻, NO₃⁻, SO₄²⁻, PO₄³⁻, Ni⁺, Cu²⁺, Zn²⁺, and Cd²⁺ which might influence the adsorption capacity, which indicated a good ability to adsorb selectively and good regeneration ability [134].

The use of graphene aerogels for the removal of heavy metals from water shows great promise and has the potential to provide an effective and sustainable solution to heavy metal pollution in water.

Graphene aerogels have also been considered for the removal of organic pollutants such as dyes, pesticides, and pharmaceuticals which are often found in wastewater and can be harmful to the environment and human health if not properly treated [85].

Chen et al. [135] investigated the absorption efficiency of lignin-modified graphene aerogels (LGAs) for removal of petroleum oils and toxic solvents such as toluene, chloroform, and carbon tetrachloride from water. The LGAs showed superior absorption efficiencies of up to 350 times of their own weight, which is one of the highest amongst the previously reported adsorbents [135]. The LGAs were able to maintain a 96% of their absorption capacity after several compressive and release cycles. A maximum absorption capacity of 522 times their own weight was achieved by the LGAs after treatment by carbonation. Regeneration of the LGAs could be achieved by the repeated heat treatment and squeezing method, which yields approximately a full release of adsorbates [135].

Xiao et al. [136] developed a multifaceted graphene oxide/nanofiber aerogel, by a novel introduction of cellulose nanofibers in graphene aerogels in order to avoid over-stacking of the sheets and improve connectivity of cell walls. The aerogel was highly stable under water and had an adsorption capacity above 800 mg g⁻¹ towards cationic dyes [136]. The density, pore structure and mechanical properties of the aerogel could be influenced by altering the composition and compression deformation [136].

Amongst the other pollutants, antibiotics are also a serious concern with regards to water pollution, because of their imminent release during production and use, which poses a toxic threat to animals, humans and aquatic organisms [85]. A porous graphene oxide-chitosan aerogel (PGO-CS) used as a recyclable adsorbent for tetracycline removal, was reported by Zhao et al. [137]. The PGO-CS aerogel had an adsorption capacity of around 1.47 × 10³ mg g⁻¹, ranking it among the most effective adsorbents for tetracycline. It was well described by the pseudo-first-order kinetics model [137].

The use of graphene aerogels for the removal of organic pollutants from water shows great potential. The ability to mass-produce, modify the mechanical properties and regeneration, could provide a cost-effective and sustainable solution to water pollution.

The desalination process involves the removal of salt and other impurities from seawater or brackish water to make it safe for consumption or agricultural use. Conventional desalination methods, such as reverse osmosis and distillation, require large amounts of energy and are often expensive. Graphene aerogels offer a potential solution to these challenges because they have a high surface area-to-volume ratio, which allows them to efficiently adsorb and remove ions and other impurities from water. Additionally, they have a low thermal conductivity, which can help reduce energy costs in desalination processes.

One example of using graphene aerogels for water desalination involves incorporating them into a membrane. The membrane can selectively filter out salt ions while allowing water molecules to pass through. This approach has the potential to be more efficient and cost-effective than conventional desalination methods.

Ma et al. [138] synthesized a self-floating composite graphene aerogel doped with Enteromorpha and modified polyethylene glycol (PEG), (referred to as PEGA), and used solar energy for desalination, in order to explore functional materials as a solar-energized evaporator with an enhanced evaporation rate and pollutant tolerance. Their analyses showed that modifying PEG enhanced the hydrophilic functionality of PEGA, which increased the evaporation rate and photothermal conversion efficiency up to $2.55 \text{ kg m}^{-2} \text{ h}^{-1}$ and 105.71%, respectively [138]. In this study, the seawater ion removal rate exceeded 99.99% via the PEGA conducted solar evaporation [138].

4.3 Graphene and cellulose composite aerogels

Graphene-cellulose composite aerogels (GCCA) represent an interesting field of research with a high potential for further development. These composite aerogels have been shown to exhibit improved mechanical and electrical properties compared to aerogels fabricated from the individual components, due to the combination of their unique properties. For example, the addition of graphene to cellulose aerogels can increase their electrical conductivity, while the presence of cellulose can improve the mechanical strength and thermal stability of the graphene aerogels [139]. A study conducted by Takeno et al. [140] on the effect of cellulose nanofibers (CNFs) on the mechanical and structural properties of poly (vinyl alcohol) (PVA) hydrogels cross-linked dually by a freezing/thawing method and borax as a cross-linker, showed that the addition of cellulose nanofibers and crosslinking with PVA and borax significantly improved the tensile performance of the aerogels [140].

The large surface area and high porosity of GCCAs make them ideal materials for use in adsorption. Shadkam et al. [141] investigated the adsorption performance of reduced graphene oxide-cellulose nanocrystal hybrid aerogels reinforced with waste paper extracted cellulose fibres, for adsorption of toluene. Using the Langmuir isotherm, a maximum adsorption capacity of 454 mg g^{-1} for toluene was achieved from aqueous medium [141].

The unique properties of GCCAs makes it possible to develop highly flexible and super-lightweight materials, due to the ability of cellulose to prevent volumetric shrinkage of graphene aerogels [142]. Illera et al. [143] demonstrated that aqueous dispersions of graphene can be stabilized by cellulose nanocrystals colloids, which allows the preservation of the intrinsic properties of graphene nanosheets by advancing noncovalent interactions between cellulose and graphene. Table 5 compares the significant properties of cellulose based aerogels and graphene oxide-cellulose composite aerogels.

4.3.1 Synthesis of graphene-cellulose composite aerogels

Various methods have been developed for synthesizing cellulose-graphene composite aerogels, such as the reduction of graphene oxide in the presence of cellulose, the incorporation of graphene sheets into cellulose aerogels, and the assembly of graphene-coated cellulose fibres.

Due to its simplicity and scalability, the hydrothermal method is a popular technique for synthesizing graphene-cellulose composite aerogels [87]. The fabrication method involves several key steps. Initially, graphene oxide (GO) is prepared using either the Hummers method or a modified version as outlined by Ghazitabar et al. [144]. The synthesized GO is then dispersed in a liquid to achieve a homogeneous solution. Concurrently, cellulose nanofibers (CNFs) are obtained by mechanical or chemical treatment of cellulose fibres, followed by their dispersion in a liquid to create a uniform solution. Subsequently, the solutions of GO and CNFs are combined in a predetermined ratio and thoroughly mixed, ensuring a consistent mixture through sonication. This uniform mixture is subjected to hydrothermal treatment within a Teflon-lined autoclave, operating at temperatures between 180 and 220 °C and pressures of 5–15 bar for a

Table 5 Comparative analysis between graphene oxide-cellulose composite aerogels and cellulose aerogels

Property	Graphene oxide-cellulose composite aerogels	Cellulose aerogels
Porosity	High porosity with interconnected pores, resulting in enhanced surface area and adsorption capacity	High porosity with interconnected pores, contributing to high surface area and adsorption capacity
Mechanical strength	Enhanced mechanical strength and stiffness due to the reinforcing effect of graphene oxide	Moderate to high mechanical strength, depending on the cellulose source and processing methods
Surface area	Higher surface area compared to pure cellulose aerogels due to the addition of graphene oxide sheets	High surface area attributed to the porous structure of cellulose aerogels
Density	Slightly higher density compared to pure cellulose aerogels due to the presence of graphene oxide	Low density, often among the lowest of all solid materials, contributing to lightweight properties
Thermal conductivity	Improved thermal conductivity compared to pure cellulose aerogels, attributed to the presence of graphene oxide	Good thermal insulation properties, with low thermal conductivity and high temperature stability
Transparency	Reduced transparency compared to pure cellulose aerogels due to the opaque nature of graphene oxide	Some cellulose aerogels can be transparent to visible light, allowing for optical applications
Chemical modification potential	Both materials offer versatility for chemical modification, enabling tailored properties for specific applications	Similar chemical modification potential, with opportunities for functionalization and customization
Processing complexity	More complex processing methods, such as incorporation of graphene oxide and optimization of composite formation	May involve simpler processing methods compared to composite aerogels, depending on the cellulose source

specific duration. During this process, the CNFs and GO self-assemble, forming a 3D network structure. Following the hydrothermal treatment, the resulting hydrogel is extracted from the autoclave after cooling to room temperature. The hydrogel undergoes multiple washes with deionized water to eliminate any unreacted chemicals and is subsequently freeze-dried to achieve a porous aerogel. For enhanced electrical conductivity of the aerogel, the GO component can be reduced to obtain a reduced graphene oxide (rGO) aerogel. This reduction process involves treating the aerogel with a suitable reducing agent, such as hydrazine hydrate or sodium borohydride [144]. Figure 9 indicates the synthesis of graphene oxide-cellulose composite aerogel using the mild chemical reduction method [144].

Besides the methods used in the reduction of graphene oxide and the drying technique used to obtain the aerogels, when producing cellulose-graphene composites aerogels, there are two crucial factors that must be considered. The first consideration is whether to maintain the intact structure of the original cellulose network. If the cellulose network remains undisturbed, the resulting composite aerogels can benefit from the inherent mechanical strength of the original cellulose. However, it can be challenging to integrate a second phase into the cellulose network, and therefore disintegrating the cellulose has been employed as a strategy to combine cellulose and graphene [145]. The second factor to consider, is the method of incorporating graphene platelets into the cellulose network. Troncoso and Torres [145] synthesized bacterial cellulose-graphene based nanocomposites. In their study, graphene was introduced after the synthesis of bacterial cellulose fibres or during the bacterial cellulose synthesis. The graphene platelets were incorporated by immersing a pristine cellulose pellicle in a graphene dispersion [145].

In the first and second approaches used in the study by Troncoso and Torres [145] described above, cellulose was produced in an unmodified medium resulting in a cellulose pellicle. The cellulose pellicle was either preserved or disintegrated. To disintegrate the cellulose, homogenization or pulping equipment was utilized to obtain a cellulose wet gel which was then combined with a GO dispersion. To ensure a homogeneous dispersion, sonication or another method was used. The resulting cellulose-GO dispersion was then dried, pressed or filtered to form an aerogel or solid film [145]. In cases where the network structure of cellulose was preserved, the GO was integrated through an impregnation step. Subsequently, the cellulose pellicle with GO was dried or pressed [145]. Alternatively, a GO dispersion was poured onto a cellulose pellicle and vacuum-dried to create a film in a single step [145].

In another study, flexible and ultra-light graphene—recycled cellulose nanofibre aerogels were produced by Hovidoost et al. [139] using a similar method described by Ghazitabar et al. [144]. Graphene oxide pre-adsorbed with zinc

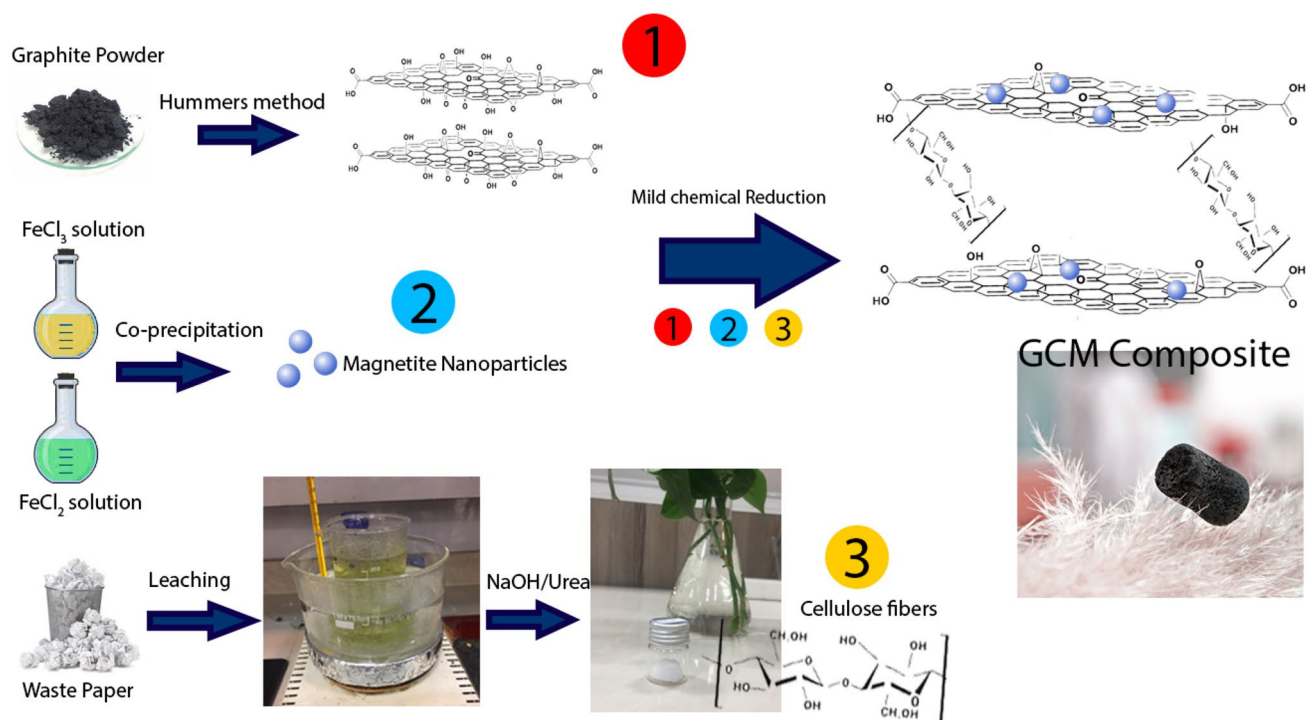


Fig. 9 Schematic of graphene oxide-cellulose composite aerogel synthesis by the mild chemical reduction method [144]

oxide (ZnO) was reduced using a mixture of L-Ascorbic Acid (LAA) and sodium bisulphite (NaHSO_3). A hydrogel was then formed after heating the mixture at 90 °C for 6 h, which was then freeze-dried to form the aerogels [139].

Moharram et al. [146] formed aerogel-like beads from porous graphene oxide crosslinked with cellulose, by air-drying the beads at ambient conditions for two days. In another study by Pinto et al. [147] a novel method which includes addition of dimethyl sulfoxide (DMSO) to the mixture during gelation and using reduction by thermal treatment with ammonia gas to induce crosslinking, was used to form aerogels from bacterial cellulose and graphene oxide. The composite aerogels exhibited considerable improvements in both thermal stability and electrical conductivity. As a result, these versatile aerogels have great promise as environmentally-friendly and sustainable substitute materials for a range of applications such as lightweight packaging, atmosphere and water treatment filters, and energy-related uses [147].

In a study conducted by Pircher et al. [148], fragile, ultra-lightweight, open-porous, and transversally isotropic bacterial cellulose (BC) aerogels were strengthened with biocompatible polymers such as polylactic acid (PLA), poly-caprolactone (PCL), cellulose acetate (CA), and poly(methyl methacrylate) (PMMA) at varying BC/polymer ratios. To incorporate the secondary polymer into the BC matrix and transform the resulting composite organic gels into aerogels, the techniques of supercritical carbon dioxide anti-solvent precipitation and simultaneous extraction of the anti-solvent using super critical CO_2 were employed [148].

Xia et al. [149] conducted a study on a straightforward and scalable bottom-up approach for producing 3D porous hybrid aerogels (HAs) composed of one-dimensional cellulose nanofibres (CNFs), multiwalled carbon nanotubes (MWCNTs), and two-dimensional graphene oxide (GO). Following the incorporation of Fe_3O_4 nanoparticles (~ 200 nm) into the HA substrate, a high-performance flexible electrode made of CNF/MWCNT/reduced-GO (RGO)/ Fe_3O_4 was created. This electrode exhibited a large capacitance of 1193 mF cm^{-2} , outstanding rate capability, and high durability [149].

4.3.2 Application of graphene-cellulose composite aerogels in waste water treatment

Recently, GCCAs have emerged as promising materials for water treatment due to their unique properties, including high porosity, large surface area, and electrical conductivity. The hydrothermal process used to manufacture GCCAs promotes the formation of a strong interfacial interaction between cellulose and graphene, resulting in improved mechanical properties and enhanced adsorption performance of the composite aerogels [139].

The ability of GCCAs to adsorb different types of water pollutants, such as heavy metals, organic dyes, and pharmaceuticals, has been the subject of investigation. For instance, Arabkhani and Asfaram [150] developed a novel 3D magnetic bacterial cellulose nanofibre/graphene oxide polymer aerogel comprising of bacterial cellulose nanofibres, Fe_3O_4 nanoparticles, GO nanosheets, and poly(vinyl alcohol), which achieved 93% adsorption efficiency, in the removal of malachite green (MG) dye from aqueous solution [150].

In another study, Ghazitabar et al. [151] reported the use of novel graphene aerogel/cellulose fibres/magnetite nanoparticles (GCM) composites, in the removal of Au from an industrial alkaline cyanide solution. The GCM composites showed a high gold adsorption capacity of 130 mg g^{-1} , along with a mechanical strength of 7 kPa at 50% strain, which indicates the possibility of using the GCM composites in industrial applications [151].

The sources of cellulose are abundant. Joshi et al. [152] synthesized GCCAs using cellulose derived from fruit-waste to study its adsorption efficiency for the removal of dyes. The composite aerogels achieved an adsorption removal efficiency of 98%, which was much higher than the adsorption efficiency of GO powder, activated carbon, cellulose aerogels, and graphene aerogels [152].

Results from a study conducted by Shadkam et al. [141] displayed promising potential for graphene-cellulose based aerogels to perform as a new class of adsorbents. Reduced graphene-oxide/cellulose nanocrystal hybrid aerogels reinforced with waste-paper extracted cellulose-fibres resulted in a maximum adsorption capacity of 454 mg g^{-1} for the removal of toluene from an aqueous medium [141].

One of the advantages of cellulose-graphene composite aerogels is their regeneration ability and reusability, which is crucial for practical applications [139]. Several regeneration methods have been investigated, including thermal regeneration, chemical regeneration, and bio regeneration. In one approach, the regeneration of the aerogels involved an initial stage in which metal ions from the aerogels were removed by dehydrating the material using mechanical compression, followed by a continuous addition of ethylenediaminetetraacetic acid (EDTA) solution until the original form is restored [153].

A study by Hoviatdoost et al. [139] also demonstrated that GCCAs could be effectively regenerated using either mechanical (squeezing) or thermal desorption (heat treatment). The aerogels maintained a high absorption capacity of 84%, after 10 cycles of being compressed and released [139]. Another study by Joshi et al. [152] also demonstrated the

ability to regenerate graphene-fruit waste derived cellulose aerogels for removal of dyes, by rinsing in a NaOH solution [152]. This further indicates the sustainability of GCCAs.

Mi et al. [142] fabricated a highly compressible ultra-light anisotropic cellulose/graphene aerogel by bidirectional freeze drying for selective oil absorption. The aerogel's remarkable elasticity allows for quick and effective retrieval of absorbed oil through straightforward mechanical squeezing. This feature ensures exceptional performance consistency even after multiple uses [142].

The unique combination of high compressibility and efficient oil recovery through mechanical squeezing sets modified GCCAs apart from other aerogels. Consequently, modified GCCA shows great potential as a highly effective material for targeted oil absorption and recovery. Table 6 represents the advantages and disadvantages of other water treatment methods, compared to using aerogels.

5 Discussion: future prospects of cellulose and graphene-based aerogels in wastewater treatment

5.1 Cellulose based aerogels

Cellulose-based aerogels have shown great potential for water treatment applications due to their high surface area, porosity, and ability to adsorb contaminants. However, as outlined in the sections above, there are still some challenges in their application for water treatment. It was noted that cellulose-based aerogels have limited stability in water, which tends to limit their effectiveness for long-term water treatment applications. This can be addressed through the development of crosslinking methods or the incorporation of other materials into the cellulose aerogels that improve their stability.

Furthermore, while cellulose-based aerogels have shown good adsorption capacities and removal efficiencies for certain contaminants such as heavy metals and organic pollutants, their adsorption capacity for several other emerging contaminants such as pharmaceuticals has not been fully investigated. Further research is needed to develop cellulose-based aerogels with improved adsorption properties for emerging and a wider range of contaminants. Further research is also necessary to develop nanocellulose-based adsorbents that can simultaneously remove multiple species from a multicomponent contaminant system, similar to those found in actual polluted water and wastewater.

Also, the scalability of the production of cellulose-based aerogels for water treatment is still a challenge. Current methods for the production of cellulose-based aerogels are generally expensive and time-consuming, limiting their practical use on a large scale. Investigating the scale-up of production processes is essential to meet a potential increase in the industrial demand for cellulose-based aerogels. Therefore, research efforts should focus on developing cost-effective and scalable methods for the production of cellulose-based aerogels to make them more accessible for water treatment applications.

Additionally, the post-treatment regeneration or disposal processes of nanocellulose-based aerogels for long-term use require further study. This review showed that many research studies on nanocellulose aerogels lack multiple adsorption-desorption runs or effective biodegradation procedures. Magnetic nanocellulose composites in the form of aerogels could be explored as they would allow for easy separation from sludge and an extended lifespan. Furthermore, research on nanocellulose aerogels for the removal of emerging contaminants could be expanded. Additionally, impregnating nanocellulose aerogels with photocatalytic nanomaterials for synchronous adsorption and photodegradation can be explored further for water remediation applications.

Several other critical aspects require further investigation to advance the field of cellulose-based aerogels. For instance, there is a pressing need to develop more cost-effective production methods for cellulose-based aerogels to make them commercially viable. Moreover, enhancing their mechanical properties, particularly stiffness and strength, is crucial to broaden their applicability. Functionalization of these aerogels involving the integration of nanoparticles or chemical groups presents a promising pathway to enhancing specific properties which for instance could hence their removal capacities and efficiencies for pollutants from water. Studies on the environmental impact of the production, utilization and disposal of cellulose-based aerogels are imperative to ensure sustainability.

Besides their potential effectiveness in water treatment applications, there is wide scope to utilise cellulose based aerogels in other applications such in energy storage, catalysis, and biomedical engineering, which requires further research to unlock their full potential. By addressing knowledge gaps on cellulose based aerogels outlined in this study, a

Table 6 Advantages and disadvantages various water treatment methods

Property	Polymer foams	Activated carbon	Ion exchange resins
<i>Advantages</i>			
High surface area	Can be engineered for high surface area [154]	Exhibits high adsorption capacity [155]	Selectively removes specific ions
Tunable properties	Flexibility in design for specific applications [156]	Versatile with tailored pore sizes and chemistries [157, 158]	Consistent performance over time
Mechanical stability	Depending on the polymer, can exhibit good mechanical stability	Regenerable, extending lifespan	Compact design suitable for various scales
Cost-effective	Some are cost-effective to produce	Well-established technology	Regenerable, reducing operational costs
<i>Disadvantages</i>			
Limited adsorption capacity	May have limited capacity compared to materials with higher surface area [159]	Expensive, especially for high-quality grades [160]	Finite capacity, requiring regeneration [161]
Limited selectivity	May lack selectivity towards specific contaminants	Particle leaching may pose health concerns	Specificity may lead to incomplete removal [162]
Durability issues	Some may degrade over time, reducing effectiveness	Bulky and heavy, requiring significant space	Chemical handling risks during regeneration
Environmental concerns	Additives or residues may leach into water, posing risks	Adsorption kinetics may be slow [163]	pH sensitivity requires careful monitoring [164]

new field of study can emerge, focusing on high-performance and functionalized cellulose aerogels tailored for advanced industrial-scale applications.

The future of cellulose aerogels in the water field holds significant promise for addressing water purification and desalination challenges. With their unique properties and tuneable characteristics, cellulose aerogels offer versatile solutions for enhancing water treatment processes. Moving forward, research efforts should focus on optimizing aerogel synthesis methods to improve cost-effectiveness, mechanical properties, and functionalization capabilities. Integration of cellulose aerogels into advanced water treatment technologies, such as membrane filtration systems and capacitive deionization devices, is anticipated to drive innovations in water purification and desalination. Additionally, exploring the scalability of cellulose aerogel production and investigating their performance in real-world applications will be critical for realizing their full potential in addressing global water scarcity issues.

Cellulose, as a raw material for aerogel production, offers several compelling attributes. Cellulose is abundantly available and renewable, being the most prevalent organic polymer on Earth, sourced from various forms such as wood, cotton, and agricultural waste. This abundance ensures a sustainable supply chain for aerogel production, aligning with environmentally friendly practices. Additionally, cellulose is highly biocompatible and biodegradable, making it suitable for applications where environmental impact and biocompatibility are crucial considerations, such as biomedical engineering and water treatment. Its biocompatibility ensures minimal adverse effects on living organisms, while its biodegradability ensures environmentally responsible end-of-life disposal. Furthermore, cellulose-based aerogels can be engineered to possess high surface area and porosity, characteristics vital for efficient water treatment processes. These aerogels offer ample sites for the adsorption of contaminants in water, enhancing the overall effectiveness of water treatment technologies. Overall, cellulose stands as a versatile and sustainable material choice for aerogel production, offering a combination of abundance, biocompatibility, and high surface area for diverse applications, including water treatment.

5.2 Graphene based aerogels

Graphene-based aerogels have shown great potential for water treatment applications due to their high surface area, mechanical strength, and excellent electrical and thermal conductivity. However, similar to cellulose based aerogels, their application in water and wastewater treatment is fraught with challenges.

From the studies reviewed, one of the major challenges is the scalability and cost-effective production of graphene-based aerogels. The current methods for producing graphene-based aerogels are often expensive and time-consuming, which limits their practical use on a larger scale. Therefore, there is a need to develop more scalable and cost-effective production methods.

Another challenge is the limited selectivity of graphene-based aerogels for certain contaminants. While graphene-based aerogels have excellent adsorption properties for a wide range of contaminants such as heavy metals, organic pollutants, and dyes, their selectivity for specific contaminants is limited. Further research is needed to develop graphene-based aerogels with enhanced selectivity for specific contaminants and adsorption efficiency of other emerging pollutants.

Additionally, the long-term stability of graphene-based aerogels in water is another challenge. Graphene-based aerogels (without functionalization) tend to aggregate and lose their structure in water over time, which limits their effectiveness for long-term water treatment applications. Strategies such as surface functionalization and the incorporation of other materials can improve the stability of graphene-based aerogels in water.

Also, the potential environmental impacts of graphene-based aerogels are not yet fully understood. The release of graphene-based nanomaterials into the environment may have unintended consequences and potential adverse effects. Further research is needed to understand the potential environmental impact of graphene-based aerogels and develop strategies to mitigate any negative effects.

Graphene, with its remarkable properties, holds significant promise for enhancing aerogel performance in various applications. Graphene exhibits exceptional mechanical strength and stiffness, making it an ideal candidate for reinforcing aerogel structures. This reinforcement enhances the mechanical properties of aerogels, ensuring durability and resilience in demanding environments. Additionally, graphene's high electrical conductivity is advantageous for applications requiring efficient electron transport, such as capacitive deionization for water desalination. The conductivity of graphene facilitates rapid charge transfer within the aerogel matrix, improving the efficiency of water treatment processes. Moreover, graphene's excellent adsorption properties are noteworthy, attributed to its large surface area and high adsorption capacity. These properties enable graphene to effectively adsorb diverse contaminants, including heavy metals, organic pollutants, and microorganisms, from water sources. By leveraging graphene's exceptional mechanical

strength, electrical conductivity, and adsorption properties, graphene-based aerogels offer promising solutions for various environmental and water treatment challenges, demonstrating their potential for advancing sustainable and efficient water purification technologies.

Overall, graphene-based aerogels have great potential for water treatment applications, but there are still some challenges that need to be addressed to fully realize their potential.

5.3 Graphene-cellulose composite aerogels

This review showed that graphene-cellulose composite aerogels (GCCAs) have gained significant interest for water treatment applications due to the combined properties of both materials. However, there are also still some challenges in their application for water treatment.

One of the main challenges is the production of uniform graphene-cellulose composite aerogels. The production of these composite materials often requires the use of harsh chemicals and high temperatures, which can result in non-uniform materials with reduced properties. Developing more efficient and uniform production methods for graphene-cellulose composite aerogels is crucial for their practical application.

The potential environmental impacts of graphene-cellulose composite aerogels need to be explored further. As with graphene-based aerogels, there is a need to understand the potential environmental impact of the graphene-cellulose composite aerogels on the environment and develop strategies to mitigate any negative effects.

While graphene-cellulose composite aerogels have shown improved properties and removal capacities and efficiencies for pollutants from water compared to aerogels prepared from the individual materials, further research is needed to fully understand the mechanisms behind their adsorption processes and optimize their performance for a wide range of contaminants. The regeneration ability and reusability of the composite aerogels make them an attractive option for practical applications.

Combining cellulose and graphene in a composite aerogel offers a compelling solution for advancing water treatment technologies. This synergistic approach harnesses the unique properties of both materials to achieve enhanced performance. Cellulose provides structural integrity, biocompatibility, and a high surface area for adsorption, while graphene contributes exceptional mechanical strength, electrical conductivity, and adsorption properties. As a result, the composite aerogel exhibits superior performance compared to its individual components, boasting improved adsorption capacity, mechanical stability, and conductivity. Moreover, the composite aerogel's versatility allows it to be tailored to specific water treatment needs by adjusting the composition and structure. It can be further functionalized with specific groups or nanoparticles to target particular contaminants, making it adaptable for various water treatment applications, including desalination, heavy metal removal, organic pollutant adsorption, and microbial disinfection. By leveraging the synergistic properties of cellulose and graphene, composite aerogels offer promising solutions for addressing complex water treatment challenges, demonstrating their potential to enhance the efficiency and sustainability of water purification processes.

5.4 Developmental trends in aerogels for water treatment applications

Aerogels for water treatment applications have undergone significant evolution over time, reflecting advancements in scientific understanding, technological innovations, and emerging challenges. In the past research efforts on aerogels were mostly exploratory and primarily focused on investigating the potential of novel materials for water treatment, with an emphasis on understanding basic principles and feasibility. Early studies often involved proof-of-concept experiments demonstrating the efficacy of materials such as cellulose and graphene-based aerogels in water purification and desalination. Additionally, method development, including synthesis techniques and prototype device construction, laid the foundation for subsequent advancements.

Presently, research efforts on aerogels have shifted towards optimization and scale-up of synthesis processes, enhancement of material properties, and interdisciplinary collaboration among researchers from various fields to address complex challenges. There is a noticeable transition towards application-oriented research, targeting specific water quality issues such as heavy metal contamination, organic pollutants, and microbial pathogens. Furthermore, integration of advanced materials such as aerogels with existing water treatment technologies, for instance membrane filtration and ion exchange, is gaining traction to enhance treatment efficiency.

It is envisaged that future work on aerogels will prioritize sustainability and environmental impact, with a focus on developing eco-friendly materials and processes for water treatment. Advanced functionalization of aerogels tailored

for targeted pollutant removal, antimicrobial activity, and selective ion exchange is anticipated to drive future research directions. Moreover, the development of aerogel based smart materials and sensing technologies capable of real-time monitoring and adaptive response to changing water quality conditions holds promise for innovation. Exploration of emerging applications, such as decentralized water treatment systems and resource recovery from wastewater, is likely to shape future research and development efforts in the field.

5.5 Future direction in the application of aerogels in water treatment

In the realm of water treatment using aerogels, several promising avenues for future research and development emerge. Firstly, it is imperative to prioritize sustainability and green chemistry principles in material synthesis and water treatment processes. This necessitates the development of eco-friendly materials and methods aimed at minimizing energy consumption, waste generation, and environmental impact. Additionally, further exploration of advanced functionalization techniques is warranted to tailor material properties for specific water treatment applications. This includes incorporating functional groups, nanoparticles, and nanocomposites to enhance adsorption capacity, selectivity, and antimicrobial activity. Moreover, integrating smart materials and sensing technologies into water treatment systems holds significant promise. This integration enables real-time monitoring, adaptive control, and predictive analytics, ultimately improving efficiency and performance.

Continued advancements in nanotechnology and the development of novel nanomaterials offer opportunities for enhancing water treatment capabilities, particularly in targeted pollutant removal, membrane technology, and catalysis. Addressing the challenges of decentralized water treatment systems for rural and remote communities is essential, emphasizing the need for research focused on the design, optimization, and implementation of small-scale, low-cost technologies suitable for decentralized applications.

Furthermore, exploring resource recovery from wastewater streams and promoting circular economy principles are integral to sustainable water management. Future research endeavours should investigate methods for recovering valuable resources such as energy, nutrients, and materials from wastewater while minimizing environmental impact. Lastly, encouraging cross-disciplinary collaboration among researchers from diverse fields will foster innovation and address complex water challenges holistically.

5.6 Future applications of aerogels in small scale and large-scale water purification systems

Water purification systems span the whole spectrum of small-scale point-of-use applications to large-scale municipal systems, each posing unique challenges and opportunities for the utilization of advanced materials such as aerogels. Historically, the trends in the application of aerogels for water purification have evolved from exploratory laboratory studies to pilot-scale demonstrations, showcasing their potential across different scales. Looking ahead, the future of aerogels in water purification is projected to witness a continued transition towards practical implementation in both small-scale and large-scale systems.

At the small-scale point of use, aerogels offer compact, efficient, and cost-effective solutions for household water purification. Aerogel-based filters and cartridges can effectively remove contaminants such as heavy metals, organic pollutants, and microbial pathogens from drinking water, ensuring safe and potable water for individual households. Future trends in this domain may include the development of portable and user-friendly aerogel-based filtration devices tailored for diverse settings, including rural and resource-constrained areas.

On the other hand, in large-scale municipal systems, aerogels hold promise for augmenting existing water treatment processes and addressing specific challenges such as pollutant removal, desalination, and resource recovery. Integration of aerogel-based adsorbents, membranes, and catalysts into conventional treatment systems can enhance efficiency, reduce energy consumption, and improve water quality. Future projections in this realm may involve the scaling up of aerogel production, optimization of manufacturing processes, and deployment of aerogel-based technologies in municipal water treatment plants.

Commercially available water purification systems based on graphene and cellulose aerogels are gradually emerging, albeit still in the early stages of development and commercialization. Companies and research institutions are actively exploring the scalability and viability of aerogel-based water purification technologies for various applications. While specific commercial products may not yet be widespread, ongoing research and development efforts are paving the way for the future commercialization of aerogel-based water purification systems.

5.7 Scaling-up and the environmental impact of producing aerogels

Scaling up the production of aerogels and ensuring the environmental friendliness of the processes used are critical aspects in advancing their practical applications. Scaling up production involves several key steps. Firstly, optimization of synthesis methods is essential to achieve consistent quality and high yield. This includes fine-tuning process parameters such as precursor concentrations, reaction conditions, and drying techniques to ensure reproducibility and efficiency on a larger scale.

Batch and continuous processing methods are employed, depending on the specific synthesis route, enabling either larger volume production in single batches or continuous production with a steady output rate. Specialized equipment design and engineering are necessary to handle larger material volumes and maintain precise control over process parameters.

Assessing the economic feasibility of scaled-up production is crucial, considering factors such as capital investment, operational costs, and market demand. Implementing robust quality control measures and characterization techniques ensures the consistency, purity, and performance of aerogels produced on a larger scale. Additionally, environmental friendliness is paramount in aerogel production processes.

Developing green synthesis routes, enhancing energy efficiency, implementing waste management and recycling strategies, conducting lifecycle assessments, and adhering to regulatory compliance are vital considerations to minimize environmental impact and promote sustainability. Developing green synthesis routes involves exploring solvent-free or low-toxicity solvents, eco-friendly catalysts, and renewable precursors to reduce the use of hazardous chemicals and promote sustainability.

Enhancing energy efficiency in aerogel production processes can significantly reduce carbon emissions and environmental footprint. Implementing energy-efficient heating, cooling, and drying techniques, as well as utilizing renewable energy sources, can contribute to reducing energy consumption and greenhouse gas emissions.

Proper waste management practices and recycling strategies are important considerations in aerogel production. Minimizing waste generation, reusing by-products or waste materials, and implementing recycling programs for unused or defective aerogel products can help mitigate environmental impact and promote circular economy principles.

Conducting lifecycle assessments (LCAs) of aerogel production processes can provide valuable insights into their environmental footprint, from raw material extraction to end-of-life disposal. Assessing environmental impacts such as energy consumption, water usage, greenhouse gas emissions, and waste generation can inform decision-making and guide efforts towards improving environmental sustainability.

Adhering to regulatory requirements and environmental standards is essential to ensure compliance with environmental regulations and mitigate potential risks to human health and the environment. This may include obtaining permits, conducting environmental impact assessments, and implementing pollution prevention measures to minimize environmental harm.

By addressing these considerations, scaling up the production of aerogels and ensuring environmental friendliness can contribute to the widespread adoption of aerogel-based technologies in various applications while minimizing their environmental footprint.

6 Conclusions

This study sought to bring out to the forefront the headways made in the development of high-performance and functionalized cellulose and graphene-based aerogels and their applications in advanced industrial-scale applications, particularly water treatment. The study also sought to highlight the existing challenges in the development and utilization of these aerogels in water treatment, and to explore the proposed solutions to overcome these. Thus, in this study, the state-of-the-art with respect to the synthesis methods, properties, application, effectiveness and performance in water and wastewater treatment of cellulose, graphene and their composite aerogels was critically reviewed.

It can be concluded that there is need to develop more cost-effective production methods for cellulose-based aerogels to make them commercially viable. Large scale production methods for cellulose based aerogels still need

to be developed. The mechanical properties of the cellulose-based aerogels also need to be enhanced to for their applicability to be more useful. Cellulose based aerogels offer exciting prospects in water treatment due to their ability to be functionalised to target specific pollutants. However, the impact of production, utilization and disposal of cellulose aerogels on the environment still needs to be studied.

The effectiveness of cellulose based aerogels in water treatment, is hampered by their limited long-term stability in water. There has also been limited studies on their effectiveness in the simultaneously remove multiple pollutants from multicomponent contaminant systems, similar to those found in actual wastewater. Also, the removal capacities and efficiencies of the cellulose based aerogels for emerging pollutants such as pharmaceuticals is still yet to be studied. Post treatment regeneration and disposal of cellulose based aerogels for long term use still needs to be studied.

Regarding graphene-based aerogels, there is need to develop more cost effective and scalable production methods. The selectivity for graphene-based aerogels for specific pollutants needs to be further enhanced through further studies. The long-term stability of graphene-based aerogels needs to be further studied, together with the potential environmental impact.

Cellulose-graphene composite aerogels have shown improved properties, removal capacities and efficiencies for pollutants from water compared to aerogels prepared from the individual materials. However, their production processes require the use of harsh chemicals and conditions, limiting practical application. Their pollutant removal mechanisms still need to be understood. The performance of these composite aerogels in the removal of specific pollutants also needs to be optimized for a wide range of pollutants. Although the regeneration ability and reusability of the composite aerogels make them an interesting prospect for practical applications, the potential impact of the graphene-cellulose composite aerogels on the environment also need to be understood and strategies developed to mitigate any negative effects.

Overall, graphene-cellulose composite aerogels have great potential for water treatment applications, particularly in the removal of heavy metals and organic pollutants. However, the challenges outlined in this study need to be addressed to fully realize their potential.

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Declarations

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