REVIEW

Preparation of aerogel and its application progress in coatings: a mini overview

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Abstract

Aerogels are predominantly mesoporous, extremely lightweight, low density ($\sim$ 0.003 g/cm^3) and thermally insulating materials. Over the years, aerogels have gained increasing attention due to their extraordinary properties (light, heat, sound, electricity and force) and application potentials in varieties of fields. Several studies have been carried out regarding aerogel preparation and its applications in coatings on different substrates. In this review, an overview of aerogels preparation and their application progress in coatings of most common substrates is presented. Attention is paid to aerogel coatings of textiles, leather, and substrates other than leather and textiles for special functionalities that could address the application progress in coatings. This review will help to inspire scientists and engineers towards novel aerogel materials and technologies to boost the industrial fabrication of flexible advanced materials.

Keywords: Aerogel, Hydrogel, Sol–gel technique, Aerogel coatings, Silica aerogel

1 Introduction

Aerogels are the special materials with porosity, extremely lightweight, low-density and thermal insulation [1, 2]. They are predominantly mesoporous solids [3]. Aerogels are generally known as solid air, solid smoke, frozen smoke, or blue smoke [4]. Based on IUPAC, the aerogels are defined as the gels that comprise a microporous solid in which the dispersed phase is gas [5]. Also, the aerogels can be defined as the gels in which the liquid has been replaced by air, while the solid network shrinks very moderately [6]. An aerogel is prepared using a sol–gel method which generally involves the conversion of a liquid system (sol) into a gel phase through chemical reactions. After a special drying, the solvent will be removed from the gel structure, followed by filling the network with air, as such the dried product is called aerogel. As is well known, silica, alumina and carbon, are the main sources for fabricating the aerogels, especially when the synthetic polymers, biopolymers, and other organic precursors are emerging sources. Low thermal conductivity is the typical feature of aerogel investigated most widely, which has been prominently used in high-performance thermal insulation. For instance, silica aerogel is a super thermal insulator in possession of a thermal conductivity lower to 12 mW/(mK). Compared with conventional insulation, this silica aerogel had an obviously lower thermal conductivity due to their small pore size that could limit the gas phase conduction [7].

Aerogels can also be used in heavy metal removal, gas absorption, radioactive waste confinement [8–10], drug delivery [11], sensors [12], nuclear particle detection (Cherenkov) [13], optics (light guides) [14, 15], Knudsen pump [16], electronic devices (solar cells, capacitors) [17, 18], catalysis [19, 20] and surface coatings (self-cleaning coatings, chemical resistant coating, etc.) [21, 22]. However, aerogels also have some disadvantages, such as fragility, rigidity, dust formation, bulkiness, et al., which limit their extensive applications. Synthesizing fiber-reinforced aerogel composites is one of the most effective methods to reduce the fragility of the aerogels [23]. However, the

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fiber-reinforced aerogel composite has a major problem that is dust-releasing from the fiber assemblies [24]. The coating of aerogels on different substrates is among the solutions to rectify the aforementioned problem. The surface coatings of aerogels on different substrates can be carried out by a sol–gel coating technique. The sol–gel method produces a thin film coating of nanoparticle clusters or nanoporous aerogels/xerogels on a substrate in a single coating step [25]. In the process, the sol is coated on a substrate and the dispersing medium constituting the sol is evaporated to form the gel layer. Through the sol–gel strategy, several researchers have synthesized silica aerogels as functional agents to coat on fibers and fabrics with multifunctionality including super-thermal insulation, super-hydrophobicity, UV radiation protection, color-fastness of materials as well as antimicrobial preservation [26, 27]. Other aerogel coating approaches mainly include the dipping, spraying and spinning coatings as well as the bar and flow coatings.

In this work, we attempt to review the general preparation methods and properties of aerogels. Their application progress in coatings are well summarized with focusing more on the flexible substrates (textile and leather) on account of the rapid development of flexible wearable devices.

2　Preparation of aerogels

As mentioned before, an aerogel is generally synthesized via a process called sol–gel process. The synthesis process of aerogels is fairly common between almost all types of aerogels. The sol–gel synthesis procedure encompasses three vital steps: (1) preparing the gel; (2) aging the gel; (3) drying the gel. The general route is illustrated in Fig. 1.

2.1　Preparing the gel

The gel phase is commonly obtained by the sol–gel method. In the process, the required precursor is dispersed in a solvent first and then a catalyst is introduced to facilitate the gel formation. For silica gel preparation, for example, the precursor tetramethoxysilane (TMOS) is dissolved in ethanol/water (4:1) mixture and then 0.1 M HCL is added to catalyze the gelation [29]. Generally, the gels are classified based on the dispersion medium, as examples, water for hydrogel or aquagel, alcohol for alcogel and air for aerogel.

2.2　Aging the gel

The obtained gel in the first step will be further aged in its mother liquor. During the aging process, two different mechanisms, including Neck growth and Ostwald ripening, will operate simultaneously to influence the structure and features of a gel at different rates. During the process, the material will be transported to the neck
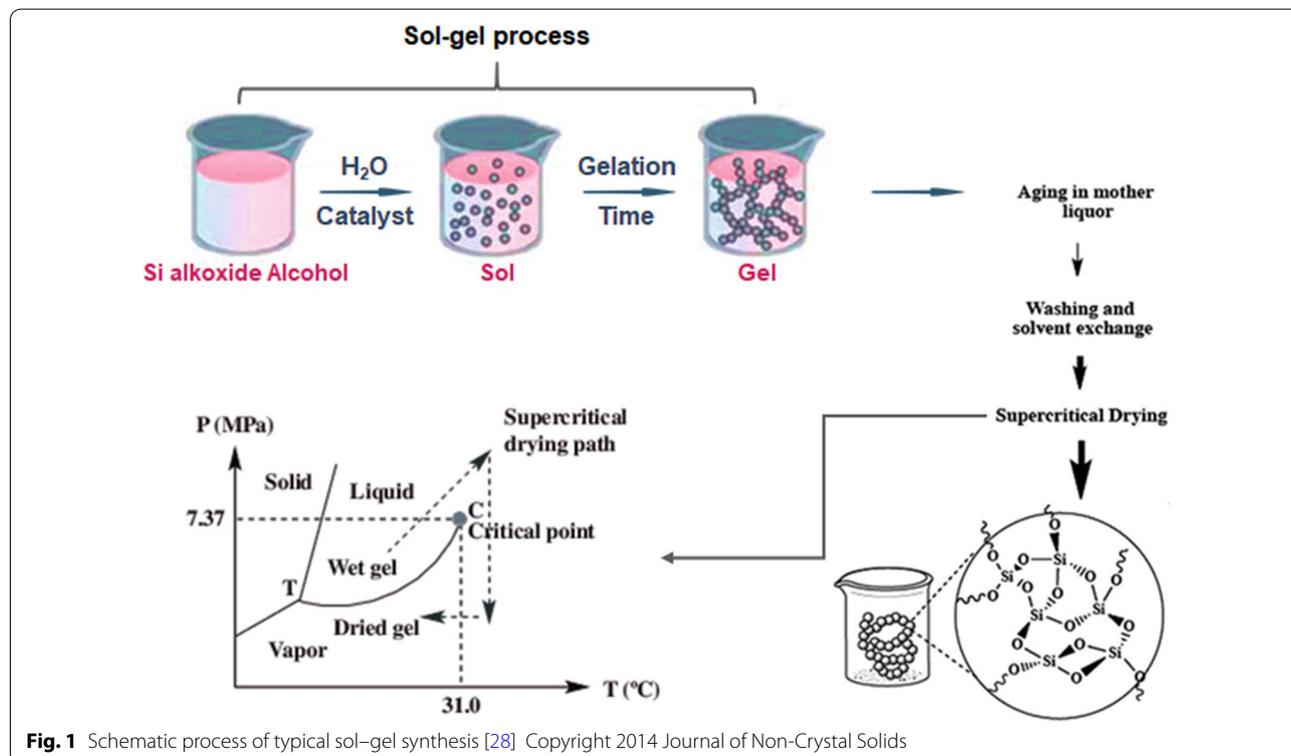
Fig. 1 Schematic process of typical sol–gel synthesis [28] Copyright 2014 Journal of Non-Crystal Solids

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region where the colloidal particles gives a firmer network. The difference in solubility (*S*) for surfaces with different curvatures (*r*) drives the transportation of material, which can be represented by the Kelvin Equation (Eq. (1)) [30].

$$S = S_0 \exp\left(\frac{2\gamma_{sl} V_m}{RTr}\right) \tag{1}$$

where S_0 is the solubility of a flat surface of the solid phase, V_m is the molar volume of the solid, γ_{sl} is the energy of solid–liquid interface, T is the temperature and R is the ideal gas constant. The neck region between particles has a negative curvature ($r < 0$) and low-solubility. The materials will be transported from the concave surface of a particle, followed by their agglomeration in these convex areas. Owing to larger solubility of the smaller particles, the driving force will also act to dissolve the smallest particles, and then the precipitation onto larger particles will further take place.

Aging action endows the wet gel with higher strength and stiffness, and it is of vital importance to prevent the gels from pore collapsing under the strong capillary forces that occur during drying such as ambient pressure drying (APD). Several works have shown that the aging of the gel for prolonged periods (e.g., multiple days or even weeks) makes the gel more rigid, and maximally prevents the aerogels from shrinking during the supercritical drying [31, 32].

The effect of aging conditions used during the preparation of silica aerogel has also been investigated in several studies [33–36]. The studies showed that reinforcing wet-silica gels via a solvent exchange into a silane precursor containing TEOS or TMOS, and a similar process to the surface modification step, could prevent the aerogels from cracking during the supercritical drying process [33, 35, 37].

Suh et al. studied the effect of aging process on the microstructure of ammonium hydroxide and NH_4F-catalyzed gels [38]. Results showed that the structure could be reorganized via the aging process, endowing the resultant aerogels with a single-peaked pore size distribution, an increase in the pore volume and mean pore diameters as well as the virtually unaffected surface areas.

During the aging of gel, it is customary that the solvent evaporation could make the network slightly shrink before completing the aging process. Nevertheless, this little shrinkage can be avoided by using the solvents with a low vapor pressure. Besides, ionic liquids can also be used for this purpose [39], because they are a class of solvents that have extremely low vapor pressure and a variety of solvent properties. Consequently, they can provide appealing approaches to make the gel network do not shrink under prolonged gelation time. As a result, the as-prepared gels are supposed to be more stable, even conventional drying will cause a little shrinkage.

2.3 Drying the gel

As the critical step, drying the gel is governed and driven by the capillary pressure (P_c), and it can be displayed by Eq. (2).

$$P_c = \frac{-\gamma lv}{\gamma p - \delta} \tag{2}$$

where γlv is the surface tension of the pore liquid, γp is the pore radius, δ is the thickness of a surface adsorbed layer.

$$\gamma p = \frac{2V_p}{S_p} \tag{3}$$

where V_p and S_p are pore volume and surface area, respectively.

Several drying techniques have been developed in the past decades in search of a well-defined 3-D networked aerogel structure. Among which the most common techniques include lyophilization, ambient pressure drying (APD), and supercritical drying (SCD).

(1) In the lyophilization, the phase boundary between the gas phase and liquid does not exist and thus the P_c does not play a vital role. The solvent must be interchanged using a low expansion coefficient and a high sublimation pressure, usually by freezing and subliming under vacuum.

(2) In the APD, the surface tension between vapor and liquid is unavoidable. The stress in the gel is directly proportional to the drying rate and the pore liquid viscosity, whereas it is inversely proportional to the permeability of the wet gel.

(3) In the SCD, the liquid in the pore is extracted above the critical temperature (T_{cr}) and critical pressure (P_{cr}) of the concerned liquid. In this case, no liquid–vapor interface and P_c exist [30].

In the preparation of aerogels, depending on the primary precursor used to fabricate the aerogel, the conventional sol–gel technique can be a one-step, two-step or multi-step process. This can be observed in the preparation of the common aerogels.
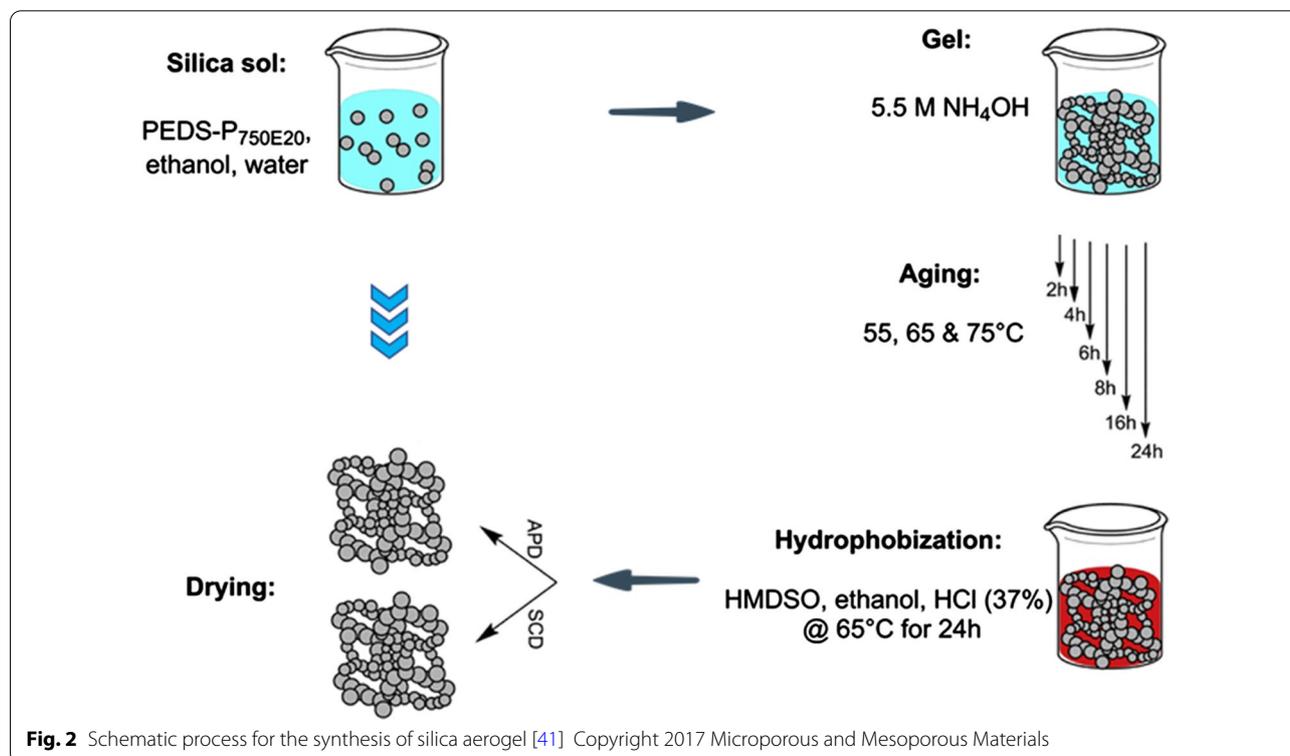
3 Common aerogels

3.1 Silica-based aerogels

Since the invention of the sol–gel process by Kistler, silica-based aerogels (SBA) are the first inorganic aerogels known to the world. SBA are a kind of materials with unique features, e.g., high porosity (80–99.8%) and specific surface area (500–1200 m^2/g), low density (about 0.003–0.5 g/cm^3) and thermal conductivity (0.005–0.1 W/mK) as well as ultra-low dielectric constant (k = 1.0–2.0) and low index of refraction (about 1.05) [7]. More than 96% of the SBA matrix is occupied by air. The remaining portion is the wispy matrix of SiO$_2$ [40]. Up to now, the synthesis of SBA has been accomplished mainly through the sol–gel method with the controlled condensation of small colloidal particles in an alcohol aqueous solution, followed by a supercritical drying process. The synthesis procedure generally takes three steps: (1) gel preparation; (2) aging; (3) drying (as seen in Fig. 2).

As reported, the most widely used precursors in the preparation of SBA are silicon alkoxides that can be acquired with a high purity. With diversified properties and functionalities, the precursors for preparing SBA, such as tetramethoxysilane (TMOS), tetraethoxysilane (TEOS), polyethoxydisiloxane (PEDS), methyltriethoxysilane (MTES), methyltrimethoxysilane (MTMS) and their mixtures, have been extensively investigated by various research teams [42]. SBA can be synthesized into multiple forms, for instance, monoliths, grains, powders, and films, conforming to their desired applications.

3.2 Polymer-based aerogels

Unlike SBA being fragile and hygroscopic, polymer-based aerogels (PBAs) usually perform higher stabilities for various applications, especially in extreme environments. PBAs typically include cellulose-based, resin-based, polyimide-based and poly(vinyl alcohol)-based aerogels. These diversified PBAs can exhibit differentiated structures and properties. Affected by the type of polymer and the preparation conditions, the structure of PBAs can vary from colloidal nanoparticles to nanofibril/microfibril networks and even sheet-like skeletons. Besides, the structure parameters (including order, pore size and shape) will greatly influence the final macroscopic performance of PBAs. The synthesis of most PBAs generally takes a two-step process. Mekonnen, et al. reported a synthesis procedure of electrically conducting collagen/polypyrrole (C/PPy) hybrid aerogels through in-situ polymerization of pyrrole and the fibrous protein collagen, followed by lyophilization [43]. The as-prepared C/PPy aerogels were considerably light with calculated apparent density ranged from 25 to 135 mg cm^{-3} via raising the dosage of PPy. Compared with the biopolymer-based aerogels, the C/PPy aerogels had comparable density (20–40 mg/cm^3), which was fairly lower than the

Fig. 2 Schematic process for the synthesis of silica aerogel [41] Copyright 2017 Microporous and Mesoporous Materials

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conventional aerogels derived from carbon (100–800 mg/cm^3) or graphene (60–150 mg/cm^3) [44, 45]. As confirmed by the scanning electron microscopy (SEM) analysis, the pore size in the as-synthesized hybrid aerogel network decreased as the concentration of the PPy polymer increased, which leaded the hybrid composite to be denser, fragile, and loses its aerogel property (as seen in Fig. 3a–f).

Zhou et al. proposed a synthesis method of polyaniline (PANI)-decorated cellulose aerogel nanocomposite with enhanced photocatalytic activity [20]. The author reported a facile method that was using ionic liquid as the solvent to prepare cellulose aerogels decorated with PANI through a dissolution/regeneration route. In this system, the cellulose aerogel acted as a scaffold material for PANI nanoparticles to avoid their aggregations. The synthesized PANI/cellulose aerogel could perform outstanding photocatalytic activity to degrade methylene blue. It was suggested that this strategy could be employed to fabricate various nanofibrillated cellulose-supported nanocomposites for catalysis and other potential applications via utilizing the renewable natural resources.

In addition to the above-mentioned polymeric aerogels, other powdered aerogel particles with a size ranging from submicrometer to several hundred micrometers could also be fabricated using the sol–gel method from polymeric precursors [46, 47]. The most widely investigated materials were the resorcinol–formaldehyde and melamine–formaldehyde based aerogels. Through the polycondensation between resorcinol or melamine and formaldehyde in a weakly alkaline aqueous solution usually catalyzed by NaOH or NaHCO$_3$, these aerogels could be successfully prepared [46, 47]. Generally, these aerogels were nontransparent and possessed a specific surface area of approximately 300 m^2/g and a volume density of 0.24 g/cm^3 [48, 49].

3.3 Carbon aerogels

Carbon aerogels (CAs) are generally prepared via carbonizing the polymeric aerogels, which are prepared by the sol–gel route from the polycondensation of different organic monomers. They are considered to be a class of attractive functional materials for high temperature insulation in non-oxidizing atmospheres or vacuum [50]. The most widely studied polymeric aerogels for carbonization are those fabricated from resorcinol and formaldehyde aqueous mixtures containing a basic or acid catalyst [51, 52]. In general, four main stages can be classified in the synthesis of CAs: (1) preparing the sol mixture and further gelling; (2) curing the gel; (3) Drying the wet gel; (4) carbonizing the dried gel and sometimes further activating the carbonized derivative.

CAs are open, porous solids consisting of a three-dimensional network of the spherical, interconnected primary particles. By varying the synthesis conditions, the average pore diameter and particle size can be
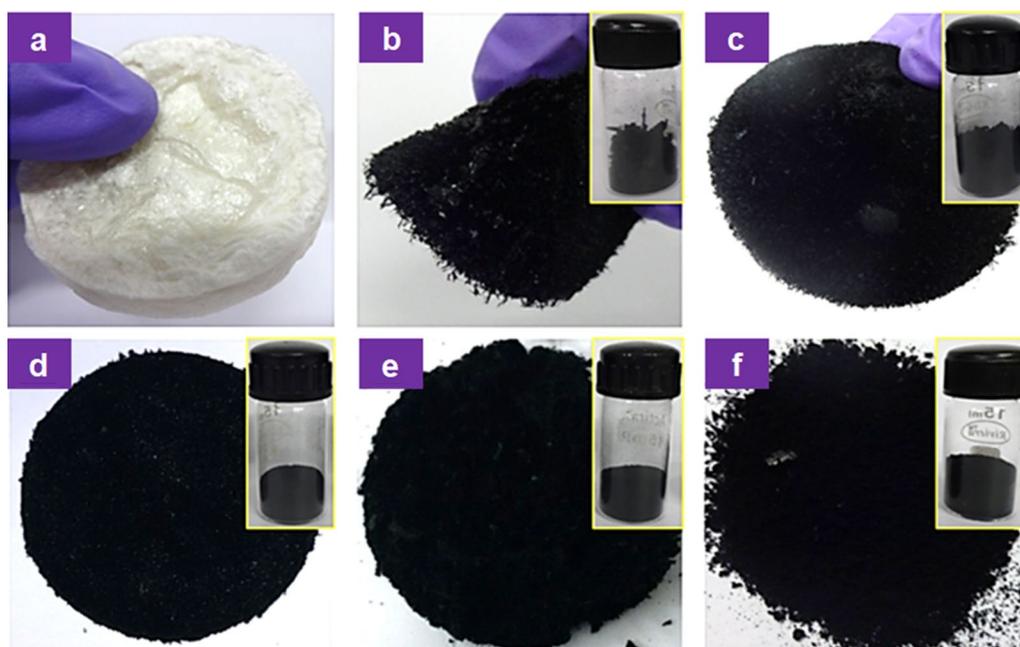
Fig. 3 Illustrations of C/PPy aerogels: **a** 100/0; **b** 100/25; **c** 100/50; **d** 100/75; **e** 100/100 and **f** 0/100. Insets showed the status of aerogels after physically grinding in a mortar and pestle [43] Copyright 2016 RSC Advances

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specifically regulated in the range of several nanometers to several micrometers, and a porosity of up to 99% for CAs can be acquired, thus leading CAs to matching the thermal insulation applications, especially at high temperatures, and also for catalysis, wearable devices, and electrodes in supercapacitors [51–55].

Other than carbonizing polymeric aerogels, direct synthesis of carbon aerogels from graphite (graphene oxide, GO) has been reported in recent years [56–58]. Tao et al. reported a simple one-step method of synthesizing a three-dimensional graphene aerogel from graphene oxide [19]. The GO was allowed to be reduced into rGO with Cu nanoparticles as a reductant and self-assembled to a graphene hydrogel and lyophilized to obtain highly porous three-dimensional graphene aerogel that is promising for various applications such as photocatalytic degradation of different dyes and oil removal.
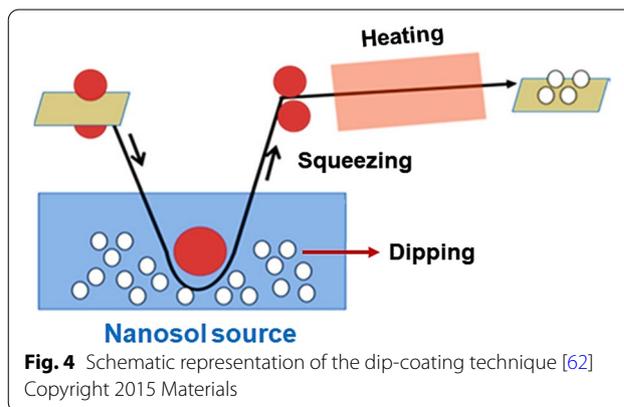
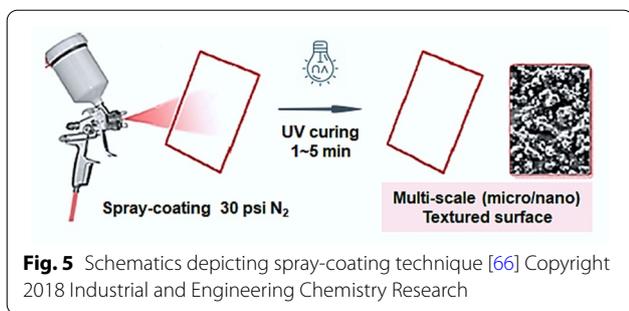## 4 Application progress in coatings

According to literature reports, aerogels can be used to develop diverse new high-performance products, such as jackets used to protect human body from extreme cold weather, constituents of space suits, specialized buildings, insulations for pipe and sound as well as heat-insulated blankets and aerogel-based textile composites, etc. Aerogel coatings on different substrates for different applications are quite common in recent years. The coating of aerogel on different substrates is conducted by a sol–gel process including techniques such as dip-coating, spin-coating, spray-coating, etc.

(1) Sol–gel coating process is a well-recognized method to prepare the gels and nanoparticles [59, 60]. When the sol is coated on a substrate, the dispersing medium constituting the sol is evaporated to form the gel layer. Many researchers have applied silica aerogel coatings onto the fibers and fabrics as functional agents through sol–gel method [26, 27]. In fabricating hydrophobic surface coatings with the sol–gel method, the surface roughness obtained can be easily regulated by varying the method conditions and the formula of the reactants. The sol–gel coating process to construct superhydrophobic surfaces has been studied widely in the last few decades. For example, a transparent and self-cleaning superhydrophobic coating can be fabricated using a very simple one-step sol–gel approach via the processing of long-chain fluoroalkylsilane [60]. This coating layer could exhibit a rough, wrinkled, hill-like surface morphology. Besides, the as-prepared coating could not only have super-hydrophobicity, but also show outstanding self-cleaning property. After the attack of a high-speed water jet, the

superhydrophobic wetting state could be also well reserved.

(2) Dip-coating is the most commonly used and versatile approach to fabricate super-hydrophobic coatings on different textile substrates with a layer of hydrophobic micro-nano materials, i.e. titanium dioxide, silicon dioxide and zinc oxide [61]. It usually needs three key processing steps, including dipping in the coating slurry, drying and curing. Generally, the coating slurry is composed of: (1) nano/micro-scale dual-scale particles that can increase the roughness of the coating layer; (2) organic solvent that can wet the fabric and disperse the particles; (3) polymer that can particularly improve the bonding strength of the coating layer. Sometimes, the coating slurry may also contain a hydrophobic agent, such as fluorocarbon silane that can reduce the surface energy of the coating layer. Due to the toughness of the polymer binder and the layered roughness of the stable particle coating, some ultra-wet resistant fabrics made by the dip-coating process can perform outstanding mechanical durability. The schematic diagram illustrated in Fig. 4 shows the typical dip-coating process.

(3) Spray-coating is a simple, rapid and universal way to construct a rough multi-scale hierarchal structure, and this technique can be used in coating a layer of low surface energy polymer on any substrate [63, 64]. For example, the superhydrophobic or superoleophilic epoxy/attapulgite nanocomposite coatings on the stainless steel meshes could be constructed via a simple spray-coating process [65]. The coated mesh was also proved to maintain a highly superhydrophobic property after treating in a variety of harsh conditions, such as high temperature, mechanical scratch, corrosive substance, and humid atmospheres. A simple and typical spray-coating scheme is illustrated in Fig. 5

Fig. 4 Schematic representation of the dip-coating technique [62] Copyright 2015 Materials

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Fig. 5 Schematics depicting spray-coating technique [66] Copyright 2018 Industrial and Engineering Chemistry Research
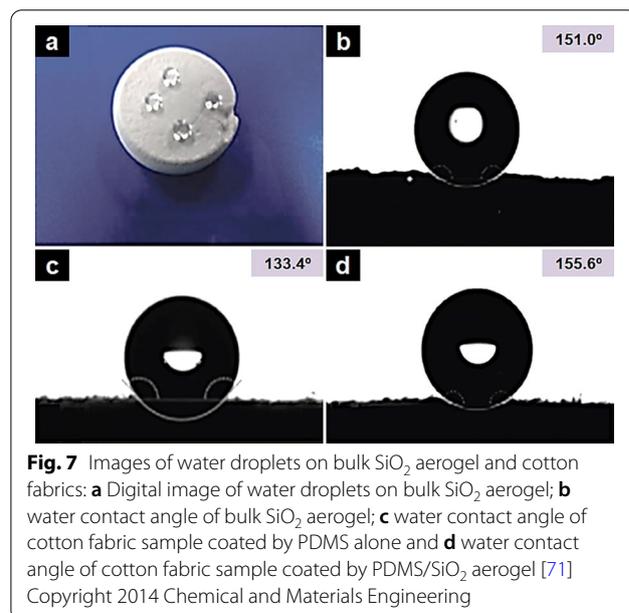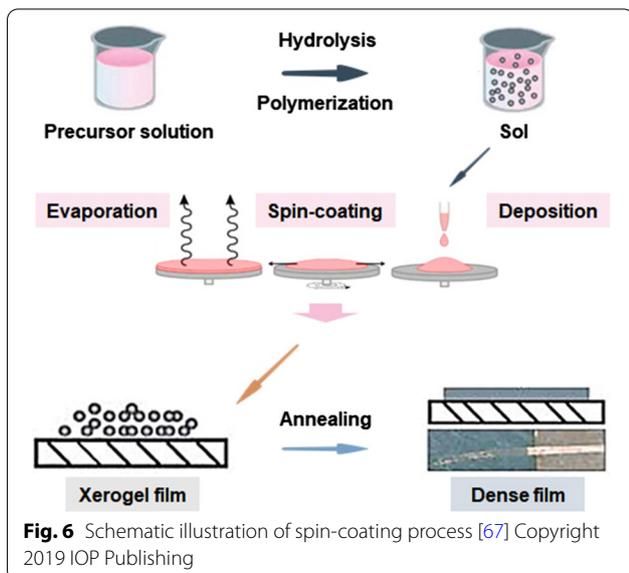
(4) Spin-coating is the simplest technique to construct coating layers on a substrate. The spin-coating can be used in the deposition of sol-gels on different substrates. Firstly, a gelatinous network on the substrate surface can be formed using this technique. Subsequently, a solid film can be obtained by removing the solvent from the gel. The typical spin-coating process on the substrate surface is illustrated in Fig. 6.

4.1 Aerogel coatings on textiles

Aerogels can be used for developing novel anti-wetting, self-cleaning and thermal insulating coatings on textiles. For instance, the silica biopolymer coatings can impart a high hydrophobicity degree to the fabric or impart flame retardancy to the fabric. In practice, most oxide-based aerogel coatings have poor durability due to their extreme brittleness, and adhesive or binder are often used to overcome these issues. However, the coating process with polymeric dispersions containing

aerogel often results in the aerogel fragmentation and loss of insulation properties due to excessive pore wetting. As a result, the versatile functionality of oxide-based inorganic aerogel coatings on textiles is generally limited by the poor durability [68]. This has been the main issue and challenge in aerogel coating. Some reports show that constructing the hybrid organic–inorganic aerogel composite coatings is an effective way to enhance the coating durability [69, 70]. Thus, in recent years, several studies have been conducted on the coating of aerogels on fabrics to induce hydrophobicity through different coating techniques. For instance, a superhydrophobic cotton fabric with a water contact angle of $155.6 \pm 0.9°$ for a 5 µL water droplet could be prepared via a dip-coating process using the PDMS/SiO$_2$ aerogel composite solution [71]. Briefly, the authors prepared the PDMS/SiO$_2$ aerogel dip-coating solution by ultrasonically dispersing SiO$_2$ aerogel powder (4%), PDMS (4%) and curing agent (PDMS/curing agent weight ratio of 10:1) in isopropyl alcohol for 30 min with an ultrasonic frequency of 50 kHz and ultrasonic power of 130 W. The raw cotton fabrics were immersed in the as-prepared PDMS/SiO$_2$ aerogel coating solution for 30 min and then compressing using an automatic padder with a nip pressure of 2 kg/cm^2 and finally dried at 80 °C for 5 min and cured at 160 °C for 60 min in an oven. The excellent hydrophobicity of the coated fabric could be attributed to the combination of SiO$_2$ aerogel particles with porous rough microstructure, high specific surface area and PDMS adhesive layer with low surface energy. As a confirmation, the

Fig. 6 Schematic illustration of spin-coating process [67] Copyright 2019 IOP Publishing

Fig. 7 Images of water droplets on bulk SiO$_2$ aerogel and cotton fabrics: **a** Digital image of water droplets on bulk SiO$_2$ aerogel; **b** water contact angle of bulk SiO$_2$ aerogel; **c** water contact angle of cotton fabric sample coated by PDMS alone and **d** water contact angle of cotton fabric sample coated by PDMS/SiO$_2$ aerogel [71] Copyright 2014 Chemical and Materials Engineering

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authors made a comparison of the ability to induce hydrophobicity on the fabric surface with PDMS and SiO$_2$ alone and in combination (as seen in Fig. 7). The cotton fabric coated by PDMS alone (as seen in Fig. 7c) showed a water contact angle of $133.4 \pm 0.6°$, indicating the difficulty in achieving superhydrophobicity only by reducing the surface tension without specific surface topography. Whereas, the cotton fabric sample coated by PDMS/SiO$_2$ composite aerogel displayed excellent superhydrophobicity with a water contact angle of $155.6 \pm 0.9°$ in (as seen in Fig. 7d). The effect of PDMS/SiO$_2$ aerogel coating treatment on the mechanical strength properties of the cotton fabrics was negligible. This simple approach may pave the potential way for practical applications. In addition to the aerogel composites prepared by using the organic binder, some inorganic substrates, such as attapulgite, ceramic fibers, glass fiber, carbon et al., could also be used to reinforce the silica aerogel structure [72], and the resultant aerogel composites could be used in the fabrication of protective clothing, endowing it with high-temperature stability and excellent thermal insulation property [73, 74]. However, the use of inorganic aerogels to construct promising flexible coatings while reserving their natural characteristics still requires in-depth research.

Multiple special functionalities can be induced to textiles through sol–gel spin coating of aerogel nanoparticles [75]. In this regard, Shaban, et al. reported the sol–gel spin coating of cotton fibers with ZnO aerogel nanoparticles to induce both self-cleaning property and photocatalytic removal of Methyl Orange dye [75]. As the authors briefed, zinc acetate dihydrate and monoethanolamine (in 1:1 molar ratio) in 2-methoxyethanol were stirred at 60 °C for 2 h to prepare the ZnO nanoparticles sol. Then the as-prepared ZnO sol was aged for 24 h and spin-coated onto cotton fibers at 1100 rpm for 60 s; finally dried at 50 °C for 30 min. The coating process was repeated for 10 times and finally annealed in a furnace at 150 °C in air for 2 h. As per the report, the ZnO-coated cotton showed self-cleaning property against methyl orange dye and decomposed the dye by 73% and 30.7% under the sunlight and lamp illumination, respectively.

Recent years, there are few studies involving the use of aerogels to coat wool-aramid blended fabrics to provide thermo-physiological comfort of protective clothing for firefighters [76]. The silica gel composite reinforced by aramid fibers with high thermal insulation property have been successfully fabricated. Its thermal conductivity was 22 mW/mK and its fiber content was 1.5–6.6% [77]. Furthermore, it has been reported that a doubling of the thermal insulation could be observed in silicon dioxide and fabric composite based on polyurethane [78]. The method of needle-free electro-spinning and

electro-spraying of polymer mixture onto textiles has also been tried to overcome the difficulties associated with other coating methods on textile fabric substrate [79].

Farzaneh et al. reported a hydrophobic and thermal insulating polyester woven fabric using an electro-spraying coating of nanoporous silica powder [80]. As reported, a scoured polyester fabric could be coated using a mixture containing silica aerogel (3%, w/v) in RUCO-COAT FC 9000 (20%, w/v) via an electro-spraying device (as seen in Fig. 8). The coating mixture was prepared by adding silica aerogel powder and a small amount of emulsifier (for better dispersion) into the aqueous solution of the fluorocarbon material and stirring for 12 h using a magnetic stirrer. The parameters of electro-spraying process mainly include the voltage, flow rate, and distance from the needle tip to the collector, which could be set as 17 kV, 0.85 mL/h, and 5 cm, respectively. A needle with 0.6 mm inner diameter was used for the electro-spraying coating of the sample fabrics. Finally, the coated fabrics were dried at 100 °C for 5 min and then cured at 170 °C in a lab dryer.

In light of the reported results, it was observed that the reduction in heat transfer from 71.65% of the unsprayed fabric to 38.35% and 30.99% of the electro-sprayed fabric required 16 and 24 h, respectively. A similar change trend for air permeability was also observed. The samples with higher content of aerogel had lower air permeability. Moreover, the presence of aerogel particles could be in favor of improving the hydrophobicity of electro-sprayed samples. After being electro-sprayed with the aerogel/fluorocarbon mixture for 24 h, the samples exhibited the highest hydrophobicity with a contact angle of 152.2°. In addition, after the abrasion test, the weight loss of the sprayed sample was negligibly less than 5%, which resulted in low dust-removal performance of the aerogel particles from the fiber surface.

As previously reported, thermo-physiological comfort was related to the thermal properties of the fabric, moisture permeability, sweat absorption and drying capacity
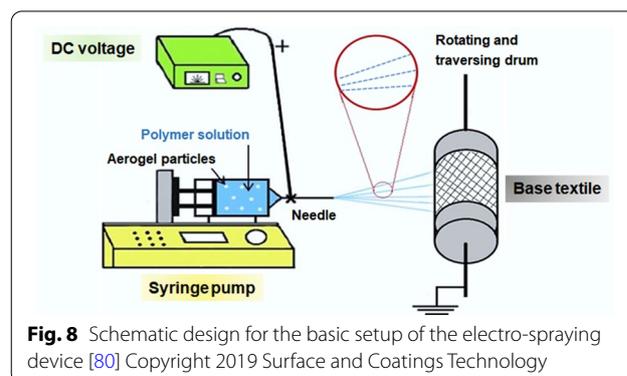
Fig. 8 Schematic design for the basic setup of the electro-spraying device [80] Copyright 2019 Surface and Coatings Technology

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[81]. These characteristics are affected by many main parameters, such as molecular structure, fiber geometry, fabric structure, cross-section, pore-distribution, channels, surface tension, thickness, density, etc. [82, 83]. Similarly, the functionality and durability of coated fabric are also facilitated by the finishing treatments [84]. Many researchers have studied aerogel coatings on fabrics to inherit thermo-physiological comfort through different coating techniques including the famous sol–gel method [26, 27].

Jabbari, et al. reported the fabrication of a kind of polyester woven fabric coated by a novel lightweight and highly thermal insulative silica aerogel-doped poly(vinyl chloride) using blade-coating method [85]. As per the report, the coating solution was prepared by mixing different proportions (0, 2, 3 and 4 wt.%) of silica aerogel with PVC using a mechanical mixer having 3-blade propellers at different speeds. The as-prepared homogenous coating solution was coated on both sides of the polyester fabric with a laboratory-scale blade-coating method. Finally, both sides (first side and second side) were cured at 180 °C and 190 °C for 2.5 min and 1.5 min, respectively. A general illustration of the blade-coating technique is shown in Fig. 9. The report showed that thermal insulation performances were improved by 26% (from 205 to 152 mW/mK) compared with the unmodified coated fabric. The report investigated the dependency of the thermal insulation characteristics of the coating on the percentage of the aerogel content. For this purpose, four proportions of PVC/aerogel composite coatings such as 0, 2, 3 and 4 wt.% were prepared and coated on the fabric. The analyses proved that composite coating with 4 wt.% of PVA/aerogel showed the best heat-insulating property. According to the report, the dosage of 4 wt.% was the critical percentage. Besides, when the aerogel content exceeded 4 wt.%, the preparation of composites would be limited due to the high viscosity. The authors suggested the potential applications of the as-prepared coated fabric in developing textile bioreactors for the production of ethanol/biogas based on waste materials, tents, exterior wall coverings, temporary houses, tarpaulins and container linings, etc.

In high heat protection clothing, e.g., firefighter's protective clothing (FFPC), aerogel coatings can both resist the incoming heat fluxes and block the dissipation of heat from the body. Therefore, the body temperature of the wearer will increase. Shaid, et al. [87] attempted to rectify the situation by introducing phase change materials (PCMs) into the aerogel coating. The work investigated the simultaneously coating of facial clothing linings using aerogel and PCM for firefighters' protective clothing. The ambient side of the thermal interlining was coated using silica aerogel particles. Meanwhile, the PCM/aerogel composite powder was coated on the side close to the skin. The new thermal insulation lining had excellent thermal insulation protection and comfort. It prolonged the time to reach the pain threshold and increased the time for pain prompts. When the composite powder was heated to a temperature three times higher than the melting temperature of pure PCM, no dripping or deformation was observed.

The thermal insulation characteristics of the aerogel coated fabrics depend on the aerogel microstructure and concentration on one hand and the fabric weave or knit structure on the other [88, 89]. Rosace, et al. investigated the impacts of weave structures and silica aerogel coatings obtained by the sol–gel method on the thermal insulation performances of woven cotton textile fabrics [90]. To this end, three kinds of important cotton fabric weaving structures were selected, including plain weave, satin weave and pile, which had different yarn counts, threads per centimeter and mass values per square meter.
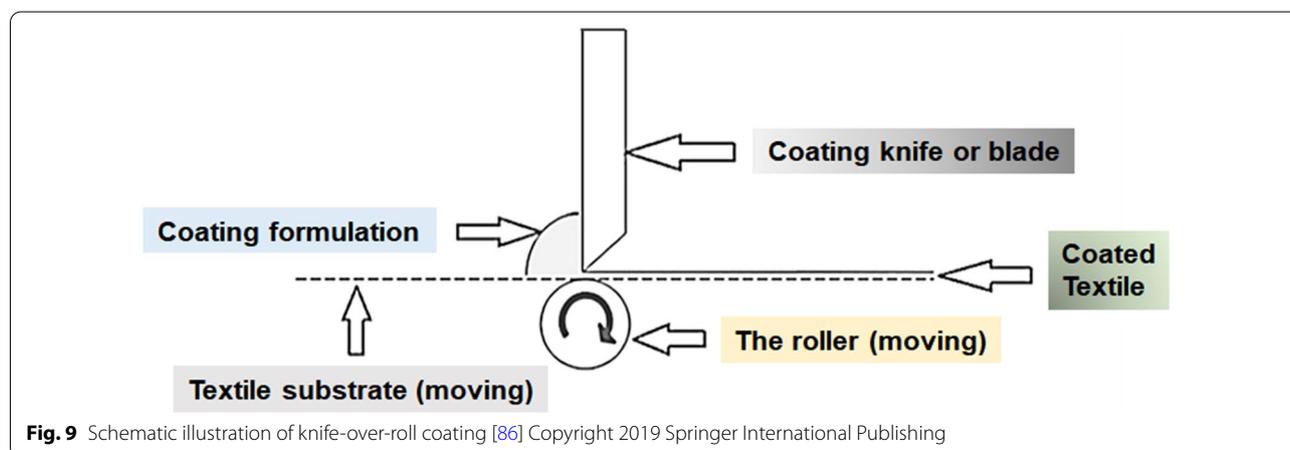
Fig. 9 Schematic illustration of knife-over-roll coating [86] Copyright 2019 Springer International Publishing
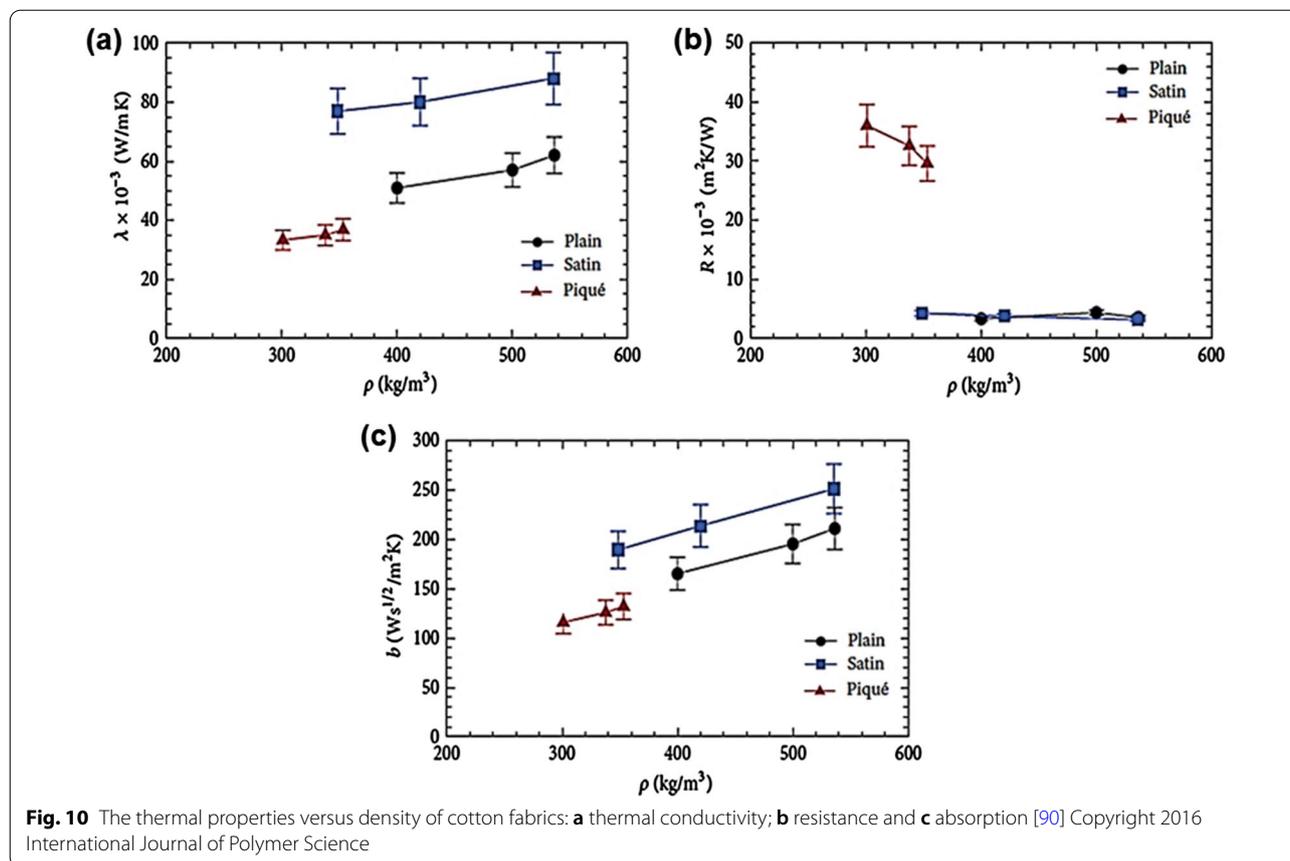
Fig. 10 The thermal properties versus density of cotton fabrics: **a** thermal conductivity; **b** resistance and **c** absorption [90] Copyright 2016 International Journal of Polymer Science

As it is observed from the thermal property versus density profile of the three weave structures in Fig. 10, the fabric weave and density strongly influenced the thermal properties as such: pique always showed the lowest value, while satin showed the highest value, and plain weave lied in between. Pique thermal resistance Ř values (as seen in Fig. 10b) decreased linearly with increasing the density, while the data for satin and plain weave fabrics actually remained the same and decreased by an order of magnitude. Therefore, the Pico cotton fabric had quite favorable thermal insulation performances, accounting for its higher thickness and weight, however, disadvantaged from its lower density, when compared to the plain and satin cotton fabrics. The thermal absorption Ь (as seen in Fig. 10c), on the other hand, increased from pique (warmer feeling) to plain weave up to satin (cooler feeling) as expected.

Imparting fire-retardant characteristics on fabrics is also a recent experience as many studies have been reported in recent years [91–94]. A good fire retardant (FR) fabric generally have the following features: (1) it will not ignite easily when exposed to flame or continue to burn after removing ignition source; (2) it will provide a barrier or an insulating layer on exposure to intense

heat; (3) it will not melt or shrink when exposed to high heat flux and have good anti-static property.

Halogen, phosphorus and phosphorus-nitrogen compounds as traditional flame retardants used for almost all the textiles, and metallic salts applicable to protein fibers [95, 96] are limited by the toxicity and bioaccumulation and environmental persistence [97]. Hence, the development of environmentally benign fire retardants is an urgent need. In the textile industry, heat-stable condensates of tannic acid and terephthaloyl chloride were used to construct environmentally friendly flame-retardant coatings on nylon 66 fabrics, and the fabrics obtained exhibited rapid self-extinguishing properties [98]. Aerogel coatings for flame retardancy applications are alternative, nano-sized objects that can be easily synthesized via a bottom-up approach such as the sol–gel process [91]. The modification of eco-friendly fire retardants with aerogels enhances the flame resistance of the coatings. The preparation of aerogel modified fire retardant coating consisting of montmorillonite, epoxy resin, and tannic acid is a recent experience in this regard [99].

Aerogel coatings are also promising in the preparation of protective clothing. Bhuiyan, et al. developed a novel protective fabric accompanied with a favorable

resistances to radiant heat and chemiosmosis of liquid [22]. The protective fabric can be tailored to achieve both barrier performance and thermal comfort by integrating the electrospun polyacrylonitrile (PAN)-silica aerogel nanofiber membrane with the needle punched viscose non-woven fabric. As demonstrated in Fig. 11, the PAN-Silica aerogel solution of different aerogel concentrations was electrospun coated on a viscose non-woven fabric. Next, another layer of non-woven material of similar thickness was used to cover the coated non-woven film. Under the temperature of 140 ℃ and the pressure of 6.0 kPa, the edge of the fabric was heat-set with an adhesive interlining for 3 min.

The incorporation (coating) of the extremely hydrophobic silica aerogel with exceptional porosity on to the naturally hydrophobic PAN non-woven fabric enhanced the overall hydrophobicity of the film to resist the penetration of chemicals with high surface tension (as seen in Fig. 12 b1, c1 and d1). When the liquid had a low surface tension, the chemicals were first absorbed on the surface of the fabric through the fibers, and then absorbed through the pores of the aerogel particles embedded in the film (as seen in Fig. 12 c2 and d2). Silica aerogels have pore sizes widely ranging from 5 to 100 nm. The interconnected network of open pores allows the chemicals to flow from one pore to another through limited restrictions, thereby being able to absorb liquids. Due to the adsorption of aerogel particles, the liquid chemicals are mainly dispersed on the surface of the film and cannot penetrate the nanofibers. More aerogel particles are adsorbed, the overall liquid adsorption capacity will be stronger, so more liquid will be absorbed, resulting in more chemicals being retained in the fabric. Finally, the protection against the penetration of liquid chemicals will be greatly improved.

The newly developed fabric also showed improved heat and moisture vapor transmission rates, and a higher evaporative cooling index indicated that the thermal comfort of the clothes was acceptable due to the reduced accumulation of sweat on the material. In addition, the proper air permeability and moisture management property suggested the diffusion of sweat vapor through the film, thereby affording good thermal comfort in the clothing-skin microenvironment.

In another study, Bhuiyan, et al. demonstrated PU-Silica aerogel coated cotton fabric having simultaneous thermal comfort and protective performance through a knife over roll coating method [100]. The protective properties, including the surface wettability and chemical resistance of the coated fabric, were evaluated based on the measured values of water contact angle, water repellency level and chemical resistance. Due to the increased hydrophobicity, PU-silica aerogel coated fabrics could exhibit higher water repellency rating. Similarly, after integrating porous aerogel particles, the chemical resistance was observed without a decrease in protective performance.

4.2 Aerogel coatings on leather
Until recently, little has been reported on the introduction of aerogels in the conventional leather finishing chemicals to impart special properties on the leather
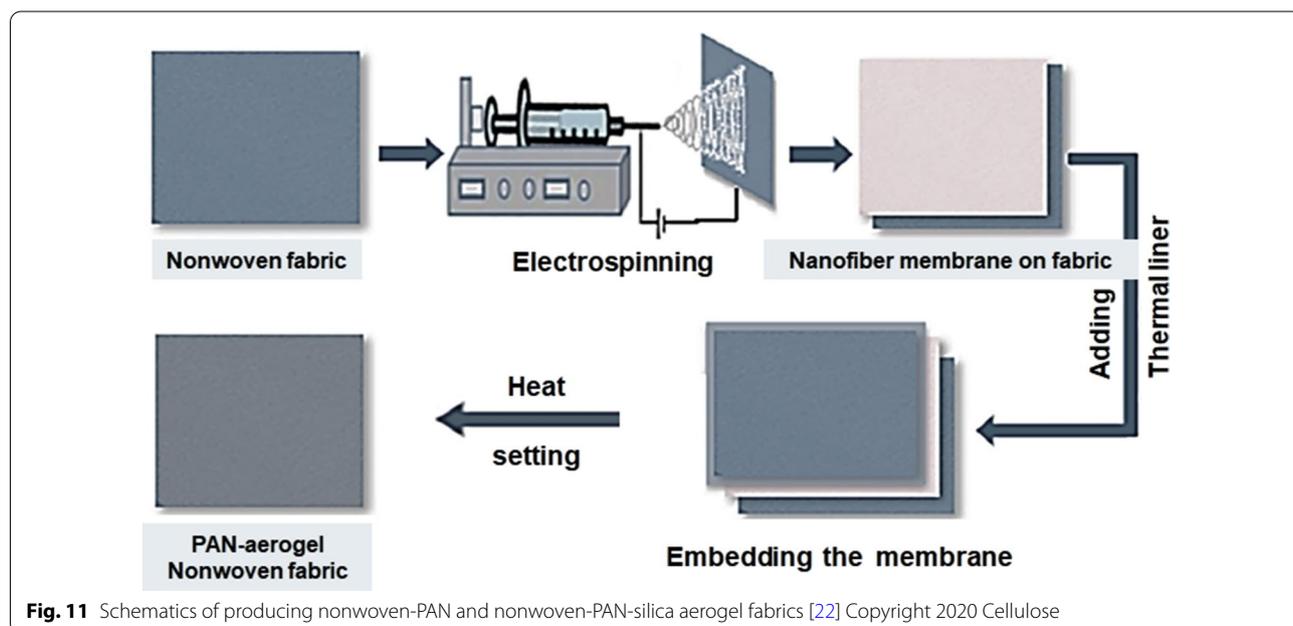
Fig. 11 Schematics of producing nonwoven-PAN and nonwoven-PAN-silica aerogel fabrics [22] Copyright 2020 Cellulose
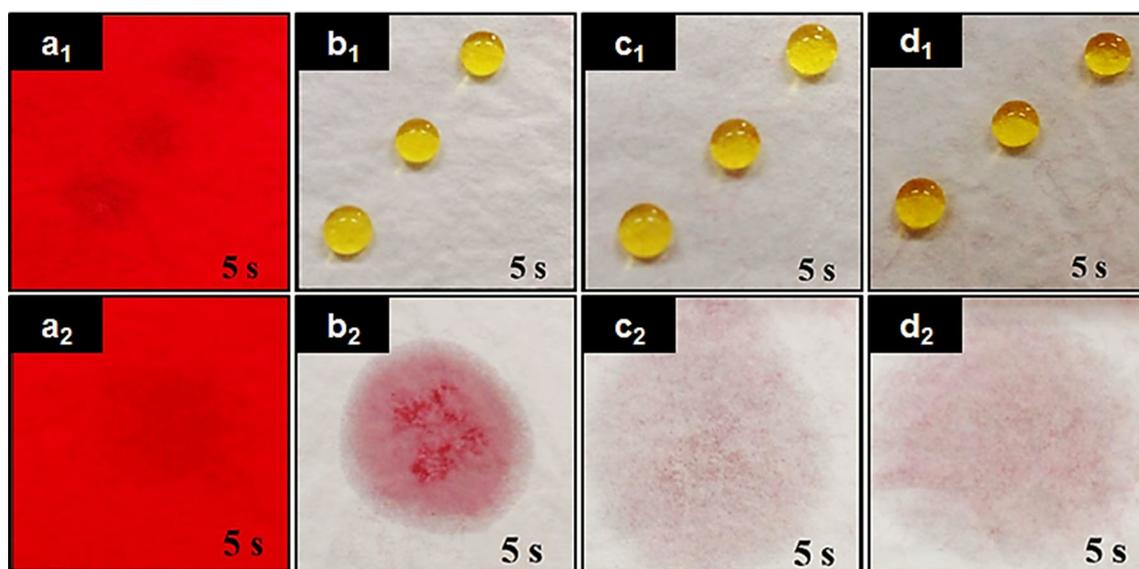
Fig. 12 Illustration of the chemical resistance of sample after removing the top layer fabric-control fabric **a** PAN-nonwoven; **b** PAN-aerogel nonwoven (2.0%); **c** and PAN-aerogel nonwoven (4.0%) fabrics (**d**). NaOH solution for high surface tension, represented by a_1, b_1, c_1, d_1, respectively. Methanol for low surface tension, represented by a_2, b_2, c_2, d_2, respectively [22] Copyright 2020 Cellulose

surface as has been tremendously reported on textile fabrics. Efforts to utilize the exceptional properties of aerogels by modifying acrylic resin in the preparation of exceptional leather finishing agents was made by Hu and associates [101, 102]. As the authors reported, the acrylic resin (AR)/nano-SiO$_2$ leather finishing agent could be prepared by physically blending AR and nano-SiO$_2$ sol. This nano-SiO$_2$ sol was prepared via the TEOS sol–gel method catalyzed by ammonia or HCl [101]. Furthermore, the AR/nano-SiO$_2$ leather finishing agent can also be prepared through the emulsion polymerization without adding emulsifier [102]. The water vapor permeability of leather treated with the AR/nano-SiO$_2$ finishing agent was increased by 9.15% and the finish adhesion was increased by 10.35%. The modified AR finishing agents (nano-coatings) exhibit unusual properties [103, 104], which were related to the uniform dispersion of nano-SiO$_2$ in the polymer, thus probably improving the resistances to abrasion, aging, climate, and the strength of the polymer. It has also been reported that the polyacrylate (PA)/nano-SiO$_2$ composite could be synthesized via semicontinuous emulsion polymerization stabilized with polymerizable surfactant [105]. The application of this composite in leather finishing showed that, compared with conventional surfactants containing latex, the latex stabilized with polymerizable surfactants showed significantly higher water–vapor permeability (increased by 2.06%) and water absorption resistance (increased by 7.88%).

At present, the aerogel-modified coatings for leather are still in the early stages of exploration, and tremendous studies on advanced leather finishing agents to induce special properties have been widely carried out, for example, preparing novel sulfanilamide-conjugated polyurethane coatings with enzymatically-switchable antimicrobial capability for leather finishing [106], fabricating nano-scale core−shell type particle of caprolactam-butylacrylate co-modified casein (CA-CPL-BA) leather finishing agent with enhanced hydrophobicity as well as higher thermo-stability and biodegradability [107]. Moreover, photosensitive silicone-containing polyurethane acrylate leather finishing agent [108], alkali-soluble butyl acrylate/acrylic acid copolymer leather finishing agent with excellent air permeability, water-resistance and resistance to wet rub fastness [109] are being explored extensively. Hence, more studies on exploiting the special functionalities of aerogels coatings on leather substrates shall be expected in future, and there is also much work to be done in the future if the various functional features of aerogels are to be brought into play on leather coatings.

4.3 Aerogel coatings on other substrates

Aerogel coatings on different substrates for advanced applications such as chemical protection, orthopedic application, microwave absorption, etc. have been achieved in recent years. For instance, Horvata et al. proposed a strategy to prepare a new type of high methoxy pectin-xanthan gum aerogel-coated medical grade

stainless steel via ethyl alcohol induced gelation followed by supercritical drying for the orthopedic applications [110]. In this process, diclofenac sodium and indomethacin were incorporated into the aerogel coating as two non-steroidal anti-inflammatory drugs. According to the author's report, all samples performed biocompatibility on the human bone-derived osteoblasts cells, thus showing the potential for the orthopedic applications. The release of the two non-steroidal anti-inflammatory drugs was completed after 24 h, which was confirmed by the IR spectra after the stationary phase of the drug release curve and the final release point. Interestingly, carbon aerogel (CA) could also be used to construct a multifunctional beta-tricalcium phosphate bioceramic platform for osteosarcoma therapy and enhanced bone regeneration [111]. The CA coated-bioceramic platform exhibited attractive photothermal and tumor ablation abilities as well as enhanced bone regeneration. In another work, a lightweight three-dimensional polystyrene (PS) scaffold dip-coated with multiple layers of TiO_2 aerogel had found potential applications in the X-band microwave absorption applications [112]. As per the report, the 10-times dip-coated TiO_2 aerogel-PS composite enhanced the microwave absorption up to 70%, and this value reached 80% for the 15-times dip-coated sample than the pristine PS scaffold which showed 40% microwave absorption in the X-band region. Furthermore, there are also some other recent progress in aerogel coatings. For example, ionic liquid-functionalized silica aerogel and CA can be used as coating materials to construct microextraction platform, and these aerogel coated-platform exhibited high analytical potential for microextraction applications [113, 114]. Polypropylene aerogel coated sponges could be used for oil–water separation, and the oil absorption capacity was not reduced much after 10th repeated use [115]. Moreover, organically modified silica aerogel film with high transparency, flexibility, superhydrophobicity and thermal stability can be used to construct functional coating layers used in solar cells, waterproof textiles, flexible electronics, and lab on papers [116]. These works revealed the high potential of functional aerogel materials used for coating applications in the fields of energy, environmental protection and health, which can provide more research inspiration for us.

5 Conclusions and outlook
Aerogel shows great promise for use in varieties of technological areas due to its extreme properties. It is synthesized through the sol–gel technique which involves the conversion of a liquid system (sol) into a gel phase. The sol–gel synthesis of aerogels comprises three major stages: preparation of the gel, aging of the gel and drying of the gel. Drying of the gel is a critical stage and

requires a special condition to avoid the influence of capillary pressure so that shrinkage of the gels is minimum. The typical features of aerogel are low density solids, refractive index, thermal conductivity, speed of sound through the material and dielectric constant as well as high surface area. The most widely investigated property of aerogels is the low thermal conductivity causing prominent applications as high-performance thermal insulation. Aerogels also can be used in other applications, such as catalysis, drug delivery, radioactive radiation detection, heavy metal adsorption, etc. Apart from their numerous advantages, aerogels have been found to possess their own set of disadvantages, including fragility, rigidity, dust formation, and so on, which will limit their extensive applications. One way of rectifying the problem is the coating of aerogels on different substrates. Over recent years, substantial progress has been made in the development, fabrication and characterization of aerogel coatings on different substrates for special functions. The sol–gel coating process produces a thin film coating of aerogels on different substrates in a single coating step. Other common coating techniques suitable for the coating of aerogels on varieties of substrates for different functionalities are dip-coating, spray-coating, spin-coating, etc. By utilizing these techniques, aerogels have the potential to be used in the coatings on textiles, leather, etc. for special functionalities such as superhydrophobicity (self-cleaning), thermal insulation, flame retardancy, chemical protection, etc. This is of great significance to provide high-performance commodities for people's daily life. Moreover, the works carried out on utilizing the special functionalities of aerogel coatings as finishing materials for textiles are incomparable to that of leathers due to the difference in fiber composition and weaving forms. Hence, this review can provide guidance to motivate readers to carry out studies on exploiting the versatile functionality of aerogel coatings as leather finishing materials hereafter.

Authors' contributions
BTM and WD conceived the original idea and wrote the paper; HL, SG, XP and ZD reviewed the manuscript; MHS performed the formal analysis. The author(s) read and approved the final manuscript.

Availability of data and materials
Not applicable.

Declarations

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