Facile and green fabrication of cellulose based aerogels for lampblack filtration from waste newspaper

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Abstract:

In this study, the lightweight, hydrophobic and porous cellulose-based aerogels (CAGs) were synthesized through a freeze-drying process using waste newspaper as the only raw material. After crosslinking with glutaraldehyde and treatment with trimethylchlorosilane (TMCS) using a simple thermal chemical vapor deposition process, the resulting CAGs became hydrophobic and oleophilic. Furthermore, the as-prepared CAGs exhibited a low density (17.4–28.7 mg cm⁻³) and mesoporous inner-structure. All these properties attributed the novel aerogel not only with a good adsorption capability of oils and organic solvents, including kerosene, nitrobenzene, and chloroform, but also an excellent filtration capacity of lampblack.

Keywords: Waste newspaper; Cellulose; Aerogel; Cellulose based aerogel

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1. Introduction

Inhalation of particulates matter is recognized as the 9th most powerful environmental risk hazard for development and aggravation of diseases, including various lung diseases like asthma and COPD, i.e., chronic obstructive pulmonary disease. (Lim et al., 2012). Recent toxicological (Elsaesser & Howard, 2012) and epidemiological studies (Meng et al., 2013; Strak et al., 2012) suggest that nanoparticles ($D_p \leq 100$ nm) fraction in ambient particulate air pollution is considered as the most harmful component when it comes to pulmonary uptake. They may cause serious health risks because of their high mobility and increased toxicity due to their large specific surface areas. In addition to health concerns, when emitted into the environment, they may potentially lead to new hazard compounds with increased risks to the environment (Maynard & Pui, 2007; J. Wang, Kim, & Pui, 2011). Recent experimental data pointed out that oleosols are more pernicious than solid particles. In particular, the lampblack issued from routine cooking cause great concern in populous and developing countries, such as China. A recent study suggested that oil smoke issued from food handling accounts for 16% of exhaust pollution. (Maynard & Pui, 2007; Oberdorster, Oberdorster, & Oberdorster, 2005) To overcome the problem, aerosol filtration process is therefore used in diverse applications, such as air pollution control, reduction of emission, human respiratory protection, and processing of hazardous materials (Hinds, 2012).

Another driving force for aerosol filtration processes is the growing necessity of clean air environments utilized by many advanced industries, such as electronics, medical, pharmaceuticals, biological research, gas turbine and nuclear energy installations, automotive applications, among others (P. Tronville & R. Rivers, 2005; P. Tronville & R.

D. Rivers, 2005). In regard to this, the filtration of airborne nanoparticles is becoming an important and urgent issue to harness as they are produced in large quantities from several sources, including material synthesis, combustion, emission, and even daily life. HEPA (High-efficiency particulate air Filter) mainly produced from materials like polypropylene filter, glass microfiber, composite PP/PET filter, melt-blown terylene non-woven fabric and melt-blown glass fiber, have been demonstrated to be efficient in reducing the hazards of particulates to health and the environment. However, the high energy consumption, high cost and the elevated emitted pollutants involved in the production of HEPA tremendously limited its wide and sustainable applications.

As the most abundant organic polymer, cellulose is regarded as an almost inexhaustible bio-resource, which can be transformed into various useful products. This was witnessed by the numerous investigations performed during the last decade, notably in ultra lightweight cellulose aerogels due to their sustainability, biodegradability and potential applications in fields of tissue engineering, controllable delivery system, blood, and water purification (Ma, Mi, Yu, He, & Zhang, 2014; Moon, Martini, Nairn, Simonsen, & Youngblood, 2011). Since its first report in 2001 (Tan, Fung, Newman, & Vu, 2001), cellulose-based aerogel has attracted extensive interest as a novel kind of green biodegradable nanoporous materials, combined unique features of native cellulose and the excellent properties of traditional silica aerogels, such as ultralow density, high porosity and high specific surface area. To the best of our knowledge, a few of methods, including the critical point drying, directional freeze-casting and freeze-drying (Dash, Li, & Ragauskas, 2012; Fumagalli, Sanchez, Boisseau, & Heux, 2013; Heath & Thielemans, 2010), had been employed to prepare the aerogels, whereas distinct deficiency presented,

such as low robustness, low flame retardancy (Moon, Martini, Nairn, Simonsen, & Youngblood, 2011).

Compared to expensive cotton linters and wood pulp, lignocellulose is available in abundant amounts and at low-cost, thus became an attractive alternative. In this respect, the underutilized agricultural wastes, woody biomass, and related resources supply are considered as promising feedstock for cellulose industry. For example, waste newspapers (WNPs), contributing to about 7% of municipal solid waste, is a typical waste biomass material containing low-cost cellulosic material (Subhedar, Babu, & Gogate, 2015). In China alone, for example, millions of tons of WNPs are generated annually. Currently, most WNPs are simply used as conventional and low-value products, including recycled newspaper, boxboard, compost, paper insulation, and animal bedding. Therefore, it would be valuable if commonly available biomass resources like WNPs could effectively be employed to prepare high-value cellulose-based materials.

During the past years, few attempts were made to prepare cellulose materials from WNPs. Jin et al. (Jin, Han, Li, & Sun, 2015) successfully prepared cellulose-based aerogels by treating WNPs with trimethylchlorosilane (TMCS). The resulting aerogels exhibited good absorption performance for oils and organic solvents. Using waste newspaper as the only raw materials, Han et al (Han, Sun, Zheng, Li, & Jin, 2016) synthesized the lightweight, hydrophobic and porous carbon aerogels through freeze-drying and post-pyrolysis processes. The synthesized aerogel showed excellence ability in harvesting organic pollutants, including gasoline and methylbenzene. Recently, Xia et al (Xia et al., 2016) fabricated transparent films from ionic liquid-regenerated WNPs.

Herein, the lightweight, hydrophobic oleophilic and porous aerogels were fabricated by a freezing-drying method using WNP as the raw material. After being treated with trimethylchlorosilane (TMCS) via a simple thermal chemical vapor deposition process, the resulting CAGs were rendered both hydrophobicity and oleophilicity. Meanwhile, the CAG's robustness could be improved by crosslinked with the glutaraldehyde solution. Impressively, the obtained aerogel not only can absorb typical organic solvents and oil, but also exhibit comparable filtration capacity of the commercial HEPA membrane for lampblack.

2. Experimental

2.1 Materials and reagents

Waste newspapers (WNPs) were obtained from the market. Fully bleached, commercial kraft softwood pulp with degree of polymerization of 220 was purchased from Beijing Fengli Jingqiu Commerce and Trade Co., Ltd. All the cellulose samples used were dried in vacuum at 100°C for 12 h before using. The ionic liquid 1-Allyl-3-methylimidazolium chloride (AmimCl) was kindly supplied by Prof. Jun Zhang, Institute of Chemistry, Chinese Academy of Sciences. Glutaraldehyde (GA), trimethylchlorosilane (TMCS), from Aladdin Shanghai, China. All other chemicals were of analytical grade and used as received.

PP nonwovens (basis weight 50 g/m²) were provided by Tianjin Teda Co., Ltd. China. HEPA (High efficiency particulate air Filter), AP1101, 3M, US.

2.2 Pretreatment of waste newspaper and the Preparation of cellulose based aerogel (CAGs):

Fig.1 present the fabrication process of the cellulose-based aerogel from the waste

newspapers. The WNP were pretreated by a series of chemical extraction, in order to remove ink and glue (H. Y. Wang, Li, & Zhang, 2013). Initially, WNP were cut into small strips (1–2 cm) and treated with 2 wt% sodium hydroxide (NaOH) solution at 90 °C for 2 h. Subsequently, the samples were cyclically treated using the acidified sodium chlorite (NaClO₂) solution at 75 °C for 2 h. Then, the pretreated WNP (2 g) were dispersed into the AmimCl (18 g), and the dispersion was subject to vigorous mechanical

Fig. 1. Schematic illustration of CAGs preparation.

stirring at 80 °C for 3 h. The resultant was thoroughly rinsed with DI water, ethanol, respectively, for at least 6 times to remove the residual AmimCl completely. For every rinsing step, the dispersion was centrifuged (12000 rpm, 10 min) to remove the upper liquid. The gray-green suspension obtained after last centrifugation step was placed inside dialysis membrane tubes (12000-14000 molecular weight cut-off) and dialyzed against slow running DI water for 1-2 days. The membrane tubes containing the extracted cellulose were placed periodically in DI water, and the procedure was continued until a

neutral pH was reached. Finally, they were frozen below $-18\text{ }^{\circ}\text{C}$ and then transferred to a lyophilizer (FD-1CE, Boyikang Co. Ltd., Beijing) with a condenser temperature of $-55\text{ }^{\circ}\text{C}$ and inside pressure 20-30 Pa. After 2-day lyophilization process **CAG0** were obtained.

2.3 Preparation of crosslinked CAGs:

The resultant CAGs hereinbefore is flexible but fragile. Therefore, to enhance the mechanical robustness, 20 uL 37% glutaraldehyde aqueous solution was added dropwise to the regenerated and dialyzed cellulose while stirred in water bath at $50\text{ }^{\circ}\text{C}$. After 2-day lyophilization, the CAG was obtained. (**CAG1**, hereinafter).

2.4 Hydrophobic modification of CAGs:

The hydrophobicity of the CAGs is improved further via the surface silanation reaction procedure as reported by Jin et al. (Jin et al., 2015). Briefly, a small open glass vial containing TMCS (1 mL) was placed in a vacuum desiccator together with CAGs. Then the desiccator was heated and sealed in a vacuum oven at $70\text{ }^{\circ}\text{C}$ for 12 h for the silanation reaction. In order to remove the excess amount of unreacted silane and the HCl byproduct, the coated CAGs were kept in a vacuum desiccator under vacuum for more than 1 h (**CAG2**, hereinafter).

2.5 Characterization

Density and porosity: The density was calculated by weighting the sample and measuring its volume and the porosity of the sample (P , %) was calculated by the following equation:

$$P(\%) = (1 - \frac{\rho}{\rho^*})$$

where ρ is the density of obtained aerogel, ρ^* is the skeleton density of cellulose,

which is about 1.59 g cm⁻³ (Heath & Thielemans, 2010). Each sample was measured in triplicate and the average value was adopted.

Sorption of organic solvents and oils by the CAGs: A series of organic reagents and oils were used to study the sorption capacity (Q_t , %) of the aerogels, and the Q_t can be assessed through the following equation:

$$Q_t = \frac{M_t - M_0}{M_0}$$

where m_t (g) is the weight of the CAGs after sorption in the selected solvent or oil, and m_o (g) is the initial weight of the CAGs.

Powder X-ray diffraction (XRD) data were collected by a Bruker D8-Advance with CuK α radiation ($\lambda = 0.1542$ nm) at a scan rate (2θ) of 2 °/min with the accelerating voltage of 40 kV and the applied current of 30 mA ranging from 5° to 40°. Scanning electron microscopy was performed on a field emission SEM (Hitachi S-4800) at an accelerating voltage of 10 kV.

Fourier transform infrared spectroscopy was carried out on a Nicolet VERTEX 70 (Bruker Corp.) spectrometer using the attenuated total reflectance (ATR) method at a resolution of 4 cm⁻¹. The spectra were generated and collected 16 times and corrected for the background noise.

Nitrogen physic-sorption measurements were performed by a Quantachrome NOVA 2000e at 77 K, and the BET analysis was done for relative vapor pressure from 0.05 to 0.3. From the obtained isotherms the BET surface area and the *BJH* (Barrett–Joyner–Halenda) pore size distribution was calculated from the desorption branch of the isotherm.

The filtration efficiency and air flow resistance (pressure drop in mm H₂O column

height) were measured by an automated filter tester TSI 8130 (TSI incorporation, Shoreview, MN, US). The tester could deliver neutralized micron monodisperse oil aerosol (with mass median diameter of ca. 0.33 micron, count median diameter of ca. 0.2 μm , and a geometric standard deviation not exceeding 3.12), and oil aerosols were fed into a filter holder with an effective area of 100 cm^2 . All filtration tests were conducted with a continuous air flow of 32 l/min at room temperature. The total aerosol concentration ranges from $10^6/\text{cm}^3$ to over $10^8/\text{cm}^3$. Structurally, the tester uses polydispersed oil particles and two photometers to measure total mass concentrations up- and downstream of the filter. The photometers rely on light scattering from multiple particles to obtain a relative concentration measurement. The signal voltage is proportional to the mass of aerosol sampled by the photometer.

3. Results and discussion

Cellulose is a polysaccharide consisting of β -1,4-*D*-linked glucose chains, in which the glucose units are joined by single oxygen atoms (acetal linkages between the *C-1* of a glucose unit and *C-4* of the next unit) (Klemm, Heublein, Fink, & Bohn, 2005). Some of the characteristic IR peaks of cellulose attribution are located at $\sim 900\text{ cm}^{-1}$ assigned to the β -glucosidic linkage between the glucose units, $\sim 1160\text{ cm}^{-1}$ of C-O symmetric stretching of primary ethanol, and $\sim 2900\text{ cm}^{-1}$ attributed to stretching vibration of C-H stemmed from the CH_2 group.

Fig. 2. FTIR spectra of the CAG0, CAG1 and CAG2 (A), and comparative FTIR spectra of CAG0 and neat cellulose.

As shown in the FTIR spectra, the dissolution and subsequent regeneration induced a broadening and shifting of the peaks towards higher wavenumbers, particularly the characteristic absorption band of O–H stretching vibration located at $\sim 3343\text{ cm}^{-1}$ (**Fig. 2A**). This implied the AmimCl-induced breakage of the hydrogen bonds in the preparation of the cellulose-based aerogels (F. C. Wang, Fève, Lam, & Pascault, 1994). It is noteworthy that, comparing with the neat cellulose, the absence of the semicellulose indicative ($\sim 1634\text{ cm}^{-1}$, C=O stretching vibration) and the lignin indicative ($\sim 1661\text{ cm}^{-1}$, $\sim 1622\text{ cm}^{-1}$, all C=O stretching vibration) suggest all three aerogels are comprised mostly of cellulose (**Fig. 2B**) (Cao, Li, Zhang, Zhang, & He, 2010; Himmelsbach, Khalili, & Akin, 2002; Kaparaju & Felby, 2010). Moreover, the weakening and downshifting of the $\sim 1451\text{ cm}^{-1}$ band indicated that there was a crystal form transformation (from cellulose I to cellulose II) during the dissolution and regeneration process (Oh et al., 2005). For crosslinking aerogel CAG1, $\sim 1705\text{ cm}^{-1}$ was assigned to the characteristic absorption band for glutaraldehyde; however, the same assignment was appeared at $\sim 1671\text{ cm}^{-1}$ and $\sim 1621\text{ cm}^{-1}$ for CAG2. For hydrophobically modified CAG2, a series of strong peaks appeared at $\sim 798\text{ cm}^{-1}$, $\sim 1257/1436\text{ cm}^{-1}$ and $\sim 2926\text{ cm}^{-1}$. These peaks were attributed to the Si-O symmetrical stretching, methylene's symmetric/or asymmetric bending, and methylene's stretching vibration, respectively (Xia et al., 2016).

In order to figure out the crystalline phases, the aerogels were analyzed by X-ray

234 diffraction and the results are shown in **Fig. 3**. The XRD profile of CAG0 exhibited a

235

236 **Fig. 3.** XRD curves of CAG0, CAG1 and CAG2.

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238 distinct peak at $2\theta = 22.2^\circ$, corresponding to the (002) lattice planes of cellulose I.

239 Moreover, two overlapping peaks at $2\theta = 14.0^\circ \sim 17.8^\circ$, corresponding to the (101) and

240 $(10\bar{1})$ lattice planes of cellulose I crystalline structure could be noticed (Zheng, Cai, Ma,

241 & Gong, 2015). On the other hand, the XRD profile of CAG1 and CAG2 displayed broad

242 peaks ranging from 20° to 21.5° , stemmed from overlapped two peaks of cellulose II

243 centered at 20.1° (110) and 21.9° (200), as well as the amorphous cellulose peak at $\sim 17.3^\circ$

244 (Oh et al., 2005). For CAG1, a small but well-resolved peak located at $\sim 20^\circ$ assigned to

245 (102) diffraction was also noticed (Morandi, Heath, & Thielemans, 2009). It is

246 noteworthy that for CAG2, the distinct peak located at $\sim 12.0^\circ$ (2θ) was attributed to the

247 lattice planes of SiO_2 inorganic additives (Xia et al., 2016).

Fig. 4. SEM images with low (×5000) and high magnification (×30000) of CAGs. (A-B) CAG0; (C-D) CAG1 and (E-F) CAG2. EDS element mapping of CAG2 which clearly demonstrated the silicon element.

The SEM images of the aerogels are shown in **Fig. 4**. At both low (×5000) and high magnification (×30000), the aerogels showed smooth morphology with no obvious

distinguished difference. In other words, all samples exhibited interconnected, highly porous and nanofiber-like structure. This implied that the dissolution, subsequent regeneration, and even the hydrophobic modification, was failed to bring about a detectable influence on microstructure of the cellulose aerogel. For TMCS modified CAG-2, the presence of a signal of silicon element (ca. 1.74 Kev) (Fig. 4G) demonstrated its incorporation in the composite matrices, which is well consistent with the FTIR data.

The internal structure of the aerogel samples was characterized by N₂ adsorption/desorption experiments (Fig. 5). The isotherms shape points of neat cellulose aerogel, CAG0, and modified aerogels CAG1 and CAG2 could be categorized to type IV character according to the IUPAC classification with a small hysteresis loop (Fig. 5A). This may suggest that the mesoporous structures of the obtained aerogels and the N₂ hysteresis have some pore blocking and percolation within the porous network (Shopsowitz, Hamad, & MacLachlan, 2011).

Fig. 5. Nitrogen adsorption/desorption isotherms (A) and Barrett–Joyner–Halenda (BJH) pore-size distribution (B) of neat cellulose aerogel, CAG0, and CAG1, CAG2 calculated from the desorption branch of the isotherm.

The estimated average density (mg cm^{-3}) of the CAGs varied from 17.36 to 28.68, consistent with densities reported for other cellulose aerogels (Heath & Thielemans, 2010; Liebner et al., 2010). On the other hand, the porosity decreased **slightly** as the density increased (**Table 1**). In addition, the amount of nitrogen adsorbed between 0.05 and 0.3 was utilized to calculate the BET surface area with the assumption of adsorbed monolayer. Interestingly, the values of BET surface areas of CAG1 and CAG2 were greater than that of CAG0, while the pore volume of CAG0 was slightly larger when compared to those of CAG1 and CAG2 (**Fig. 5B and Table 1**). The distribution of pore size calculated from the desorption branch of the isotherm using Barrett-Joyner-Halenda (BJH) model yielded pore sizes ranging from 8 nm to 13 nm with the following order: CAG0 > CAG1 > CAG2 (**Fig. 5B**).

Table 1 Characteristic parameters for CAGs

Sample	Density (mg cm^{-3})	Porosity (%)	S_{BET} ($\text{m}^2 \text{g}^{-1}$)	Pore volume ($\text{cm}^3 \text{g}^{-1}$)	Pore size (nm)
CAG0	17.4	98.9	296	0.97	13.1
CAG1	23.1	98.5	412	0.78	7.6
CAG2	28.7	98.2	405	0.82	8.1

After glutaraldehyde crosslinking and crosslinking/TMCS treated simultaneously, the CAG1 and CAG2 displayed enhanced mechanical properties (in strength and modulus) and hydrophobicity comparing with the CAG0 (**Fig. 6**). Specifically, the CAG2 indicates a water contact angle of ca.135°, which is markedly higher than that of CAG0 and CAG1. The lightweight, porous structure and the excellent hydrophobicity/oleophilicity of CAG

Fig. 6. The stress-strain curves (A) and water contact angles (B) of CAG0, CAG1 and CAG2.

make it an ideal candidate for highly efficient separation/extraction of organic pollutants and oils. In order to quantitatively assess the sorption capacity of CAGs, the weight gain (wt%) was defined as the weight of absorbed substances per unit weight of the original CAGs. A series of organic solvents and oils were investigated, including chloroform, benzene, nitrobenzene, as well as some commercial oils, such as kerosene, gasoline, lubricant, used engine oil, and soybean oil. When compared to CAG0, the crosslinked and chemically modified CAG2 exhibited an outstanding sorption property, with weight gain as high as 2600–4500 wt% (**Fig. 7**). Obviously, CAG2 exhibited a rather high absorption capacity of all investigated oils and organic solvents.

Fig. 7. Sorption capacity of CAGs for oils and various organic solvents. The tests are performed with a duration of 1 min in the selected solvents or oils at room temperature (22°C).

In addition to its excellent sorption capability towards organic pollutants and oils, the resulting CAGs also displayed a good filtration performance. As a proof of concept, the total mass concentrations up- and downstream of the filter were examined using polydispersed oil particles. As shown in **Fig.8**, all CAGs exhibited better filtration performance than PP nonwovens in term of filtration efficiency (%). CAG2 showed a

Fig. 8. The filtration performance of PP nonwovens, HEPA and three kinds of cellulose based aerogels, CAG0, CAG1 and CAG2.

filtration efficiency of about 82.4 %, which was comparable to that of HEPA filter (~86.3 %). Meanwhile, the CAGs possessed lower pressure drop (ca. 32.4-41.6 mm H₂O) as compared to that of HEPA filter (~44.0 mm H₂O). The filtration efficiency and pressure drop are key parameters of fibrous filters, and a combination of high filtration efficiency with low-pressure drop is an indication of a good filter (Jiao et al., 2015; Li, Wang, Zhang, & Wei, 2014).

4. Conclusions

A lightweight, hydrophobic and porous cellulose-based aerogels (CAGs) were successfully synthesized through freeze-drying process of waste newspaper as the only raw materials utilized instead of cost prohibitive and high-grade pulp. With further crosslinking with glutaraldehyde and surface treatment using trimethylchlorosilane under simple vapor deposition process, the resulting CAGs were bestowed with enhanced

robustness, hydrophobic, and oleophilic properties.

Systematic characterizations yielded more insights about the (inner-) structure and the fabrication route. The nitrogen sorption/desorption analysis revealed mesoporous structures of the obtained aerogels with pore sizes ranging from 8-13 nm, while XRD and FITR measurements showed a crystal form transformation from cellulose I to cellulose II during the dissolution and regeneration processes. The resulting aerogels presented not only prominent adsorption for oils and organic solvents, but also exhibited excellent filtration capacity comparable to that of HEPA membrane for lampblack. Therefore, this study provided a simple, cost-efficient, and scalable method for fabricating highly flexible with elevated performance aerogels based on abundant natural cellulose.

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References and notes

- Cao, Y., Li, H. Q., Zhang, Y., Zhang, J., & He, J. S. (2010). Structure and Properties of Novel Regenerated Cellulose Films Prepared from Cornhusk Cellulose in Room Temperature Ionic Liquids. *Journal of Applied Polymer Science*, 116(1), 547-554.
- Dash, R., Li, Y., & Ragauskas, A. J. (2012). Cellulose nanowhisker foams by freeze casting. *Carbohydrate Polymers*, 88(2), 789-792.
- Elsaesser, A., & Howard, C. V. (2012). Toxicology of nanoparticles. *Advanced Drug Delivery Reviews*, 64(2), 129-137.
- Fumagalli, M., Sanchez, F., Boisseau, S. M., & Heux, L. (2013). Gas-phase esterification of cellulose nanocrystal aerogels for colloidal dispersion in apolar solvents. *Soft Matter*, 9(47), 11309-11317.
- Han, S. J., Sun, Q. F., Zheng, H. H., Li, J. P., & Jin, C. D. (2016). Green and facile fabrication of carbon aerogels from cellulose-based waste newspaper for solving organic pollution. *Carbohydrate*

388 *Polymers*, 136, 95-100.

389 Heath, L., & Thielemans, W. (2010). Cellulose nanowhisker aerogels. *Green Chemistry*, 12(8), 1448-1453.

390 Himmelsbach, D. S., Khalili, S., & Akin, D. E. (2002). The use of FT-IR microspectroscopic mapping to study
391 the effects of enzymatic retting of flax (*Linum usitatissimum* L) stems. *Journal of the Science of*
392 *Food and Agriculture*, 82(7), 685-696.

393 Hinds, W. C. (2012). *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*,
394 2nd Edition.

395 Jiao, K. Y., Zhu, T., Li, X. H., Shan, M. J., Xu, Z. W., & Jiao, Y. N. (2015). Assembly of graphene oxide on
396 nonconductive nonwovens by the synergistic effect of interception and electrophoresis. *Journal*
397 *of Nanoparticle Research*, 17(9), 15.

398 Jin, C. D., Han, S. J., Li, J. P., & Sun, Q. F. (2015). Fabrication of cellulose-based aerogels from waste
399 newspaper without any pretreatment and their use for absorbents. *Carbohydrate Polymers*, 123,
400 150-156.

401 Kaparaju, P., & Felby, C. (2010). Characterization of lignin during oxidative and hydrothermal pre-treatment
402 processes of wheat straw and corn stover. *Bioresource Technology*, 101(9), 3175-3181.

403 Klemm, D., Heublein, B., Fink, H. P., & Bohn, A. (2005). Cellulose: Fascinating biopolymer and sustainable
404 raw material. *Angewandte Chemie-International Edition*, 44(22), 3358-3393.

405 Li, P., Wang, C. Y., Zhang, Y. Y., & Wei, F. (2014). Air Filtration in the Free Molecular Flow Regime: A Review
406 of High-Efficiency Particulate Air Filters Based on Carbon Nanotubes. *Small*, 10(22), 4543-4561.

407 Liebner, F., Haimer, E., Wendland, M., Neouze, M. A., Schlufte, K., Miethe, P., . . . Rosenau, T. (2010).
408 Aerogels from Unaltered Bacterial Cellulose: Application of scCO₂ Drying for the Preparation of
409 Shaped, Ultra-Lightweight Cellulosic Aerogels. *Macromolecular Bioscience*, 10(4), 349-352.

410 Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., . . . Ezzati, M. (2012). A
411 comparative risk assessment of burden of disease and injury attributable to 67 risk factors and
412 risk factor clusters in 21 regions, 1990-2010: a systematic analysis for the Global Burden of
413 Disease Study 2010. *Lancet*, 380(9859), 2224-2260.

414 Ma, S. R., Mi, Q. Y., Yu, J., He, J. S., & Zhang, J. (2014). Aerogel Materials Based on Cellulose. *Progress in*
415 *Chemistry*, 26(5), 796-809.

416 Maynard, A. D., & Pui, D. Y. H. (2007). *Nanotechnology and occupational health*: Springer.

417 Meng, X., Ma, Y. J., Chen, R. J., Zhou, Z. J., Chen, B. H., & Kan, H. D. (2013). Size-Fractionated Particle
418 Number Concentrations and Daily Mortality in a Chinese City. *Environmental Health Perspectives*,
419 121(10), 1174-1178.

420 Moon, R. J., Martini, A., Nairn, J., Simonsen, J., & Youngblood, J. (2011). Cellulose nanomaterials review:
421 structure, properties and nanocomposites. *Chemical Society Reviews*, 40(7), 3941-3994.

422 Morandi, G., Heath, L., & Thielemans, W. (2009). Cellulose Nanocrystals Grafted with Polystyrene Chains
423 through Surface-Initiated Atom Transfer Radical Polymerization (SI-ATRP). *Langmuir*, 25(14),
424 8280-8286.

425 Oberdorster, G., Oberdorster, E., & Oberdorster, J. (2005). Nanotoxicology: An emerging discipline evolving
426 from studies of ultrafine particles. *Environmental Health Perspectives*, 113(7), 823-839.

427 Oh, S. Y., Yoo, D. I., Shin, Y., Kim, H. C., Kim, H. Y., Chung, Y. S., . . . Youk, J. H. (2005). Crystalline structure
428 analysis of cellulose treated with sodium hydroxide and carbon dioxide by means of X-ray
429 diffraction and FTIR spectroscopy. *Carbohydrate Research*, 340(15), 2376-2391.

430 Shopsowitz, K. E., Hamad, W. Y., & MacLachlan, M. J. (2011). Chiral Nematic Mesoporous Carbon Derived
431 From Nanocrystalline Cellulose. *Angewandte Chemie-International Edition*, 50(46), 10991-10995.

432 Strak, M., Janssen, N. A. H., Godri, K. J., Gosens, I., Mudway, I. S., Cassee, F. R., . . . Hoek, G. (2012).
433 Respiratory Health Effects of Airborne Particulate Matter: The Role of Particle Size, Composition,
434 and Oxidative Potential-The RAPTES Project. *Environmental Health Perspectives*, 120(8),
435 1183-1189.

436 Subhedar, P. B., Babu, N. R., & Gogate, P. R. (2015). Intensification of enzymatic hydrolysis of waste
437 newspaper using ultrasound for fermentable sugar production. *Ultrasonics Sonochemistry*, 22,
438 326-332.

439 Tan, C. B., Fung, B. M., Newman, J. K., & Vu, C. (2001). Organic aerogels with very high impact strength.
440 *Advanced Materials*, 13(9), 644-646.

441 Tronville, P., & Rivers, R. (2005). International standards : filters for buildings and gas turbines. *Filtration &*
 442 *Separation*, 42(7), 39-+.
 443 Tronville, P., & Rivers, R. D. (2005). International standards: filters for vehicular applications. *Filtration &*
 444 *Separation*, 42(9), 24-27.
 445 Wang, F. C., Feve, M., Lam, T. M., & Pascault, J. P. (1994). FTIR ANALYSIS OF HYDROGEN-BONDING IN
 446 AMORPHOUS LINEAR AROMATIC POLYURETHANES .1. INFLUENCE OF TEMPERATURE. *Journal of*
 447 *Polymer Science Part B-Polymer Physics*, 32(8), 1305-1313.
 448 Wang, H. Y., Li, D. G., & Zhang, R. R. (2013). Preparation of Ultralong Cellulose Nanofibers and Optically
 449 Transparent Nanopapers Derived from Waste Corrugated Paper Pulp. *Bioresources*, 8(1),
 450 1374-1384.
 451 Wang, J., Kim, S. C., & Pui, D. Y. H. (2011). Carbon Nanotube Penetration through a Screen Filter: Numerical
 452 Modeling and Comparison with Experiments. *Aerosol Science and Technology*, 45(3), 443-452.
 453 Xia, G., Wan, J., Zhang, J., Zhang, X., Xu, L., Wu, J., . . . Zhang, J. (2016). Cellulose-based films prepared
 454 directly from waste newspapers via an ionic liquid. *Carbohydrate Polymers*, 151, 223-229.
 455 Zheng, Q. F., Cai, Z. Y., Ma, Z. Q., & Gong, S. Q. (2015). Cellulose Nanofibril/Reduced Graphene
 456 Oxide/Carbon Nanotube Hybrid Aerogels for Highly Flexible and All-Solid-State Supercapacitors.
 457 *Acs Applied Materials & Interfaces*, 7(5), 3263-3271.
 458