Corresponding Author: Professor Fatang Jiang, PhD

Corresponding Author's Institution: Hubei University of Technology

First Author: Yixin Wang

Order of Authors: Yixin Wang; Yuehong Su; Weiling Wang; Ying Fang; Saffa Riffat; Fatang Jiang, PhD

Highlights

1. Polysaccharide-based aerogels can be prepared with supercritical or freeze drying.

2. Polysaccharide-based aerogels may have nm - μm pore size distribution.

3. Aerogels with open or close pores have different functions and applications.

4. The formula and drying methods can change the pore structure and functions.

5. Advances and future challenges of polysaccharide-based aerogels are clarified.

1   **The advances of polysaccharide-based aerogels: preparation and potential application**

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3   Yixin Wang [a], Yuehong Su [a], Weiling Wang [b], Ying Fang [b], Saffa B. Riffat [a] and Fatang Jiang
4   [*b,a]

5   [a] Faculty of Engineering, University of Nottingham, University Park, Nottingham, NG7 2RD,
6   UK;

7

8   [b] School of Bioengineering and Food Science, Hubei University of Technology, Wuhan
9   430068, China;

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11   **Abstract:**

12   Polysaccharide-based aerogels have high application value as one kind of unique functional

13   materials. Not only has it high porosity and low-density, but also the non-toxicity and

14   biodegradability. In recent decades, a variety of natural raw materials and their combinations

15   along with various preparation technologies have been investigated to develop

16   polysaccharide-based aerogels with different functions for diverse applications. This review

17   aims to clarify a general approach in the development of polysaccharide-based aerogels

18   regarding pore structure design, polysaccharide selection and drying methods. The relevant

19   researches and reports of polysaccharide-based aerogels have been also classified according

20   to the applications in environmental engineering, buildings, medical practice, packaging and

21   electrochemistry. Furthermore, some statistical graphs have been produced to summarize

22   those publications during the past ten years, with an aim to indicate the distribution and

23   research trend. Finally, the approaches to improve the quality of the aerogels are discussed

24   and some perspectives are put forward to provide a reference for the future development of

25   polysaccharide-based aerogels.

26   **Keywords**: polysaccharide; aerogel; pore structure; drying method; application

1. Introduction

Aerogels were firstly fabricated by Kistler. He obtained it from gels, in which the liquid was replaced by gas. Some other practitioners hold the views that only the materials with meso- and macropores with diameters up to a few hundred nanometres and porosity of more than 95% can be called as aerogels (Ziegler, et al., 2017). João P. Vareda (Vareda, Lamy-Mendes, & Durães, 2018) reconsidered about the definition of aerogels and present the view that aerogels need to be redefined as the recent development of aerogels with multiple drying methods. With the widening of the definition, more and more studies on aerogels have been carried out.

By summarizing researches with two drying methods, this review is inclined to the view that aerogels have high porosity, high surface area and low density, and pore size distribution is mainly from nano to micro scale. One kind of new and sustainable polysaccharide-based aerogels stood out and attracted a lot of interests from researches.

In the past decade, a large research effort worldwide has been devoted to developing polysaccharide-based aerogels. This has been stimulated by the fact that the raw materials of conventional aerogels come from inorganic or petrochemical-based materials such as those used in silica aerogels, graphene aerogels (Jiang, Chowdhury, Balasubramanian, 2019), titanium aerogels (Zhang, Liu, Qi, Cui, Yang, 2018), or their oxides aerogels. In response to the environmentally friendly requirement, development of polysaccharide-based aerogels has attracted extensive interest from researchers (Rudaz, et al., 2014). The polysaccharide-based aerogels are formulated from natural ingredients, they have an excellent eco-friendly biodegradable feature and hence maximizing the polysaccharide ingredients in aerogels has become a tendency.

Polysaccharide-based aerogel as the precursor has abundant natural sources, such as cellulose, starch, chitosan, alginate, carrageenan and pectin. As one of the most abundant natural polymers, cellulose widely exists in nature wood (Bauli, Rocha, De Oliveira, & Rosa, 2019), bamboo (Sheng, Zhang, Qian, & Fontanillo Lopez, 2019), cotton (Cheng, et al., 2017), banana fiber (Harini, Ramya, & Sukumar, 2018) and coconut husk (Rosa, et al., 2010). It may become a key ingredient in novel functional materials because of their unique properties, including biocompatibility, sustainability, low toxicity and renewability. As for the application as packaging foam, polysaccharide-based aerogels have overwhelming superiority to accelerate the promotion of plastic ban. The functional biocompatibility and low toxicity properties of polysaccharide-based aerogels also can be applied in health and medicine fields to improve the stability problem of drugs and some limitation of drugs delivery to benefit mankind.

This review aims to describe and discuss the structure design, raw materials selection, preparation and application of biodegradable polysaccharide-based aerogels. In Section 2, two drying methods of aerogels are compared especially on the aspect of pore structure to provide a reference for aerogels structure design. Then with excellent advantages, polysaccharide-based aerogels have attracted wide interests in many fields, such as environment engineering, buildings, medicines, electrochemical components and food packages. This is discussed in Section 3. In Section 4, a statistical survey showing popular research tendency of polysaccharide-based aerogels from 2011 to the beginning of 2019 can be seen from the number of publications and the area distribution of all publications from the pie chart. Finally, the challenges, perspectives and concluding marks are presented in Section 5.

75   Hopefully, this review could help to explore the formulation, preparation and

76   application for polysaccharide-based aerogels. It also presents challenges for the future

77   development of polysaccharide-based aerogels.

78

79   **2. Preparation methods and carbohydrates for polysaccharide-based aerogels**

80   **2.1. Pore structure design**

81   In all porous solid materials, a variety of channels and cavities exist, which are regarded

82   as pores. Aerogel materials stand out from porous materials due to their unique pore structure,

83   high specific surface area and low density which could be used in many applications (Hüsing

84   & Schubert, 1998). Depending on the application requirements, the meaningful functional

85   properties which is affected by pore structures could be introduced in different aerogels. The

86   shape of aerogel pores could be divided into open pores and close pores according to the gas

87   fluid flow property presented in the theory of Rouquerol, et al (Rouquerol, et al., 1994). In

88   our previous studies, relatively close and open pores could be observed in SEM images as

89   shown in **Fig. 1**. Open pores have the opportunity to communicate with others and the

90   external surface of the materials. The close pore is defined as the relatively independent one

91   separating from any nearby pores. For example, aerogels are often used in different fields,

92   such as drug encapsulation, generative medicine and water pollutants adsorption. In these

93   application fields, the requirement of the material structure is quite different. When the drug

94   is being carried, the drug needs to be encapsulated by the carrier, porous aerogels with

95   relatively closed pores (De Marco, Baldino, Cardea & Reverchon, 2015), and then released

96   under specific conditions. In the field of regenerative medicine, aerogels as the scaffold

97   structure, require uniform open pores (Martins, et al., 2015). As an adsorbent material,

98   aerogels require a certain amount of open pore structure to inhale pollutants as well as close

99   pore structure to store the pollutants. Therefore, the significance of aerogels structure design

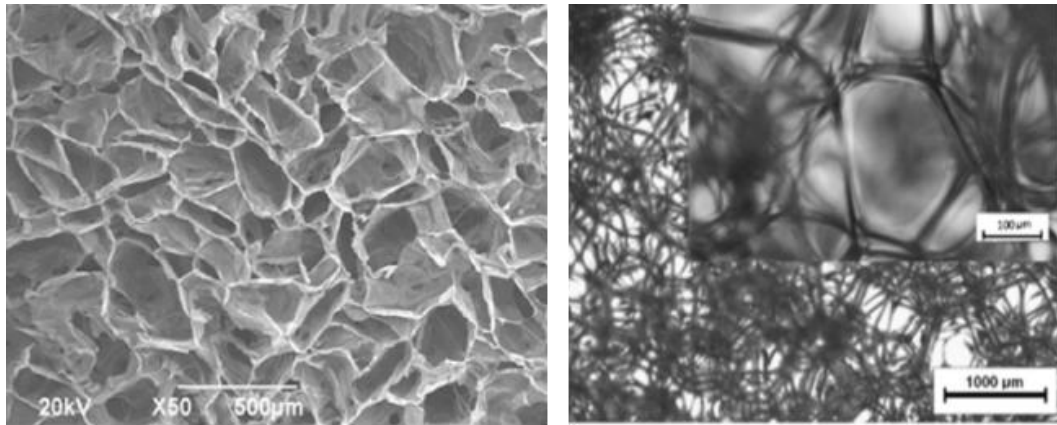100   is to be better adapted to the different application requirement.

101

102   **Fig. 1** SEM observations of relatively close (left) (Wang, et al., 2018) and open pores

103   (right) (Cuadros, Erices, & Aguilera, 2015)

104

105   According to previous researches (Oschatz, et al., 2017; Zhu, Li, Pang, & Pan, 2018),

106   different preparation technologies including raw materials, gel process and drying methods

107   lead to different aerogel pore structure. With different special molecule chain structure,

108   polysaccharide could directly affect aerogels physical properties which will be given a

109   detailed introduction in Section 2.2. Meanwhile, physical properties could be also changed by

110   adjusting aerogels pore structure. Raw materials could affect the pore structure due to its

111   unique properties. As presented in our previous work (Wang, et al., 2018), with a high

112   concentration of starch, the pore wall becomes thicker which benefit the formation of close

113   pores in aerogels. With the thicker wall, it can bear certain pressure to protect the pores in

114   aerogels forming as the close pores. Oppositely, from the optical microscopy images

115   comparison of a biopolymeric porous matrix with three different solutions (alginate solution,

116   gelatin solution and mixture solution), more interconnected pores will be formed with the

117   addition of gelatin (Cuadros, Erices, & Aguilera, 2015). Gelatin, sucrose and paraffin wax

118   have been confirmed to be used as porogens to increase the porosity of materials. (Cuadros,

119   Erices, & Aguilera, 2015; Liu, Manesis, Chan, & Yu, 2015; Pircher, et al., 2015).

120   On the other hand, drying methods of aerogels can obviously control the pore structure

5

121 of aerogels. Supercritical drying produces pores dominantly at the nanoscale. Sophie Groult,

122 et al. (Groult & Budtova, 2018b) have studied various facts on aerogel pore structures impact

123 of external conditions. With the supercritical drying, a large number of cylindrical rods are

124 crossed together as the skeleton to form a three dimension network. The pore size distribution

125 and network intensive degree can be adjusted during supercritical drying. Recent years,

126 freeze drying is commonly used due to its safety and low cost. Choosing a different way to

127 pre-freeze samples will lead to different size of ice crystal presenting pores in aerogels which

128 will be introduced in Section 2.3.2.

129 **2.2. Polysaccharide and pore structures of aerogel**

130 Polysaccharides are widely used in aerogels for their bio-sustainability and

131 biocompatibility. The polysaccharides selected in this section are commonly used in

132 recent years. Described below are the molecular structure features with or without

133 modification and pore structures of aerogels (as summarised in **Table 1 and Table 2**)

134 of polysaccharides and the characteristics of polysaccharide-based aerogels:

135

136 **Table 1** Summary of polysaccharide modification influence on performance

Name	Modification methods	Performance improvement	References
Starch	Hybrid	Improvement of aerogel hardness, anti-fungal property surface area, thermal insulation property	(Abhari, et al., 2017), (Miao, et al., 2008) and (Wang, et al., 2018)
Chitosan	Grafting, monomethyl-modification	Improvement of solubility coagulation	(Hsan, et al., 2018) and (El Knidri, et al., 2018)
Konjac glucomannan	Grafting, oxidation	Hydrophobic interactions ability, good swelling ability and appropriate water retention capacity	(Wu, et al., 2013) and (Luo, et al., 2018)
Alginate	Grafting and targeted modification	Thermos-responsiveness, pH-responsiveness and hydrophobicity	(Shao, et al., 2018) and (Cheng, Lu, Zhang, Shi & Cao, 2012)
K-Carrageenan	Carboxymethylation	Targeted release in the intestine.	(Leong, et al., 2011)
Pectin	Oxidation, carboxymethylation,	Intrinsic viscosity decrease, thermo-sensitive	(Gupta, et al., 2013) and (Işıklan, et al., 2018)

137

138 **Table 2** Summary of polysaccharide influence on aerogel pore structure

Name	Influence on aerogel pore structure	Reference

		(Jin, Nishiyama, Wada & Kuga, 2004)
Cellulose	(NFC) Formation of small pore size of aerogels	
	(CNC) Formation of rigid structure of aerogels	(Heath & Thielemans, 2010)
Starch	Influence on specific surface area and density of aerogels;	(Zhu, 2019)
Chitosan	Formation of more random orientation aerogel pore structure	(Takeshita& Yoda, 2015)
Konjac glucomannan	Formation of open and close pores with gelatin and starch	(Wang, et al., 2018)
Alginate	Impaction on porosity and pore size of aerogels	(Dekamin, et al., 2018)

139

140   Cellulose: As one of the most abundant organic resources on the earth, cellulose has been

141   transformed into aerogels with excellent performance and value of industrial field

142   applications during the last decades. It is commonly obtained from plants (Somerville, 2006)

143   and microorganisms (Moon, Martini, Nairn, Simonsen, & Youngblood, 2011; Somerville,

144   2006). Cellulose is a biopolymer composed of D- glucose and β-1, 4-glucosidic bond. Three

145   kinds of cellulose aerogels, including nanofibrillated cellulose aerogels, bacterial cellulose

146   aerogels and cellulose nanocrystal aerogels (Wan, et al., 2019), are distinguished through

147   different ingredient and synthetic methods. These three general types of cellulose are divided

148   depending on the different resource, process and morphology (Abdul Khalil, et al., 2015).

149   (1) Nanofibrillated cellulose: After a fibrillated process, cellulose fiber reached to microfibril

150   units that can be considered as nanofibrillated cellulose (NFC) with 5-70 nm diameter,

151   several micrometer lengths (Cherian, Paulose, & Vysakh, 2018; Blanco, et al., 2018).

152   After forming aerogels, NFC presents the pores of about 10-100nm with supercritical

153   drying. Some researchers use this particular structure to construct the properties of

154   aerogel. Shaoliang Xiao et al. (Xiao, Gao, Lu, Li, & Sun, 2015) reported fabrication of

155   NFC from natural pine needles. Using nano-level NFCs interconnecting with each other

156   (diameter range from 30 to 70 nm), NFC aerogel fabricated with three dimensional

157   polymeric networks has been introduced to increase the surface area. The specific

158   structure of nanofibrillated cellulose forms its unique properties. The high content of

159   nanofibrillated cellulose will hinder the ice crystals growth during freezing, contributing

160   to the small pore size of aerogels, which could be used in particle separation in gas and

161   liquid phases (Jin, Nishiyama, Wada & Kuga, 2004).

162   (2) Cellulose nanocrystal: Cellulose nanocrystal (CNC) aerogels are also known as

163   nanowhiskers. After hydrolysis reaction and mechanical treatment, lignocellulose fibers

164   were disposed into a few nanometers in length with strong acids (De Oliveira, et al., 2019;

165   Seabra, Bernardes, Fávaro, Paula, & Durán, 2018; Bhat, Khan, Usmani, Umapathi, & Al-

166   Kindy, 2018). It has been proved by Heath & Thielemans (Heath & Thielemans, 2010)

167   that hydrogen bonds collapse could be avoided due to the rigid structure resulting from

168   the high modulus of the highly crystalline cellulose nanowhiskers. To pursue the high

169   mechanical properties of aerogels, it is an option to consider the cellulose nanocrystal. As

170   reported by Quan Yong Cheng et al. (Cheng, Guan, Wang, Li, & Zeng, 2018), a super

171   hydrophobicity CNC coated cotton fabric was fabricated to separate oil and water with

172   good mechanical handleability.

173   (3) Bacterial cellulose: Unlike NFC and CNC, bacterial cellulose (BC) is produced by

174   acetobacter xylinum (Bodin, Bäckdahl, Petersen, & Gatenholm, 2017; Foresti, Vázquez,

175   & Boury, 2017). (Lee & Bismarck, 2016). Since bacterial cellulose is from microbial

176   metabolism, it has good biocompatibility and nontoxicity (Foresti, et al., 2017). BC

177   aerogels with good biocompatibility, high mechanical strength in wet environment, high

178   stimulative epithelialization ability, good liquid and gas permeability, and inhibition of

179   skin infections could be used in medical, such as skin care (Picheth, et al., 2017;

180   Amnuaikit, Chusuit, Raknam, & Boonme, 2011; Keskin, Sendemir Urkmez, & Hames,

181   2017), topological wound healing (Petersen & Gatenholm, 2011) and drug delivery

182   (Sheikhi, et al., 2018). Hadi Hosseini (Hosseini, Kokabi, & Mousavi, 2018a) took full

183   advantage of entangled BC nanofibers to fabricate a new class of BC/reduced graphene

184   oxide nanocomposite aerogels to be used as sensor in some components and parts.

185   *Starch:* Starch is the main storage carbohydrate in higher plants. Starch contains two D-

186   glucan biopolymers, i.e., amylose, a relatively linear 1,4-α-D-glucan with a small number of

187   long branches; and amylopectin, mainly a 1,4-α-D-glucan containing high-density branches

188   (ca. 5% of glycosidic bonds are α-1,6) (Maningat, Seib, Bassi, Woo, & Lasater, 2009). With a

189   mass of starch branched structure, it is easy to stretch into three dimensional network when

190   the chains of starch meet water (Ubeyitogullari & Ciftci, 2016). According to the diverse

191   constitution of amylose and amylopectin, the performance of starch presents totally different.

192   Pure amylose could not form as aerogels because the heterogeneous structure could not stand

193   the pressure during the freezing process (Druel, Bardl, Vorwerg & Budtova, 2007). However,

194   appropriate addition of amylose can increase the specific surface area of the aerogel but

195   reduce the density. And the type of starch and ratio of amylose and amylopectin are important

196   for the microstructure of aerogels, resulting in different properties. For example, the amylose

197   content of starch could determine the mechanical properties of aerogels (Zhu, 2019). Despite

198   of the outstanding advantages of starch aerogel, there remain some drawbacks, such as low

199   hardness. However, the mechanical property is able to be improved through start materials

200   modified in the preparation process of aerogels (Abhari, Madadlou, & Dini, 2017; Miao, et

201   al., 2008; Wang, et al., 2018). Negar Abhari et al. (Abhari, et al., 2017) fabricated trisodium

202   citrate cross-linked starch aerogels to improve the hardness and decrease the adhesiveness.

203   Chitosan: Extracted from various organisms commonly found in arthropod shells, chitin

204   has β-linked N-acetyl-D-glucosamine carbohydrate polymer presenting insoluble

205   performance (Ziatabar, et al., 2018). To have a certain extent solubility of chitin, chitosan can

206   be obtained through deacetylate treatment (El Knidri, Belaabed, Addaou, Laajeb, & Lahsini,

207   2018). The properties of chitosan can be enhanced by chemical modification, such as grafting

208   reaction (Hsan, Dutta, Kumar, Bera, & Das, 2018), monomethyl-modification (El Knidri, et

209   al., 2018), O-alkylated reaction, etc. In the research of Takeshita & Yoda (Takeshita& Yoda,

2015), the comparison between cellulose aerogels and chitosan aerogels with same density shows that chitosan aerogels have more random orientation pore structure contributes to the good thermal insulation properties. Meanwhile, formed in chitosan aerogels, three dimension hierarchical porous scaffold benefits to the improvement of electrochemical cyclability and rate capability (Hassan, Suzuki & El-Moneim, 2014; Ji, et al., 2013).

Konjac glucomannan: Konjac glucomannan (KGM) is a β-1, 4 glycosidic bonds linked polysaccharide composed of D- mannose and D- glucose with a molar ratio of 1.5:1-1.6:1 and 5–10 % acetyl substitution. It has abundant free carboxyl and hydroxyl groups in the skeleton unit, contributing to excellent ability to attract the multivalent cations and form desirable cross-linking structure. It has a high viscous property (30,000 mPa s, 1%, w/ v), and molecular weight (6.8×10^5–9×10^6 Da) (Crosby, 2002; Li, et al., 2019), making it easier to be considered as a framework material in aerogels (Zhu, Hu, Jiang, Liu, & Li, 2019). In our previous studies (Wang, et al., 2018), KGM molecular chain as a skeleton can be composited with gelatin and starch, which could promote the formation of open and close pores, respectively. Currently, some physical and chemical methods are commonly used to modify the native konjac glucomannan getting wonderful functions. Chen Xin et al. (Xin, et al., 2017) studied the effect of different deacetylation interaction degrees on intramolecular and intermolecular forces in KGM system and thereby lead to various hydrophobic ability. Simultaneously active hydroxyl groups in KGM structure do favor of some other chemical reaction, including grafting (Wu, Deng, & Lin, 2013; Xia, et al., 2010) and cross-linking (Ratcliffe, Williams, English, & Meadows, 2013), oxidation (Luo, et al., 2018).

Alginate: Having free carboxyl and hydroxyl groups in alginate linear structure as well as the G-blocks, alginate has an excellent capacity to attract the multivalent cations and then form a novel "egg-box" structure. The block content of β -d-mannuronic acid or α -l-guluronic acid impacts on the structure of hydrogel and the aerogels pore structures including

porosity percentages and pore size (Dekamin, et al., 2018). Sodium alginate possesses the characteristic of pH sensitivity with the different solubility. At pH < 4, the sodium alginate is insoluble while it has a viscous and stable characteristic when pH increased to 6-9. Chemical modification is a universal method to introduce the functional groups into molecular. Lin Shao et al. (Shao, et al., 2018) have obtained a thermos-responsiveness and pH-responsiveness alginate composite aerogel by grafting with N-isopropylacrylamide (hydrophobic block) and N-hydroxymethylacrylamide (hydrophilic block).

Carrageenan: 3-linked-β-d-galactopyranoses and 4-linked-α-d-galactopyranoses constitute the carrageenan alternatively and repetitively. According to different sulfation degree and position, carrageenan can be distinguished from different variants and is mainly divided into κ-carrageenan, ι -carrageenan and λ-carrageenan (Anderson, Dolan, & Rees, 1973). The difference between these three types of carrageenan is the binding ions as forming helical structures. κ-, ι- and λ-carrageenan are easier to strengthened with the presence of K^+, Ca^{2+} and no need for sodium salts, respectively (Derkach, et al., 2018). Located out the double helix, the sulfonate groups of κ-carrageenan were interacted with the cations (K^+, Ca^{2+}, Co^{2+} and Fe^{3+}) by ionic force. This specific binding could contribute to dope the conductive polymer, which promotes the aggregation to junction areas and the cross-linking of conductive polymer with the carrageenan-cation, resulting in the improvement of mechanical properties of aerogels (Zamora-Sequeira, Ardao, Starbird & García-González, 2018). Chemical modification of carrageenan is considered as an effective route to improving properties. Carboxymethylation process has been used in kappa carrageenan to produce carrageenan derivatives which have promising potential to apply in drug delivery (Leong, et al., 2011).

Pectin: Pectin is composed of different degrees of esterified residues of galacturonic acid (Groult & Budtova, 2018a). Depending on various degrees of pectin esterification, it can be

260 classified into two categories: high-methoxy pectin and low-methoxy pectin. As verified by

261 Victor J. Morris et al. (Morris, Belshaw, Waldron, & Maxwell, 2013), pectin based materials

262 present various biological and chemical properties with diverse chemical structure. In some

263 specific aerogels, pectin were used as gelata and stabilizer due to its high solution viscosity

264 and gel speed. Moreover, it is promising to use pectin aerogels in oral drug delivery because

265 of their gastro-resistant properties (Groult & Budtova, 2018b). When the pH is near or higher

266 than PK_a (3-3.5), pectin can combine with divalent cations, also known as "egg-box"

267 structure, to form an intermolecular junction zone mainly through the connection of ionic

268 bonds. An affinity with alginate has been compared and ranked as follows: Pb (II) > Cu (II) >

269 Cd (II) > Ba (II) > Sr (II) > Ca (II) > Co (II), Ni (II), Zn (II) > Mn (II) (Mørch, Donati, &

270 Strand, 2006). Commonly, other direct chemical modifications have been introduced into

271 pectin, including oxidation (Gupta, Tummalapalli, Deopura, & Alam, 2013), functional

272 copolymerization (Işıklan & Tokmak, 2018) and carboxymethylation (Muthukumaran, et al.,

273 2018), etc.

274 **2.3. Drying methods for aerogels preparation**

275 Concluded from past researches, there are two mainstream drying methods to prepare

276 aerogels with different mechanism. Supercritical drying is the most efficient method to

277 produce small size pores. Utilizing the characteristics of supercritical fluids, supercritical

278 drying needs high pressure. It is dangerous and expensive, and the process is not easy to be

279 big scale commercialized production. The freeze drying is simple, economical,

280 environmentally-friendly, and easy to operate, large scale production. However, its drying

281 cycle is long. Since atmospheric drying is rarely used (Vareda, Lamy-Mendes, & Durães,

282 2018), it will not be discussed in this review.

283 **2.3.1. Supercritical drying.**

284 The fabrication process of Supercritical drying can be classed into two approaches, high

temperature supercritical drying (HTSCD) and low temperature supercritical drying (LTSCD). In HTSCD, the water in hydrogel needs to be replaced with an organic solvent (alcohol, acetone) and then placed in an autoclave for heating and pressurization. The solvent in the gels reaches the supercritical state, and then vents out at constant temperature. In LTSCD, soluble CO_2 is used as a drying medium to replace the organic solvent (alcohol, acetone) in gels, and can be transferred into supercritical CO_2 at critical temperature close to room temperature, followed by the formation of aerogels. However, Supercritical drying can eliminate capillary pressure and maintain the original shape of materials because the pressure and temperature can be controlled to get critical point at which gas and liquid co-exist with same density and the interface disappeared to achieve phase transfer (Kistler, 1931). This high efficient hydrogel drying method has the advantage that surface tensions in pores can be avoided to maintain the pore structure of aerogels. The transformation from gels to aerogels in supercritical drying, hydrogel aqueous phase will be substituted by ethanol with the purpose of reducing surface tension and thereby eliminate capillary pressure to prevent from pore collapse (Liu, Fang, Oderinde, Zhang, & Fu, 2017). Finding suitable and safe supercritical fluid was considered as a challenge in the previous research work until CO_2 being used as drying medium for its relative high security, temperature (31 °C) and mild pressure (74 bar) (Ciftci, et al., 2017) during preparation process in industry. After ethanol substituting (the preparation of alcogels), the supercritical extraction of ethanol assisted by supercritical fluids (such as CO_2, CH_4) was carried out and then aerogels could be collected.

Amount of ethanol: Certificated by porosity, density and scanning electron microscope, the surface area increased with the increase of ethanol content (Estella, Echeverría, Laguna & Garrido, 2008). And the excessive flow rate of ethanol extraction caused an increase in the diffusion rate of ethanol between the pores. This will lead to more cracking and affect the structure inside the gel, which is not conducive to the formation of high specific surface area

310   in aerogels.

311   Drying time: It has been reported that the structure of the materials was influenced by

312   different mass transfer resistances with time (Brunner, 2013). García-González et al. (García-

313   González, Camino-Rey, Alnaief, Zetzl, & Smirnova, 2012) showed there is a difference in the

314   density of the starch aerogels with different drying time (drying time after 30 min, the density

315   is 0.40 g/cm^3; after 1 h, the density is 0.27 g/cm^3 and after 2 h and longer, the density is 0.21

316   g/cm^3). And relative long drying time led to high surface area. However, the optimal drying

317   time need to be controlled due to the comprehensive economic benefits.

318   Drying pressure: The supercritical drying pressure increases contributed to decreasing of

319   specific surface area of aerogels and increasing of the density. It has been reported that high

320   pressure produced aerogels with small pores and the rate of depressurization affects the pore

321   growth, leading to the formation of larger pores (Tai, et al., 2017).

322   **Table 3** showed a microstructure comparison between different ingredient aerogels with

323   same supercritical drying methods. It can be found that the pore size distribution is on the

324   range of nano level. Low density and high porosity are observed from previous researches.

325   They are the typical characteristic of aerogels from supercritical drying method.

326

327   **Table** 3 Physical properties of different type polysaccharide-based aerogels

Samples	Density (g/cm^3)	Surface area(m^2/g)	Pore size(nm)	Porosity (%)	Reference
Pectin aerogel	0.04-0.05	270-350	-	-	(Nešić, et al., 2018)
barley and yeast β-glucan aerogels	0.03-0.12	89.4-173.1	23.7-16.1	-	(Salgado, et al., 2017)
Alginate aerogel	0.15-0.17	126.9-173.3	15.48-17.34	-	(Tkalec, Kranvogl, Perva Uzunalić, Knez, & Novak, 2016)
Ag nanoparticle/cellulose nanofiber aerogel	0.021	31.5		98.6	(Matsuyama, et al., 2019)
Nano-cellulose aerogels	-	260.87-353.83	7.81-9.38	-	(Wang, et al., 2016)
starch aerogels	-	93	24-25	87.7	(Goimil, et al., 2017)

|					
Chitosan aerogels	-	257-479	12.6-15.0	96.8	(López-Iglesias, et al., 2019)
Tungsten/alginate aerogels	-	381	31	96	(Paraskevopoulou, et al., 2018)
chitosan/lanthanum hydroxide aerogel	-	172.74	19.79	-	(Lin, Li, Song, Jiao, & Zhou, 2018)

328

2.3.2. Freeze drying.

330   Because of low cost and high safety, it is a tendency that the freeze drying is extensively

331 applied for aerogel preparation. First, gel sol is frozen, and then the ice of gel is sublimated

332 during which the liquid in gels is replaced by gas to generate pores under high vacuum. (Ni,

333 et al., 2016; Wang, et al., 2019). Aerogel structure is determined by ice crystal growth process

334 of the gel solution. Shunli Liu et al. (Liu, Yao, Oderinde, Zhang, & Fu, 2017) fabricated

335 gum/graphene oxide aerogels and found that there is a force pushing and flocking the solute

336 (XG/GO hybrid) together between two ice blocks during ice crystalizing, and then after

337 sublimation the aerogel walls formed (schematic diagram as shown in **Fig. 2**). Therefore, the

338 structure of pores is directly determined by ice crystal frame. Using low temperature

339 polarizing microscopy, Xuewen Ni et al. (Ni, et al., 2016) observed the ice crystal growing

340 process with different temperature conditions and different ingredient concentration. The

341 SEM images and figures of pore size distribution show that temperature and ingredient

342 concentration can significantly affect the growth of ice crystal and further the pore structure.

343 Under different freeze drying conditions, aerogel pore size can be adjusted (as summarized in

344 **Table 4**).

345

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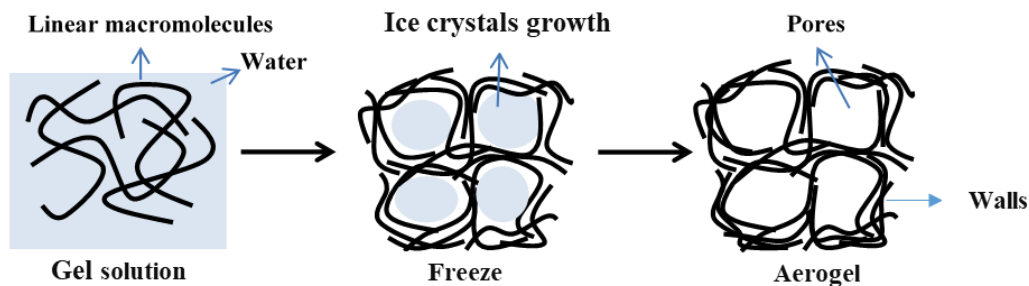
347   **Fig. 2** Schematic of aerogel formation (YX. Wang, et al., 2017)

348

349   *2.3.2.1. Ice crystal growth theory.* From the freezing profile provided by Dimitris

350   Zaragotas et al. (Zaragotas, Liolios, & Anastassopoulos, 2016), who studied the ice

351   nucleation and crystal, it can be seen that there are two phases of crystallization: nucleation

352   and crystal growth. The first short step of crystallization is nucleation. During this period,

353   when solvent cools down to reach supercooling status, the crystal center as well as crystal

354   nucleus formed with some certain stimulus. As previous studies (Searles, Carpenter, &

355   Randolph, 2001) have suggested, it is vital to control the ice nucleus because the initial

356   nucleation temperature and crystallization rate depend on it so that the size of ice crystal was

357   determined. The second step is the growth of ice crystal. There is a competitive relationship

358   between growth of ice nucleus and ice crystal (as shown in **Fig. 3**). Under the condition that

359   ice nucleus are easy to form, it will grow rapidly, condense into a block and finally decrease

360   the growth chance of ice crystal contributing to form relatively small and uniform crystal.

361   However, if the temperature lower down slowly, the ice nucleus near cold side have the

362   tendency to derive to other regions resulting in the formation of large size ice crystal.

363

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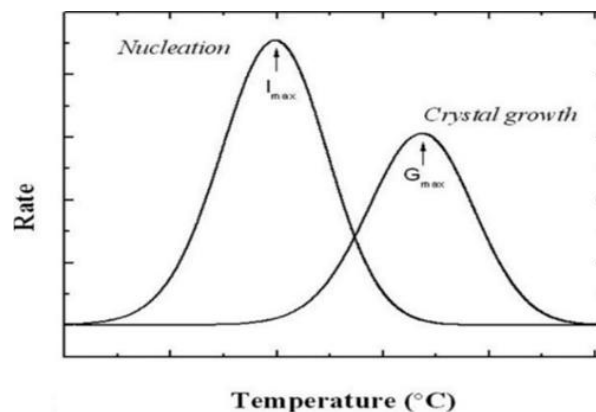
365   **Fig. 3** Temperature influence on nucleation rate and ice crystal growth rate. (Lorenzo, et al.,

366   2008)

367

2.3.2.2. Ice crystal growth controlling.

Temperature: Generally, temperature and slurry concentration are considered as the major factors in affecting the ice crystal growth (Chen, et al., 2018). By low temperature polarizing microscopy, the morphology of ice crystal was observed under different temperatures (-15, -25 and -40°C), and the results showed that a high correlation between temperature and the ice crystal structure. From -15 to -40∘C, the mean diameter of ice crystal decreased significantly, while pore size had the same tendency (Ni, et al., 2016). An extreme lower temperature freezing can be achieved by liquid nitrogen freezing. Alginate/gelatin aerogel was fabricated via liquid nitrogen bath at -196 ∘C with the pore size about 4-8 μm (Li, Chen, & Chen, 2019).

Pressure: During freezing process, high pressure can short the time of cooling and form small and regular ice. According to the theory, some researches (Zhu, Ramaswamy, & Le Bail, 2005) has studied the high-pressure shift freezing (HPSF). The stable structure of H_2O molecules will be destroyed at high pressure, and the broken bonds have a tendency to be more compact with each other (Zhang, et al., 2015). Under the high pressure, the freezing point will be decreased, inducing the super-cooling and rapid ice nucleation rates producing small ice crystals. M.T. Kalichevsky-Dong et al. (Kalichevsky-Dong, Ablett, Lillford, & Knorr, 2000) found the HPSF technique can produce smaller and uniform ice only under -15°C. It has been applied in some food products to promote the formation of more regularity ice crystal (Martino, Otero, Sanz, & Zaritzky, 1998; Sequeira-Munoz, Chevalier, Simpson, Le Bail, & Ramaswamy, 2005).

Concentration: According to the simulation of ice crystals in sugar solutions (Van der Sman, 2016), it can be concluded that ice crystal sizes depend on solute concentration. During the crystals growing process, the concentration will be continuously increased, while

392 the solute is pushed to the higher concentration area, which inhibits further growth of ice

393 crystals. The three dimensional of ice crystals growing in trehalose solution with different

394 concentrations can be obtained by Mach-Zehnder interferometer. With the rise of trehalose

395 concentration (8 wt%), the growing rate of ice crystal increases to a top point (2 wt%), and

396 then decreases (Shimada & Furukawa, 2018). Additionally, the existence of suspension solids

397 also has a great influence on the shape of ice crystals. During the freezing process of locust

398 bean and xanthan gums solution, LT-SEM micrographs have been used to verify that ice

399 crystals growth have been limited by suspension solids (Fernández, Martino, Zaritzky,

400 Guignon, & Sanz, 2007).

401

402 **Table** 4 Polysaccharide-based aerogels pore size under different Freeze-drying conditions

Aerogel type	Pore size	Freezing temperature	Freezing pressure	Concentration	Reference
Nanofibrillated cellulose aerogel	2-50 nm	liquid nitrogen bath (-196°C)	< 5 Pa	0.1 wt%	(Gupta, Singh, Agrawal, & Maji, 2018)
cellulose aerogel	20-600 μm	-80℃	20 Pa	2.0 wt%	(Geng, 2018)
nanofibrillated cellulose aerogel	50.6-74.3 μm	-196°C (liquid nitrogen)	< 20 Pa	3.0 wt%	(Li, et al., 2018)
nanofibrillated cellulose aerogel	47.3-68.1 μm	-55°C (dry ice)	< 20 Pa	3.0 wt%	
	11.8-19.2 μm	-18°C (certified freezer)			(Li, et al., 2018)

403

3. Potential applications of polysaccharide-based aerogels with performance assessment

3.1. Environmental engineering

406 With advances of industrialization and globalization, pollution problems follow the

407 emission of organic liquid, leakage of oil. It will destroy the ecological environment. Aerogel,

408 the ultralight porous material, is useful for removing the pollutants (Maleki & Hüsing, 2018).

409 **3.1.1. Water pollutants adsorption (phosphate and metal ions).** Water pollution is a

410 serious problem which threats the health of human and animals in the world. The

combination properties of low density, abundant porous structure and large surface area can endow aerogels with transcendental absorption performance. There is a large volume of published studies regarding aerogels as a water pollutants adsorption material as shown in **Table 5**. Recent studies on phosphate uptake ability have revealed that the remarkable adsorption capacity could reach 189.06 mg·/g as well as the fast absorption rate with Zr-GO/Alg (zirconium-crosslinked grapheme oxide/alginate) aerogels beads (Shan, Tang, Zhao, Wang, & Cui, 2019). There are also nanoporous chitosan/lanthanum composite aerogel beads prepared for phosphate adsorption due to their high surface area and large pore volume of 172.74 m^2/g and 1.05 cm^3/g, respectively (Lin, et al., 2018). The nanocellulose-based aerogels are able to remove the Cu (II) and Pb (II) with the maximum capacity of 175.44 mg/g and 357.44 mg/g, respectively (Zhang, Li, Shi, Chen, & Fan, 2018). For metal ions absorption, polysaccharide composite aerogels have been considered as one of the most effective ways to remove water metal ions pollution.

Table 5 Water pollutants adsorption performance of different types of polysaccharide-based aerogels

Polysaccharide type of aerogels	Form of aerogels	Pollutant adsorption mechanism	Preparation method	Pollutant	Reference
Alginate-graphene metal oxide)	Beads	Electrostatic interaction and ligand exchange effect	Freeze drying technique	Phosphate	(Shan, et al., 2019)
Nanoporous chitosan/lanthanum	Beads	Lanthanum has a good binding ability to phosphorus	Supercritical carbon dioxide drying	Phosphorus	(Lin, et al., 2018)
Cross-linked chitosan aerogel	Cylindrical	Chelation of amino and hydroxyl groups with heavy metal ions	Cross-linked process and freeze drying technique	Cu ion	(Li, et al., 2016)
Nanocellulose-polyethyleneimine Aerogels	Cylindrical	Amine groups combine with metal ions	Freeze drying technique	Cu (II) and Pb (II)	(Zhang, et al., 2018)

3.1.2. Oil separation from water.

The natural water resource has been polluted severely by the accidental oil leakage, petroleum spills and industrial emission (Liu, et al., 2018). Water protection and purification as a detective task accelerated the development of porous materials, such as the activated carbon, CNTs (Yan, et al., 2019), polypropylene sponge (Wang, et al., 2018), polyurethane foam (Kong, et al., 2018; Nikkhah, Zilouei, Asadinezhad, & Keshavarz, 2015) and wood fibers (Likon, Remškar, Ducman, & Švegl, 2013). Porous aerogels have been proved to effectively separate the water and oil mixture. As studied by Changzhou Chen et al. (Chen, et al., 2018), the brittle graphene aerogels modified by cheap and renewable carbohydrate resources lignin were created as compressive, ultralight and fire-resistant oil absorption materials. In the research of Hongyuan Zhang et al (Zhang, et al., 2019), nanocellulose and sodium dodecylsulfate (SDS) aerogels which have high pump oil absorption capacity about $145.20 \, \text{g} \cdot \text{g}^{-1}$ (aerogel density is $1.50 \, \text{mg/cm}^3$) were fabricated through a high speed mechanical whipping combined with liquid nitrogen freezing and freeze drying method. Many studies focused on biomass aerogels, including chitosan/cellulose (Zhang, Li, Shi, Chen, & Fan, 2018), banana peel and waste paper (Li, et al., 2018) to promote water protection.

3.1.3. Remove organic pollutants. With the development of industrial manufacture and chemical production, toxic chemical leakage aggravates water contamination and resulting the world water shortage. In 2005, the Songhua River in China was seriously polluted by benzene chemical industry (Wang, Feng, Zhao, & Li, 2012). However, this pollution was not a simple local short term pollution problem, because many tributaries of the Songhua River diverted the polluted water. This lead to the pollution impact on nearly 70% of Heilongjiang Province, with a total population of more than 20 million. To improve this water quality, some urgent solutions need to be utilized to deal with it. Some plant waste has been processed to a novel carbohydrate-based aerogels with excellent sorption ability. Lin Zhu et

453 al. (Zhu, et al., 2017) prepared the carbon aerogels using waste inedible pomelo peels as the

454 polysaccharide precursor for the application of organic pollutants removal. Y. Wang et al

455 focused on carbon aerogels using raw materials derived from waste durian shell (DSCA) for

456 removing the organic solvents (Y. Wang, et al., 2017).

457 **3.2. Buildings**

458 **3.2.1. Thermal and sound insulation.** Since the advent of aerogels, NASA's Kennedy

459 Space Center and NASA's Glenn Research Center have focused on aerogels' thermal

460 insulating function to be applied into space industry, such as space radiator and space suit

461 (Fesmire, 2006). As the improvement of technologies and reduction of cost, the civilization

462 of aerogels in buildings has been widely achieved contributing to the improvement of energy

463 saving. In order to avoid drawbacks of traditional aerogels including environment pollution

464 during the preparation process as well as degradation period after being abandoned,

465 carbohydrate ingredients are being introduced into aerogel insulation materials. Konjac

466 glucomannan (KGM)/starch based aerogels strengthened with the waste of agriculture (wheat

467 straw) presented good thermal insulation and mechanical properties (Wang, et al., 2018). An

468 optimized thermal conductivity of 0.046 W/(m·K) and compression modulus of 67.5 kPa was

469 measured for assessing potential application prospect in thermal insulation. The main goal

470 and advantage of this konjac glucomannan (KGM)/starch based aerogels is the biodegradable

471 performance after being abandoned which will not increase the burden on the environment. In

472 another work, Pragya Gupta et al. (Gupta, et al., 2018) invented a nanofibrillated cellulose

473 aerogels based on pinewood cell wall for thermal insulation application. The lowest thermal

474 conductivity of nanofibrillated cellulose aerogels was 0.025 W/(m·K).

475 Noise pollution were commonly caused by the industrial machines, home appliances,

476 vehicles and buildings (Arenas & Crocker, 2010; Zannin, Diniz, & Barbosa, 2002). It was

477 considered as the most widespread and hardest controlled environment pollution (Bronzaft &

478 Hagler, 2010; Chen, Chang, & Jiao, 2014). It will not only affect the human hearing system

479 but also cause some health problems such as high blood pressure (Schmidt, et al., 2015) and

480 increased physiologic stress (Seidman & Standring, 2010; Tzivian, et al., 2015). Apart from

481 the influence on human health, noise can accelerate the aging of buildings and machinery,

482 affecting the accuracy of the equipment. Therefore, acoustic insulation materials are urgently

483 needed. Porous aerogels with special open pore structure are commonly used in the acoustic

484 design of buildings and aircraft so that the sound wave could be absorbed. Jingduo Feng et al.

485 (Feng, et al., 2016) carried out a silica/cellulose hybrid aerogels for acoustic insulation. The

486 sound absorption coefficients reached 0.39-0.50 with 10 mm thickness.

487 **3.2.2. Fire resistant.** It is urgent to enhance the fire resistance of the building insulation

488 materials. Liang Wang et al (Wang, Sánchez-Soto & Abt, 2010) had fabricated gum

489 Arabic/clay aerogels from gum Arabic (GA) and sodium montmorillonite (Na$^+$-MMT) clay

490 offered sustainability, thermal stability and flame retardancy. The fire combustion behavior

491 was tested with cone calorimeter, which presented that alginate/clay aerogels with the

492 addition of p-toluenesulfonic acid can reduce the total heat release of aerogels and toxic

493 combustion. HongBing Chen et al (Chen, Li, Chen, He & Zhao, 2019) reported a self-cross-

494 linked melamine-formaldehyde-pectin aerogels with good thermal stability, excellent water

495 resistance and low flammability. Pectin acts as the structure materials during the cross-linking

496 reaction of melamine-formaldehyde. The results showed that composite aerogels have the

497 time to ignition of 24-27 s, peak of heat release rate of 80.1-116.6 kW/m.

498 **3.3. Healthy and medical applications**

499 As follows, some common polysaccharide aerogels are taken as examples.

500 **3.3.1. Chitosan aerogels.**

501 Chitosan has a lot of beneficial properties in the medical field. It can accelerate blood

502 clotting, improve antibacterial ability and reduce pain in patients with nerve endings (López-

503 Iglesias, et al., 2016; Lodhi, et al., 2014; Okamoto, et al., 2003). The healing process is a

504 complex process, requiring the collaborative efforts of many different tissues and cell

505 lineages. And wound repair is the process by which scar tissue replaces normal skin.

506 However, the physical self-repairing ability of many critically injured and patients with

507 chronic wounds is relatively poor and challenged by many hampered, such as the diabetic

508 foot ulcers and leg pressure ulcers which will maintain a long–term inflammatory phase.

509 Polysaccharide-based aerogels can play an important role to absorb high amounts of aqueous

510 medicine. With the chitosan aerogels loading with vancomycin, Clara López-Iglesias et al.

511 (López-Iglesias, et al., 2019) devoted to treat and prevent infections at the wound site.

512 Combined with cotton fibre, highly absorbable composite chitosan aerogels were developed

513 to stop bleeding (Duong, et al., 2018). The main function of this composite aerogels is to

514 counteract the systolic blood pressure in the wound cavity preventing the blood loss.

515 **3.3.2. Pectin aerogels.** Polysaccharide is preferred raw materials in aerogels for drug delivery

516 due to their biocompatibility, diverse functionality and low toxicity. Pectin attracts the

517 attention of researchers because of its gelation, stability and low toxicity. In the research of

518 Tkalec et al. (Tkalec, Knez, & Novak, 2015), a high-methoxyl pectin aerogels were prepared

519 to be carriers to enhance the dissolution of some drugs. For example, when used to release

520 Nifedipine, 100% drug release was achieved within 12 hours. And a core-shell structure

521 polysaccharide-based aerogels based on alginate (as the shell) and amidated pectin (as the

522 core) were fabricated to prolong the drug activity (Horvat, Pantić, Knez, & Novak, 2018).

523 **3.3.3. Cellulose aerogels.** Cellulose has already been used in wound healing due to their

524 excellent high stability, porosity and non-allergenic. With ultrapure lignocellulosic nanofibers,

525 cellulose aerogels were prepared and applied as dressing material for wounds.

526 Beclomethasone dipropionate nanoparticles were introduced in cellulose aerogels, which

527 could be used to deliver drugs (Valo, et al., 2013). Using waste wheat straw, polypyrrole and

528   silver nanoparticles to prepare a special functional composite cellulose aerogels to achieve

529   the ability to kill E. coli, S. aureus and L. monocytogenes at direct contact (Wan & Li, 2016).

530   **3.3.4. Alginate aerogels.** Alginate is an anionic polysaccharide with linear molecule structure.

531   Combined with divalent cations, a special "egg-box" structure formed with unique properties.

532   Cristiano Agostino Bugnone et al. (Bugnone, et al., 2018) researched silica/alginate aerogel

533   beads with different coating affecting the release rate of poorly-water soluble ketoprofen. To

534   utilize the pH-sensitive release of alginate aerogel, it could be used as drug delivery to

535   simulate different pH values, such as gastric environment (pH 1.2) and intestinal environment

536   (pH 6.8). Lin Shao et al. (Shao, et al., 2018) have grafted alginate with N-

537   isopropylacrylamide and N-hydroxymethylacrylamide to design a dual responsive

538   polysaccharide aerogels for delivery of hydrophobic drug model, Indomethacin. With dual

539   thermo- and pH-responsive intelligent, alginate composite aerogels will be erosion at low

540   temperature and structure shrinkage characteristic at high temperature as well as faster drug

541   release in neutral solution.

542   **3.4. Electrochemistry applications**

543   **3.4.1. Sensor.** Regardless of the high popularity of advanced graphene technology,

544   environmental friendly, unexceptionable flexibility polysaccharide materials are still of acute

545   interests to be combined in the electrochemistry applications. As the rapid development of

546   electronic skin converting the environmental stimuli to the electronic signal, the high

547   sensitive sensor materials are required. Recent years, bacterial cellulose has been chosen as

548   the combination with high electrical and magnetic materials, such as carbon nanotube (CNT)

549   (Hosseini, Kokabi, & Mousavi, 2018a) and graphene oxide (Hosseini, Kokabi, & Mousavi,

550   2018b). Hadi Hosseini et al. (Hosseini, Kokabi, & Mousavi, 2018a) reported an assessment

551   about the synthesis of a novel sensing material based on bacterial cellulose (BC)/reduced

552   graphene oxide (rGO) aerogels via super critical drying method as strain sensor to detect

553 human motion.

554 **3.4.2. Capacitor electrodes.** Super capacitors are promising candidates of energy storage,

555 could be of interest for various industrial application. With the rapid charging or discharging

556 rate and long service life, it presents great potential performance rather than traditional

557 capacitors. As the requirement of good electrical conductivity for capacitors, porous carbon

558 composite aerogels have attracted the attention of many researchers. Heteroatom doping

559 especially using most fascinating nitrogen (N) heteroatom derived from seaweed is

560 considered as one effective way to further improve the electrical capacity of carbon aerogels

561 (Bing, et al., 2017). Jing Han et al. (Han, et al., 2017) have demonstrated 3D porous glucose-

562 based carbon aerogels created more connection opportunity for good electrochemical

563 behavior. Furthermore, to overcome the poor mechanical strength, hypotoxicity and

564 fabricating pollution, many polysaccharide materials with low cost, economic friendly, and

565 excellent mechanical properties have been added in carbon aerogel such as cattail (Yu, Han,

566 Li, Li, & Wang, 2018), nanocellulose (Yang, et al., 2018), etc.

567 **3.5. Packaging**

568 To prevent food from getting metamorphic or moldy, the function of food package is

569 isolating air and humidity to reach the request from customers and manufacturers. The plastic

570 and expanded polystyrene are widely used in food packaging with the risk of toxic hazard to

571 the customer, pollution to our environment (Ali & Wahab, 2017) and harm to animal's health

572 (Rochman, et al., 2013), such as marine plastic debris (Mendenhall, 2018) and toxic additives

573 (Raj & Matche, 2011) remain in plastic package. As used for food package, many expanded

574 polystyrene boxes present effective thermal insulation properties in food storage whereas this

575 can be achieved by aerogels with significant insulation performance. To replace expanded

576 polystyrene, biodegradable polysaccharide-based aerogels are common used as food

577 packaging materials. In 2013, Kirsi S.Mikkonen et al. (Mikkonen, Parikka, Ghafar &

578   Tenkanen, 2013) has presented that the application of polysaccharide-based aerogels in food

579   section would be food packaging rather than food edible productions. Some polysaccharides

580   are incorporated into aerogel food packaging, since they present the excellent degradable

581   properties. Safoura Ahmadzadeh et al (Ahmadzadeh, et al., 2015) fabricated cellulose

582   nanocomposite aerogels with surface-modified montmorillonite as substitute for foamed

583   polyurethane materials for dry food packaging. Aleksandra Nešić et al. (Nešić, et al., 2018)

584   developed pectin-TiO_2 nanocomposite aerogels with thermal insulation and antimicrobial

585   properties, which could be used as packaging for temperature-sensitive food. In the recent

586   studies of Jean Paulo De Oliveira et al. (De Oliveira, et al., 2019), hybrid

587   PVA/cellulose/nanocellulose aerogels have the ability to control the release of bioactive

588   extracts, which is conductive to the application in the production of active food packaging

589   materials.

590

591   **4. Current research status of polysaccharide-based aerogels**

592   According to an advanced search in ScienceDirect.com, the statistics data of published

593   papers on polysaccharide-based aerogels with respect to the publication year present the

594   research status in recent years. As can be seen, the data were sorted by polysaccharide type,

595   different fields and countries. In this work, the countries of literature were determined

596   according to the first author's first affiliation institution. If there are co-first authors, the first

597   one in the author list will be considered as the first author. **Fig. 4 (a)** shows the popular

598   tendency of various categories of polysaccharide-based aerogels from 2011 to the beginning

599   of 2019. Although aerogel has been invented in 1931, polysaccharide-based aerogels have

600   attracted the attention of researches in recent years for the demand of sustainable

601   development. It can be seen that there is an obvious increase in 2014, and then followed with

602   sustainable growth until now. For the first half of 2019, 36 publications can be found. In

addition, another important data can be found that cellulose aerogels occupied the majority of the total number of published papers during the period from 2013 as shown in **Fig. 4 (a)**. Cellulose is one of the most abundant natural polysaccharide which could be derived from cotton, straw and plant. It has been demonstrated to be nontoxic and good-biocompatible in both animal and human, which attracts the attention of researchers. In addition, the excellent chemical and mechanical properties contribute to the wide spectrum use of cellulose. Cellulose has been converted into green aerogels and assumed the structure roles in polysaccharide-based aerogels with extremely low density and high surface area. According to different application fields, the literature numbers ratio of polysaccharide-based aerogels are presented in **Fig. 4 (b)**. **Fig. 5** was a statistical comparison of publications on polysaccharide-based aerogels with respect to countries. Polysaccharide-based aerogels was paid a greater attention to by the Chinese researchers and it took a large proportion (49.5%) of the total areas. The major proportion of countries of research institutes distribution is taken by USA, France, Germany and Slovenia, with 6.9%, 5.3%, 4.7% and 3.7%, respectively. Country contribution could put forward the needs and inputs analysis of polysaccharide-based aerogels in the world. These information could provide the researchers with a reference on the tendency.
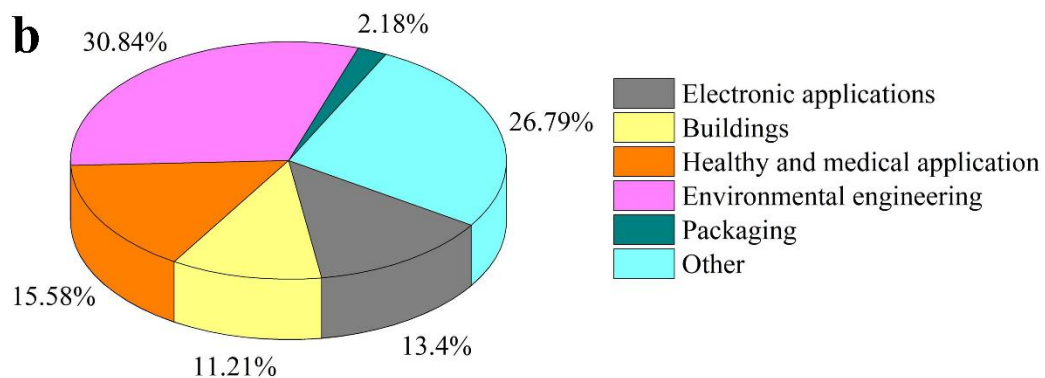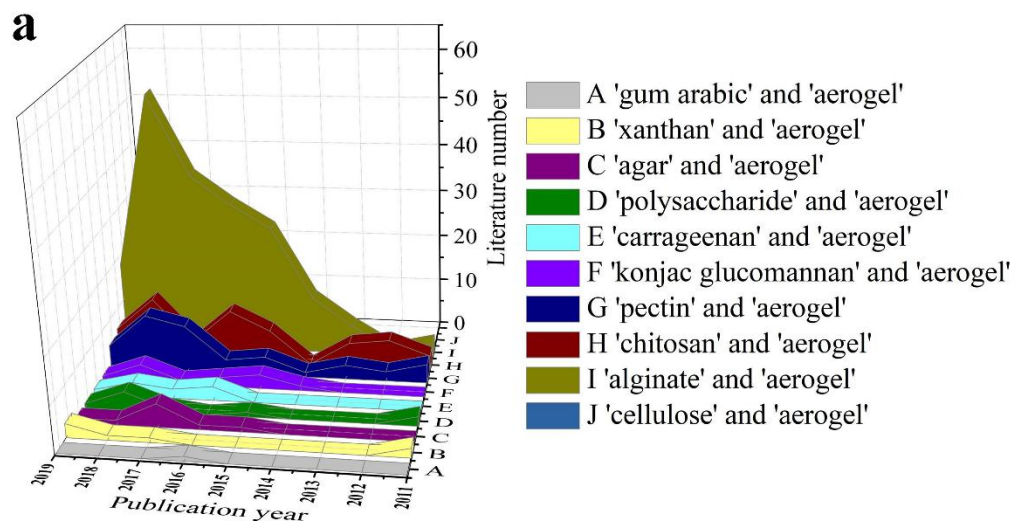
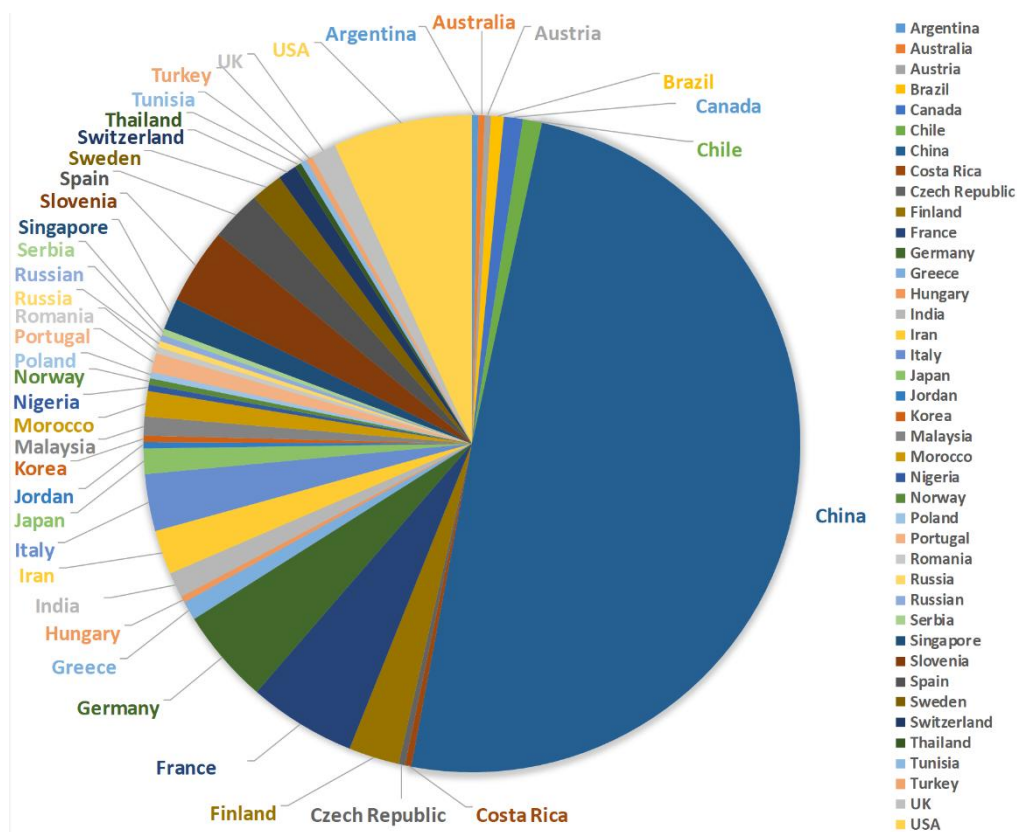**Fig. 4** Polysaccharide-based aerogel studies according to publication year, polysaccharide type (a) and different fields (b)

Fig. 5 Polysaccharide-based aerogel studies according to countries of research institutes (total number of publications: 321)

5. Conclusion, challenges and perspectives

Polysaccharide-based aerogels belong to the new generation of porous materials and have been regarded as the most sustainable aerogel material because of its advantages of sustainability, low toxicity, biocompatibility and renewability. The procedure include extraction of raw materials (polysaccharide), modification of raw materials, preparation of hydrogel and dehydration (supercritical drying and freeze drying method). Polysaccharide-based aerogels have significant application prospects in fields including environmental engineering, buildings, medicine, electrochemical components and packagings. The numbers of published papers with respect to publication years, regions and applications have been counted and analyzed. The publication number on polysaccharide-based aerogels keeps a stable growth from 2011 to 2014 and then from 2014 to the beginning of 2019, a significant

640 increase appeared. Regional distribution of those published papers is presented by a pie chart,

641 which shows China is the main research region of polysaccharide-based aerogels.

642   By summarizing the research on polysaccharide-based aerogels in recent years, it can be

643 seen that low-density, high specific surface area polysaccharide-based aerogels are widely

644 used in various fields due to their excellent functions. Optimizing the performance of

645 polysaccharide-based aerogels that meet the needs of the current society will be a hot spot for

646 future research. With the increase of environmental pollution, polysaccharide-based aerogels

647 can also exert their specific advantages, to replace traditional materials for benefiting

648 mankind. However, there is still improvement space for further researches on polysaccharide-

649 based aerogels. Based on the literature above, several recommendations can be put forward

650 which may provide clear guidance for the future development of polysaccharide-based

651 aerogels:

652   *(1) Use of natural agricultural waste as raw materials and improving performance.* Most

653 polysaccharide are combined with some conventional inorganic and organic materials to

654 prepare polysaccharide-based aerogels with the advantages of both natural and synthetic

655 materials. However, we should promote the use of agricultural waste (e.g., straw and leaf) to

656 prepare polysaccharide-based aerogels. The agricultural waste has large output, wide sources,

657 cheap price and specific natural physical structure. Some of this structure could be obtained

658 at micro-scale or even nano-scale, contributing to the performance improvement of

659 polysaccharide-based aerogels. In recent years, the promotion has been reflected in some

660 publications, such as the use of discarded banana peels, straws with the natural network,

661 porous cavity to prepare polysaccharide-based aerogels, although the qualities and

662 performances should be enhanced.

663   *(2) Increase the hydrophobicity.* In practical applications, the use of polysaccharide-based

664 aerogels is far from enough. There is some challenges in developing polysaccharide-based

aerogel materials, such as the hydrophobicity. Generally, polysaccharides are hydrophilic materials. The problem from weather, water and microorganism will follow, contributing to destroy and degrade polysaccharide-based aerogels. So researchers need to pay attention to increase the hydrophobicity. In recent years, some chemical methods have been found to increase the hydrophobicity, but most of these methods will do harm to our environment. We need to get a balance between hydrophilic and hydrophobic which could refer to the bionics, such as lotus leaves with specific structure non-polar methyl groups. When the balance between hydrophilic and hydrophobic could be adjusted as wanted, the use of scope and shelf life of aerogels will be expanded.

(3) Production scale expansion. The preparation is easy to be achieved in the lab, but it is a challenge in industrialization. There is still a problem in preparing large scale supercritical drying equipment and there is a risk to operate for production in large scale. Therefore, in order to achieve the mass production in industry and application in broad fields, it is urgent to find ways to fabricate and optimize equipment as well as adjust the operating parameters to achieve large capacity and continuous production

(4) Product form/shape design. Polysaccharide-based aerogels have been used in many fields as new functional materials. In order to meet the requirement of the market, polysaccharide-based aerogel products with different size and shape need to be designed and manufactured. The shape of aerogels could be roughly divided into the following categories, block, sheet panel, balls and beads. In the buildings, block aerogels are needed, and it is difficult to prepare directly. When producing large-sized aerogels, it is prone to cracking due to stress concentration. How to prevent cracking will be a challenge in the future. In some certain specific applications, a folded aerogel with high surface area is required, such as an air filter. It is necessary to produce aerogels with a certain degree of softness for folding. Finally, for spherical and bead aerogels, continuous production is a challenge that depends on

690   the design of the production mold and the production line.

696

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