Recyclable Bacterial Cellulose Aerogel for Oil and Water Separation

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Abstract

Bacterial cellulose (BC) aerogel has great potential in treating oil spill and organic pollutant. However, its inherent hydrophilicity and poor rigidity limit its practical application and recyclability. In this study, elastically compressible and high oil-absorbing aerogels were developed by freeze-drying aqueous suspensions with appropriate BC concentrations, followed by a chemical vapor deposition of methyltrimethoxysilane with ammonia as catalyst. The modified aerogel shows high water contact angle of 142° and enhanced compression resistance. The effect of BC concentration on the adsorption capacity and recyclability of aerogel has been investigated. The results show that the aerogel prepared with 0.3 wt% BC exhibits simultaneously high absorption capacity (121.8–284.1 g/g) and excellent recyclability. Furthermore, the aerogel could also separate chloroform-water mixture by gravity-driven filtration, giving the separation efficiency of 96.7 %. Therefore, this economical green aerogel provides a feasible strategy for solving oil leakage in industry.

1. Introduction

Oil pollution has become a big environmental problem as frequent oil spills and widespread discharge of large quantities of industrial waste oil have caused serious pollution to oceans and lakes [1–4]. Therefore, it is of great significance to explore an effective route to recover waste oil and organic solvents in water without causing secondary environmental pollution.

Aerogel is a kind of lightweight material with high porosity, and considered as one of the most ideal absorbents [5–7]. Various aerogels made of gelatin[8], graphene [9], silica [10], cellulose nanofibers [11–13], chitosan [14, 15], have already been studied for oil absorption. Among them, the bacterial cellulose (BC) aerogels have attracted intensively attention. BC is a kind of natural nanocellulose produced by microorganisms, identical in chemical formula to the plant-derived cellulose. However, BC exhibits many superior properties over the plant cellulose, such as higher purity and crystallinity, better mechanical properties and degradable [16–18]. Therefore, the aerogels made of BC possess higher holding capacities and tensile strength than that made of plant cellulose, representing a promising material for oil/water separation [19]. As BC aerogel is inherently hydrophilic due to the presence of abundant hydroxyl groups in BC [5, 20], a hydrophobic surface modification is necessary to realize the selective adsorption for oil from oil/water mixture. The modification process is usually performed using modifiers with low surface energy. Besides, the hydrophobic and oleophilic nature can also be obtained by directly carbonizing BC aerogel. Wu et al. [21] reported that the BC aerogels carbonized at 1300°C became hydrophobic and exhibited high adsorption capacities for organic liquids (up to 312 g/g). However, the high cost and energy consumption of carbonization process is not conducive to the practical application of aerogels. Comparatively, the latter modification method is inexpensive. Using modifiers, the surface hydrophobization of cellulose aerogels were realized mainly via two different routes, gas phase reaction, and aerogel or hydrosol immersed in liquids containing modifiers. It is noted that most of the plant-derived cellulose aerogels were hydrophobic via gas phase reactions [22–24], while the reported BC aerogels were often hydrophobic modified in liquids containing modifiers. Wang et al.[25] prepared a

superhydrophobic BC aerogel by immersing BC hydrogel in a modifier solution containing stearic acid, giving the maximum adsorption capacity for soybean oil of 48.2 g/g. Similarly, Sai et al. [26] immersed blocky BC aerogel in a solution containing trimethylchlorosilane and triethylamine, and the prepared hydrophobic BC aerogel showed a maximum absorption capacity of 185 g/g for chloroform. He et al. [27] prepared a superelastic and superhydrophobic BC/silica aerogel by immersing BC aerogel in flexible silica sol with methyltriethoxysilane (MTES) as the precursor, and the adsorption capacities for the organic solvents and oils were 8–14 g/g. Although the surface modification of aerogels by chemical vapor deposition (CVD) is easy to be heterogeneous that restricts preparation conditions [26, 28], the CVD method can reduce the amounts of modifiers and solvents needed compared with the modification in liquid. Thus, the CVD modification is adopted in this work.

Adsorption capacity and recyclability are two important properties that affect the practical application of BC aerogels. From the researches mentioned above [21, 25–27], we note that the modified BC aerogels with modifiers need to further improve their adsorption capacities compared with the carbonized BC aerogels. Normally, hydrophobic aerogel with higher porosity owns higher adsorption capacity for oil [26]. And the porosity is critically determined by the preparation conditions during freeze-drying process, such as solvent [25], feature of suspension [29], along with cooling rate and temperature gradient [30]. Here, only BC concentration and surface energy of the mold are considered. The mold with lower surface energy tend to increase the freezing delay time and degree of supercooling due to a smaller contact area with the aqueous suspension. This will result in smaller ice nuclei uniformly formed in the whole suspension [31]. In addition, it is understandable that the suspension with low BC concentration tends to obtain a highly porous aerogel. On the other hand, low BC concentration in suspension may reduce the mechanical strength of aerogel, which is harmful to its recyclability in practical application. Therefore, it is necessary to explore an appropriate BC concentration for preparing aerogels to aspire excellent performance. To the best of our knowledge, the effects of BC concentration on adsorption capacity and recyclability of aerogels have rarely been studied.

Here, elastically compressible and highly oil-absorbing aerogels were developed by freeze-drying the suspension with low BC concentration (0.2, 0.3 and 0.4 wt%), followed by a CVD modification with methyltrimethoxysilane as the modifier and ammonia as the catalyst. The influence of BC concentration on the performances of aerogel were investigated. A polytetrafluoroethylene (Teflon) mold possessing low surface energy was used for freeze-drying so as to form BC aerogels with homogenized porous structure. This recyclable and hydrophobic BC aerogel displays great advantages in cost and performances.

2. Experimental

2.1 Materials

The bacterial cellulose hydrogel with 2.1 wt% BC concentration was purchased from Guilinqihong Technology Co., LTD. NH_4OH ($\geq 25\%$), anhydrous ethanol ($\geq 99.7\%$), and chloroform ($\geq 99\%$) were all

purchased from Guoyao Chemical Reagent Co., LTD. Methyltrimethoxysilane (MTMS, $\geq 98\%$), dimethyl silicone oil (AR) and NaOH (AR) were purchased from Aladdin. Pump oil was purchased from Lichen Technology Co., LTD and engine oil was purchased from Mobil Universal 4T. Dichloromethane, petroleum ether, castor oil, and soybean oil were all purchased from the local market. All experiments were conducted using deionized water (DI).

2.2 Preparation of BC aerogel

The hydrogel of bacterial cellulose was cut into small pieces, and then purified through soaking in DI at room temperature for 5 h and subsequently in 1% NaOH solution at 80°C for 8 h. The purified cellulose block was washed repeatedly with alcohol and DI water until the solution was neutral. Three types of uniform cellulose suspensions with BC concentration of 0.2 wt%, 0.3 wt% and 0.4 wt%, were obtained by stirring the mixture of BC block and DI at a high speed of 20000 r/min. 10 g suspension was poured into a Teflon cylindrical mold ($\varphi = 3\text{cm}$), pre-frozen at -60°C for 12 h, and then freezing-dried for 48 h. The BC aerogels prepared at different BC concentrations were denoted as DB-0.2, DB-0.3 and DB-0.4.

2.3 Modified BC aerogels

The surface of the BC aerogel was modified via the CVD method. The BC aerogel (30 mg) was put into a sealed dryer together with two small glass bottles containing 5 mL MTMS and 7 mL NH_4OH , respectively, and then heated at 75°C for 3.5 h to complete the vapor deposition. Later, the bacterial cellulose aerogel was taken out of the dryer and maintained at 60°C for 12 h. The modified BC aerogels were denoted as DBS-0.2, DBS-0.3 and DBS-0.4.

2.4 Characterization

The morphology of aerogel was characterized by using field emission scanning electron microscope (Sigma-500). Fourier transform infrared spectrometer (Nicolet 5700) with a resolution of 4 cm^{-1} , was used to determine the change of the groups in the modified samples. X-ray diffractometer (Miniflex-600) was used to analyze the crystal structure of the samples at a scanning speed of $2^\circ/\text{min}$. The surface chemical properties of the samples were identified by using X-ray photoelectron spectrometer (XPS, Thermo Scientific™ K-Alpha™). The static contact Angle was measured by the contact angle analyzer (SL200B). The stress-strain curves of the BC aerogels were tested by the universal testing machine (CMT4202).

3. Results And Discussion

3.1 The structure and morphology of BC aerogels

The SEM images of DB-0.3 and DBS-0.3 are shown in Fig. 1a-d. It can be seen that the original morphology and the pore structure of the aerogel almost remain the same after modification. The SEM images at low magnifications (Fig. 1a and 1c) show that both DB-0.3 and DBS-0.3 have macroscopic porous structures, containing a large number of micron-sized pores with the diameter of 50-200 μm ,

which would endow the aerogel with high adsorption capacity. The SEM images at high magnifications (Fig. 1b and 1d) show that DB-0.3 and DBS-0.3 aerogels exhibit a large amounts of entangled BC fibers with the diameter of 50-100 nm, forming a three-dimensional network porous structure, which enables the aerogel to have good mechanical properties. Moreover, the modified DBS-0.3 exhibits many worm-like SiO₂ nanoparticles with the length of 400-800 nm, which greatly improves the surface roughness of DBS-0.3 aerogel. The energy dispersive X-ray energy spectrum (EDS) of DBS-0.3 in (Fig. 1e, f) shows that the Si element as well as C and O elements were uniformly distributed on the surface of DBS-0.3, proving that the aerogel was successfully modified by MTMS.

The XPS test was carried out to explore the surface chemical constituent of DB-0.3 and DBS-0.3, as shown in Fig. 2(a-d). The DB-0.3 is composed of C and O elements, while DBS-0.3 contains C, O and Si elements. For the DBS-0.3 aerogel, the spectrum of O 1s shows two peaks at 533 eV and 534 eV, respectively, corresponding to C-O and Si-O components. In addition, the Si-O bond at 103 eV and Si-C bond at 104 eV were observed in the Si 2p spectrum of DBS-0.3 (Fig. 2d) [11]. These results demonstrate the presence of the silanization reaction between MTMS and hydroxyl group of cellulose.

The changes of the chemical groups of aerogels after modification were analyzed by the infrared spectroscopy. As shown in Fig. 3a, both DB-0.3 and DBS-0.3 aerogels show four vibration adsorption bands in the range of 3348-3350 cm⁻¹ (-OH), 2915-2917 cm⁻¹ (C-H), 1631-1634 cm⁻¹ (C=O) and 1059-1060 cm⁻¹ (C-O), revealing that BC has abundant oxygen containing groups such as hydroxyl groups and carbon chains [25]. It can also be observed that the intensity of the adsorption band around 3349 cm⁻¹ significantly decreases after modification. This should be due to the decrease of hydroxyl groups in BC induced by the reaction between MTMS and the hydroxyl of cellulose. In addition, the modified aerogel DBS-0.3 exhibits new vibration bands at 781 cm⁻¹ (Si-O-Si) and 1274 cm⁻¹ (Si-CH₃), which also means the Si-CH₃ groups were grafted on BC [32]. Owing to low surface energy, these Si-CH₃ groups on the surface of BC are benefit to enhancing the surface hydrophobicity of aerogel. Fig. 3b shows the X-ray diffraction spectra of DB-0.3 and DBS-0.3. The original aerogel DB-0.3 shows three typical diffraction peaks at 15 °, 17 ° and 22.8 °, corresponding to the (10), (110) and (020) reflection planes of the cellulose crystal, respectively [26]. In the X-ray diffraction spectrum of DBS-0.3, two diffraction peaks at 15 ° and 17 ° disappear and the peaks at 22.8 ° become weakened, indicating that the crystallinity decreases after modification. This is because the silanol from the hydrolyzed MTMS could been easily grafted and polycondensed on the surface of the aerogel, due to the presence of abundant hydroxyl groups in BC.

3.2 Compressibility of BC aerogels

Fig. 4(a, b) give the compressive stress-strain curves and the loading-unloading

resilience test curve of the aerogels. As shown in Fig. 4a, the compressive stress first presents a gentle rising with increasing strain, and then rises abruptly when the compressive strain exceeds 60%. This is because the interlaced structure of BC became densified due to the squeezed and constricted pores. Thanks to the macroscopic porous and microscopic interleaving network structure of the aerogels, the

force required to compress the aerogels to perform 50 % strain is less than 2 kPa, indicating the good flexibility of these aerogels. Compared with the modified samples, DB-0.3 shows much lower stress in the strain range of 60-80%, and its poor rigidity is detrimental to the mechanical recycling. Among the three modified aerogels, DBS-0.3 exhibits the highest stress within the abruptly increasing stress region, indicating the strongest ability to bear compressive deformation. Fig. 4b shows that the compressed DBS-0.3 aerogel can be immediately restored to the original height after unloading the weight. Moreover, about 98% of the original height can be restored after 300 cyclic loading-unloading resilience tests. Therefore, DBS-0.3 has been proved to have excellent flexibility and elastic recovery ability, which makes it possible to recycle the adsorbed aerogels by squeezing.

3.3 Adsorption for various oils and organic solvents

The wettability changes of aerogel after modification are shown in Fig. 5(a-d). Due to the large amount of hydroxyl groups on the surface of pristine BC, the water and engine oil droplets are able to quickly spread over the aerogel surface, suggesting that the original aerogel has high affinity to engine oil and water (Fig. 5a). For the modified aerogel, the surface hydrophobicity was observed (Fig. 5b). In order to further investigate whether the interior shows oleophilic-hydrophobicity, the modified aerogel was cut along the diameter direction, and the wettability of the transverse section is shown in Fig. 5(c-d). It can be seen that oil droplets spreads over the aerogel surface, but water droplets remain the spherical shape and the water contact angle is 142° . These results indicate the oleophilic-hydrophobicity of the whole aerogel instead of just the surface, has been achieved by the facile CVD of MTMS.

By virtue of good hydrophobicity, the modified BC aerogel can realize adsorption for oil from water. As shown in Fig. 6a, DBS-0.3 can absorb engine oil (dyed red) floating on the water surface within 10 s. Fig. 6b shows that DBS-0.3 can rapidly absorb chloroform and repelled water when being immersed in a mixed aqueous solution containing chloroform (dyed red) at the bottom. Therefore, DBS-0.3 exhibits highly efficient oil-absorption. The adsorption capacity of DBS-0.3 for various oils and organic solvents was showed in Fig. 6c. The DBS-0.3 aerogel exhibits good adsorption ability and the obtained adsorption capacities were 121.8-284.1 g/g. Moreover, Fig. 6d shows that the adsorption capacity of DBS-0.3 is linearly positively correlated with the density of the absorbed liquid, indicating that the adsorption ability of aerogel is mainly determined by its pore volume. As the pore volume of a given aerogel is constant, the liquid with higher density will present higher mass per unit volume, resulting in increasing the adsorption capacity of the aerogel. As given in Table 1, it can be seen that the adsorption capacity of the aerogel DBS-0.3 prepared in this work is superior to most of the previously reported aerogels due to low cost and high adsorption capacity, and so it is easier to achieve large-scale waste oil treatment in industry.

3.4 Separation for oil/water mixture by filtration

A test to separate oil/water mixture by filtration was carried out via plugging DBS-0.3 into the bottom of a funnel, in which dyed chloroform (10 g) and DI (10 g) were poured. As shown in Fig.7, driven by the gravity, the dyed chloroform can quickly flow through the pores of DBS-0.3 aerogel to the beaker below, while the DI was holded in the funnel due to the good hydrophobicity of DBS-0.3 aerogel. The separation

efficiency can be calculated by the ratio of the mass of separated chloroform (M_t) in the beaker to the original mass (M_0) of chloroform. In this work, the obtained separation efficiency is up to 96.7 %.

3.5 Effect of BC concentration on the adsorption and recyclability of aerogels

The excellent recyclability of aerogel is very important for practical applications. In this work, the recyclabilities of DBS aerogels with different BC concentrations were investigated. As shown in Fig. 8a, the oil adsorbed was released by simply squeezing the saturated adsorbed DBS aerogel, and then the flattened DBS aerogel was re-immersed in the engine oil. After a few seconds of adsorption, the re-saturated DBS aerogel was restored to its original shape. The adsorption capacity of the recycled DBS aerogel can be calculated based on the following equation:

$$Q_r \text{ (g/g)} = \frac{M_b - M_a}{M_0} \quad (1)$$

Where M_0 is the mass of the original DBS aerogel, M_a and M_b are the mass of the flattened aerogel and saturated aerogel in the n th squeeze-adsorption cycle, respectively. The adsorption capacity of DBS-0.2, DBS-0.3 and DBS-0.4 for engine oil are shown in Fig. 8(b-d). It can be seen that the maximum adsorption capacity and the recyclability are quite different for the three DBS aerogels with different BC concentrations. Although the DBS-0.2 has the highest maximum adsorption capacity, some breakage of aerogel can be observed in the cycling process, resulting in rapidly-decreasing absorption capacity with increasing cycling times. Compared with DBS-0.2, the DBS-0.3 aerogel could maintain its integrity of appearance, showing lower maximum adsorption capacity and better recyclability with an adsorption capacity maintained above 150.0 g/g throughout the 10 squeezing-absorbing cycles. This is consistent with the result of stress-strain test that DBS-0.3 has a higher ability to bear compressive deformation. Among the three DBS aerogels, DBS-0.4 shows the most stable adsorption capacity throughout the 10 cycles. However, its adsorption capacity is far lower than that of DBS-0.3, attributed to the lower porosity caused by higher BC concentration. As a consequence, the DBS aerogel with 0.3 wt% BC concentration exhibits the best comprehensive performance, simultaneously its high adsorption and excellent recyclability provide a feasible strategy for solving oil leakage in industry.

4. Conclusions

The lipophilic-hydrophobic aerogels have been successfully developed by freeze-drying the aqueous suspensions with different BC concentrations (0.2, 0.3 and 0.4 wt%), followed by a simple modification via chemical vapor deposition. The adsorption capacity of aerogel for oil increases with decreasing BC concentration, but the recyclability shows an opposite trend. The DBS-0.3 aerogel achieves high absorption capacity (121.8-284.1 g/g) for various oils and organic solvents (soybean oil, castor oil, pump oil, engine oil, dichloromethane, chloroform, dimethyl silicone oil, petroleum ether and anhydrous ethanol). Owing to its excellent elastically compressibility, DBS-0.3 can be recycled through a simple

squeezing, and the adsorption capacity for engine oil is above 150 g/g during 10 cycles. It can also be used to rapidly separate the chloroform-water mixture with a separation efficiency of up to 96.7 %. This economical green aerogel is promising for treating oil leakage and purifying water.

Declarations

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Conflicts of Interest

There are no conflicts to declare.

Authors' contributions

Zhuofeng Yan: Conceptualization, Verification, Writing manuscript. Kaixiao Zhu: Resources. Xiangqi Li: Supervision, Writing - review. Xiao Wu: Reviewing & editing manuscript. All authors read and approved the final manuscript.

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Tables

Table 1. Comparison of the adsorption capacities of various aerogels for oils

Aerogels		Adsorption capacity (g/g)		Cost	Reference
Starting materials	Modifier	Pump oil	Chloroform		
Poplars Catkins	carbonized in N ₂ atmosphere	101	161	medium	[33]
Bacterial cellulose	TMCS and TEA	118	185	low	[26]
EVOH nanofibers	GA	66.3	91.4	low	[34]
Polyimide	MXene-3	57.8	45.1	high	[35]
Graphene oxide/nanofiber	vapor deposition of hexadecyltrimethoxysilane	384	641	high	[36]
sisal	Cu nanoparticles	91	164.5	medium	[37]
Bacterial cellulose	vapor deposition of Methyltrimethoxysilane	153.4	284.1	low	This work

Figures

Figure 1

SEM images of (a-b) DB, (c-d) DBS-0.3 at different magnifications, (e-f) EDS spectrograms of DBS-0.3.

Figure 2

(a) XPS spectra of DB-0.3 and silane modified DBS-0.3. (b) XPS C 1s spectrum, (c) XPS O 1s spectrum and (d) XPS Si 2p spectrum.

Figure 3

(a) FTIR spectra and (b) XRD spectra of modified aerogel DBS-0.3 and original aerogel DB-0.3

Figure 4

(a) The stress-strain curves of aerogels and (b) the loading-unloading resilience test curve of DBS-0.3

Figure 5

Wettability of engine oil (dyed red) and water (dyed blue) droplets on (a) original aerogel, (b) silane modified aerogel, (c) the transverse section of silane modified aerogel and (d) the water contact angle of the transverse section

Figure 6

(a) Adsorption for engine oil (dyed red) on water surface; (b) Adsorption for chloroform (dyed red) under water; (c) Absorption capacity of DBS-0.3 for various oils and organic solvents; (d) Absorption capacity marked by solvent density.

Figure 7

Oil-water mixture filtration test

Figure 8

The squeeze-adsorption cycle process (a) and cyclic adsorption capacity of (b) DBS-0.2, (c) DBS-0.3, and (d) DBS-0.4 for engine oil.