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Title: Fabrication and characterization of a novel konjac glucomannan-based air filtration aerogels strengthened by wheat straw and okara

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Abstract: The konjac glucomannan (KGM)-based aerogel as an air filtration material was fabricated through sol-gel and freeze-drying methods. Results showed that gelatin and starch addition could increase the filtration efficiency and compressive strength of aerogel significantly, due to the appearance of more microporous structure and the formation of dense structure in aerogel. The addition of wheat straw could decrease the filtration resistance and increase the breathability of KGM-based aerogel, which was attributed to the multi-cavities of wheat straw. The aerogel with wheat straw had a filtration efficiency of 93.54% for particle matters $\geq 0.3 \mu\text{m}$, a filtration resistance 29 Pa, and an air permeability 271.42 L/s·m². Okara addition could increase the hydrophobicity of KGM-based aerogel by increasing the water contact angle and decreasing the equilibrium water content. The water contact angle of the aerogel containing okara reached 105.4°, and the equilibrium water content was decreased by 17.03%-81.10% compared with that without okara, with relative humidity 0%-80%. The results demonstrated that the KGM-based aerogel had good performance on filtration, mechanical and hydrophobic properties, indicating high potential application as an air filtration material.

1. KGM-based aerogel with good filtration and hydrophobic properties was prepared.
2. Starch and gelatin addition enhanced mechanical and filtration property of aerogels.
3. Wheat straw addition improved filtration resistance and gas permeability of aerogels.
4. Okara addition could improve aerogel hydrophobicity.

1 **Fabrication and characterization of a novel konjac glucomannan-based**
2 **air filtration aerogels strengthened by wheat straw and okara**

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28 ABSTRACT

29 The konjac glucomannan (KGM)-based aerogel as an air filtration material was fabricated through
30 sol-gel and freeze-drying methods. Results showed that gelatin and starch addition could increase the
31 filtration efficiency and compressive strength of aerogel significantly, due to the appearance of more
32 microporous structure and the formation of dense structure in aerogel. The addition of wheat straw
33 could decrease the filtration resistance and increase the breathability of KGM-based aerogel, which
34 was attributed to the multi-cavities of wheat straw. The aerogel with wheat straw had a filtration
35 efficiency of 93.54% for particle matters $\geq 0.3 \mu\text{m}$, a filtration resistance 29 Pa, and an air
36 permeability $271.42 \text{ L/s}\cdot\text{m}^2$. Okara addition could increase the hydrophobicity of KGM-based aerogel
37 by increasing the water contact angle and decreasing the equilibrium water content. The water contact
38 angle of the aerogel containing okara reached 105.4° , and the equilibrium water content was decreased
39 by 17.03%-81.10% compared with that without okara, with relative humidity 0%-80%. The results
40 demonstrated that the KGM-based aerogel had good performance on filtration, mechanical and
41 hydrophobic **properties**, indicating high potential application as an air filtration material.

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43 Keywords: Konjac glucomannan; Air filtration aerogel; Wheat straw; Okara; Mechanical property;
44 Hydrophobic property

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55 **1. Introduction**

56 In recent decades, the fast economic growth of modern society has accompanied with serious
57 environmental air pollution, threatening humans' health and life (Chow, & Judith, 2006). As the main
58 cause of air pollution, harmful particles in the air come from many aspects and are mainly divided into
59 three categories according to their types: physical pollution (particulate matter, dust, pollen, etc.),
60 chemical pollution (SOX, nitrogen oxides and volatile organic compounds, etc.) and biological
61 contamination (bacteria, mold spores, viruses, etc.) (Landrigan, 2017; Pope, et al., 2002; Anderson,
62 Thundiyil, & Stolbach, 2012). PM_{2.5} (particle sizes < 2.5 μ m) is the main cause of air pollution
63 (Brunekreef, & Hoffmann, 2016), and could seriously threaten people's health (Cohen, et al., 2005;
64 B.R. Gurjar, et al., 2010; Russell, & Brunekreef, 2009).

65

66 Air filtration is the most effective way to solve air pollution problems (Sutherland, 2008). Various
67 filter materials have been used for air filtration, such as fiber filter materials, composite filter materials,
68 and functional filter materials (Antonicelli, Bilò, Pucci, Schou, & Bonifazi, 1991). Fiberglass and
69 quartz fiber are used for air filtration with 95% filtration efficiency (Akbarnezhad, Amini, Goharrizi,
70 Rainey, & Morawska, 2017). Nano-TiO₂ photocatalytic materials have shown attractive application
71 prospects in air purification, and it can **absorb harmful gases in air** (CO, SO₂, NH₃, NO_x, and VOC),
72 achieving the purpose of sterilization and air filtration without secondary pollution (Suarez, et al.,
73 2011). Activated carbon fibers, nanofibers, and photocatalytic materials are often used as air filtration
74 materials in air conditioning systems (HVAC) (Tang, et al., 2018; Park, Yoon, & Hwang, 2011;
75 Pigeot-Remy, et al., 2014). These air filter materials not only have a limited source of raw materials
76 but are also not environmentally friendly. Therefore, there is an urgent need to develop new
77 environmentally friendly air filter materials.

78

79 As classical porous materials, aerogels are considered to be good air filtration materials due to their
80 continuous three-dimensional network structure, adjustable density, high specific surface area, and

81 high porosity (Kim, Chase, & Jana, 2015). Plant polysaccharide aerogels, such as cellulose aerogels
82 (Shi, Lu, Guo, Liu, & Cao, 2015; Xu, Bao, Xu, Wang, & Sun, 2015), starch aerogels
83 (García-González, Uy, Alnaief, & Smirnova, 2012) and sodium alginate aerogels (Wang, et al., 2016),
84 not only have the physical properties of aerogel, but also **their** raw materials have abundant resources,
85 good biosafety, and environmentally friendly advantages. However, the problems of poor mechanical
86 and hydrophobic **properties** limit the application of plant polysaccharide aerogels for air filtration
87 purposes (Zhu, Hu, Jiang, Liu, & Li, 2018). Konjac glucomannan (KGM) is a high molecular weight
88 water-soluble polysaccharide (Fang, & Wu, 2004; Davé, & McCarthy, et al., 1997), and it was
89 suitable for aerogel preparation with high specific surface area (as high as 51.8 m²/g) (Jiang, 2013;
90 Wang et al., 2017). Gelatin is rich in hydroxyl, carboxyl and amino groups in its molecular chain,
91 making it easy to gel and functionalize, and so it can be the starting material for constructing a 3D
92 structure (Wang, et al., 2016). Porous gelatin networks for tissue engineering, flame retardancy,
93 oil/water separation, and contaminant adsorption have been developed (Kang, Tabata, & Ikada, 1999;
94 Huang, et al., 2017; Li, et al., 2016), and incorporating biobased gelatin to poly (vinyl alcohol) / clay
95 aerogels could improve aerogel strength and flame retardancy (Wang, et al., 2017). Starch has a
96 special retrogradation phenomenon **that the starch molecules will rearrange into ordered crystals when**
97 **fully gelatinized starch is cooled at a lower temperature or slowly dehydrated and dried**
98 (Jiamjariyatam, Kongpensook, & Pradipasena, 2014). As a by-product of wheat, wheat straw is
99 usually incinerated and causes environmental pollution, however, it can be also used to produce air
100 filtration materials (Wang et al., 2017). Okara is a by-product of soy milk or tofu and contains a large
101 amount of insoluble dietary fiber residue (Mateos-Aparicio, Redondo-Cuenca, Villanueva-Suárez,
102 Zapata-Revilla, & Tenorio-Sanz, 2010). In the okara, the dietary fiber content reaches 50% to 70%, fat
103 content is 8% to 11%, and protein content is 19% to 23% (Redondo-Cuenca, Villanueva-Suárez, &
104 Mateos-Aparicio, 2008). Appropriate addition of different polymers to the composite material could
105 improve functional properties (Corobea et al., 2016), and therefore aerogels with air filtration function
106 may be produced with these environmentally friendly materials (KGM, gelatin, starch, wheat straw,
107 okara). **The study aimed to investigate the pore structure, mechanical and filtration properties (DEHS**

108 (dioctyl sebacate) as an aerosol for filtration property) of KGM/gelatin/starch aerogels, and the
109 filtration and hydrophobic properties of KGM/gelatin/starch-based aerogels strengthened by wheat
110 straw and okara. This study can contribute to the research and application of KGM-based aerogels as
111 air filtration material.

112

113 2. Materials and method

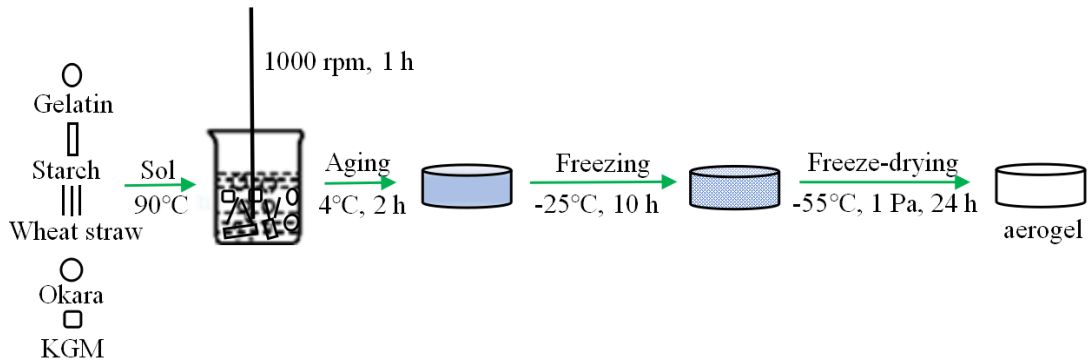
114 2.1. Materials

115 Konjac glucomannan (KGM) was supplied by Licheng Biological Technology Co., Ltd. (Wuhan,
116 China). Potato starch (S) was obtained from Wuhan Lin He Ji Food Co., Ltd. (Wuhan, China). Gelatin
117 (G) was purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). Raw wheat straw
118 (WS) and okara (O) were obtained from farmhouses in Wuhan. Both the raw wheat straw and okara
119 were ground into flours by a grain pulverizer and screened through a 160 mesh sieve before use.

120

121 2.2. KGM-based aerogel preparation

122 The preparation of KGM-based aerogel was based on the previous research (Wang, 2018) with minor
123 modification as illustrated in Fig. 1. Gelatin, starch, wheat straw, okara, KGM were dissolved in
124 double-distilled water (90 °C) in order and stirred at a speed of 1000 rpm for 1 h to mix the entire
125 solution. And then the sol was injected into two different sizes of cylindrical mold (diameter 34.8 mm
126 and height 18 mm, diameter 142 mm and height 10 mm) and placed in a 4 °C refrigerator for aging for
127 2 h, after that, it would be placed in a -25 °C ultra-low temperature refrigerator for 8 h. **The frozen**
128 **samples were put in a vacuum freeze dryer (Modulyod-230, Thermo Electron Corporation, USA)**
129 **(-55 °C, 1 Pa) for 24 h to be completely freeze-dried.** Aerogel samples were coded as the form of
130 K0G0S0WS(O)0, and the number after the letter indicates the mass percentage of the component. All
131 aerogel samples were stored in a drying vessel (50 °C) for 12 h before use.



132

133

Fig. 1. Schematic procedure of preparing KGM-based aerogels.

134

135 2.3. Characterization

136 2.3.1. Filtration performance test

137 The filtration efficiency, filtration resistance and breathability of samples were tested using a LZC-K1
 138 type filter comprehensive performance test bench (LZC-K1, Suzhou Huada, China). The test bench
 139 mainly included test channels (including filter fixtures), flow control units, atomized aerosol
 140 generators, particle counters, pressure gauges, fans, and other components, as well as control units and
 141 data acquisition software. The effective test area was 15 cm × 15 cm, and the filtration efficiency and
 142 filtration resistance were tested by feeding an electrically neutral monodisperse DEHS aerosol to
 143 samples at a median diameter of 0.3-10 μm. The filter piezoresistive force was coordinated by the
 144 flowmeter and two electronic pressure sensors. The breathability test was performed by measuring the
 145 air flow rate through the sample per unit area under a pressure of 200 Pa and converted it into the air
 146 permeability. All filtration tests were performed at 24 ± 2 °C, the flow rate of filtration was 32 L/min
 147 for resistance and filtration efficiency tests, and the time for each filtration test was 1 min.

148

149 2.3.2. Microstructure and pore size distribution

150 Prior to test, aerogel samples were cut into small pieces (5 mm × 5 mm × 1 mm). The samples were
 151 fixed on a stainless steel sample stage with conductive paste before sputtered with gold for 80 s (JFC
 152 1600, JEOL Ltd, Japan). Then the surface microstructure was observed by a scanning electron

153 microscopy (SEM) (JSM6390LV, JEOL, Japan) at magnifications of $\times 50$, $\times 100$, $\times 500$, $\times 850$, $\times 1000$.
154 The pore size distribution was evaluated by Image Pro Plus software (Media Cybernetics Inc,
155 Maryland, America), and for each aerogel sample, six representative SEM images were used.

156

157 2.3.3. Mechanical property

158 The mechanical property test of aerogel samples was determined by a Texture analyzer (TA. XT Plus,
159 Stable Micro Systems, Surrey, UK) equipped with a flat bottom probe (No. 10585), based on the
160 method in previous research (Wang, 2018) with minor modification. Double compression mode was
161 adopted with compression percentage 30% and compression rate 1.00 mm/s, and the trigger force was
162 1.00 N. The parameter of hardness was determined, which was the maximum force (F) during the first
163 cycle of compression. S represents the initial area (mm^2) of samples in contact with the probe, so the
164 stress (σ) was calculated by the following standard equation (Eq. (1)):

$$165 \sigma = \frac{F}{S} \quad (1)$$

166

167 2.3.4. FTIR analysis

168 Attenuated total reflection was collect at 25 °C by using a **fourier transform infrared spectroscopy**
169 **(FTIR)** spectrometer (VERTEX 70, Bruker Co., Ltd Germany) equipped with a horizontal **attenuated**
170 **total reflectance (ATR)** in the range of $4000\text{-}650 \text{ cm}^{-1}$. Data were collected in 32 scans at a resolution
171 of 4 cm^{-1} .

172

173 2.3.5. Water contact angle

174 The water contact angle measurements of aerogel samples were tested at 25 °C by a contact angle
175 analyzer (DSA25, Krüss Co., Ltd, Germany) equipped with a **charge coupled device (CCD)** camera
176 and an image analysis software. The contact angle was measured after the water droplets ($5.0 \mu\text{L}$)
177 were deposited on the aerogel samples surface ($2.0 \text{ cm} \times 1.0 \text{ cm}$) for 10s (Jin, Han, Li, & Sun, 2015).
178 The angle was measured from 0° to 180° with a measurement accuracy of $\pm 0.3^\circ$. The drop image was
179 recorded by the CCD camera.

180

181 2.3.6. Moisture adsorption isotherm

182 **Dynamic vapor sorption (DVS)** apparatus (Surface Measurement Systems, London, UK) was used to
183 obtain the moisture adsorption curve of aerogel samples at 25 °C. A weight change (dm/dt) of less than
184 0.002%/min over 10 min was chosen as the criterion for reaching equilibrium at each relative
185 humidity (RH) step and then increasing to the next rise or descending RH.

186

187 2.3.7. Dry density and porosity estimation

188 The obtained aerogel weight (m) was determined by an analytical balance (ME204, METTLER
189 TOLEDO, China), and the volume (v) was calculated by its size determined by a vernier caliper. The
190 density (ρ) of the aerogel is calculated by the following formula (Eq. (2)):

191
$$\rho = \frac{m}{v} \tag{2}$$

192 Aerogel porosity was estimated based on the method in previous research (Kim, Park, Kim, Wada, &
193 Kaplan, 2005) with minor modification. The aerogel sample was first immersed in ethanol of known
194 volume V1 for 5 min. The volume of the aerogel impregnated with ethanol and ethanol was recorded
195 as V2, and the aerogel impregnated with ethanol was removed. The volume of ethanol is V3, and the
196 porosity (ϵ) is obtained by the following formula (Eq. (3)):

197
$$\epsilon \quad (\%) = \frac{(V1-V3)}{(V2-V3)} \times 100\%$$

198 (3)

199

200 All experimental data points were analyzed and drawn figures using Origin 2017 (Originlab
201 Corporation, Northampton MA) and Microsoft Excel 2010. One-way analysis of variance (ANOVA)
202 was performed using **statistical product and service solutions (SPSS)** (21th edition, Endicott, NY,
203 USA) and the significance of each average property value was determined by measuring Tukey's
204 multi-range test ($p < 0.05$).

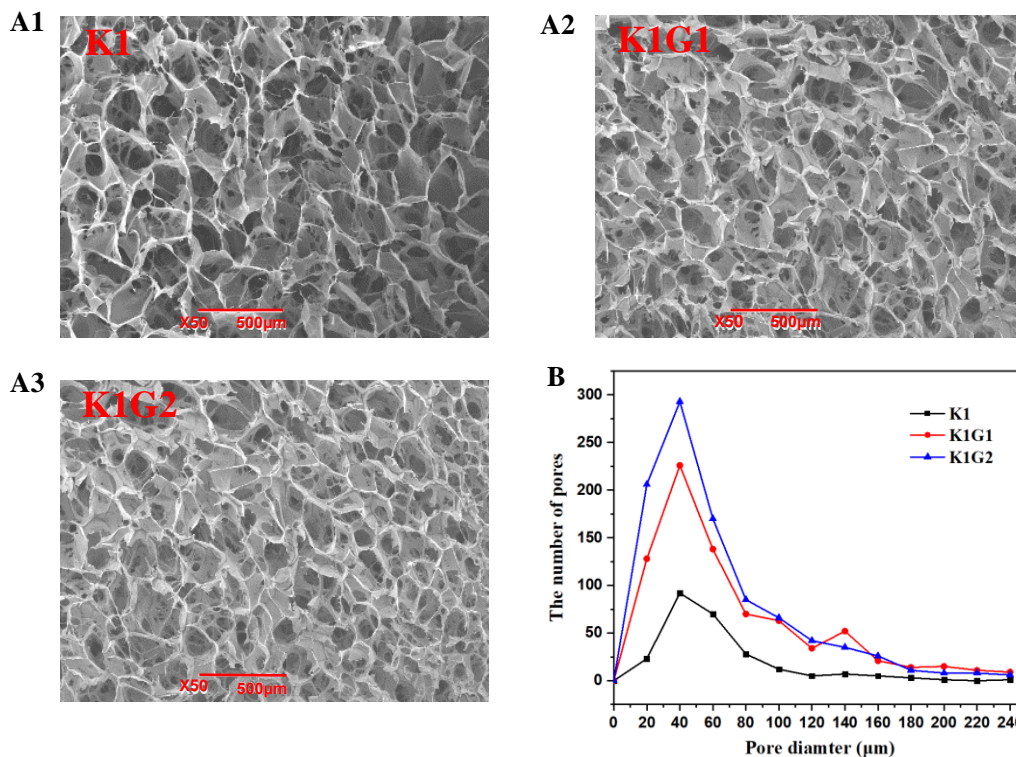
205

206 **3. Results and discussion**

207 3.1. Impact of gelatin on the structure, filtration and mechanical properties of KGM/gelatin aerogel

208 The formation of ice crystals in the sol led to concentration and aggregation of the solute molecules,
209 and aerogel sample shape was maintained by the aggregated solute molecules during ice crystal
210 sublimation in the lyophilization process, forming a porous network structure (Gutiérrez, Ferrer, & del
211 Monte, 2008). Different network structures might be formed with different solute. As shown in Fig.
212 1(A1), the SEM image indicated pure KGM aerogel (K1) had a porous three-dimensional network
213 structure, consistent with the previous report (Ni et al., 2016). To demonstrate the impact of gelatin on
214 the change of the pore structure of KGM-gelatin aerogel, SEM images and size distribution (0-240 μm)
215 curves of pores were drawn (Fig. 1(A, B)). Compared with K1, gelatin addition of 1% (K1G1), 2%
216 (K1G2) could bring more micropores and increase pore numbers with pore sizes 0-80 μm by 316.19%,
217 387.044%, respectively. Therefore, the higher the concentration of gelatin, the higher the number of
218 aerogel pores (0-80 μm) in the range of 0-2%. Gelatin gels changed from disordered single-stranded
219 structure to ordered structure during the formation process with the intrachain hydrogen bonds and
220 interchain hydrogen bonds as the main force, however, the presence of KGM disordered the gelatin
221 coil-helix transition, and this might cause the system to be loose, leading to more pores in the
222 KGM/gelatin aerogel (Khomutov, Lashek, Ptitchkina, & Morris, 1995; Kuijpers, 1999; Jin, Xu, Ge, Li,
223 & Li, 2015).

224



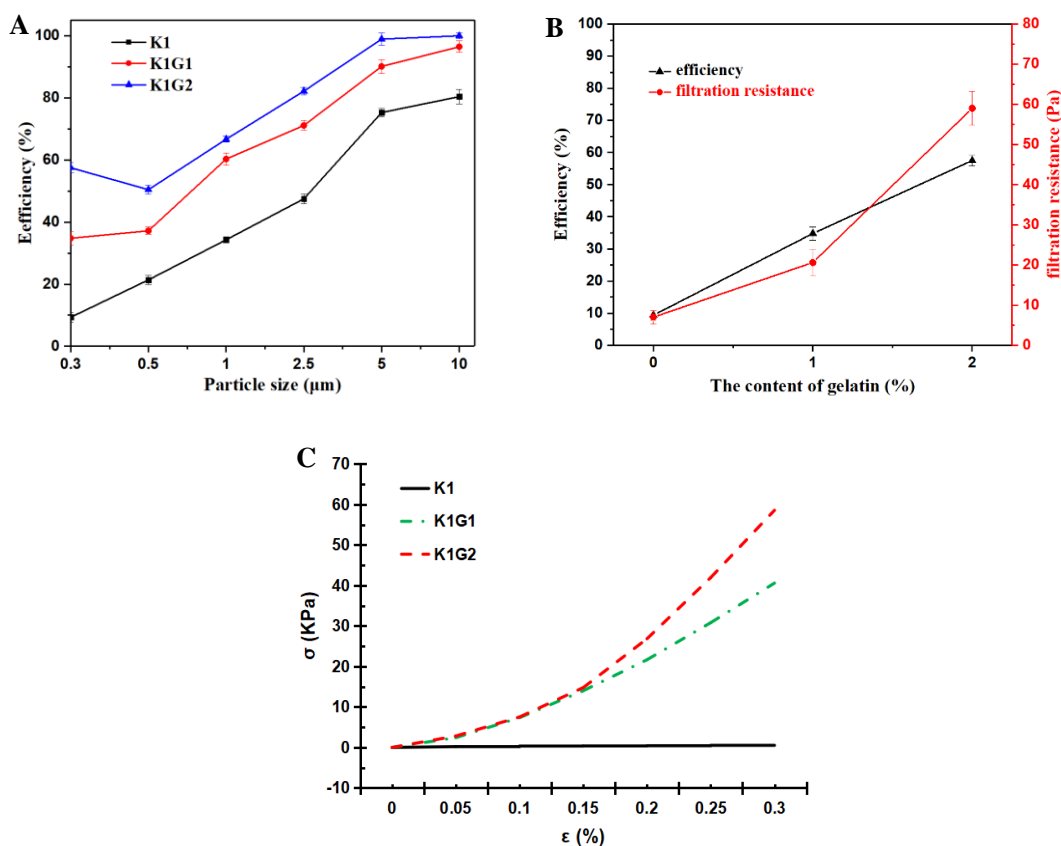
225

226 **Fig. 2. (A1-A3) SEM images of KGM/gelatin aerogels under magnification 50×; Size distribution**
 227 **(0-240 μm) of KGM/gelatin aerogels pores with different gelatin concentration.**

228

229 The effect of gelatin addition on KGM/gelatin aerogel filtration efficiency is shown in Fig. 3A. With
 230 increased addition of gelatin (1%-2%) (w/v), the filtration efficiency of KGM/gelatin aerogel
 231 gradually increased. When further gelatin addition increased to 2%, the filtration efficiency of K1G2
 232 aerogel increased to 57.511% (particle size $\geq 0.3 \mu\text{m}$). Fig. 3B showed the filtration resistance of
 233 KGM/gelatin aerogel. The filtration resistance of K1 aerogel without gelatin was 7.015 Pa, and with
 234 the addition of gelatin, the filtration resistance gradually increased, e.g. **the filtration resistance of**
 235 **aerogel with 2% gelatin increased to 59 Pa (Fig. 3B).** This was due to the fact that the addition of
 236 **gelatin could increase the number of small holes (0-80 μm) on the pore wall of KGM/gelatin aerogel**
 237 **(Fig. 2), which might increase the probability of internal inertial collision and Brownian motion of**
 238 **particles (Hutten, 2007), improving the filtration efficiency (Wang & Shen, 2004) and filtration**
 239 **resistance.** Improvement in mechanical property is very important for filter materials (Calis Acikbas et

240 al., 2017), and the stress-strain curve (strain 0-30%) of KGM/gelatin aerogel is shown in Fig. 3C.
 241 When the addition amount of gelatin was increased from 0% to 1%, the compressive strength was
 242 significantly increased, and then it increased slowly with further gelatin addition from 1% to 2% (w/v).
 243 The stress of gelatin-added aerogels increased significantly, e.g. from 0.6142 kPa (K1) to 40.5777 kPa
 244 (K1G1) and 58.5590 kPa (K1G2). This might be explained by that gelatin and KGM formed an
 245 interpenetrating network, and the gel network was enhanced via covalent cross-linking between the
 246 complexes (Suo et al., 2018; Liu, Li, Zhang, Li, & Hou, 2018). Therefore, the addition of gelatin not
 247 only improved the filtration efficiency of KGM-based aerogel but also increased the compressive
 248 stress, facilitating the practical application of KGM-based aerogel as a filter material.
 249



250
 251 **Fig. 3. (A) Filtration efficiency of aerogels with different gelatin concentration for various**
 252 **particle sizes; (B) Filtration efficiency and filtration resistance of aerogels (K1Gn, n=0, 1, 2) for**
 253 **particle matters of 0.3 μm and beyond; (C) Stress-strain curves for KGM/gelatin aerogels with**

254 **different gelatin concentration.**

255

256 3.2. Impact of starch on the structure, filtration property of KGM/starch aerogel

257 SEM images of KGM/starch aerogels with different starch concentration are shown in Fig. 4. All

258 aerogel samples exhibited a complete, uniform three-dimensional network structure. With increased

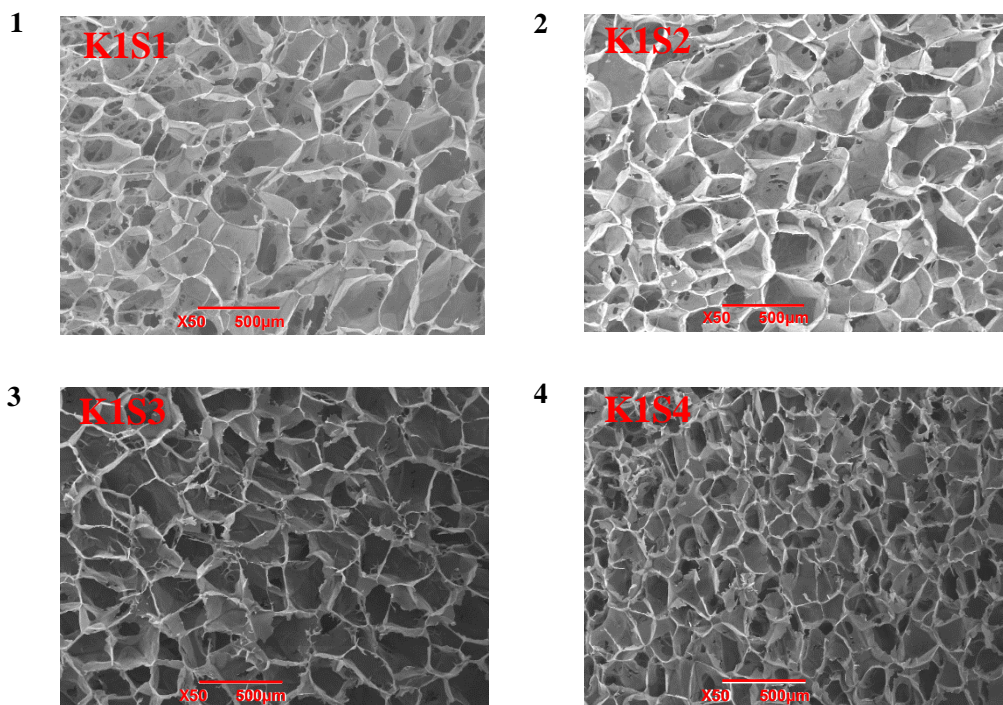
259 starch concentration (1%-4%), the pores became smaller, and pores on the pore wall became fewer.

260 The pores were the smallest and the structure was densest when starch concentration was 4%. This

261 could be interpreted as the starch concentration increased, the molecular distance of the system

262 became smaller, reducing spaces for ice crystal growth, and therefore aerogel structure became denser

263 with smaller pores (Qian, Chang, & Ma, 2011).



264

265

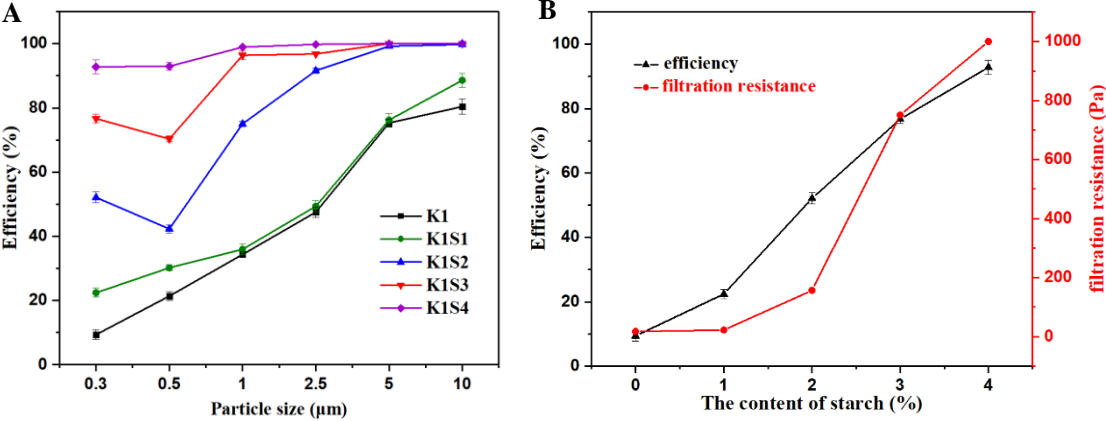
266 **Fig. 4. (1-4) SEM images of KGM/starch aerogel under magnification 50×.**

267

268 The effect of starch on the filtration efficiency and filtration resistance of KGM/starch aerogel is

269 shown in Fig. 5(A-B). The filtration efficiency of KGM/starch aerogel (starch concentration: 1%-4%

270 (w/v) was gradually increased (Fig. 2(A1)), and the filtration efficiency was maximized when starch
 271 addition reached 4% (92.78%), but the filtration resistance was overload (>1000 Pa). Based on
 272 previous research, the addition of starch could increase the pores with pore sizes range 10-50 μm
 273 (Wang et al., 2018), and this might cause an increase in the probability of particles colliding in the
 274 aerogel, consuming the kinetic energy of the particles to achieve interception (Lifshutz, & Pierce,
 275 1997). Considering the high resistance is not conducive to the practical application of air filtration
 276 material (Wang, Yu, Lai, & Chung, 2018), starch addition was $\leq 3\%$ in the following experiment.
 277



278
 279
 280 **Fig. 5. (A) Filtration efficiency of KGM/starch aerogels with different starch concentration for**
 281 **various particle sizes; (B) Filtration efficiency and filtration resistance of aerogels (K1Sn, n=0, 1,**
 282 **2, 3, 4) for particle matters of 0.3 μm and beyond.**

283
 284 3.3. Filtration property of KGM/starch/gelatin aerogel

285 To optimize the component ratio of KGM/gelatin/starch aerogel based on the filtration efficiency, an
 286 $L_9(3^3)$ orthogonal array was tested and an optimized aerogel formulation was obtained (Table 1). The
 287 highest filtration efficiency was 94.41% (K1G1S3), and the lowest filtration efficiency was 20.40%
 288 (K1S1). According to the filtration efficiency, k and range values were calculated, and the results
 289 showed the following sequence: starch > gelatin > KGM. The optimized aerogel formula was

290 K1G1S3 and was used in the following experiments. Its filtration efficiency was 94.41%, and the
 291 compression stress was 241.698 kPa.

292

293 **Table 1**

294 **Analysis of $L_9(3)^3$ test results about filtration efficiency.**

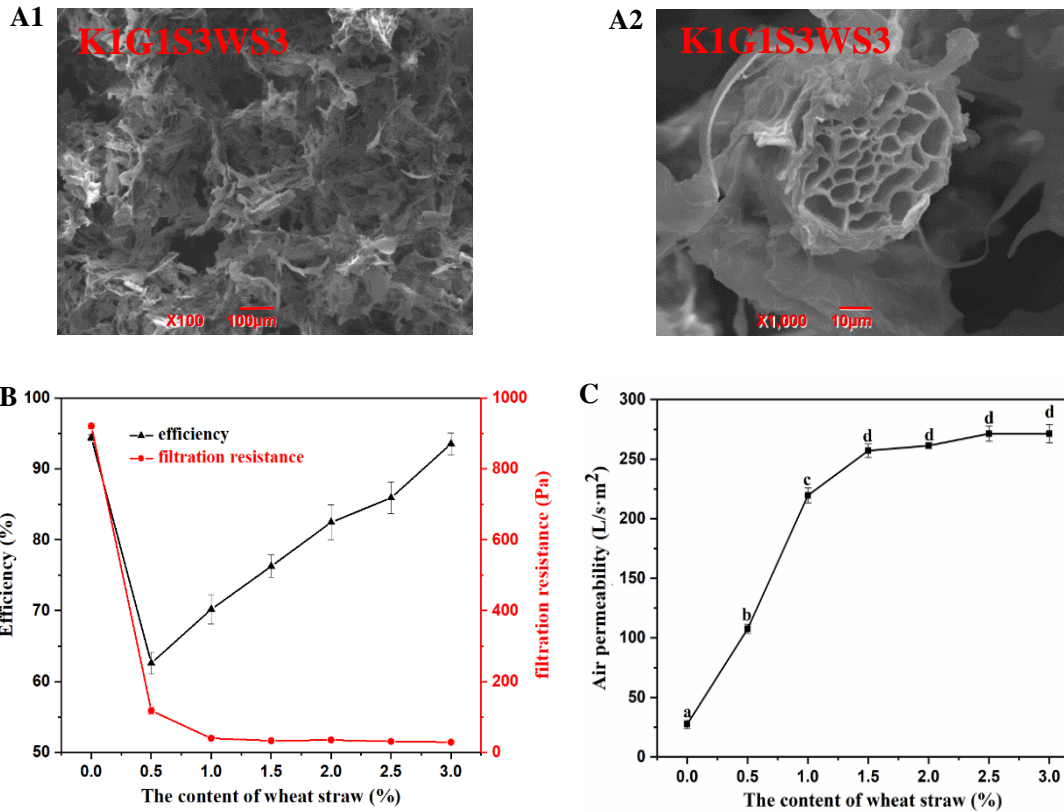
Sampel code	KGM	Gelatin	Starch	Filtration Efficiency
	(g/100mL)			(Mean \pm SD) (%)
K0.5G2S1	0.5	2	1	62.74 \pm 1.6015
K0.5G1S2	0.5	1	2	76.47 \pm 0.5950
K0.5S3	0.5	0	3	88.63 \pm 0.7204
K1S1	1	0	1	22.40 \pm 1.4300
K1G2S2	1	2	2	81.35 \pm 0.4800
K1G1S3	1	1	3	94.41 \pm 0.3953
K1.5G1S1	1.5	1	1	68.73 \pm 0.8265
K1.5S2	1.5	0	2	73.89 \pm 0.3955
K1.5G2S3	1.5	2	3	82.04 \pm 0.3869
k1	75.95	75.38	51.29	
k2	66.05	79.87	77.24	
k3	74.89	61.64	88.36	
range	9.9	18.23	37.07	
Optimal level		S > G > K		
Major factor (w/v)	1%	3%	1%	
Optimized formula		K1G1S3		94.41 \pm 0.3953

295

296 3.4. Impact of wheat straw on the structure and filtration property of KGM/gelatin/starch aerogel

297 Wheat straw in aerogel (K1G1S3WS3) had multi-cavities structure and the pore structure was
 298 irregular in SEM images (Fig. 6(A1-A2)). The filtration property of KGM/gelatin/starch/wheat straw
 299 aerogel is shown in Fig. 6B. As wheat straw concentration increased from 0% to 0.5% (w/v), the
 300 filtration efficiency was reduced from 94.41% to 62.59% (particle matters \geq 0.3 μ m), and the filtration

301 resistance was reduced from 921 Pa to 117.67 Pa. The filtration efficiency of aerogel (K1G1S3WS2.5)
302 was increased to a maximum value of 93.54% (particle matters $\geq 0.3 \mu\text{m}$). The filtration resistance
303 was continued to decrease until below 50 Pa when wheat straw concentration $\geq 0.5\%$ (w/v). Air
304 permeability is also an important indicator of filter materials, affecting the filtration efficiency of filter
305 materials (Woudberg, Theron, Lys, & Le Coq, 2018). The air permeability of aerogel with wheat
306 straw addition is shown in Fig. 6C. With increased addition of wheat straw (0%-1.5% (w/v)), the air
307 permeability started to increase significantly ($27.33\text{-}257.02 \text{ L/s}\cdot\text{m}^2$), and then it became to change
308 slightly when the wheat straw addition was further increased from 2% to 3% (w/v). The highest air
309 permeability ($271.42 \text{ L/s}\cdot\text{m}^2$) of aerogel (K1G1S3WS3) was reached with 3% wheat straw addition,
310 and the density and porosity were $0.1050 \pm 0.0008 \text{ g/cm}^3$ and $92.13 \pm 0.04\%$, respectively. Similar to
311 wood cells, wheat straw is also a porous material with the micro cellular structure (Strømdahl, 2000),
312 thus the pore structure of K1G1S3 aerogel might be affected due to cavity structure of wheat straw,
313 resulting in a decrease in the filtration efficiency of the aerogel. However, the micro cellular structure
314 also increased microchannel inside aerogel, so the filtration efficiency (Liu et al., 2019) and air
315 permeability (Wang, Cai, Yang, & Yang, 2018) increased with increased wheat straw concentration.
316



317

318 Fig. 6. (A) SEM images of K1G1S3WS3 under magnification 100× (A1), 1000× (A2). (B)

319 Filtration efficiency and filtration resistance of aerogels (K1G1S3WS_n, n=0, 0.5, 1, 1.5, 2, 2.5, 3)

320 for particle matters of 0.3 μm and beyond; (C) Air permeability of aerogels (K1G1S3WS_n, n=0,

321 0.5, 1, 1.5, 2, 2.5, 3), data points with the different letter are significantly different.

322

323 3.5. Impact of okara on the structure and hydrophobic property of KGM/gelatin/starch aerogel

324 The impact of okara addition on the hydrophobicity improvement of aerogel was studied based on

325 K1G1S3 aerogel sample. The pore shape of K1G1S3O₂ aerogel was more disordered than K1 (Fig.

326 2(A1)), and a special structure of agglomeration occurred in Fig. 7(A2), by the fact that the special

327 lumpy structure of insoluble dietary fiber in okara (Mateos-Aparicio, Mateos-Peinado, & Rupérez,

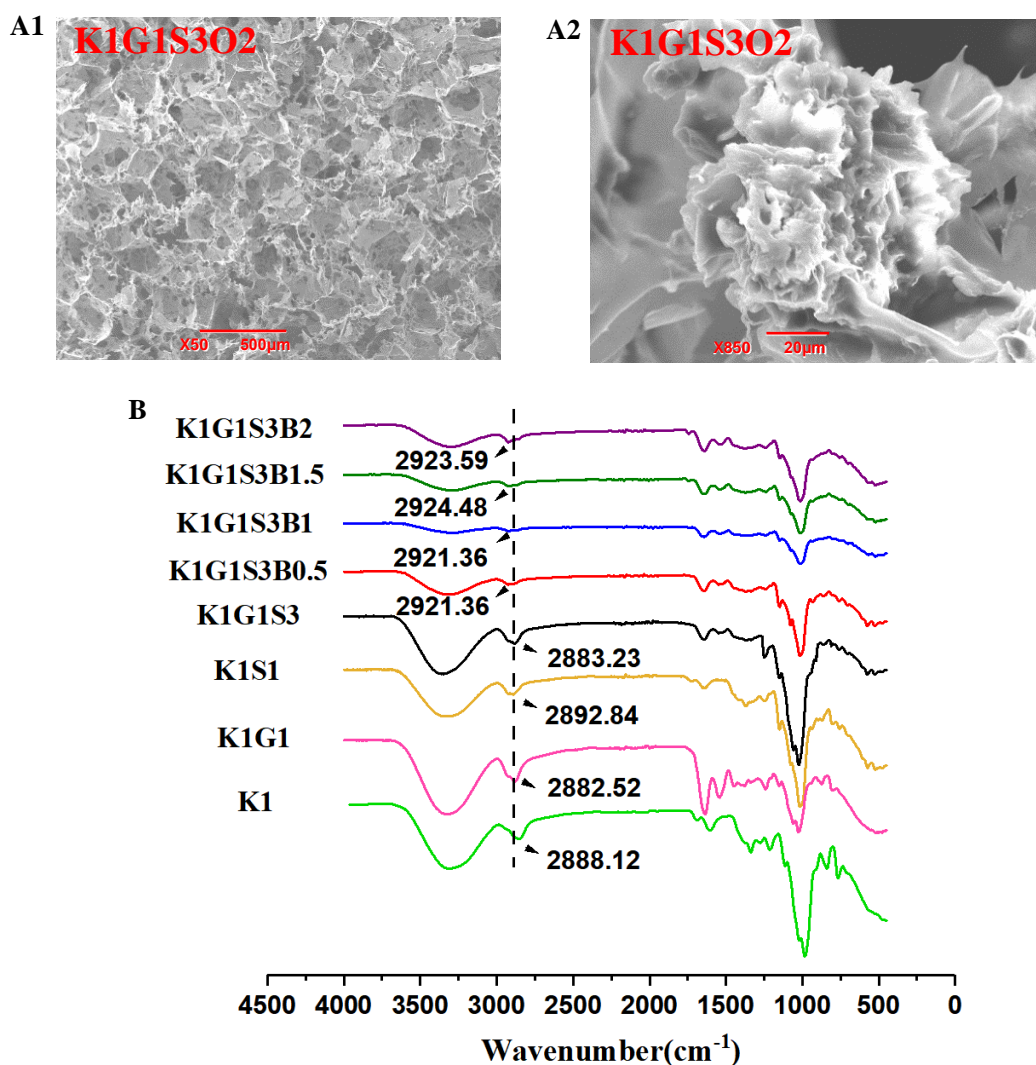
328 2010) was uniformly dispersed in the aerogel and caused shape changes of the pore structure of the

329 aerogel (Kiani & Sun, 2011). The analysis of the FTIR spectra is shown in Fig. 7B. The stretching

330 bands of 2923.59, 2924.48, 2921.36, 2925.63, 2883.23, 2892.84, 2882.52, and 2888.12 cm⁻¹ were

331 assigned to C-H. Comparing the spectra of K1, K1G1, K1S1, K1G1S3 and K1G1S3O_n aerogels

332 (n=0.5, 1, 1.5, 2), the addition of okara caused a shift of the C-H stretching bands to the higher
 333 frequencies (“blueshift”), which may be caused by hydrophobic interaction of the methyl groups
 334 (Schmidt, Dybal, & Trchová, 2006). The insoluble components in the okara might act as a special
 335 structure in Fig. 7(A2) in aerogel and the aerogel might be therefore hydrophobic. The density and
 336 porosity of K1G1S3O2 were $0.0752 \pm 0.0009 \text{ g/cm}^3$ and $90.30 \pm 0.05\%$, respectively.
 337

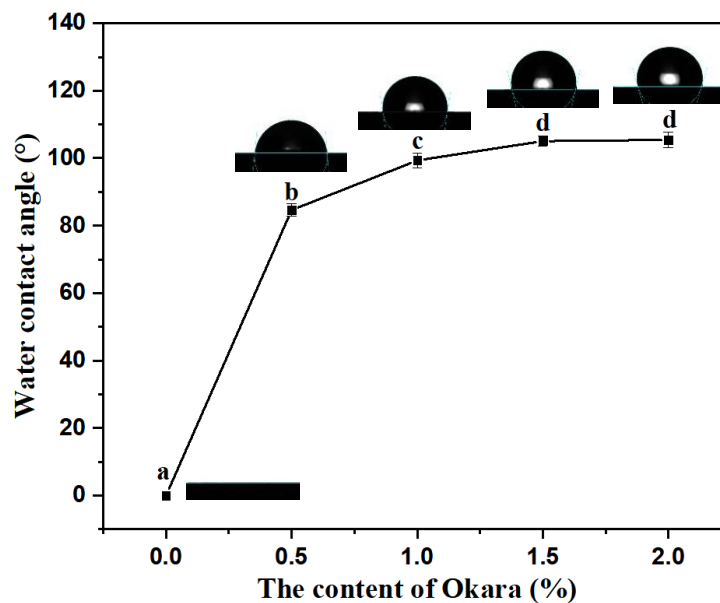


338
 339 **Fig. 7. (A) SEM images of K1G1S3O2 under magnification 50× (A1), 850× (A2); (B) FT-IR**
 340 **spectra of aerogels (K1, K1G1, K1S1, K1G1S3, and K1G1S3On, (n=0.5, 1, 1.5, 2)).**

341

342 Generally, the greater the water contact angle, the higher the surface hydrophobicity (Yin et al., 2014;

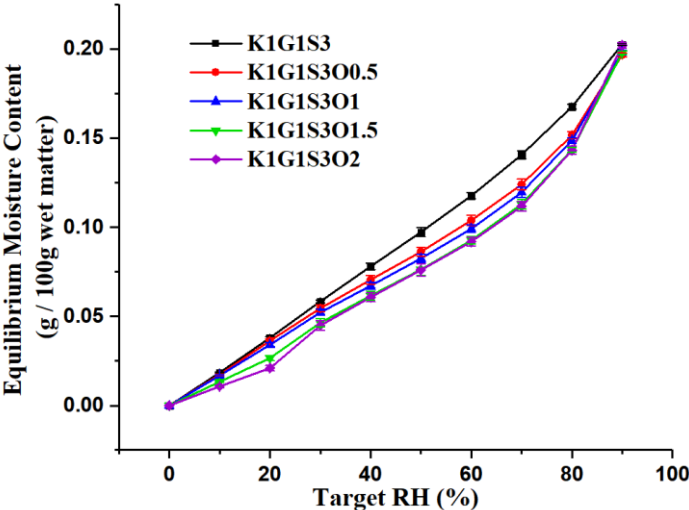
343 Escamilla-García et al., 2013). The effect of okara addition on the water contact angle of
344 KGM/gelatin/starch aerogel is shown in Fig. 8. The water contact angle of the aerogel without okara
345 addition (K1G(1-2), K1S(1-4), and K1G1S3) was 0°. K1G1S3 aerogel is composed of polysaccharide
346 and proteins with high polar groups, which easily destroyed the cohesion of water molecules and
347 resulted in a low water contact angle (Kaity et al., 2013). With okara concentration increased from 0%
348 to 1.5% (w/v), the water contact angle began to significantly increase. Further increase of the okara
349 concentration (1.5% to 2.0%) resulted in a slight increase of the water contact angle till reaching the
350 maximum value 105.4° (2% (K1G1S3O2)). The material with water contact angle $\geq 90^\circ$ is
351 hydrophobic and has good hydrophobicity (Chen, Wang, & Shi, 2017; Wu et al., 2017; Scaffaro,
352 Sutera, & Botta, 2018). The presence of okara containing insoluble protein might increase the amount
353 of non-polar substances on the surface of aerogel, which increased the water contact angle.



354
355 **Fig. 8. Water contact angle of aerogels (K1G1S3On, n=0.5, 1, 1.5, 2), data points with the**
356 **different letter are significantly different.**

357
358 The moisture adsorption isotherms (Fig. 9) showed Type II-b shape according to Blahovec and
359 Yanniotis's research classification (Blahovec, & Yanniotis, 2009), which was consistent with the
360 moisture adsorption isotherms of most materials (Mohammadi Nafchi, Moradpour, Saeidi, & Alias,

361 2014; Bingol, Prakash, & Pan, 2012). The experiment results showed that the aerogel with different
362 content of okara all exhibited less equilibrium water concentration compared with K1G1S3 aerogel in
363 the ranges of RH 0%-80%, and the equilibrium water content of K1G1S3O2 was reduced by
364 17.03%-81.10% compared with K1G1S3. This further demonstrated that hydrophobicity of
365 KGM-based aerogel with okara was improved.
366



367
368 Fig. 9. Water adsorption isotherms of aerogels (K1G1S3O_n, n=0.5, 1, 1.5, 2) at 25°C determined
369 by DVS.

370

371 **4. Conclusions**

372 The KGM-based aerogel with enhanced filtration, mechanical and hydrophobic properties was
373 prepared. Gelatin and starch components caused the appearance of more microporous pore structure
374 and the formation of the dense structure of KGM-based aerogel network, which could improve the
375 mechanical and filtration properties of KGM-based aerogel. The addition of wheat straw could
376 decrease the filtration resistance and increase the breathability of KGM-based aerogel, which was
377 attributed to the multi-cavities of wheat straw. Okara addition could make KGM-based aerogel more
378 hydrophobic by increasing surface water contact angle and decreasing equilibrium water content of
379 aerogel. The data revealed that aerogel containing 3% wheat straw (K1G1S3WS3) has a filtration

380 efficiency $93.54 \pm 1.5450\%$ (particle matters (DEHS) $\geq 0.3 \mu\text{m}$), a filtration resistance 29 Pa, an air
381 permeability $271.42 \text{ L/s}\cdot\text{m}^2$, and a compressive strength 241.698 kPa. The water contact angle of the
382 aerogel containing 2% (w/v) okara (K1G1S3O2) reached the maximum value 105.4° , and the
383 equilibrium water content of K1G1S3O2 was 17.03%-81.10% lower than K1G1S3, with RH 0%-80%.
384 This study enhanced the practicality of KGM-based aerogel as air filtration material.

385

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390

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