Hydrogels and Aerogels of Carbon Nanotubes

Anju Paul and Arunima Reghunadhan

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Abstract

Aerogels and hydrogels are materials having high surface area, adsorption capacity, and mechanical properties. These gel materials are employed in various fields such as water electronics, smart materials, drug delivery, biomedicine, etc. Carbon nanotubes are much famous for their extraordinary mechanical strength and aspect ratio. Incorporation of carbon nanotubes (CNTs) generally impart enhanced properties to the matrix. CNTs can be employed in the fabrication of aerogels and hydrogels, and the resultant materials can be employed in various

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applications. The chapter discusses in detail about the basics of hydrogels and aerogels, their structure, properties, and applications.

Keywords

Aerogels · Hydrogels · Carbon nanotubes · Applications

Introduction

Gel is a common terminology used for semisolids or solid-like liquids. Gels were assigned the name in connection with gelatin. The gel state is considered as a colloid of liquid dispersions of solids. The chief component is a liquid in which a suitable solid will be dispersed. Even though they appear to be like liquids, they have a three-dimensional arrangement of solid particles. In the case of macromolecules, when certain substances (solids or liquids) are heated or cooled from their respective physical state, instead of giving a sharp change will undergo a process called gelation, where the viscosity of the material will rise. Gelation can be described as a bond percolation transition. At the gelation point, an infinite cluster is created that spans the whole system. They have been employed nowadays in many fields due to their diversity in their properties and synthetic sources. They can be very soft and smooth and at the same time hard and tough. They have been employed in energy storage devices such as batteries and fuel cells, in food and pharmaceuticals, coatings, in sanitary napkins and diapers, and so on (Farjami and Madadlou 2019; Draper and Adams 2017; Padmasri et al. 2020).

Gels can be classified mainly into four categories

(a) Aerogels.
(b) Hydrogels.
(c) Organogels.
(d) Xeroxes.

Aerogels and hydrogels are an emerging class of materials, which has very high degree of applications. Both these materials are largely exploited in the biomedical research owing to their superior properties and biocompatibility. Aerogels and hydrogels are porous structures with capacity to absorb or hold other materials and thus they can act as host materials in many applications, especially in drug-related studies. The era is mainly of biopolymers, biomaterials, biotechnology, and biomedicine. All these fields will use the aerogels and hydrogels in one way or the other. Aerogel and hydrogels can be synthesized from a variety of substances like polyelectrolytes, biomaterials, polymers, nanomaterials, etc. This chapter is dealing with the gels of carbon nanotubes, which are materials with versatile properties. Carbon nanotubes from their discovery stage to till date are inevitable materials in many fields. They have been the most favorite filler for the polymers as they can give strength and other superior properties to the matrix materials in which they are incorporated (De France et al. 2017; Jung et al. 2014; Bigall et al. 2009). The basic things such as the structure, classification, properties, and general applications of the CNTs are widely discussed in many reports.

Aerogels and Hydrogels: Structure and Properties

Aerogels have been a popular term since 1960s for the aerospace applications, but nowadays their applications have been widened to many areas. The discovery of the aerogels was reported in the 1930s by Samuel Stephens Kistler. Aerogels are gels which maintain their porosity after fabrication. Aerogels are a broad category of porous, solid materials that exhibit a bizarre range of extreme material properties. Aerogels, in particular, are notable for their extremely low densities ranging from 0.0011 to 0.5 gcm^{-3}. Aerogels, in fact, are the lowest density solid materials yet created, including a silica aerogel that was only three times heavier than air when it was created.

Structure

Structure of Aerogels

All aerogels have open porous structure. This means that the gas or air in the aerogel is not entrapped within solid compartments, and porosity varies in size from 1 to 100 nanometers, with a typical diameter of 20 nm. Aerogels are a type of dry substance. The term “aerogel” alludes to the fact that aerogels are made from gels, which have essentially the same solid structure as a wet gel but with gas or vacuum instead of liquid in their pores. An aerogel by definition is a solid foam that is open-celled, mesoporous, and consists of a network of interconnected nanostructures with a porosity (nonsolid volume) of at least 50%. Aerogels are commercially available in many forms (shapes). The materials involved and the synthetic strategy makes the difference in their shapes. The commonly available ones are particles, powders, monoliths, thin films, composite blankets, etc.

A simple schematic structure of the aerogel is given in Fig. 1.

Structure of Hydrogels

Hydrogels possess a three-dimensional network of materials, mainly polymers, which are capable of swelling when in contact with water and entrap a large amount of water in them. The polymers can be both natural and synthetic, which are essentially hydrophilic. They were first reported in 1960s by Wichterle and Lím. For a substance to be classified as a hydrogel, it must contain at least 10% water by weight (or volume). Because of their high water content, hydrogels have a similar degree of elasticity to real tissue. The hydrophilicity is owed to the presence of functionalities like amine ($-NH_2$), carboxylic (-COOH), hydroxyl (-OH), amide ($-CONH_2$), etc. Hydrogels are classified as cationic, anionic, or neutral based on

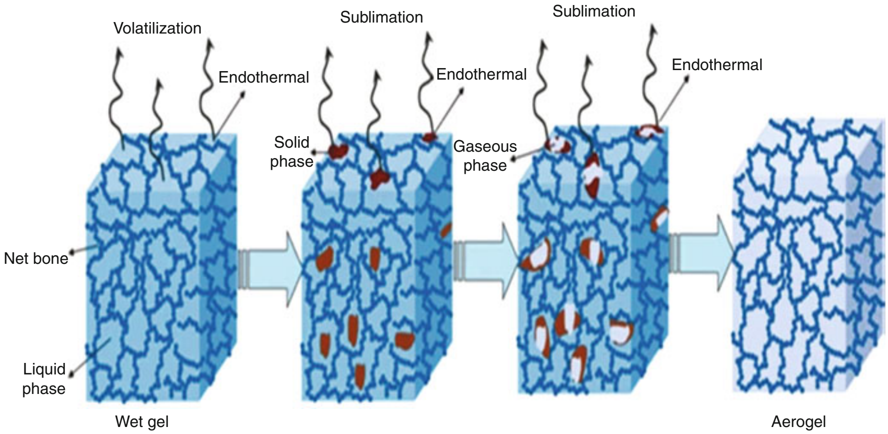

Fig. 1 Schematic structure of aerogel. (Reproduced with permission from Alwin and Sahaya Shajan (2020) under CC By license)

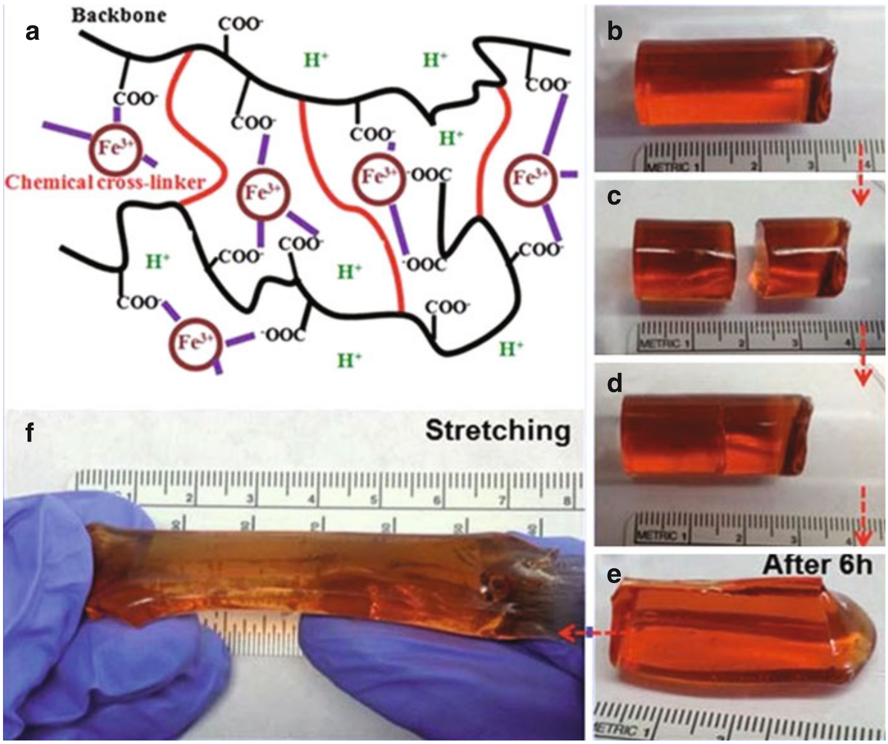

Fig. 2 Images of a self-healing hydrogel and the internal chemical bonding. (Reproduced with permission from Wei et al. (2013), Copyright (2013), RSC)

the ionic charges of the bonded groups. A schematic image of the hydrogel structure is given in Fig. 2.

Properties

Properties of Aerogels

Quite a lot of aerogels have a unique set of material features that no other substance has. The properties of aerogels mainly depend on the materials from which the aerogel is obtained. Many of an aerogel's qualities can be altered by customizing the manufacturing process. The lowest bulk density of any known material is held by specific aerogel compositions. Commonly they have very high damping, electrical conductivity, adsorption capacity towards water and oil, elasticity, catalytic activity, luminescence, etc. Also they are known for their low refractive index, low dielectric constant, high specific surface area, low mean free path, etc. (Najjar 2012).

Properties of Hydrogels

Like the aerogels, the properties of the hydrogels are also dependent on the material involved. Despite considerable advances, a basic understanding of gel characteristics is still insufficient for the logical design of new gel systems. Since they have been mostly employed in the biomedical field, they obviously possess biocompatibility. They possess tunable biodegradability, which make them superior over the other materials that are used in the biomedical applications. Since they can hold water, swelling is the most important quality of a hydrogel. The ingested liquid acts as a selective filter, allowing some solute molecules to diffuse freely, while the polymer network acts as a matrix, holding the liquid together. Whenever a dry hydrogel absorbs water, the initial water molecules will wet the polar, hydrophilic groups, resulting in primary bonded water. The network swells when the polar groups are hydrated, exposing hydrophobic groups, which connect with water molecules as well, resulting in hydrophobically bound water, or secondary bound water. Whenever a dry hydrogel absorbs water, the initial water molecules will wet the polar, hydrophilic groups, resulting in primary bonded water. The network swells when the polar groups are hydrated, exposing hydrophobic groups, which connect with water molecules as well, resulting in hydrophobically bound water, or secondary bound water. The total bound water is typically referred to as a combination of primary and secondary bound water. The measurement of total water and the excess water will predict the efficiency of swelling (Yahia 2015; Thakur et al. 2018). Similar to aerogels, they have high mechanical strength and porous structure. The natural polymer-based hydrogels are, however, less strong than their synthetic ones. To attain extended life span, high water absorption capacity, and high gel strength, naturally produced hydrogels are increasingly supplanted by synthetic hydrogels (Chai et al. 2017; Bahram et al. 2016).

Synthetic Aspects of Aerogels and Hydrogels

Aerogels

Aerogels can be derived from metal oxides like zirconia, titania, quartz, lanthanide, and actinide oxides, etc., from polymers, from carbon derivatives such as graphite, grapheme, CNTs, amorphous carbon, from biopolymers such as starch, cellulose, alginates, from chalcogenides, carbides, and so on. Depending on the synthesis, aerogels are classified as normal aerogels, xerogels, cryogels, and aerogel-related materials.

Synthetic Strategy of Aerogels

Aerogels are chiefly synthesized by the sol-gel method. Aerogel fabrication is a multistep process and generally limited to three stages. They are – gel preparation, aging of the gel, and drying.

(a) Gel preparation: Solid nanoparticles form a three-dimensional solid network with solvent-filled pores when they cross-link. A gel is first formed in solution, and then the liquid is carefully removed to leave the aerogel intact; first, a colloidal suspension of solid particles known as a sol is created.
(b) Aging of the gel: The gel's structure is strengthened by this process. The gel that was previously made is aged in its mother solution. The gel is strengthened during the aging process, resulting in little shrinking during the drying phase. The gel is left undisturbed in the solvent after gelification to finish the process. The aerogel product is created after the reaction is completed. Sol-gel manufacturing, which involves dissolving alkoxides or metal salts in alcoholic or aqueous solutions and supercritical drying, can be used to make inorganic aerogels.
(c) Drying: The solvent must be removed while the solid aerogel network is preserved. This can be accomplished either through supercritical drying or at ambient temperatures. In most cases, aerogel materials are made by extracting the solvent from a gel matrix in a supercritical fluid media. This can be performed by raising the gel solvent system's temperature and pressure over their critical points, then releasing pressure above the critical point until only vapor remains (Thapliyal and Singh 2014).

Schematic representation of the aerogel manufacturing is given in Fig. 3.

Hydrogels

As stated above, the hydrogels are fabricated from many materials, polymers, and polyelectrolytes. Depending on the polymer involved, the type of interaction, and type of functional groups, the hydrogels are classified to many types. As per the polymer involved, hydrogels can be homopolymeric, copolymeric, and

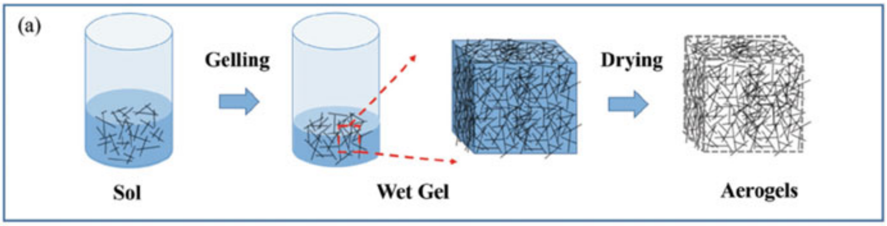

Fig. 3 Fabrication of aerogels from sol-gel synthesis. (Reproduced with permission from Zou et al. (2010), Copyright (2010), American Chemical Society)

multipolymeric or interpenetrating. Depending on their physical structure, there can be amorphous, crystalline, and semicrystalline hydrogels. As per the interaction involved, they are further classified as chemically cross-linked or physically cross-linked. The shape or the appearance is also a factor to classify them. Hydrogels are generally classified or termed as per the functionalities involved. Hence, there are anionic hydrogels, cationic hydrogels, nonionic hydrogels, zwitterionic, etc. (Ahmed 2015; Nguyen 2018; Ahmed 2013).

Synthetic Strategy of Hydrogels

Different strategies are employed for the abovementioned different categories of hydrogels. Simply we can say that a hydrogel can be made using any process that can be used to make a cross-linked polymer (Madduma-Bandarage and Madihally 2021). The available methods for the synthesis of hydrogels are:

A. Physical cross-linking: Physical or reversible gels have gained popularity because of their relative simplicity of manufacturing and the lack of cross-linking agents. These methods are further categorized into a number of ways as follows (Gulrez et al. 2011; Nautiyal et al. 2020).
   (a) Varying temperature of polymeric solution.
   (b) Ionic cross-linking.
   (c) Hydrogen bonding.
   (d) Maturation (heat-induced agitation).
   (e) Freeze-thawing.
   (f) Complex coacervation.
B. Chemical cross-linking: This involved bond formation and extension to three-dimensional cross-linked structure between the components (Akhtar et al. 2016). It can be introduced in the following ways.
   (a) Using cross-linking agents.
   (b) Grafting.
   (c) Radiation cross-linking.
   (d) Bulk polymerization.
   (e) Solution polymerization.

Carbon Nanotube Hydrogel Composites

The hydrogels are considered as smart materials which respond towards the external stimuli. The stimulus can be pH, temperature, light, etc. Carbon nanotube hydrogels finds their application in actuators and sensors (Chang et al. 2019; Berke et al. 2018; Manek et al. 2016; De Volder et al. 2011), biofuels (Choi et al. 2015; Yin et al. 2019; Moehlenbrock et al. 2012; Wen et al. 2012), tissue engineering (Vashist et al. 2018; Ahadian et al. 2016; Shin et al. 2012; Liu et al. 2020; Shin et al. 2017), water treatment (Wang et al. 2020; Zhao et al. 2021), energy harvesting (Zhang et al. 2019), etc. Most of these smart materials are employed either in the electronic applications or in biomedical applications. Carbon-based hydrogels can be of either biopolymers or synthetic. They are generally classified as hydrogels involving or reinforced with carbon nanotubes, CNT-hybrid hydrogels, composite hydrogels of CNTs, and functionalized CNT hydrogels. Hybrid hydrogels based on carbon nanotubes (CNTs) have emerged as promising possibilities for regenerative medicine, tissue engineering, drug delivery, implantable devices, bio-sensing, and bio-robotics. Because of their great mechanical strength, efficient surface area, and dielectric strength, CNT-based hybrid hydrogels have gotten a lot of interest. CNTs have been shown to have excellent bioavailability, stability, and electromagnetism, making them one of the most potential nano-fillers for a diversity of applications (Ravanbakhsh et al. 2019, 2020). Inclusion of nanotubes normally contributes towards the enhanced the mechanical strength.

Liu and coworkers reported the random copolymerization of acryloyloxyethyltrimethylammonium chloride and acrylamide in aqueous suspensions of carbon nanotubes to yield multifunctional carbon nanotube composite hydrogels having mechanical improvement. The produced composite hydrogels have a high toughness of 1.6 MJ m^3 and Young's modulus of 0.2 MPa, and can be stretched to 1017 percent strain and compressed to 80 percent strain. The composite hydrogels had a strong self-healing capacity, with a mechanical self-healing efficiency of greater than 90% (Pan et al. 2020). The properties can be tuned by varying the concentration of CNTs. Furthermore, the quantities of CNTs and cross-linking agents in the hydrogel matrix have a big impact on the mechanical behavior. The concentrations of CNTs and cross-linkers injected in the hydrogel matrix were optimized to provide good mechanical performance to the pure polymer solution, which causes easy network formation (Klink and Ritter 2008).

Carbon nanotube hydrogels can be of conductive nature and thus can be employed in the electrical and electronic applications. By electrodepositing CNTs and chitosan onto a carbon paper electrode that functioned as an anode electrode in a bioelectrochemical system, Liu et al. created a portable conductive CNTs nanocomposite hydrogel. The outcomes of their microscopic, spectroscopic, and electrochemical investigations were reported, and they suggested that the hydrogels possess excellent electrical properties (Liu et al. 2014). In one of their research, Servant et al. created an electro-responsive delivery system that can distribute drug molecules in a pulsatile way and can be regulated by the application of an electric voltage on/off. The inclusion of pristine Mints in the polymeric network enhanced the electrical

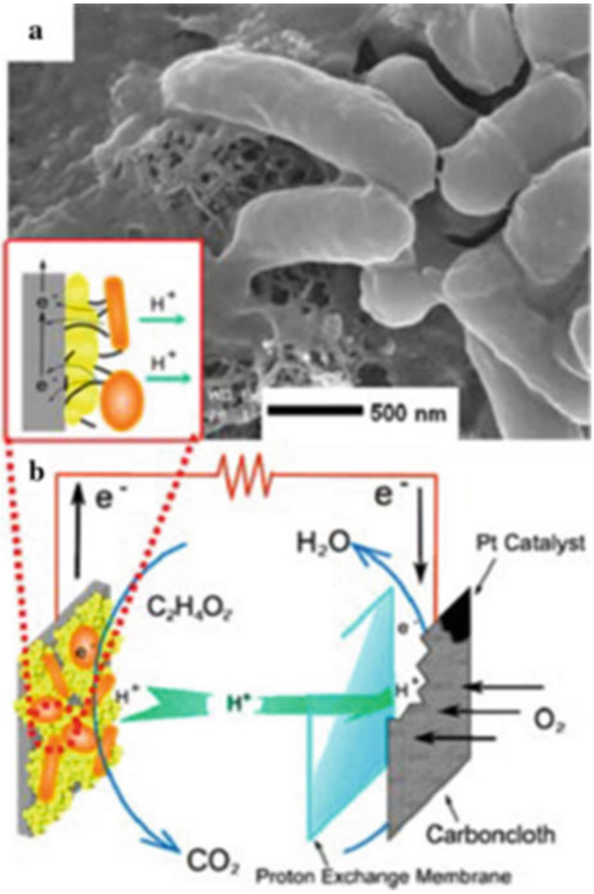

Fig. 4 Carbon nanotube-based conductive composite hydrogel paper (**a**) the morphology and (**b**) the working of electrode based on this composite hydrogel. (Reproduced with permission from Ref. (43), Copyright (2014), American Chemical Society)

characteristics of the produced hybrid hydrogels, according to their findings (Servant et al. 2013) (Fig. 4).

Polysaccharides contribute towards a chief part in the CNT-based hydrogels. Alpha and beta cyclodextrins can be employed in the hydrogel fabrication, and these gels are very much important in the supramolecular chemistry or the host-guest chemistry (Hui et al. 2010; Deng et al. 2018). Ritter and Klink suggested that the dispersion between the cyclodextrin and CNTs increase the stability of the system and increase in the viscosity of the mixture compared. The biomedical applications of composite hydrogels of CNT with biopolymers are endless (Ahadian et al. 2014; Venkatesan et al. 2014; Imaninezhad et al. 2017; Kuche et al. 2018). Single-walled and multiwalled nanotubes are commonly associated with PNIPAAm (poly(N-isopropylacrylamide)) for various biomedical applications. In an interesting report by Chen and his colleagues, PNIPAAM hydrogels were created having interpenetrated structure with MWNTs and served as a substrate for the production and attachment of cell sheets. The increased mechanical strength and hydrophobicity of the MWNTs-PNIPAAM nanocomposite samples were credited with the effective cell attachment. Their research also demonstrated that MWNT impregnation in PNIPAAM hydrogels resulted in a high cell density and a hydrophilic surface that allowed for efficient cell sheet recovery (Y. S. Chen et al. 2013). Multiwalled nanotube-carrageenan hydrogel composites were fabricated for drug delivery applications. The properties like remote drug carrying ability showed variation as a function of nanotube concentration (Estrada et al. 2013). A large number of works also exploit the nanotube hydrogels for medical applications.

Carbon Nanotube Aerogel Composites

Aerogels have a prominent role in the scientific realm, as they are one of the exciting materials with unique properties. They are derived from gels composing networks of nanomaterials. Currently aerogels have entered the field of nanotechnology by the incorporation of numerous nanomaterials into aerogel networks, among which carbon materials are gaining attraction. Carbon nanomaterials such as graphene, carbon nanofibers, carbon nanotube to name a few have been incorporated into aerogels to form composites resulting in enhanced functional properties of aerogels (Araby et al. 2016).

Carbon nanotubes have remarkable electrical conductivity and known to possess outstanding mechanical properties and stability. They were utilized in intense research for a variety of applications since its discovery (Kim et al. 2009). Sol-gel assembly is widely employed to transform carbon nanotube powders to aerogels. In this technique, initially carbon nanotube powders are dispersed in aqueous or organic solvents to obtain a colloidal solution. This solution is then hydrolyzed to form a gel with numerous homogeneous cells. Air drying or freeze drying may also result in carbon nanotube aerogels. Reinforced aerogels can be synthesized by binding with specific polymers, thereby improving strength and structural stability. When polyvinyl alcohol was mixed with single-walled carbon nanotube by freeze-drying, it was seen that strength and elasticity were improved (Bryning et al. 2007). Such reinforcing effect can be provided by other binders like hexagonal boron nitride and polydimethane siloxane. These carbon nanotube composite aerogels can be mold into a variety of size and shape. Nevertheless, the addition of such binders increases the impurity content, which may further reduce porosity.

Another significant way of improving mechanical properties is to graft carbon nanotube with functional groups, and such aerogels are found to contain less impurity with an additional advantage of larger surface area and better electrical conductivity. Compression and heating of fluorinated multiwalled carbon nanotube produced strong interlinked covalent bonds between individual nanotubes and aerogels with superior properties were obtained (Sato et al. 2008).

Chemical bonding can also tailor the morphology of aerogels. Honeycomb structure was attained by freeze-drying technique, and these aerogels are found to possess unique properties like large surface area, light density, and high electrical conductivity. Surface of carbon nanotube has to be modified in order to prevent agglomeration while forming a gel. Surfactants, conjugated polymers, and other nanomaterials like graphene oxide are used for this purpose. Carbon nanotube aerogel composite with graphene oxide was fabricated by hydrothermal treatment resulted in enhancement of mechanical properties. The aerogels can withstand a strain load of 80% with complete structural recovery (Lv et al. 2016). Vacuum infusion technology was employed to synthesize shape memory polymer composite from carbon nanotube and graphene, epoxy resin being the matrix. The composite exhibit very high conductivity owing to the three-dimensional network of reduced carbon nanotube and graphene aerogel. The high conductivity together with low density makes this material wonderful candidate in the field of thermal sensors, electromagnetic shielding, and actuators (X. Liu et al. 2015).

A conductive, cross-linked, lightweight, and highly stable aerogel films of carbon nanotube and copper oxide was fabricated by two-step chemical vapor deposition to act as electrode for rechargeable zinc-air batteries. Nitrogen-doped carbon nanotube synergically act with spinel Co_3O_4 nanoparticles for the catalysis of both oxygen reduction and oxygen evolution reactions. These catalyst supporting scaffolds were found to be highly bendable, showed stable cyclic performance, and displayed low resistances, and state promising applications in energy storage (Zeng et al. 2017). Both single-walled and multiwalled carbon nanotube were composited with cellulose to develop thermoelectric materials. The obtained films were lyophilized to get aerogels, and it was found that no significant change in Seebeck coefficient for composite of multiwalled carbon nanotube, and an elevated Seebeck coefficient was observed with single-walled carbon nanotube. Thermal conductivity was raised by the addition of carbon nanotube, and their cost-effectiveness and flexibility make these aerogels as superior candidates for thermoelectrical applications (Gnanaseelan et al. 2018). Qi and coworkers developed electrically conductive aerogels from carbon nanotube and cellulose, and these aerogel composites were analyzed for their capacity to sense vapors of volatile organic compounds such as ethanol, methanol, and toluene. Both polar and non-vapors were sensed by these composites with prompt response and high sensitivity. The results were reproducible and the work provides a pathway to utilize carbon nanotube with its three-dimensional porous matrix to act as novel class of chemical sensors (Qi et al. 2015).

In order to overcome poor solubility and low dispersion of carbon nanotube, it was composited with polyacrylonitrile to obtain aerogel via thermally induced phase separation method. It was found that there was enhancement in thermal, electrical, and mechanical properties with significant increase in compressive strength which can be attributed to the three-dimensional nanotube and nanofibers network in matrix (Dourani et al. 2019).

Shen et al. synthesized carbon nanotube aerogels using sol-gel technique. These aerogels exhibited varying and controllable density, good electrical conductivity, and high specific surface area. These cost-effective carbon nanotube aerogels were found to have a variety of applications. Multifunctional thin aerogel films were synthesized by freeze-drying from aramid nanofibers, carbon nanotube, and hydrophobic fluorocarbon. These hybrid aerogel films demonstrate large specific surface area

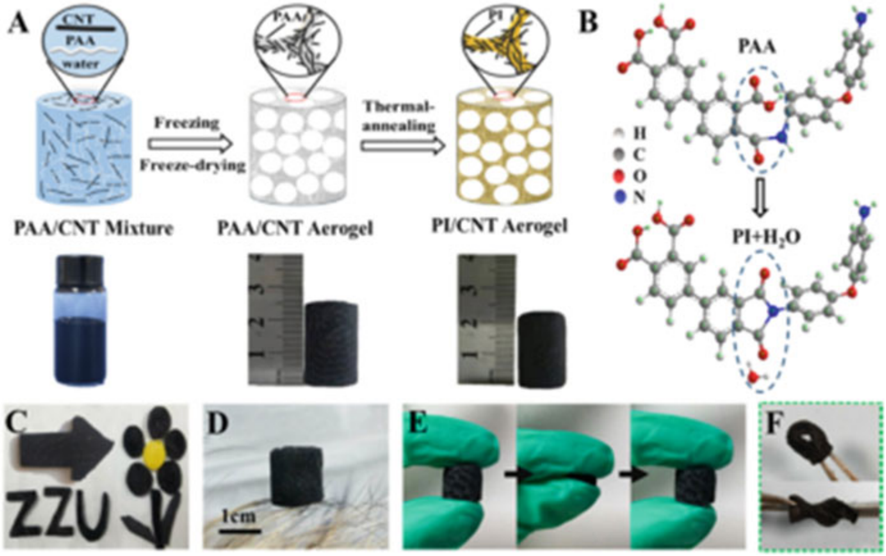

Fig. 5 CNT-based polyimide aerogels: (**a**) schematic of the stages in the aerogel production; (**b**) the possible chemical structural changes; (**c**), (**d**), (**e**) and (**f**) are the images of the aerogel material during different mechanical deformation such as compression, expansion, and bending. (Reproduced with permission from Ref. (61), Copyright (2016), American Chemical Society)

(232.8 $m^2 \cdot g^{-1}$), high hydrophobicity, and excellent electrical conductivity (230 $S \cdot m^{-1}$). The fluorocarbon resin enables the aerogel to act as self-cleaning material by improving the hydrophobicity (Fig. 5).

Highly porous, super elastic, robust, high temperature-resistant polyimide carbon nanotube composite aerogels were prepared by freeze-drying and thermal imidization technique. Efficient sensing was exhibited due to the strong chemical interactions of the components. It displays ultrahigh sensitivity (11.28 kPa^{-1}), ultralow detection limit (0.1% strain, <10 Pa) with a fast response time (50 ms) and recovery time (70 ms). In addition, these composite aerogels were stable at very high temperatures as they could sense even after annealing at different temperatures (X. Chen et al. 2019). To conclude, high porosity, ultralow density, lightweight, and high specific surface area of aerogels are accompanied by excellent mechanical strength, high electrical conductivity, reactivity, and thermal stability by the incorporation of nanomaterials like carbon nanotube. Generally, low cost, efficient methods like freeze-drying or sol-gel technique are used for the fabrication. Carbon nanotube acts as spacer in aerogels, which improves the electrical conductivity, mechanical and structural properties. Carbon nanotube incorporating aerogel composites are widely used in energy storage applications like lithium-ion batteries and supercapacitors.

Properties of Hydrogel and Aerogel Composites

Properties of Hydrogel Composites

Hydrogels can absorb large amount of water and highly biocompatible when compared with polymers owing to their porous structure which makes them potential candidates for biomedical applications. They can form cross-links among polymer molecules, and bioactive agents can be added to sustain cell viability (Xu et al. 2010). Nevertheless, mechanical properties are degraded by the presence of interstitial fluid in hydrogels compared with other polymers. Hence, structure should be modified in order to improve the mechanical response of hydrogels. Hydrophilic polymers like polyethylene glycol and polyethylene oxide are composited for tissue engineering due to their resistance to protein adsorption. However, the polymers such as polyethylene glycol, polyethylene oxide, and polyvinyl alcohol are highly inert in a biological environment. But they are restricted due to the resistance in adherence to living cells while cross-linking being the essentiality in tissue engineering (Kobayashi and Hyu 2010).

Versatility in synthesis, flexibility, diversity in composition, tunability in properties, and good shape moldability of hydrogels make them significant in industrial and academic research. They are three-dimensional networks, which can absorb large quantity of water, and are formed from hydrophilic homopolymers or cross-linked copolymers. When exposed to external stimuli like temperature, enzymatic activity, light, magnetic field, pressure, or electric field, some hydrogels act as reversible in their properties. Hence they can be treated as stimuli responsive biomaterials for various applications (Ward and Georgiou 2011).

Large number of synthetic polymers such as polyacrylic acid, polyethylene glycol, polyvinyl chloride, polyacrylamides, and polyurethanes are cross-linked with hydrogels, and these polymeric hydrogels are highly beneficial owing to their mechanical strength, flexible chemical structure, and degradation rate. However, hydrogels from natural sources are found to be more cell proliferative and undergoes efficient tissue generation. Chitosan, collagen, elastin, heparin, and hyaluronic acid are composited with hydrogels for better cell interactive and cell adhesion properties (Slaughter et al. 2009).

Biodegradability of hydrogels is utilized in biomedical applications, and many hydrogels especially natural hydrogels can undergo dissolution by hydrolysis easily. They are widely employed in drug delivery applications since they possess highly porous structure thereby improving the transportation ability. By controlling the porosity, drug diffusion and timely release can be manipulated. In case of tissue engineering, bioadhesion capability to act as scaffolds is the significant feature. For adhering tissues and cells to hydrogels, linker molecules are sometimes used to form covalent or noncovalent interactions thereby improving adhesive properties The efficiency and suitability of hydrogels for various applications mainly rely on network structure. Choice of the polymer, type of cross-linker, cross-linking density can tailor porous network architecture and mechanical properties.

Functional hydrogels are used as biosensors and actuators as they respond to external optical, magnetic, and electrical stimuli. Polymeric hydrogels are comparatively inert and nonresponsive to external changes. In such a case, hydrogels are composited with novel functional entities to attain desirable properties (Merino et al. 2015; S. Xu et al. 2016). Nowadays, nanoparticles are incorporated in hydrogels to fabricate novel nanocomposite with tunable functionalities and tailored properties.

On account of response to stimuli, they can be differentiated into various types such as thermo-responsive hydrogels, pH-responsive hydrogels, and light- and chemical-responsive hydrogels. The equilibrium among hydrophobic and hydrophilic segments decides the characteristics of a thermo-responsive hydrogel. A minute change in temperature can disturb this equilibrium and results in sol-gel transition (Bajpai et al. 2008). pH-responsive hydrogels display specific physical and chemical properties at various pH ranges. At high pH, acidic functionalities deprotonate while basic groups undergoes protonation at low pH. Binding of different ions to polymer segments may also results in hydrogel swelling. Activation of hydrogels by light can be noninvasive and remove, hence they are potential materials in drug delivery, sensors, micro lenses, etc. (Dong and Jiang 2007).

Properties of Aerogel Composites

Aerogels are nanostructured ultra-highly porous functional material synthesized via sol-gel technology. Aerogels can be treated as gel-like structures with nanoscale pores capable of forming macroscopic monolith. Generally they consist of noncrystalline matter with random cross-linking network. They are qualitatively different in bulk properties of other states of matter and possess diverse chemical constitution. These aerogels can be transparent, opaque, or colored with a variable refractive index. They are remarkably significant by their huge internal surface. Surface can be easily functionalized and can be made hydrophobic by introducing into polymeric residues. Interpenetrating aerogel architecture can be attained by embedding particles like dyes or ferroelectrics into aerogels. They are characterized by their low density, low sound velocity, ultralow modulus, low dielectric constant, and comparatively low thermal conductivity. They can form silica and other oxide aerogels as they will not give any reaction with metallic melts up to a temperature of 950 °C (Schaedler et al. 2011). They are capable of forming composites with a variety of compounds such as oxides, polymers, cellulose, starch, and carbon materials. Recently nanomaterials are incorporated into the aerogel matrix to fabricate composite aerogels and these composites find potential applications in sensors, biomedical implants, aerospace, and energy storage devices.

Incorporation of nanomaterials enhanced many functional properties of aerogels. Introduction of carbon nanomaterials like graphene, carbon nanotube, etc., into aerogels could significantly improve the electrical conductivity of the material. Mu and coworkers prepared tetradecanol/graphene aerogel form stable composite by physical absorption. Vitamin C and ethylenediamine are used to enhance the thermal conductivity and the material displayed outstanding thermal energy storage capacity.

It was seen that thermal conductivity improved with the increase of graphene aerogel content (Mu and Li 2018).

Conclusion

Aerogels and hydrogels possess high surface area that they are exploited for catalysis, water and effluent treatment, and other fields. The different techniques for the synthesis of aerogels and hydrogels are discussed in this chapter. The synthesized materials can be either composite materials, hybrids, or reinforced materials. CNTs owing to their good intrinsic properties can contribute much in the sensors and actuators, biomedical materials, and in drug delivery. CNT-based hydrogels and aerogels act as smart materials, which respond towards the stimuli such as pH, temperature, light, etc. Thus the CNTs find applications in the thermo-responsive materials and pH-sensitive materials. The properties of these materials depend on the amount of CNT incorporated.

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