PREPARATION STRATEGIES AND MULTIFUNCTIONAL APPLICATIONS OF CELLULOSE-BASED AEROGELS IN ENERGY AND ENVIRONMENTAL FIELDS

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Abstract

Aerogel is a type of material with an expanded three-dimensional network structure, assembled by consistent polymer molecules. Organic aerogels have gained attention in recent years as environmentally friendly materials due to their high porosity, low density, and similar characteristics. With the increasing interest, bio-based aerogels are regarded as key materials for sustainable studies due to their renewable sources, and biocompatible structures. In this study, the synthesis methods of cellulose-based aerogels, the structural properties of the obtained materials, and their applications are discussed. In aerogel production, cellulosic materials are generally converted via the sol-gel method and various drying techniques are applied. The pore size and structures of the produced bio-based aerogels vary depending on the precursor, synthesis method, and drying technique. To increase their mechanical strength, crosslinkers and/or hybrid structures are used, and various modifications are made. The produced bio-based find a broad spectrum of applications. Those with low density can be used as supercapacitor materials. Additionally, they are recognized as efficient materials in absorption and adsorption. As a result, bio-based aerogels have a potential that can be evaluated for the development of sustainable material technologies. Thanks to their eco-friendly characteristics and advantageous properties, they hold great potential in various fields including energy storage, CO_2 capture, and electromagnetic shielding. Future studies will focus on improving their properties and enabling industrial-scale production.

Keywords: Cellulose-based aerogels, energy, environment.

1. INTRODUCTION

Cellulose-based aerogels stand out due to their renewable origin, biodegradability, and tunable physicochemical properties, and are becoming pioneers for next-generation energy and environmental technologies [1], [2]. However, despite extensive research, a performance-processability gap persists. Structural homogeneity, mechanical robustness, moisture sensitivity, and scalable production are determined by a complex interplay between precursor raw material selection, sol-gel chemistry, and drying dynamics. Strategies such as cross-linking, composite formation, and hybrid material design are widely explored to achieve the desired properties in aerogels. However, these approaches often lead to trade-offs between sustainability, performance, and process complexity. This perspective focuses on critically evaluating current preparation strategies for cellulose-based aerogels, identifying key limitations, and highlighting open challenges. Emphasis is placed on structure-property relationships related to energy storage, adsorption, CO_2 capture, and electromagnetic shielding. Finally, future research directions necessary to advance cellulose-based aerogels towards scalable and application-oriented material design are discussed.

2. SYNTHESIS METHODS OF CELLULOSE-BASED AEROGELS

Cellulose is a natural and abundant biomass polymer [3]. While aerogel production utilizes a wide array of sources including fibrous plants and agricultural residues the choice of precursor determines the final

morphology and homogeneity [4], [5]. Lignocellulosic biomass typically requires pretreatment to remove lignin and hemicellulose, whereas recycled sources may introduce fillers and pigments that disrupt gelation kinetics [6]. These varying properties compromise reproducibility, necessitating a shift from simply utilizing green sources toward developing source tolerant fabrication protocols. The formation of a homogeneous cellulose solution remains a major production congestion, due to the significant dissolution barrier posed by the intermolecular and intramolecular hydrogen bonds of cellulose. Effective dissolution typically requires specialized solvent systems or alkali-based solutions [7], [8]. While these types of specialized solvent systems enable molecular dispersion, their industrial applications are hindered by environmental toxicity and high operating costs. However, assessment reveals that the primary challenge is not a lack of potential solvents, but a lack of process compatible green systems. These factors make reproducibility difficult and pose significant obstacles to growth on an industrial scale. The sol-gel transition represents a decisive step in network engineering; in this stage a homogeneous cellulose solution is converted into a continuous three-dimensional network, ultimately defining the pore architecture of the aerogel [9]. The transition typically proceeds through three primary mechanisms: physical entanglement, chemical crosslinking, or polymerization-induced gelation [10], [11], [12]. In physical gelation, Van der Waals forces and hydrogen bonds are involved. These networks are known to be extremely fragile and sensitive to moisture, often failing under environmental stress [13]. In chemical gelation, crosslinking agents such as glutaraldehyde or epichlorohydrin are used to obtain a more robust and stable network [14]. While effective in enhancing mechanical strength, these synthetic additives present a significant sustainability performance paradox [15]. Polymerization induced gelation offers a middle ground by improving network integrity through cellulose derivatives, but often increases process complexity [16]. Reliance on traditional synthetic crosslinkers remains a key limiting factor for the industrial application of green aerogels. While alternative methods such as linker assisted or biomass derived systems have demonstrated feasibility [17], they often involve trade-offs in terms of process simplicity or scalability. Solvent exchange and diffusion dynamics represent a key temporal obstacle in production, as the resulting wet gel cannot be dried directly without causing pore collapse. This leads to capillary tensions during evaporation, causing collapse and shrinkage [18]. To maintain the network structure, solvent exchange is essential, typically involving the gradual replacement of the aqueous phase with low surface tension organic solvents. Current protocols suggest gradual change, where the change is gradually increased over long hours or even several days [19]. While these stepwise methods reduce structural damage, they present significant challenges in terms of solvent consumption and process efficiency [20]. Studies on accelerated diffusion techniques or one pot gelation systems as alternatives to time consuming solvent changes will both shorten the time and reduce solvent waste. Drying is the most energy intensive stage in aerogel production and determines whether the delicate 3D fibrous network will survive the transition from wet gel [21]. Supercritical drying remains the reference method for maintaining high surface area and homogeneous pore structures by bypassing the liquid gas interface by operating above the critical point of CO_2 [22]. On the other hand, while cost competitive at laboratory scales (15–50 €m^{-2} for freeze-drying compared to 30€ m^{-2} for $scCO_2$ drying), the processing time, scaled by gel thickness, poses significant obstacles to mass production [23], [24]. Although freeze-drying is more environmental friendly, it often results in disordered and anisotropic pore structures due to stochastic ice crystal growth, which disrupts the fibrous network and compromises mechanical strength [25]. This disadvantage can be overcome with alternative freezing strategies. For example, cellulose nanofiber/sodium alginate aerogels with interlocking lamellar structures have achieved significant mechanical properties (340 kPa at 90% strain) and high adsorption capacities (88.91 g g^{-1}) [26]. Ambient pressure drying represents the most economically viable way for mass production [27]. However, this leads to the problem of springback effect and pore collapse due to capillary stresses during evaporation [28]. While extensive surface modifications or skeleton reinforcements can yield aerogels with low densities (0.058 g cm^{-3}) and minimal shrinkage (<10%) [29], these chemical interventions add complexity and cost. The focus of drying studies should shift to developing naturally robust cellulose scaffolds via bio based crosslinking that can withstand evaporation stresses without structural degradation and provide consistent integrity without the need for extensive chemical processing.

3. PERFORMANCE AND CHARACTERIZATION ANALYSES OF CELLULOSE-BASED AEROGELS

Most studies characterizing cellulose-based aerogels focus on high surface area or capacitance under idealized laboratory conditions, while neglecting the synergistic relationships that determine the durability of real applications. Electrochemical evaluation of cellulose-based aerogels suffers from an idealized performance gap; unprecedented measurements obtained in the laboratory cannot be replicated with commercial scale electrodes. Cycling stability, specific capacitance (F g^{-1}), and specific surface area (g m^{-2}) are important evaluation measures for energy storage [30], [31]. Cellulose-based supercapacitors have been reported to have capacitance values between 20 and 326 F g^{-1} and energy densities between 10 and 15 Wh kg^{-1} [32]. For assessing charge storage mechanisms, methods like Galvanostatic Charge-Discharge and Cyclic Voltammetry are essential [33]. However, a critical gap exists: these metrics are often measured using thin-film electrodes with low mass loadings, which fail to translate to commercial-scale systems where internal resistance scales non-linearly. Furthermore, while high capacitance retention (e.g., 82–96% after 5,000 cycles) is frequently reported [34], [35] these tests are rarely conducted under mechanical deformation or varying humidity. Structural and morphological analysis through electron microscopy imaging, and gas adsorption remains indispensable for quantifying the interconnected porosity and fibrillar network of cellulose-based aerogels [36], [37]. Likewise, spectroscopic techniques are essential for determining crystal domains and functional groups [38], [39]. There is a significant gap in research investigating the relationship between Crystal Index and long-term thermal fatigue measured by Thermal Gravimetric Analysis [40]. Characterization should shift from relying on snapshots to in-situ and in-operation measurements to understand how the pore architecture evolves during real CO_2 adsorption or electrochemical cycling. Mechanical dependability and compositional residues are important but sometimes disregarded aspects of cellulose aerogel performance. X-ray Fluorescence and elemental analysis reveal significant inorganic residues, which vary depending on the source [41], [42]. Rather than treating these as impurities to removed, they should be evaluated as potential inherent catalysts for adsorption or structural stabilizers. Furthermore, mechanical performance, evaluated through tensile or compression testing, remains a primary limiting factor [43]. Because different investigations use varying electrolyte concentrations and compression ratios, direct comparison is challenging and the existing characterization lacks integrated comparison.

4. ENERGY AND ENVIRONMENTAL APPLICATIONS OF CELLULOSE-BASED AEROGELS

Aerogels' multipurpose structures enable a range of uses, including wastewater treatment and the storage of renewable energy. Cellulose aerogels have shown remarkable adsorption capacities in environmental remediation, reaching 575.7 mg g^{-1} for dyes and exhibiting great potential in oil spill cleaning by solar powered viscosity reduction [44], [45], [46]. Although hierarchically porous cellulose matrices are theoretically perfect for CO_2 capture, their effectiveness is often assessed using a straightforward mass difference during regulated heating/desorption cycles [47], [48], [49]. Aerogels utilized in supercapacitors and capacitive ion removal systems [50] demonstrate electrochemical capabilities as high as 326.51 F g^{-1} in the field of energy storage. They scale nonlinearly with actual commercial electrodes, yet [51]. The paradox of reliance on large carbon footprint fillers eliminates cellulose's sustainability due to the loading of conductive polymers or carbon nanoparticles [52], [53] needed for aerogels to operate competitively. Cellulose aerogels have significant potential for radiative cooling and thermal control. Sustainable cellulose/ZnO composites have achieved temperature reductions below ambient temperature of up to 6.9 °C by utilizing their strong emissivity in the atmospheric transparency window [54]. However, the long term resistance of bio based cooling films to factors such as UV degradation and dust accumulation has largely remained under researched. Additionally, even though aerogel fibers have attained extremely low thermal conductivities (0.0242 W m^{-1} K^{-1}), structural integration is still limited by their mechanical fracture strength [55].

5. CONCLUSION AND FUTURE OUTLOOK

At this point, cellulose-based aerogels must satisfy demanding industrial performance requirements while maintaining their inherent sustainability. The shift to functional systems are limited by a persistent performance processability gap, even while laboratory prototypes attain remarkable metrics, adsorption or high specific capacitance. In addition to standardized benchmarking that assesses materials under realistic environmental stressors like humidity and mechanical fatigue, a fundamental shift toward process simplification through one ot synthesis and low energy drying is necessary to bridge the benchtop to market transition. Additionally, in order to overcome some technical obstacles like CO_2/N_2 selectivity and ion diffusion resistance in thick electrodes, material design must shift from being precursor-driven to application-driven. Utilizing biomass chemistry without sacrificing its ecological integrity through hazardous additions is ultimately what will determine the future of cellulose aerogels. By establishing structural property relationships, these materials can be transformed from laboratory scale into scalable, long-lasting systems for energy and environmental sustainability.

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