Review

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Incorporation of Cellulose-Based Aerogels into the Textile Structure

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Abstract: Given its exceptional attributes, aerogel is viewed as a material with immense potential. Being a natural polymer, cellulose offers the advantage of being both replenishable and capable of breaking down naturally. Cellulose-derived aerogels encompass the replenish ability, biocompatible nature, and ability to degrade naturally inherent in cellulose, along with additional benefits like minimal weight, extensive porousness, and expansive specific surface area. Even with increasing appreciation and acceptance, the undiscovered possibilities of aerogels within the textile sphere continue to be predominantly uninvestigated. In this context, we outline the latest advancements in the study of cellulose aerogel formulation and their diverse impacts on textile formations. Drawing from the latest studies, we reviewed the materials used for the creation of various kinds of cellulose-focused aerogels and their properties, analytical techniques, and multiple functionalities in relation to textiles. This comprehensive analysis extensively covers the diverse strategies employed to enhance the multi-functionality of cellulose-based aerogels in the textile industry. Additionally, we focused on the global market size of bio-derivative aerogels, companies in the industry producing goods, and prospects moving forward.

Keywords: bio-based aerogel; multi-functional properties; thermal insulation; flame retardant; textile applications

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1. Introduction

Aerogels are a fascinating substance of the twenty-first century due to its unique structure [1]. They possess remarkable properties like high porosity, low density, huge surface area, and superb heat and sound insulation. However, their low mechanical strength and high production costs restrict their usefulness [2].

Aerogels are extremely porous nanostructured materials invented by Kistler in 1931 [2,3]. Aerogels may be created using either supercritical drying or freeze-drying processes. The material's microporous structure stays intact throughout drying in both circumstances. Kistler's first aerogel, prepared by supercritical drying, was silica based. During the creation of silica aerogels, toxic precursors were used, and the aerogel formed was not biodegradable [4]. The costly process of making this type of aerogel, having restrictions described earlier confirms restricted use of this material[4].

Aerogel is also used to develop a wide range of tools, such as optoelectronics, adsorption catalysis, sound insulation, pharmaceutical materials, and aerospace materials [5–10]. However, some drawbacks, together with the huge expenses associated with fabrication, have severely limited the use of aerogels [11]. Aerogels can be formed using a wide range of materials, including inorganic ones [12], synthetic polymer-based [13], and natural polymer-based [14,15] as cellulose [16,17] depending on the starting substance used for their manufacture [2,18].

Cellulose aerogels, in particular, have cellulose's renewability, biocompatibility, and biodegradability, as well as additional benefits like small density value (0.0005–0.35 $g.cm^{-3}$), enhanced porosity (84.0–99.9%), along with a huge specific surface area, representing cellulose aerogels as the materials with the highest potential in the 21st century [11]. Cellulose aerogels represent a greater compressive strength (0.0052 -16.67 MPa) and superior biodegradability [11]. So, cellulose aerogels are an ecofriendly and versatile modern material with enormous opportunities in the application of adsorption and oil/water separation, heat separator, biomedical materials, metal nanoparticle/metal oxide carriers and carbon aerogel precursor. Cellulose aerogel as a porous solid, is typically produced through a two-step process: cellulose or cellulose derivatives are dissolved/dispersed, resulting in the formation of cellulose sol using the sol-gel method, and the cellulose sol is subsequently dried to gel preserving sol three-dimensional porous structure. [11].

This study describes the materials used in the production of many types of cellulose-based aerogels, their features, analytical techniques, and multi-functionality on textiles. For the textile sector, the different techniques for the multifunctionality of cellulose-based aerogels and analyses are comprehensively discussed [11].

2. Creating Cellulose Aerogel

Cellulose can be derived from various sources [19–21], Plants and plant-based materials, such as rice straw, are among the most commonly utilized sources for obtaining cellulose [22], cannabis [23], cotton [24,25], wood [26,27], potato tubers [28], coconut (coir) [29] and bagasse [30]. The extraction of cellulose involves obtaining it from specific plant species and employing various production processes, including pretreatment, post-treatment, and disintegration processes, determine its performance characteristics, like size, molecular chain length (degree of polymerization, DP), thermal stability, and degree of crystallinity [31,32]. As a result, the plant source significantly impacts the structure and performance of cellulose aerogels [34], [35].

Cellulose is a member of the polysaccharide's family, which is the primary building element for plants. Plants are the first or most basic link in the food chain (which details the feeding interactions of all living organisms) [33–35]. Cellulose is a key component of many natural fibers, including cotton and other plants [36,37].

Cellulose exhibits insolubility in water and the majority of common solvents [34], owing to strong intramolecular and intermolecular hydrogen bonding between individual chains [33]. Cellulose is employed in a variety of products despite its poor solubility, including composites, netting, upholstery, coatings, packaging, and paper. To make cellulose more processable and to produce cellulose derivatives, which can be customized for certain industrial purposes, cellulose is chemically modified [39,40].

Aerogel materials can benefit from the mechanical qualities and moisture affinity of cellulose and its derivatives [41–43]. To keep their solid network, the dissolving cellulose in appropriate media, such as NMMO, hydrates of some molten salt and ionic liquids is realized, and then drying using processes such as supercritical or freeze drying [44–48] are used for preparation of cellulose aerogels. There are several strategies for creating cellulose aerogels published in the literature [45,46,49–51]. The manufacturing of cellulose aerogels and their applications are depicted in Figure 1.

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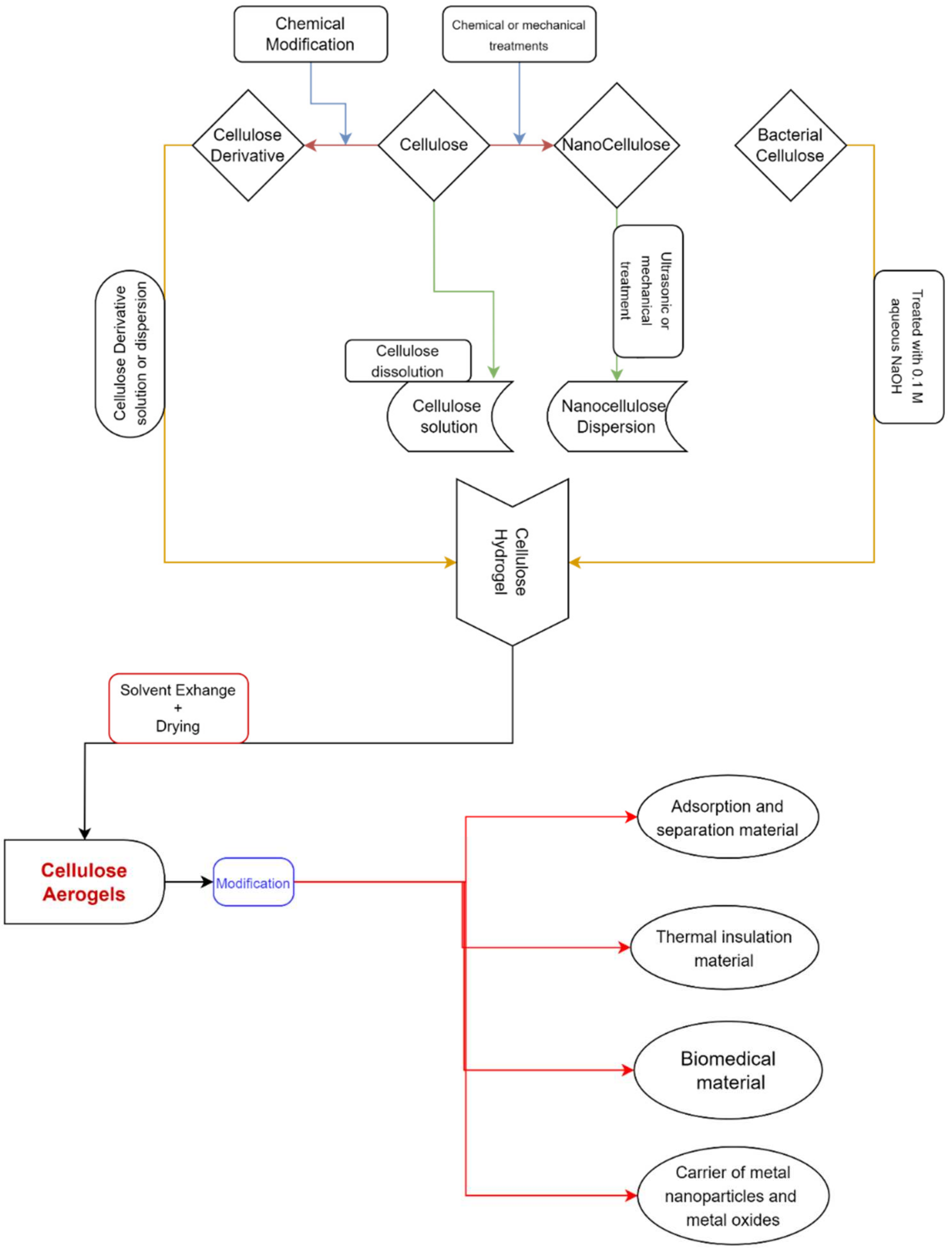

Figure 1. A graphical representation depicting the manufacture and application of cellulose aerogels [38].

Furthermore, employing cellulose as a precursor in the production of aerogels has the following benefits. (1) The supply of cellulose raw materials is infinite and renewable; (2) Because the cellulose chain contains a lot of hydroxyl groups, no cross-linking agent is needed during the aerogel production process. A stable three-dimensional (3D) network structure can be created by employing hydrogen bond physical cross-linking both intra- and -inter molecules, building the aerogel production technique incredibly easy. (3) Chemical cellulose modification is a quick and easy method for enhancing the structural integrity and mechanical strength of cellulose aerogels. The performance and concentration of the cellulose have a significant impact on the manufacturing process and structural characteristics of cellulose aerogels [11]. To create cellulose gels, the cellulose morphology and structure of the cellulose fibers must be changed. This is done by using a suitable solvent [52–54], that is capable of breaking the large hydrogen bonding network that does not degrade along the cellulose polymer chain or beginning polymer chain derivate processes [45].

Because of its numerous interior pores and effective heat insulation, cellulose aerogel is one of the most capable thermal insulating materials for construction or domestic applications (for

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example, refrigerator insulation material) and has the potential to improve their poor properties as flame retardancy, huge swelling, and antibacterial properties [55,56].

Table 1. Classification of Cellulose-based aerogel together with published examples from.

Classification of Cellulose Aerogels						
Cellulose -Aerogel Type	**Starting material**	**Solvent**	**Surface chemistry**	**Drying method**	**Application**	**Ref.**
1. Natural Cellulose	Pineapple leaf fiber, Cotton waste fiber	Poly (vinyl alcohol) (PVA)	-		Building towards sustainable development	[29]
	Raw cotton fibers and cotton stalk	Tert-butyl alcohol	-	Freeze-drying Freeze-drying	-	[57]
	Softwood cellulose pulp	TEMPO	Monocomponent endoglucanase, cupriethylendiamine		Bio-fabrication of tissues, additional health and pharmacological uses	[58]
1.a. Nano Cellulose	Cellulose nanofibers (CNFs), Graphite powder, concentrated sulfuric acid, concentrated acetic acid, solution hydrogen peroxide	Sodium hydroxide, sodium hypochlorite, MO (methyl orange), and potassium permanganate	NaOH	Freeze-drying	The treatment of domestic organic wastewater	[59]
1.b. Bacterial Cellulose	Komagataeibacter sucrofermentans H-110, TEMPO, dextrose, protein hydrolysate, yeast concentrate, disodium phosphate	Sodium hydroxide solution	NaClO, NaBr	Freeze-drying	Bio-fabrication of tissues and preparation of injury treatment materials	[4]
	Bacterial cellulose (BC) pellicles	-	Deionized water (DIW)		Pressure sensors, batteries and super-capacitors, substrates for catalysts, high-tech detectors	[60]
2. Regenerated Cellulose	Cotton and viscose-based regenerated cellulose	Imidazolium acetate ([EMIM], non-enium acetate ([DBNH][OAc])	DMSO	Supercritical CO_2, Lyophilization, ambitious drying	-	[61]
	Bamboo pulp boards	NaOH/urea aqueous solutions	Methyl-pyrrolidone (NMP), potassium hydroxide (KOH)	Freeze-drying Freeze-drying	Application of energy storage devices	[62]
	Bamboo cellulose nanofibrils (BCNF)	Polyvinyl alcohol (PVA)	Sodium tetraborate decahydrate (borax), N, N′-methylenebisacrylamide (MBA), Methyltrimethoxysilane (MTMS)		Eco-friendly wrapping in the refrigerated transportation of fresh produce	[63]

3. Cellulose Derivative	Softwood kraft pulp sheets	1,2-ethanediol, hydroxylammonium chloride monochloroacetic acid, poly-(1,4)-β-D-glucosamine	Sodium (meta) periodate, sodium chlorite	Freeze-drying	The production of advanced bio-adsorbents	[64]
	Softwood bleached kraft pulp (SBKP)	Water/tert-butyl alcohol (TBA)	(TEMPO)-oxidized cellulose nanofibril (TOCN)	Freeze-drying	High performance air filter	[65]
	Cellulose acetate	Acetone	Polymethylene polyphenylpolyisocyanate (PMDI)	$ScCO_2$ drying	Thermal insulation application	[51]

2.1. Sol - Gel Procedure

All steps of the production process influences of the gel structural characteristics, determining its characteristics and, as a result, the utilizations (Figure 2) [66]. Frequently, also other techniques are employed in improving the structural attributes and characteristics of the cellulose based gel [67].

- A colloidal suspension is produced by dispersing solid nanoscale particles formed from a reactant in a liquid.
- Adding an acidic or basic catalyst initiates crosslinking and leads to the linkage and spreading of particles, forming an interlinked network configuration.
- Gel aging: To strengthen the gel's backbone and material toughness, it is aged in its mother solution.
- To avoid gel fracture, the solvent is extracted from the pores of the gel during drying. [68].

The procedure starts with the creation of a colloidal solution, often known as a sol. A solution of reactants and solvents contains solid nanoparticles or initiator materials. Ageing, drying, densification, crystallization, hydrolysis, polycondensation, and gelation are the steps needed. Following the creation of the sol, it is commonly mixed with one of the preceding methods, resulting in a spongy framework within a wet,- semi-solid consistency [68,69].

Solvent-gel products may be manufactured from a variety of substances, including oxides such as silicon dioxide and oxide minerals, natural compounds such as large molecules like plant-derived material, and carbon-based substances such as 2D carbon allotropes and carbon nano pipes [68,70]. Pineapple fibers (PF) aerogels were successfully created by pre-treating PFs with naturally decomposable polyvinyl alcohol (PVA). PVA solution preparation was combined with PFs and freeze-dried. According to the findings, the PF have high porosities (~99%), ultra-low densities, and micro-porous formations as shown by field-emission scanning electron microscope, Brunauer-Emmett-Teller isotherm, and X-ray diffraction analysis. The exceptionally low thermal conductivities of PF aerogel demonstrate its applicability for thermal barrier uses. A thermal coat wrapped over a water bottle with a PF aerogel filling can unquestionably keep the water temperature near 0°C (just above the freezing temperature) up to 6 hours (temperature for beginning: -3 °C) and more than 40 °C up to 2.5 hours (temperature for beginning :90 °C). The thermal coat has a potential thermal barrier that is nearly three times that of a product that is currently on the market [71]. The goal of the paper waste cellulose aerogel is to develop a thermal coat for army canteens to increase the life of ice slurry for dynamic army troops in exercise or operations. However, because of the minimal stretching capacity and the ease with which the bio-based aerogel structure can be damaged, the bio-based aerogels must be sandwiched between two protective layers to make the thermal coat more durable. The paper waste was combined with deionized water and crosslinked with Kymene chemicals (crosslinker based on polyamide-epichlorohydrin resin) before being frozen overnight. After freezing, the gel was dried using the lyophilization drying technique at -91 °C to create cellulose aerogels, followed by the crosslinking process in the dryer for 3 h at 120°C. Following all measurements, the results show that the heat barrier function of the developed thermal coats is significantly better than that of marketed thermal flasks, and similar to that of vacuum flasks for the

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same duration of 4 hours and surrounding temperatures [72]. Cellulose aerogels were made by dissolvable cellulose filaments in melts of calcium thiocyanate salt hydrate, then regenerating in ethanol and drying under supercritical CO_2. It is possible to create uniform structured bio-based aerogels with minimal bulk mass. The microstructure of bio-based aerogels exhibited a continuous 3D network with a large specific surface ratio coupled with a significantly sponge-like structure (up to98%). This research enabled the examination of increased cellulose amounts of up to 6% wt. Bio-based aerogels displayed remarkable physical strength and heat transfer efficiency for textile applications at atmospheric pressure. The Young's modulus of cellulose aerogels can be reached 13.5 MPa, and the Poisson ratio is near to zero [73]. Yangyang exploited discarded cotton textiles to enhance the anti-flaming capabilities of cellulose aerogels by producing magnesium hydroxide nanoparticles in situ in cellulose gel nanostructures. In addition, three-dimensionally nano porous cellulose gels were produced by disintegrating and coagulating cellulose in an aqueous NaOH/urea solution, and these were employed as patterns for the un-clustered production of magnesium hydroxide nanoparticles. According to the findings, produced mixture -matrix aerogels have extremely porous architectures and exceptional thermal isolation characteristics with minimal heat transfer. In addition, effective flame-retardant and mechanical characteristics are obtained [74].

The sol-gel process is linked to the organic polymer type. Because molecular composition of organic polymer variants contains a restricted amount of active (e.g. hydroxyl) groups, a connecting agent is often necessary to achieve a required gel structure [11].

The creation of bio-based aerogels from nanoscale crystalline polysaccharide and dissolvable organic polymer from various materials is shown schematically in Figure 2 simply by recovering them as a coagulant from their liquid solution, followed by lyophilization and the resulting regenerated cellulose alcogel [75].

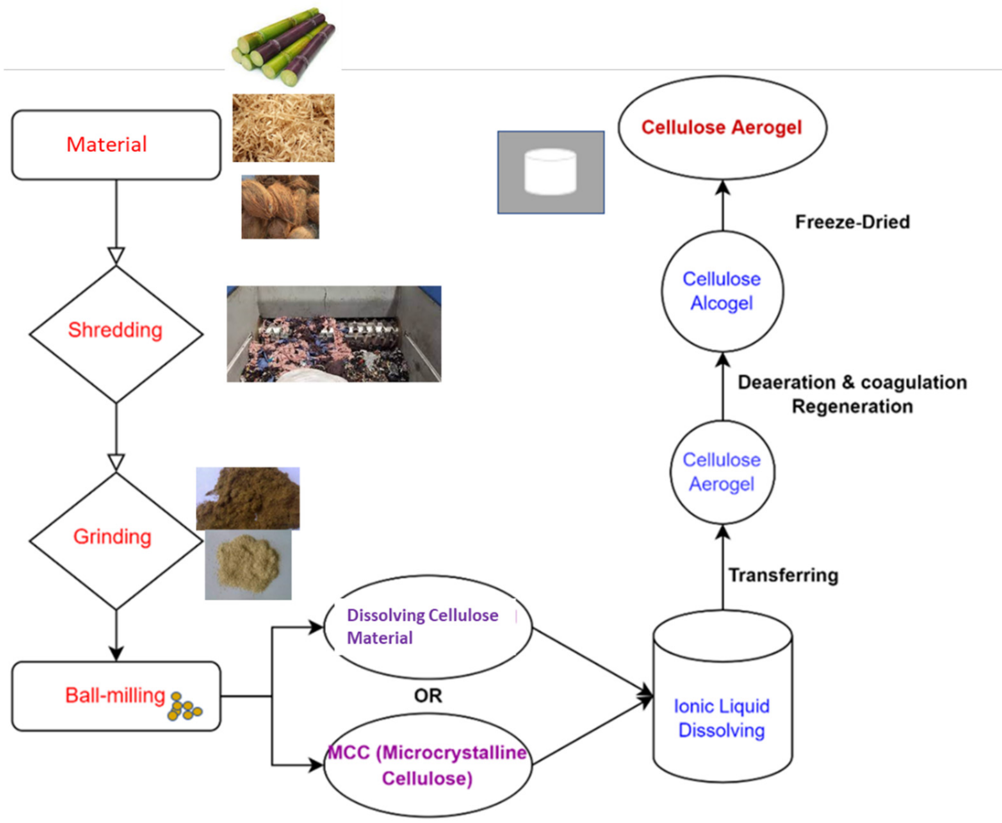

Figure 2. A schematic diagram depicting the cellulose aerogel preparation process [38].

2.2. *Drying Methods of Cellulose-Based Aerogels*

The most crucial phase in the manufacture of aerogels is drying. The drying process influences the shape of cellulose aerogels. Due to the capillary pressure, traditional drying processes can result

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in the collapse of the gel pore structure. Supercritical drying (using alcohol, acetone, or CO_2) and vacuum freeze-drying are extensively used for cellulose aerogel manufacturing procedures [76,77]. The sublimation of a solid, frozen water, from a moist precursor's pores is identified as freeze-drying. As a result of the formation of ice during the process of water freezing, freeze-drying produces a sheet-like cellulose network with large and linked holes having width in numerous micrometers [77] Under supercritical (sc) conditions, the absence of a liquid/gas meniscus results in a complete elimination of surface tension between the liquid and gas phases. ScCO_2 dried aerogels usually have a cauliflower-like cellulose arrangement: an assemblage of tiny shaggy beads.

2.2.1. Drying by Supercritical Carbon Dioxide

Aerogels are formed by drying a wet gel while keeping most of its intrinsic porosity. Silica gels are produced using the sol-gel method, which modifies the molecular structure of the gel [78]. After use and washing, the wet gel's porous silica structure is strengthened, and the pores are only partially filled with the pore liquid, which is often an organic solvent (ethanol). If the wet gel is dried in circumstances where the porous silica structure partially collapses due to capillary forces [3,79], the dry and wrinkled gels are identified as xerogels or cryogels. To maintain the gel's porous structure, the supercritical drying method can be employed, effectively eliminating capillary forces during the drying process [80–82]. The organic solvent is first eliminated from the gel by applying compressed CO_2 at operational settings above the pore liquid and CO_2 mixture critical point. With no liquid-gas interactions and hence no capillary forces, this ensures extraction in a single-phase mixing process. CO_2 can be emitted in a single-phase process during progressive depressurization at operational conditions above its critical temperature after entirely replacing CO_2 for the organic solvent. The remaining material is dried gel, the pores of which are promptly filled with CO_2 after drying. When dried gel is exposed to air, CO_2 is exchanged, and the gel is called an aerogel [78]. CO_2 is a fluid normally employed in the drying of cellulose aerogels because of a reasonable critical point (304 K, 7.4 MPa) and the benefits of low cost and great safety.

Supercritical drying is distinguished by the two-way mass transfer of scCO_2 and gel solvent into and out of the wet gel pores [83]. To begin, the drying is largely caused by a high scCO_2 dissolution in the liquid gel solvent, which results in an expanded liquid and spilling of the extra liquid volume removed from the gel network. Second, the amount of CO_2 increases over time until supercritical conditions are reached for the fluid mixture in the pores, without any intermediary vapor-liquid transitions. Finally, the presence of supercritical fluid mixtures in pores with no liquid phases causes a lack of surface tension, which precludes pore collapse in the gel structure during solvent removal [83].

The water with a high surface tension might destroy a cellulose network's delicate and extremely porous structure, which is generated during the drying process. The reasons behind this phenomenon include variances in the specific energies during the transitions between solid-liquid and liquid-gas phases, along with the generation of inward forces near the solvent menisci along the capillary walls. As a result, it is required to entirely replace the high surface tension water [84]. In an NMMO solvent system, for example, while manufacturing regenerated cellulose aerogels, The cellulose gel requires re-priming with water, followed by either ethanol and acetone exchange or solely acetone exchange [85,86]. In the case of employing an ionic liquid as the solvent system, the cellulose gel necessitates an initial re-priming step with water, followed by subsequent acetone exchanges conducted repeatedly [48]. Ethanol exchange is a popular treatment for natural cellulose aerogels [87,88]. It has been demonstrated that cellulose solvent residue reduces drying efficacy [46]. Furthermore, the surface tension of various liquids, as well as shaking is involved during the re-priming and solvent exchange procedures, may destroy the cellulose gel structure [46,89]. The solvent exchange process is exceedingly slow, requiring an average of 2-3 days. Finally, supercritical drying using scCO_2 can be helpful to reduce damage caused by capillary pressure inside the pores can be advantageous, as it promotes the production of aerogel materials with enhanced uniformity in their 3D network.

However, because a high-pressure tank is required, this method is costly [11]. Nevertheless, the SCD processes offer a notable advantage in that the choice of solvent for the gelation process is highly versatile, allowing for a wide range of options. This approach is applicable across several types of gel materials and is not limited to specific ones. Two fundamental drying strategies exist: high-temperature (HT) drying and low-temperature (LT) drying [90].

After a pre-pressurization stage, the solvent is heated over its critical point in the high temperature (HT) process. This method requires heating the wet samples and solvent in a sealed autoclave to supercritical temperatures. For commonly used organic solvents, which have a critical point above 200°C and a critical pressure ranging from 40 to 80 bar, the desired conditions can be achieved through this method. Subsequently, a slow depressurization is carried out [91–95]. The process and instrumentation involved in the high-temperature (HT) approach are straightforward, as it does not require pumps, and it enables direct surface modification to produce hydrophobic aerogels. However, one disadvantage of utilizing organic solvents is the risk of fire in the case of an unintentional or uncontrolled discharge, and the higher temperature may cause damage to heat-sensitive components. An alternative to the normal HT method is to drop the temperature slightly, maintaining it below the solvent's critical temperature [96,97].

Low temperature drying uses supercritical carbon dioxide (CO_2) since it has a low critical temperature (31°C), is non-flammable, and is ecologically friendly. To eliminate all solvents, wet gel samples are periodically, or continuously flushed, with supercritical CO_2. Heat exchangers and a liquid CO_2 pump are critical equipment components for both periodic and continuous operations. The continuous low-temperature (LT) method may need a greater volume of CO_2; nevertheless, particular drying costs can be greatly lowered via careful optimization and scaling to an industrial level [98,99]. Supercritical conditions offer the possibility of functionalizing the aerogel skeleton [100]. Considering the significance of aerogels in both scientific research and industrial applications, extensive studies have been carried out to examine the influence of drying conditions, diffusion, chemical composition, and temperature profiles on the quality of aerogels [78,80–82,101–104].

2.2.2. Vacuum freezing and drying

Cellulose aerogels can be produced using a straightforward and environmentally friendly method known as vacuum freeze-drying. At a temperature below the freezing point of the liquid medium, which is usually water, the gel is initially frozen in this process. Much of the liquid is then eliminated through sublimation, which is an essential step to avoid structural collapse and reduce shrinkage. Consequently, the pore structure of porous aerogels, including pore morphology and distribution, is influenced by the liquid crystallization process and growth behavior, which are controlled by the cooling rate and temperature. Additionally, various factors such as cellulose content, gel size and shape, and temperature affect the rate of sublimation, which is typically slow [11].

Freeze-dried aerogels that are made of nanocellulose, and its derivatives are commonly encountered, although the self-agglomeration of nanocellulose may reduce their specific surface area. Tert-butyl alcohol, on the other hand, possesses a low interfacial tension and a single hydroxyl group, enabling it to create hydrogen bonds with the surface hydroxyl or carboxyl groups of nanocellulose and its derivatives. Altogether, the presence of multiple butyl groups creates a steric barrier that inhibits the aggregation of nanocellulose. Consequently, when employed in solvent exchange, tert-butyl alcohol has the potential to preserve the gel structure of nanocellulose and its derivatives more effectively than water, thereby preventing the collapse of the cellulose aerogel structure [50,105–107].

By employing liquid nitrogen or liquid propane to enhance thermal conductivity, it is possible to quickly cool the cellulose gel. This process effectively reduces cellulose agglomeration and the formation of ice crystals, while simultaneously increasing the porosity of the resulting aerogel. Zhang et al. examined three different chilling rates in their study: liquid nitrogen (-196°C for 30 minutes), a freezer with an extremely low temperature (-80°C for 12 hours), and a standard refrigerator (-20°C for 24 hours). Their observations revealed that the use of liquid nitrogen facilitated the rapid formation of ice crystals, thereby effectively mitigating cellulose self-agglomeration and leading to the development of a more homogeneous and seamless surface structure [108]. In order to achieve uniformly structured aerogels, the utilization of anti-freezing chemicals [109] and spray freeze-drying techniques [110,111] both depend on accelerating the freezing rate. However, prior to the advancement of the solid-liquid interface, comparable freezing rates and localized temperature gradients are observed, similar to the scenario of freeze-drying small samples in a freezer while simultaneously cooling the larger sample [11]. The specific surface area and pore size distribution of a specific kind of cellulose aerogel are significantly influenced by the drying process employed [44,87]. Because of the creation of ice crystals and the high interfacial tension of water, freeze-drying typically generates fractures in the aerogel material. Another disadvantage of freeze-drying is its lengthy processing time and significant electric energy usage. In contrast, drying with supercritical carbon dioxide (scCO2) offers improved preservation of the cellulose gel structure, resulting in aerogels with minimal shrinkage, smaller pore sizes, and higher specific surface areas [47,88,112,113].

2.2.3. Ambient Drying

Atmospheric pressure drying of (ligno-)cellulose aerogels is still in its infancy. The fundamental issue impeding the development of atmospheric drying for aerogels is significant network shrinkage produced through the liquid meniscus and pressure gradient. Under the same regeneration circumstances, vacuum-dried aerogels show significant shrinkage and collapse as compared to supercritical CO_2-dried aerogels. According to ESEM images, the capillary force during vacuum drying degrades the porous structure. [114].

The structure of cellulose aerogels may be adjusted and controlled using drying processes. For this purpose, four drying procedures are outlined. $ScCO_2$ drying can result in mesopore aerogels with large specific areas and high porosities. Although t-BuOH drying can achieve comparable conclusions, the porous structure created is less homogeneous than that produced by $ScCO_2$ drying [114].

3. Characterization Methods of Cellulose-Based Aerogels

Aerogels have two unique phases: the solid backbone and the pore phase and are characterized by several essential characteristics, including phase proportion, usual extent of each phase, and connectedness. Furthermore, the broad interface separating the two phases' physical and chemical characteristics are crucial aerogel qualities [115]. Cellulose aerogel being a porous solid material represents qualities like regular aerogels. In comparison to the original aerogel, the cellulose's chemical alteration improves the aerogel's mechanical and structural properties. [11]. As a result, mechanical and structural characterization of cellulosic aerogels can be distinguished.

3.1. Characterization of Cellulose Aerogels' Structure

By applying the structural characterization to the cellulose aerogels, it can be answered that rigidness, or flexibility, physical properties of material, containing the component type such as organic or inorganic or how small or large porous it has [115].

The essential structural parameters of a porous material include the overall fraction of the pore and solid phase, the typical size of the backbone and pore phase, the connectivity between two phases, properties of interface between them, and the molecular structure of the backbone phase. These characteristics are evaluated using various characterization methodologies [115].

3.1.1. Microscopic analyses

Microscopy techniques offer valuable visual insights into the structure of aerogels at various length scales, including those as small as a few Angstroms. While light microscopy (LS) has a resolution boundary of around 500 nm, it fails to capture the properties of most aerogels due to their particle sizes typically less than 1 mm. Atomic force microscopy (AFM) theoretically has the potential to be used for aerogels but proves to be challenging in practice due to the irregular and deep surface topology found in fractured aerogels [115].

Consequently, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are the primary microscopic techniques employed in aerogel research. SEM allows for the visualization of the three-dimensional interconnected structure of the aerogel backbone, with a maximum resolution of approximately one nanometer. In contrast, TEM has better resolution capabilities and can examine the substructure of the particles that make up backbone as well as any other phases employed into the aerogel skeleton, like metal particles. The released electrons and X-rays inside the hemisphere of main beam are examined using a reflection setup in SEM mode. TEM, in contrast, investigates the sample's transmission by high-energy primary electrons. All high-resolution electron microscopes operate with a chamber vacuum of less than 10^{-4} mbar to reduce undesired scattering [115].

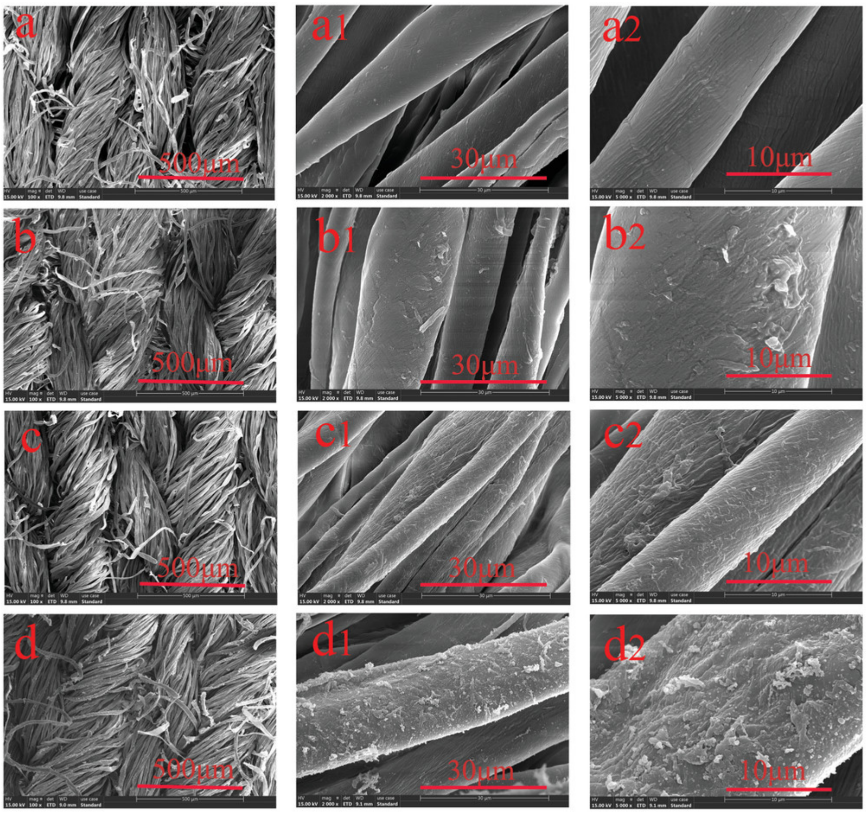

Figure 3. SEM images of various cotton samples: a) untreated cotton, b) cotton loaded with TA, c) cotton loaded with TA/B, and d) cotton coated with TA/B@PDA. The images were taken at different magnifications, specifically a) 100, a1) 2000, and a3) 5000 respectively (The permission is approved by the John Wiley and Sons, Macromolecular Materials & Engineering under License number:5570770529195)[116].

3.1.2. Scattering Techniques

Scattering techniques provide a quantitative and noninvasive means of analyzing the structure, without causing damage to the sample [117]. Moreover, it proves to be a valuable technique for assessing the transparency of aerogels, investigating their pore size, structure, and mechanical strain, as well as exploring the sol-gel evolution modes that shape their microstructure. The examination of wavelength-dependent scattering enabled by ultraviolet-visible transmission spectroscopy permits comparisons across aerogels of various sources and thicknesses, as well as analyzing the influence of residual pollutants. Infrared reflectance tests provide the actual real and hypothetical refractive indices of porous aerogel materials, allowing researchers to better understand material characteristics and radiant heat transport. Scattering measurements at a given angle are useful for quality control, locating scattering sources, and evaluating inhomogeneities [118].

The dispersed intensity of aerogels often displays radial symmetry, suggesting an isotropic material. As a result, in a certain experimental arrangement, the angle θ becomes the only variable. However, if the aerogel sample has anisotropic qualities (due to drying conditions, for example), anisotropy will be seen in the scattering pattern, necessitating the use of directional information to explain the orientation of the anisotropy. In general, scattering is classified into two types: small-angle scattering (SAS) and wide-angle scattering (WAS). When employing a reflection setup, X-ray scattering is often referred to as X-ray diffraction (XRD). SAS employs scattering angles (2θ) ranging from 0.001° to nearly 10°, with wavelengths between a few angstroms and nanometers. The equivalent q-range is 5 to 10^{-4} nm^{-1}, with the lowest q limit observed in ultra-low-angle scattering (USAXS and USANS) [118–120].

3.1.3. Thermoporometry

Cellulosic aerogels' outstanding capabilities are greatly influenced by their hierarchical pore structure. However, utilizing a single approach for examining the complete pore size distribution (PSD) might be difficult. Mercury infiltration and nitrogen sorption are the most often used techniques for evaluating the PSD of aerogels. Nitrogen sorption, on the other hand, only protects pores up to around 170 nm, which is insufficient for the vast PSDs observed in cellulosic aerogels that span the single-digit nanometer to low micrometer range. On the other hand, mercury intrusion is not suitable for characterizing fragile and ultra-lightweight cellulose aerogels as it tends to destroy the delicate nanofibrillar network of these soft materials. As a result, estimating the bulk volume decrease (sample compression) instead of monitoring mercury penetration into the pore network (non-intrusive mercury porosimetry) is the only way to calculate the PSD. Nitrogen sorption tests can be used to characterize the PSD under the greatest pore size impacted by densification after applying a given mercury pressure [121,122]. Thermoporometry is a technique that examines the freezing behavior of a liquid within pores. By analyzing the resulting data, it is possible to extract details regarding the arrangement of pore sizes and the morphology of the pores. This technique is particularly sensitive to pores within the range of 2 to 30 nm. Unlike traditional porosimetry methods that involve mercury or gas adsorption and require dry samples, thermoporometry can be applied to gels, eliminating the need for sample drying [115]. It is a proposed method that offers a different approach to characterizing the structure of porous aerogels. It depends on the utilization of the Gibbs-Thomson equation, which was introduced by Gibbs in 1928 and Thomson in 1872. This equation measures the experimental shift in an interstitial liquid's melting point caused by confinement within microscopic pores [123,124].

3.1.4. Gas Sorption

The processed macro-meso porous aerogels displayed the following characteristics concerning the flow of air passing through them in the atmosphere:

- Pressure-time curves were consistent with that of theoretical model created for pure Darcy flow, which was employed to fitting the data and get the permeability constant.
- Permeability remained consistent regardless of the difference in pressure.
- Choice of surfactant had an impact on the permeability.

Above mentioned findings display that Darcy's law may be used without considering other effects such as Knudsen effects, Klinkenberg effect or slip flow at cell walls [125–127]. The commonly employed method for characterizing aerogels is nitrogen (N_2) sorption at a temperature of 77.3 K. An extensive variety of relative gas pressures, ranging from 0 to 1, may be obtained by adjusting the gas pressure from vacuum to 0.1 MPa (1 bar). This method often yields useful data on particular surface areas as low as 0.01 m^2/g and pore diameters ranging from 0.3 to 100 nm [115].

3.1.5. Hg Porosimetry

Porosimetry using mercury (Hg) is a technique capable of investigating a broad range of accessible pore sizes, spanning approximately six orders of magnitude. This range extends from

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about 400 mm down to a few Angstroms [128]. In theory, Hg porosimetry may be used to examine the distributions of pore sizes spanning between microns and nanometers. On the other hand, when working with flexible materials like aerogels, considerable deformation effects may arise unless the sample stiffness aligns well with the pore size [129,130]. One drawback of mercury porosimetry is that it frequently leads to irreversible deformation of the sample, and a portion of infiltrated mercury becomes captured. Consequently, the sample is rendered unusable for subsequent investigations and must be disposed of as toxic waste [115]. Consequently, a comparative study on pore size measurement of cellulose-based aerogels revealed no observable damage or degradation in the aerogel produced from softwood pulp cellulose nanofibrils [131].

3.2. Mechanical Characterization of Cellulose Aerogels

Thorough mechanical characterizations are essential in assessing the suitability of aerogels for load-bearing applications, as it is crucial to understand their mechanical response under operational conditions. Mechanical characterization studies must be meticulously constructed to reproduce or nearly match the loading conditions met in the desired service environment. Since aerogels can be subjected to diverse service conditions, it is necessary to perform multiple types of loading tests to fully evaluate their mechanical response. These tests generally encompass compression, bending, tension, multiaxial stress states, and torsion under various loading circumstances such as dynamic, fatigue and quasi-static. Following approaches are routinely used in this regard [132].

3.2.1. Differential scanning calorimetry (DSC), Dynamic mechanical analysis (DMA)

DSC is a technique for measuring thermal effects during heating of materials (e.g. glass transition temperature). The relationship between the rigidity of aerogels and temperature is determined using DMA. DMA uses alternating loads at a constant frequency, generally 1 Hz, or changing frequencies ranging from 0.1 to 20 Hz. The tests are carried out at temperatures ranging from -100°C to 300°C, with the setup for a three-point bending test. Dynamic Mechanical Analysis (DMA) findings display the loss and storage moduli as they vary with temperature. The peaks observed in the loss tangent as a function of temperature can serve as indicators for identifying the glass transition temperatures [133].

3.2.2. Tension, Compression

Aerogels are not well-suited for conducting impact tests, hence the approach used to quantify strength may be problematic from an engineering standpoint. Bending or tensile testing are preferable. The test should be tailored to the material [134]. Furthermore, determination of the compressive stiffness of aerogel composites by microstructure's effectiveness in transmitting applied stress and is regulated by the solid content. The density of the composite increases with higher solid content, resulting in higher stiffness [135]. Aerogel elastic characteristics are commonly assessed using sound velocity measurements [136–138] or static methods [139–142]. It is well accepted that silica and cellulosic aerogels are "fragile materials" because of the low mechanical properties caused by the network's limited connectivity and large porosity. The existence of defects, which act as stress concentrators, has a significant impact on rupture resistance [141–146]. Dog-bone-shaped specimens (as per ASTM D638) are often used for tension testing. Similarly, compression testing may be performed on cylindrical specimens (following ASTM D695). It is critical to prepare the specimen surfaces so that they are smooth and free of flaws. The end surfaces of cylindrical compression specimens should be parallel to one other. To avoid mistakes, compression tests should be done on compression plates with extremely parallel surfaces, especially for brittle aerogels. To achieve adequate alignment, a frequently employed method is the utilization of a self-aligned compression fixture. It is necessary to calculate the entire stress-strain relationship, including the point of ultimate failure, at exceedingly low strain rates to evaluate the energy absorption capacity and strength, as represented by area under stress-strain curve. To assess the dimensional stability of samples, conducting constant deflection-compression set tests (referred to as Test-D in ASTM D3574 Standard) is feasible. This test involves measuring the changes in thickness of cylindrical specimens exposed to a range of compression strains at an elevated temperature for a specified duration [115].

3.2.3. Sound Absorption and spreading

Noise pollution, considered the second most significant environmental hazard impacting human well-being and living conditions after air pollution, necessitates the utilization of porous sound absorption materials. These materials offer remarkable attributes like a broad range of sound absorption frequencies, cost-effectiveness, and ease of shaping. With its voluminous porous structure characterized by low density, high porosity, and extensive surface area, aerogel allows acoustic waves to enter deeply into its structure, enabling diverse interactions to occur [147–150].

Acoustic performance evaluations are often performed using an impedance tube (SW422, SW477, BSWA Technology Co. Ltd., China) in accordance with ASTM E1050-10 [151]. Sound absorption coefficient is expressed as $\alpha = (E_i - E_r)/ E_i$, where E_i denotes incoming acoustic energy and E_r denotes reflected acoustic energy. For sound absorption testing, samples having diameters (100 mm and 30 mm) are used, with frequency in the ranges of 200-1000 Hz and 1000-6300 Hz, respectively [152–154]. A demonstration was also carried out to evaluate the sound insulation capabilities of silica-cellulose aerogels and their cellulose matrices. The experiment used a sound signal generator (Blesi Guardian angel anti-rape alert, 90 dB) to generate known sound signals with a specified sound strength. The coefficient of absorption of sound for the silica-cellulose aerogels and the cellulose matrices was measured using a sound meter (Amprobe SM-10, USA). The sound generator was positioned both inside and outside of an insulating box. The insulating box was built by taping a certain type of aerogel to both sides of the container, resulting in a sealed system. In all situations, the incident sound signal was measured at an equidistant position from the sound generator. The absorbed sound intensity was determined by subtracting the known sound intensity from the incident sound intensity. The coefficient for sound absorption was computed as the ratio of absorbed sound intensity to known sound intensity [155].

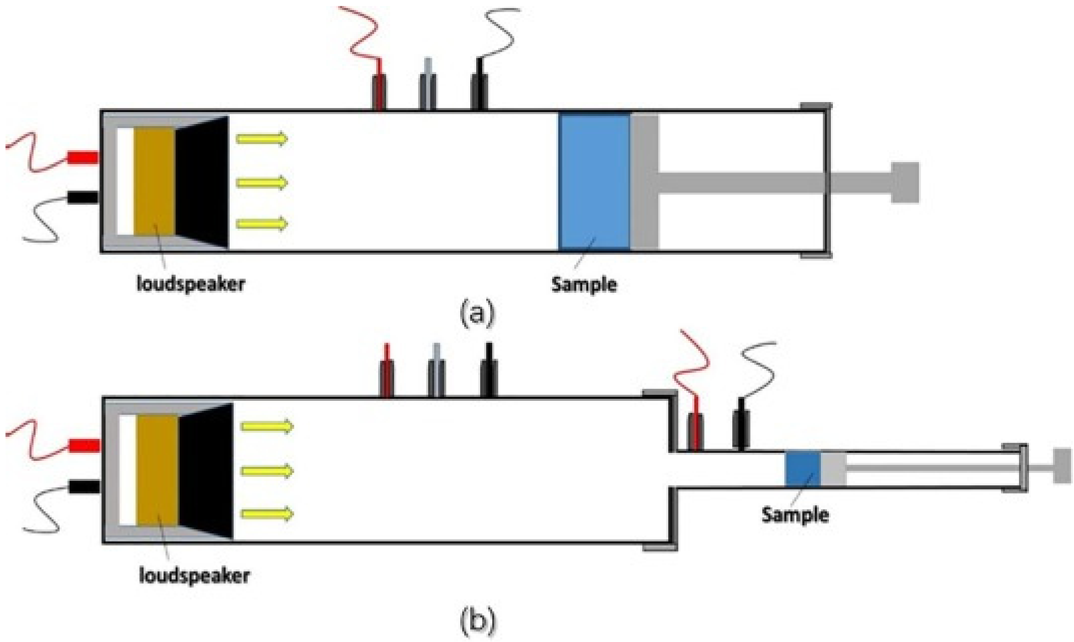

Figure 4. The schematic picture depicts the principle of measurement of sound absorption coefficient with an impedance tube at several frequency ranges. The frequencies in Figure (a) vary from 50 to 1000 Hz, whereas the frequencies in Figure (b) extend from 500 to 6300 Hz [154] (Permission is granted by Creative Commons CC-BY license).

3.3. Thermal Characterization

Cellulose aerogel has great thermal insulation capability because of its exceptionally low thermal conductivity, making it ideal for insulation material applications. As the demand for thermal insulation materials continues to grow and evolve, traditional options like polyurethane and polystyrene foam face limitations due to their non-renewable properties. As a result, cellulose aerogel emerges as a groundbreaking and ecologically friendly substitute to old thermal insulation materials, meeting the demand for long-term solutions in industry and society at large [156]. Thermal conductivity is widely recognized as the most crucial thermal property, with specific heat being of secondary importance [157,158].

The most frequent methods for investigating thermal characteristics of cellulosic aerogels are thermogravimetric analysis (TGA), transient plane source (TPS) method-thermal conductivity, and derivative thermogravimetric analysis (DTG) transient hot-wire method [156,159–161].

Due to the chemical composition of cellulose aerogel, which consists of elements such as C, H, and O, it exhibits an elevated level of flammability. This characteristic imposes significant limitations on its use in various critical areas that necessitate flame retardant materials. As a result, it becomes imperative to pursue flame retardant modifications for cellulose aerogels, aiming to enhance their fire resistance and broaden their applications [156]. Vertical burning tests (UL-94), limiting oxygen index (LOI) tests, and cone calorimetry tests are routinely employed to assess flame retardancy of cellulose nanofibril (CNF) and its composite aerogels [159–161].

4. Properties of Cellulose aerogels

Because cellulose-based aerogels have comparable porosity (84-99.9%), density (0.0005-0.35 gcm^{-3}), and specific surface area (10-975 m^2/g) to conventional silica aerogels and synthetic-based polymer aerogels, they have superior compressive strength (ranging from 5.2 kPa to 16.67 MPa) and enhanced biodegradability. Hence, a new environmentally friendly and multifunctional material called cellulose-based aerogel has been developed, showing immense potential for diverse applications. These applications encompass areas such as adsorption and separation of oil/water, thermal insulation, biomedical materials, carriers for metal nanoparticles/metal oxides, carbon aerogel production, and various other fields. The versatility of this material opens numerous possibilities for its utilization.

Table 2. Summarizing Characteristics of Cellulose-Based Aerogels.

Number	Aerogel Type	Main Properties	Application	Ref.
1	MXene composite aerogel (M−Aerogel)	Single-layered structure Conductive active material Three-dimensional porous structure Remarkable flexibility Superior compressive strength	Flexible piezoresistive sensors	[162]
2	Holocellulose nanofibrils (HCNFs) Aerogel from Bamboo pulp and birch wood blocks	Fiber form aerogel properties Exceptional self-cleaning capabilities Outstanding thermal insulation performance Washability Impressive tensile strength Biodegradability Superb mechanical properties Potential for weaving into multifunctional textiles suitable for demanding environments	Thermal management EMI shielding performance	[163]
3	Cellulose nanofibrils (CNFs) from rice straw cellulose	Amphiphilic - Hydrophobic and oleophilic nature High porosity Extremely lightweight	Selective oil removal and recovery	[164]

4	Barley-straw cellulose aerogels	Highly porous and lightweight aerogel, large surface area, high concentration of cellulose content	Oil-spillage clean-up	[165]
5	Bio-inspired tubular cellulose aerogel from kapok fibers	Exceptionally high compressive strength of 32 MPa, self-extinguishing capabilities and exhibits excellent flame retardancy, cost-effective solution	Exterior wall insulation and vehicle interior	[166]
6	Bio-based aerogel (polysaccharide cryogel) from sodium alginate and chitosan	Eco-friendly and sustainable, excellent thermal insulation, bio-based flame-retardant, ultralight porous structure, practical mechanical properties, great flexibility, facilitating continuous flexing and rotating without fragmentation	Anti-flame apparel	[167]
7	Agar aerogels	substantial surface area per unit weight, significant acceleration in wound healing in vivo, the ability to be used for skin healing, in addition to its biocompatibility, renewability, and sustainability properties.	Wound dressing	[168]
8	Novel alginate-chitosan aerogel fibers	Highly porous structure reminiscent of cotton, non-cytotoxic, making it biocompatible, strong antibacterial activity, speeding wound closure in vitro design imitating injured life-unit monolayer healing	Wound healing applications	[169]
9	Aerogels made of tempo-oxidized cellulose nanofibers and sodium algin/chitosan	Serving as an interactive extracellular fabric, derived from biological sources and the capacity to degrade naturally, highly porous structure, creating an ideal microenvironment for various applications	Wound dressing, and injury tissue maturation	[170]
10	Alg-CaCO3 composite aerogels from Sodium alginate	Cost-effective, environmentally friendly, ultralight, and fireproof, characterized by high permeability and excellent structural properties, reduced heat transfer rate, and excellent hydrophobic characteristics	Green fireproof building insulation materials	[171]
11	Kapok aerogel	Lightweight, providing insulation and robustness, reusable and decomposable, and exceptional fire protection, high filling capacity, superior compressive resilience, and remarkable heat insulating abilities	Application in emerging fields	[172]
12	Chitosan aerogel	Elevated permeability and extensive superficial expanse, enabling rapid local administration of antibiotics, Infections are efficiently prevented early after wound debridement while cell viability is maintained, absorbing substantial amounts of aqueous fluids	The management of chronic wounds	[173]
13	A novel intelligent bio-aerogel using cellulose/Salep/ anthocyanins	Maintaining structural integrity and allows for precise control over the porous structure, usage as intelligent aerogels in meat products, providing unique properties and benefits, serving as suitable matrices for pH-sensitive dyes, enabling their effective utilization	Application in beef packaging	[174]

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14	Essential oil-loaded starch/cellulose aerogel	Aerogels with antimicrobial properties made from affordable materials	Application in cheese packaging	[175]
15	Hybrid bio-aerogel with green pectin (PML) and corn stalk nanofiber (CNF)	High porosity and low density, providing excellent elasticity. It exhibits a remarkable oil sorption capacity ranging from 82 to 161 g/g.	Applications to oil pollution treatment	[176]
16	Nanofibrillated cellulose/chitosan aerogel	Lightweight and flexible, having a well-defined three-dimensional linked cellular network structure, exhibiting outstanding mechanical properties both in air and underwater, high maximum adsorption capacity, rapid adsorption rate, and offers a low-cost solution with a long lifespan	Heavy metal pollution in agriculture	[177]
17	Aerogels comprising graphene oxide (EGO) and TEMPO-oxidized cellulose nanofibril (TOCNF)	Great promise as an environmentally friendly conductive ink suitable for printing 3D objects using the direct ink writing (DIW) method, the inks exhibit a high yield stress, improved electrical conductivity, uniform distribution of micro- and nano-scale fibrils, and efficient penetration, representing a sustainable approach to produce conductive carbon-based ink	Advanced applications (EMI shields)	[178]
18	Silica- cellulose nanoclaws hybrid aerogels	A biomimetic hybrid technique that is eco-friendly, cost-effective, outstanding formability and mechanical stability, as well as substantial surface area per unit weight, strength, Lightweight, and minimal heat transfer	Structures, industrial production, air transport, and cosmic space	[179]

5. Multifunctional Application of Cellulose-Based Aerogels on Textile Structures

Due to the robust chemical reactivity of cellulose, the wide range of diverse derivatives with various functions, the adaptable construction process, and the multiple methods of modification, bio-based aerogels exhibit multifunctionality. There exist three primary methods for modifying cellulose aerogels [11]:,

- Other components can be added to the cellulose solution/suspension [11]. For example, the reaction of CNF with N-methylol-dimethyphospylpropionamide (MDPA) and further cross-linking by 1,2,3,4-butane tracarboxylic acid (BTCA) yields a flame retardant with good flexibility and self-extinguishment [180].
- Coating or adding additional substances to the aerogel structure [11], such as the polyacrylonitrile-silica aerogel coating over viscose nonwoven fabric for protection and comfort [181]. Another area of study is the application of molecular layer by layer (m-LBL) technology. This technique enables the deposition of ultra-thin layers onto a surface through sequential covalent processes. As a consequence, a precise molecular-scale coating is generated, mostly by surface oligomerization, which is not possible with bulk synthesis techniques [182–184].
- Surface modification of cellulose aerogels may be attained using a number of methods [11], including dip-coating with PDMS (Poly(dimethyl siloxane)) [185].

Cellulose aerogels are lightweight 3D porous materials. They are currently employed mostly in insulation, flame retardant [74,186,187], and biological applications [4,11]. Additionally, they find applications in carbon aerogel production and the transportation of metal nanoparticles and metal oxides [11].

5.1. Thermal Insulation Materials

Materials are classified according to their thermal conductivity as thermal conductors (λ_{eff} ≥0.1W/(mK), insulators (0.1 W/(mK)>λ_{eff} >0.025 W/(mK), and super-insulators (λ_{eff} ≤0.025 W(mK). It is known that thermal conductivity of dry air is around 0.025 W/(mK), generally slightly dependent on temperature and moisture content.

Due to their thermal conductivity levels spanning from tens to hundreds of W/(mK), metals are good thermal conductors. Expanded polystyrene, extruded polystyrene, glass wool, mineral wool, and wood exhibit thermal conductivities within the range of 0.1 to 0.026 W/(mK), making them effective insulators against heat transfer [158,188]. Silica aerogels, vacuum insulation panels and vacuum glasses are regarded as super insulating materials as their thermal conductivity is below 0.025 W/(mK) [189].

The thermal conduction of aerogel can be classified as solid-state, gas-phase, open-pore, or radiation thermal conduction. Once the pore size of a porous material approaches the average free path of the gas (which is approximately 70 nm when vented), the thermal conductivity of the substance decreases. This is attributed to the fact that the pores impede gas flow and restrict convection, thereby hindering heat transfer. The thermal conductivity of mesoporous cellulose aerogels primarily depends on two factors: solid-state thermal conduction and gas-phase thermal conduction. These factors, in turn, are closely associated with the aerogel density (determined by the initial cellulose concentration), the distribution of pore sizes, and the surface structures of the aerogel material [11].

Regenerated cellulose aerogels possess a porous structure with a relatively higher fraction of large pores compared to other cellulose aerogels. This increased presence of large holes within the aerogel structure enhances heat conductivity as it facilitates improved gas transport [11].

Antlauf et al. conducted a study in which cellulose fibers (CFs) and cellulose nanofibers (CNFs) were produced from commercially available birch pulp. The production process involved varying pressure and temperature parameters as experimental variables. For temperatures ranging from 80 to 380 °C, their results exhibited very little fluctuation in thermal conductivity with density (ϱ_{sample} = 1340-1560 kg/m^{-3}). Furthermore, temperature dependency is independent of fiber size, density, and porosity.

Figure 5 depicts their studies on thermal conductivity [190]. Thai et all studied the oil repellency and insulation properties with the sugarcane fiber by sol-gel synthesis and using freeze-drying. According to their findings, increasing sugarcane fiber content has a substantial influence on thermal conductivity ranging between 0.031 and 0.042 W/(mK) [191].

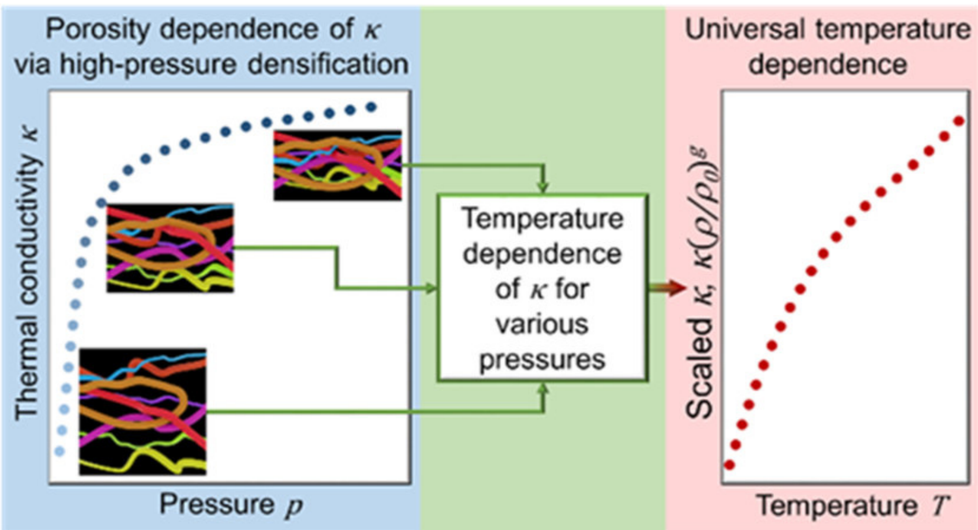

Figure 5. Thermal conductivity against pressure (Note: It has been approved by the ACS publisher, and any further permissions connected to the content excerpted should be given to the ACS) https://pubs.acs.org/doi/10.1021/acs.biomac.1c00643) [190].

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Table 3. Thermal insulation properties and application of cellulose-based aerogels.

No.	Material	Drying Method	Thermal Conductivity	Pore Size	Density	Application	Reference
1	Raw pineapple-leave fibers (PALF)	Freeze-drying	0.030-0.034 W/mK	1.38nm-2.21 nm	0.04 g/cm³	Heat and sound app.	[72]
2	Aerogels composed of bidirectional anisotropic polyimide/bacterial cellulose (b-PI/BC)	Freeze-drying	23 mW/mK- 44 mW/mK (bidirectional PI/BC aerogels) 37 mW/mK -66 mK/mK (unidirectional PI/BC aerogels)	10–20 μm	46 mg/cm³	Practical and complex thermal insulation applications in buildings and aerospace	[192]
3	Aerogels made of fibrous silica and bacterial cellulose (BC)	Ambient pressure drying	-	13.7-15.5 nm	0.164 g/cm³	Wearable substances	[193]
4	Holocellulose nanofibrils/ cellulose aerogel fiber (HCAFs)	$ScCO_2$ drying	0.048 W/mK	265.4 ± 34.5 nm	0.22 g/cm³	Wearable substances	[163]
5	Multiscale nanocelluloses (NCs)	Freeze-drying	25.4 mW/mK	32 - 48 nm	7.2 kg/m³	Thermal insulation app.	[194]
6	Textile waste fibers (TWF) aerogel	Freeze-drying	0.049 - 0.061 W/mK	-	0.040-0.096 g/cm³	Building insulation and oil spill cleanup.	[195]
7	Nanofibrous Kevlar Aerogel Threads	ScCO2 drying and Freeze-drying	0.036 W/mK	11-12.8 nm	13 g/cm³	Thermal insulation and thermal management.	[196]
8	Hydrophilic recycled cellulose aerogels	Freeze-drying	0.029 -0.032 W/mK	40-200 μm	0.040 g/cm³	Sorption of water/oil, resistance of water, and thermal insulation	[197]
9	Silk fibroin aerogel	Freeze-drying	0.031 W/(mK)	19.71 ± 8.53	0.21 g/cm³	High performance thermal insulation	[198]
10	Aerogels made of nanofibrillated cellulose	Spray lyophilization	0.018 W/(mK)	10 to 100 nm	0.012–0.033 g/cm³	Thermal super insulating material	[110]

5.2. Flame Retardancy

Aerogels with a lightweight composition derived from bio-based materials represent the interest of the academia because of their exclusive properties, some examples include being environmentally conscious, sustainability, and amazing thermal insulation effectiveness [199,200]. A fire-resistant clothing used for firefighting is a form of specific thermal-protection clothing used by firemen during firefighting operations [201,202]. As a result, advanced flame-resistant and thermally insulating materials with exceptional performance are vital in thermal protective garments to safeguard the safety of firefighters. Para-aramid polymer is now used mostly in thermal protective gear as a material that provides flame retardancy due to thermal insulation of porous fabrics created from

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them [196,203]. Using the wet-spinning procedure, Liu et al [196] revealed that the lab-scale nanofibril Kevlar aerogel exhibited a strong exhibiting a flame-suppressing property characterized by a comparatively slow combustion rate (0.013 cm/s) and the ability to extinguish itself.

It is vital to identify environmentally acceptable thermal insulation materials designed for firefighting applications apparel [167]. Researchers have lately expressed interest in flame-retardant aerogels made from low-cost biomaterials because of their sustainable nature, eco-friendliness, affordability, and lightweight properties, and strong thermal insulation properties [204–206]. Due to high porosity, low thermal conductivity, lightweight structure, and excellent thermal insulation properties, aerogels find extensive utilization in various applications such as fire resistance and thermal insulation [207,208]. Polymers derived from natural polysaccharides is a common renewable biomass resource that is more biodegradable and environmentally friendly compared to fossil-based products [209]. In consequence, several efforts have been undertaken to create aerogels based on polysaccharides that exhibit remarkable low density, porosity, non-toxicity, biodegradability and bio-sustainability [167]. Among the notable examples are magnesium hydroxide nanoparticles (MH NPs) in waste cotton fabrics-cellulose gel nanostructure [74] by freeze drying method demonstrated that the addition of magnesium hydroxide to the gel structure effectively enhanced the flame retardant properties of the aerogel in foam form. Another example is shown in Figure 6 [210].

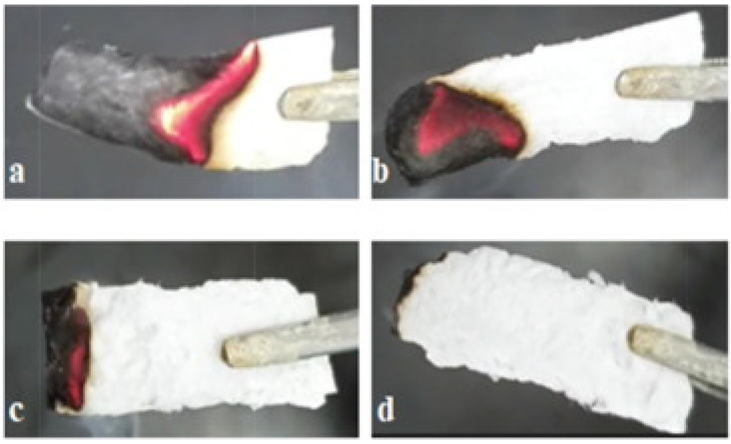

Figure 6. Images of Paper cellulose aerogel after a 10-second burn, the following samples were observed: (a) pure cellulose aerogel, (b) cellulose aerogel with 1% $NaHCO_3$, (c) cellulose aerogel with 2% $NaHCO_3$, and (d) cellulose aerogel with 3% $NaHCO_3$ [210].

5.3. Medical Applications

Because of the most common polymer on the planet, cellulose is mostly obtained from plant and microbiological sources [211]. Nevertheless, due to its unique properties, such as decomposability, compatibility with living systems, and low cytotoxicity, it is one of the most commonly used polymers in manufacturing aerogels [212]. Bio-based aerogels are widely employed in medical treatments such as biological detection, drug release systems, regenerative scaffolds, and anti-infective wound wrap materials [213]. Several publications have previously been published on the sequential evolution of aerogel formation and the therapeutic uses of nanofibrillated cellulose aerogels [17]. Nevertheless, little research has been conducted on utilization of bio-based aerogels for bactericidal administration and wound treatment in textile applications. As a result, cellulose aerogels employed in clinical applications will be described with a textile design. Medicinal applications of bio-based aerogel are listed in Table 4.

Table 4. Medical application of cellulose-based aerogels and their applied method.

Material	Drying Method	Applied Methods for Properties	Type of obtained aerogel	Application	Reference
- Komagataeibacter sucrofermentans H-110 - Hestrin and Schramm - Sodium Fusidate - NaBr - TEMPO	Freeze-drying	SEM, Shrinkage of aerogels, Porosity of aerogels, Thermal conductivity, TGA, FTIR, Antibacterial activity, AFM, Cytotoxicity Tests	Gel film (colorless, transparent)	Wound dressing	[4]
- Populus ussuriensis wood powder - Collagen - Sodium chlorite (analytical reagent) - Acetic acid - Potassium hydroxide (KOH) - Sodium hydroxide (NaOH) - Hydroxylamine hydrochloride (NH2OHHCl)	Freeze-drying	XRD, FTIR Spectra, liquid substitution method, MTT assay	Powdered dried and ultra-thin pellet	Wound bandage & Biological tissue platform	[214]
- MBGs (SiO2-CaO-P2O5-CuO) - Tetraethyl orthosilicate (TEOS), - Triethyl phosphate (TEP), - Calcium nitrate tetrahydrate - Copper (II) nitrate hemi(pentahydrate)	Ambient drying leading to self-assembly (EISA)	TEM, SEM, XRD, SXAS and N_2 physisorption, stimulated body fluids in vitro (SBF), PCR analysis, gram-negative bacteria, Escherichia.coli (for antibacterial properties)	Fine powder (combined with membrane structure later obtained composite aerogel)	Chronic wound healing dressing	[215]
- Cotton nanocellulosic crystal (CNC) - Sodium chloride (NaCl) - Potassium chloride (KCl) - Monosodium phosphate (NaH2PO4) - Hydrochloric acid (HCl) - trifluoroethanol (TFE) - Sodium hydroxide (NaOH) were	$ScCO_2$ drying	Zeta potential for surface charge, Circular dichroism (CD), X-ray Diffraction (XRD), Rietveld method to determine a crystallite size	-	Wound bandage and Bactericidal activity	[216]
- Doxycycline hyclate - Alginate - Amidated pectin - Carbondioxide (purity 99%) - Doxycycline	$ScCO_2$ drying	SEM, Sphericity coefficient (SC), UV–vis spectroscopy, Encapsulation efficiency (EE), DSC, FTIR spectrophotometer, simulated wound fluid (SWF) contact	Core-shell droplets gel (beads)	Wound healing process	[217]

6. Companies of Producing Cellulose Aerogels

Natural fiber aerogel with three-dimensional (3D) framework, lightweight, large surface to volume ratio, increased pore volume, and compatibility with living systems is generally used in ecological sediment accumulation [16,218], medical biology [219] and temperature isolation [220,221]. These characteristics make cellulose aerogel appealing for a variety of applications in a variety of sectors. Here below in Table 5 listed the current commercial cellulose aerogel providers.

Table 5. A list of current commercial cellulose aerogel providers [222].

Nation	Supplier	Chemical composition of the aerogel	Trade Name	Configuration of Aerogel	Reference
Spain	Technalia	Cellulose aerogels from wooden pulp	Inacell	Cellulosic sponge	[223]

Germany	Aerogel-it	Biomass and waste materials derived from agriculture, forestry, and marine ecosystems that are not intended for human consumption	Lignin Aerogel	Boards	[224]
Estonia	Fibenol	lignin, wood sugars, and specialty cellulose from wood residues.	Lignova	Fine and coarse ground	[225]
Switzerland	Empa	TEMPO-oxidized nanofibrillated cellulose (NFC), chitosan	-	Monolith	[226,227]
Singapore	Jios Aerogel	Ultra-light silica material into a fibrous material	Armacell/Armagel	Blankets	[228]
France	Enersens Absolute Insulation	Silica aerogel into nonwoven fibers	Skogar	Composite blankets	[229]
USA	Cabot corporation	Aerogel granules embedded in non-woven fibers	Thermal Wrap	Blankets	[230]
USA	Aspen Aerogel	Mainly silica, but also combining with reinforcing fiber	Spaceloft C	Blankets	[231]

7. Global Market Study Focused on Cellulose-based Aerogel and Their Future Aspects

The aerogel market is witnessing significant growth driven by several factors, including the growing oil and gas industry requirement and unique qualities related to aerogel, such as exceptional heat resistance, recyclable use, and recoverability. In a recent report titled "Aerogel Market by Form (Blanket, Particle, Panel, and Monolith), Type (Silica, Polymers, Carbon, and Others), End-Use Industry (Building and Construction, Oil and Gas, Automotive, Aerospace, Performance Coatings, and Others): Global Opportunity Analysis and Industry Forecast, 2022-2032," published by Allied Market Research, it was revealed that the global aerogel market reached a value of $1.3 billion in 2022. The commercial industry is projected to rise at a CAGR of 19.4% from 2023 to 2032, reaching a value of $7.5 billion. This growth is a result of the increasing applications of aerogel across various industries, including infrastructure development, petroleum and natural gas, vehicle manufacturing, aeronautics, efficiency-enhancing coatings, and others. The report highlights the diverse forms of aerogel, such as blanket, particle, panel, and monolith, as well as several types including silica, polymers, carbon, and others. The forecast indicates a promising future for the aerogel market, driven by its wide range of applications and the growing demand for innovative and sustainable materials [232]. There is a significant current focus on the utilization of both synthetic polymers and biopolymers in the production of aerogels. Natural polymers derived from diverse reservoirs, including polysaccharides including sodium alginate, plant fiber, pectic substances, poly-(1,4)-2-amino-2-deoxy-β-D-glucan, poly-(1,4)-N-acetyl-D-glucosamine, as well as lignocellulosic biomass, amino compounds, and other materials, have been employed as reactant for aerogel synthesis. For instance, the "Aerowood" project in the European Union aims to explore various fractions derived from lignocellulose, such as unlike C5 and C6 sugars, for the purpose of aerogel manufacturing[233]. The obtained aerogels demonstrate a combination of the specific functionalities inherent to the utilized biopolymers and the characteristic traits of aerogels, including an open porous structure

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having a large surface density and porosity. This synergistic blend of features presents a significant capacity for a broad range of utilization. It is important to highlight that the characteristics of the biopolymers, such as molar mass, components, and branching level, have a notable impact on both the overall properties of aerogels and the molecular-level structure of their porous network [234].

Therefore, the primary focus of analysis in natural polymer aerogels revolves around defining numerical relationships between the characteristics of aerogels and the chemical nature of raw materials, together with exploring various material mixes. Additionally, the ongoing search for alternative primary resources that offer improved eco-friendly nature and cost-effective is essential, particularly for applications with high demand [234]. Figure 7 shows where the cellulose-based aerogel takes place in the pie chart and what is the growth in general aerogel usage.

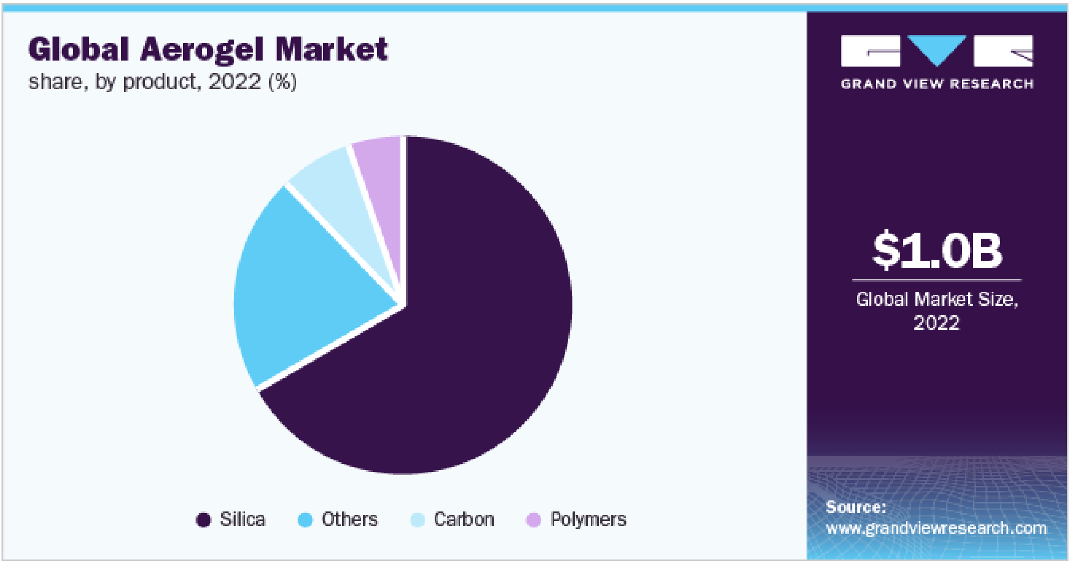

Figure 7. Global Aerogel Market size included cellulose based aerogel in the 'others' and 'polymers'[235] (Note: It has been approved by the Grand View Search, and any further permissions connected to the content excerpted should be given to the Grand View Search) (https://www.grandviewresearch.com/industry-analysis/aerogel-market).

Cellulose aerogels hold immense promise for a range of prospective applications and advancements. The following are a few examples of prospects and potential breakthroughs linked to cellulose aerogels:

Eco-friendly insulation: A building's thermal insulation is critical for reducing energy use and retaining an ideal indoor environment. Therefore, enhancing the thermal insulation properties of buildings is crucial, particularly by reducing losses of energy during heating and cooling applications, thus enabling energy savings. It is widely recognized that high-quality thermal insulation materials depend on various significant factors, including a reduced ability to conduct heat, renewability, cost-effectiveness, and environmental friendliness. Within this context, the utilization of cellulose aerogel derived from biomass emerges as an attractive material that meets these criteria more effectively compared to conventional insulation materials [236]. Bio-cellulose aerogels have a few impressive features that make them excellent as thermal insulation materials. These properties include lightweight [237], high porosity [238,239], large surface to volume ratio [240], low heat transfer [241], low heat expansion [242], high strength, elastic modulus [243], flame retardancy [244], sustainability [245], and biocompatibility. These properties offer significant advantages in terms of providing long-term sustainable solutions for effective thermal insulation materials in newly developed applications[236].

Restoration of the environment: As well as the major application of aerogel in thermal insulations in aviation and space and construction industries, it is also encouraging other applications such as the remediation of the environment, in the fields of materials and energy in which major requisite features. Among these applications, environmental remediation stands out as a prominent

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area of focus. The field of aerogel-based environmental remediation has matured considerably and encompasses various air and water treatment processes. Aerogels are used in air cleaning for CO_2 adsorption from the atmosphere along with the eliminating of pollutants of volatile organic compounds from industrial and municipal effluents. Furthermore, they play a crucial role in water treatment by adsorbing heavy metal particles, oil, and toxic organic substances, These pollutions are essential contributors to the environmental challenges faced by our world today, including global warming and threats to human health [5]. There are many studies to describe how to make adsorbent such as a technique for expediting the production process and enhancing the quality of bio-based materials derived from paper waste, employing a Kymene cross-linker enhancing formation of gels rather than the alkali/urea dissolving agents and by drying lyophilization method. This mixed reclaimed fiber aerogel is highly hydrophobic and has a highly bending structure. According to their results, the maximum capacity o absorption is 95 gg^{-1} at 50°C with less than 1 wt% bio-based aerogel due to its min lower density (0.007g cm^{-3}) and max high porosity (99.4%)[246]. Another research investigation centered on the development of lightweight and hydrophobic watermelon carbon aerogels (WCA), which exhibited remarkable selectivity in absorbing an extensive range of natural solvents and lubricants. These aerogels demonstrated a ultimate absorption limit ranging between 16 and 50 times of their own weight, and they were able to maintain their absorption and harvesting capabilities over five cycles [247]. To extract the anionic and cationic heavy metallic impurities in water, protein-infused carbon aerogel derived from cellulose (carbogel) is synthesized. Following the successful use of aerogels in the capture of several risky agents like organic liquids, lubricants, and carbon dioxide, marked by their sponge-like structure and tri-dimensional framework have lately piqued the interest of researchers for their potential in the cost-effective capture of heavy metals in waste and effluent streams [248]. The customization of gap measurement, pore range, specific face area and structure chemistry are key requirements that can be readily adjusted in aerogels to encounter the specific requirements of adsorption uses. A diverse range of synthetic, natural, and blended aerogels have been proposed for this objective [249–254].

Energy storage: As modern society and the global economy continue to advance rapidly, accompanied by a growing demographic-based consumption of exhaustible energy types like petroleum and methane has steadily risen. This has resulted in a pressing global challenge of depleting energy resources. Furthermore, the utilization of non-renewable resources unavoidably leads to environmental pollution [255]. To minimize the environmental repercussions and address the depletion of energy resources, there is an urgent necessity to develop state-of-the-art, affordable, and ecologically sustainable energy storage solutions [256]. Presently, two prominent energy storage technologies, namely supercapacitors and rechargeable batteries, have garnered significant attention as highly promising options [257]. To produce high-performance electrochemical energy storage tools, electrochemical effective components are the main indicator [258,259]. To improve their electrochemical behavior, designing the porous framework with magnified precise area for surface exposure and adjustable pore dimensions are the necessity factors [260,261]. Likewise, in situations of batteries, a larger precise area for surface exposure and appropriately sized pores creates additional pathways for the migration of Li^+ ions, resulting in increased volume. Thus, it is crucial to improve novel methodologies and renewable resources that can accomplish a substantial specific surface dimension while effectively controlling the sizes and volumes of the pores [262]. Because of the chemical and mechanical durability, superior resiliency, and porous framework of the bio-based aerogels and foams perform them excellent structure reinforcement component for energy storage devices [255,263]. In contrast to conventional metallic support substances, foam structures and aerogels composed of cellulose offer distinct advantages in terms of lower density, improved flexibility, and enhanced electrochemical performance. These materials possess water-friendly surfaces and numerous absorbent locations, which promote the absorption and transport of electrolyte ions. Additionally, their structured pore hierarchy provides ample space for efficient power conservation [255,264]. For this reason, foam structures and aerogels derived from cellulose have gained recognition as prospective and environmentally sustainable configurations for

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combining with various other active materials in the model and production of cutting-edge energy conservation equipment, including energy-storing capacitors and reusable cells [240,264].

Medical applications: The appealing candidacy of bio aerogels in biomedical applications stems from their essential features of biocompatibility, biodegradability, and non-toxicity [265,266]. These bio aerogels, designed to mimic the extracellular matrices (ECM) found within the body, have facilitated various biomedical applications. Examples include drug delivery [267–269], tissue engineering scaffolds [270–272], antibacterial agents [273–276], biomedical devices [122], biosensing platforms [277,278], and wound dressings [173,217,278,279].

Flexible electronic systems: Fibers and textiles play a significant role in various aspects of our daily lives. The incorporation of multi-functional elements into fabrics, particularly through utilized nanoparticle investigation and informatics, is an expanding field of study. These materials can adapt to environmental changes or respond towards external catalysts like mechanical, thermic, chemical, and magnetic effects [280–284]. Portable electronic devices and intelligent fabrics evolved as new communication platforms with broad uses in industries as diverse as medical sector, professional uniforms, sports, power industry, and defense forces. [286]– [290]. As a result, the development of compact and bendable wire-based electronic apparatus or fiber forms, as well as their incorporation into textile textiles, is gaining relevance [290,291]. Even though there are many studies on the aerogel regarding flexibility of systems, cellulose aerogels show highly promising and effective results [292,293,293]. Such as the synthesis of holocellulose nanofibers/cellulose aerogel filaments using microscopic holocellulose fibers and a reformed cellulose structure from a water-based LiOH-urea solution. These holocellulose nanofibers/cellulose aerogel filaments present a novel procedure for wide-ranging scope, uninterrupted creation of eco-friendly, bendable, and durable aerogel strands with outstanding properties including expansive specific surface area and significant porosity", or "steady production of decomposable, flexible, and sturdy aerogel filaments with exclusive attributes like large surface area-to-volume ratio and pore structure. To achieve this, the group combined specific pulping conditions, which help preserve the hemicellulose content, with mechanical defibrillation techniques, resulting in the fabrication of holocellulose nanofibrils. These holocellulose nanofibers/cellulose aerogel filaments possess remarkable properties including a significant dimension ratio, uniform measurement, outstanding physical strength, and exceptional ability to disperse [294,295]. Yamada et al. has successfully created a bendable energy storage device by combining molybdenum electrical contacts, carbonaceous granules, and ionic fluid blends, alongside ionic gel conductive substances and bendable plant fiber dividers. This assembled device exhibits excellent cycling stability, ensuring its long-term performance [296].

It is worth highlighting that investigation and progression efforts in the domain of bio-based aerogels are currently in progress, and these upcoming facets signify promising avenues for further investigation. Anticipated progress and revelations are poised to unlock new applications and advantages for cellulose aerogels soon.

8. Conclusion

Since the turn of the century, cellulose-based aerogels have sparked a surge in technical and scientific interest due to their sustainable and ecologically favorable origins. Their distinct morphologies and characteristics, as discussed in this study, may be tuned to provide materials well-suited for a diverse implementation. The very porous materials also offer an intriguing foundation for developing high-performance functional materials with distinct features using a template approach. The use of three-dimensional cellulose aerogel as a template can open up new avenues for the development of flexible three-dimensional devices with large surface area [297], scaffolding materials for tissue engineering [298], inorganic nanomaterials [299,300], and polymer nanocomposites [301,302]. On the other hand, it is still extremely difficult to fabricate large-scale cellulose-based aerogels through simple and cost-effective processes such as highly efficient cellulose extraction from various properties and the gathering of cellulose components and one-dimension nano units to evolve exclusive features. With ongoing efforts throughout the world, new innovations in original and recycled cellulose aerogels will give an essential richness of potential for additional improvements and findings in the biochemical, biotic, physics, and technical domains.

9. List of Abbreviations

Acronym	Description
DP	Degree of Polymerization
NaOH	Sodium Hydroxide
NMMO	N-methyl-morpholine N-oxide
3D	Three dimensional
PVA	Polyvinyl Alcohol
TEMPO	2,2,6,6-Tetramethylpiperidin-1-yl)oxyl
CNF	Cellulose Nanofibers
MO	Methyl Orange
NaClO	Sodium Hypochlorite
NaBr	Sodium bromide
BC	Bacterial Cellulose
DIW	Deionized Water
EMIM	Imidazolium acetate
([DBNH][OAc])	Non-enium acetate
DMSO	Dimethyl sulfoxide
SC CO_2	Supercritical Carbondioxide
NMP	Methyl-pyrrolidone
KOH	Potassium hydroxide
BCNF	Bamboo cellulose nanofibrils
MBA	N, N′-methylenebisacrylamide
MTMS	Methyltrimethoxysilane
SBKP	Softwood bleached kraft pulp
TBA	Tert-butyl alcohol
(TEMPO)- (TOCN)	2,2,6,6-Tetramethylpiperidin-1-yl)oxyl, oxidized cellulose nanofibril
PMDI	Polymethylene polyphenylpolyisocyanate
PF	Pineapple Fiber
CO_2	Carbondioxide
HT	High temperature
LT	Low temperature
ESEM	Environmental Scanning electron Microscope
t-BuOH	Tert butyl alcohol
LS	Light Microscopy
AFM	Atomic force microscopy
SEM	Scanning electron microscopy
TEM	Transmission electron microscopy
TA	Tannic acid
TA/B	Tannic acid/Borax
TA/B@PDA	Tannic acid/borax Polydopamine
SAS	Small-angle scattering
WAS	Wide-angle scattering
XRD	X-ray diffraction
USAXS	Ultra-low-angle scattering
PSD	Pore size distribution
N_2	Nitrogen
Hg	Mercury
DSC	Differential scanning calorimetry
DMA	Dynamic mechanical analysis
ASTM D638	American Society for Testing and Materials- Standard Test Method for Tensile Properties of Plastics
ASTM D695	American Society for Testing and Materials- Standard Test Method for Compressive Properties of Rigid Plastics
ASTM D3574	American Society for Testing and Materials-Standard Test Methods for Flexible Cellular Materials—Slab, Bonded, and Molded Urethane Foams

ASTM E1050-10	Standard Test Method for Impedance and Absorption of Acoustical Materials Using A Tube, Two Microphones and A Digital Frequency Analysis System
SW422, SW477, BSWA Technology Co. Ltd., China	SW series Impedance Tubes can accurately measure sound absorption coefficients and impedance
Amprobe SM-10, USA	Sound Meter, United States of America
Hz	Hertz
CC-BY license	Creative Commons Attribution
TGA	Thermogravimetric analysis
TPS	Transient plane source
DTG	Derivative thermogravimetric analysis
C, H, O	Carbon, Hydrogen, Oxygen
LOI	limiting oxygen index
CNF	Cellulose Nanofibril
UL-94	The Standard Tests for Flammability -Vertical burning tests
MPa	Megapascal
HCNFs	Holocellulose nanofibrils
EMI	Electromagnetic Interference
MXene	Two-dimensional (2D) layered conductive nanomaterial, composed of transition metal carbide/nitride
$CaCO_3$	Calcium carbonate
PML	Premna Microphylla
EGO	Electrochemically synthesized graphene oxide
DIW	Direct Ink Writing
TEMPO- (TOCNF)	2,2,6,6-tetramethylpiperidine-1-oxyl oxidized cellulose nanofibrils
MDPA	N-methylol-dimethyphospylpropionamide
BTCA	1,2,3,4-butane tracarboxylic acid
m-LBL	Molecular layer by layer
PDMS	Poly(dimethyl siloxane
CFs	Cellulose Fibers
PALF	Pineapple-leave fibers
b-PI/BC	Bidirectional anisotropic polyimide/bacterial cellulose
HCAFs	Holocellulose nanofibrils/ cellulose aerogel fibers
NCs	Nanocelluloses
TWF	Textile waste fibers
MH NPs	Magnesium hydroxide nanoparticles
FTIR	Fourier-transform infrared spectroscopy
AFM	Atomic force microscopy
MTT assay	3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl-2H-tetrazolium bromide assay
PCR	Polymerase chain reaction
SXAS	Small Angle X-Ray Scattering
CAGR	Compound annual growth rate
LiOH	Lithium hydroxide

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