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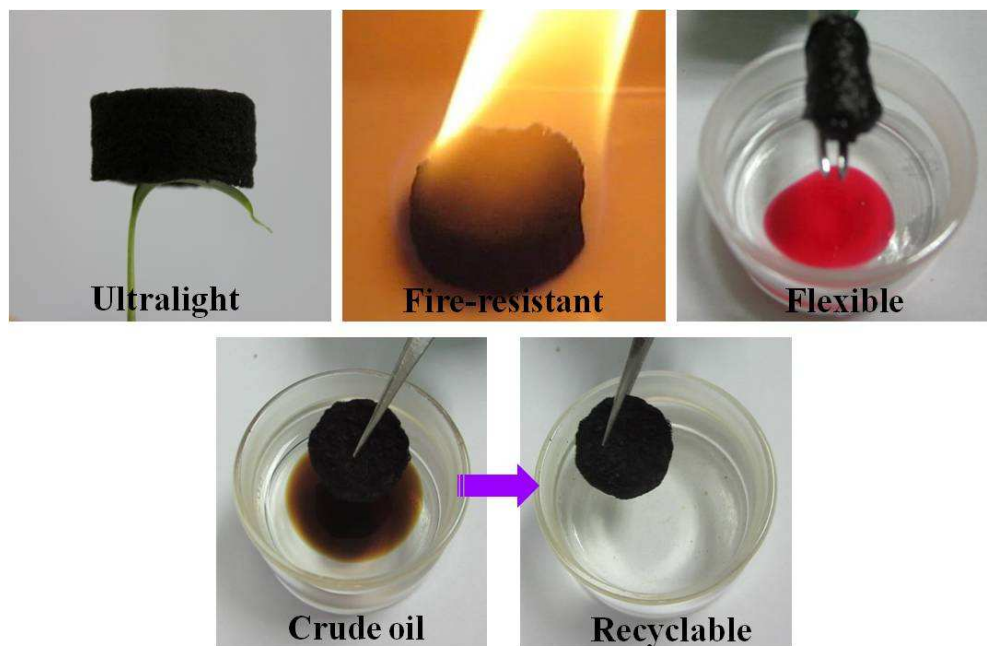
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Graphical abstract



Ultralight, fire-resistant, and flexible carbon fiber aerogels as recyclable oil adsorbents are prepared by using disposable bamboo chopsticks as precursor. The aerogel can be recycled for many times by distillation, combustion or squeezing, making the material satisfy the requirements for oil-water separation in practice.

Low cost carbon fiber aerogel derived from bamboo for the adsorption of oils and organic solvents with excellent performances

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Abstract

This paper reported the preparation of multi-functional carbon fiber (MCF) aerogel by a simple hydrothermal and carbonization process using disposable bamboo chopsticks. The developed material manifested dramatic multi-functionalities, including excellent flexibility under the mechanical compression, efficient capability to separate oily droplets from water, and high adsorption capacity for a variety of oils and organic solvents by up to 129 times of its own weight. Moreover, the MCF aerogel can be recycled for many times by distillation, combustion or squeezing, making the material satisfy the requirements for oil-water separation in practice. Coupled with economical, environmentally benign manufacturing process, sustainability of precursor and versatility of material, the MCF aerogel developed in this study will be a promising candidate to address the problems arising from the spills of oily compounds.

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Introduction

With ever-increasing production and usage of organic materials, oil spillage and chemical leakage occur frequently worldwide every year. Water pollution arising from these accidents has led to severe environmental and ecological problems¹⁻³. Therefore, materials that can effectively adsorb and transfer spilled oils and organic solvents from water are in urgently needed. Different techniques have been employed to collect hydrophobic organic liquids from water surface with varying degree of success⁴⁻¹⁰. Among them, physical adsorption by porous materials is considered to be the most effective and promising technique. Generally, an excellent adsorbent material should exhibit high adsorption capacity, high selectivity, low density, good recyclability and environmental friendliness. These requirements also acted as driving forces in developing novel materials with multi-functional properties to separate oil-water mixture effectively and efficiently.

Three-dimensional (3-D) carbon-based aerogel has been applied for oil adsorption in recent years due to its low apparent density, high porosity and specific surface area¹¹⁻¹⁴. Particularly, the inherently hydrophobic nature of carbon in aerogel makes it suitable to separate oily compounds mixed with water^{11, 15-20}. Various carbon-based aerogels, such as carbon nanotube (CNT) sponge^{5, 21}, graphene sponge^{15, 22}, graphene/CNT hybrid foam^{23, 24} and carbon nanofiber (CNF) sponge^{11, 25}, have been developed and showed very high adsorption capacities for oil and petroleum products. Nevertheless, the relatively high cost of starting materials (CNTs or graphene), expensive equipments and complicated multi-steps consisting of dispersing nanomaterials in solvent, formation of aerogel using sol-gel method and following thermal treatment, hamper the large-scale production of carbon-based aerogel for practical applications.

As an alternative, there is a trend to use sustainable and low cost materials to produce carbon-based aerogels^{10, 11}. In this regard, the employment of inexpensive biomass precursors provides the opportunity to achieve this goal and has recently gained lots of interest both academically and commercially because of the economic/process/chemistry advantages offered by either the starting materials or the preparation approaches. For example, bacterial cellulose¹¹ and raw cotton¹⁶ were employed to prepare carbon-based 3-D frameworks, and the products showed high capacity for oil adsorption and excellent fire-resistant properties.

In addition, there is a continuous effort to optimize the way to recycle/reuse the aerogels with saturated oil adsorption. It is known that the oily compounds with low boiling points can be recovered by distillation easily, and the adsorbed liquids with high boiling points undergo a direct combustion for the reuse of adsorbents. In this context, the thermal stability of aerogel is important during the practical operation. For precious or non-flammable compounds with high boiling points, the strategy of utilizing compressible materials under the adsorption-squeeze is an attractive method to recycle both the absorbed liquids and adsorbents. Unfortunately, some adsorbents face the challenges in this simple process due to their structural instability under the mechanical compression. Ideal materials for the recovery of spilled oil would be of multi-functionality integrating porous structure, hydrophobicity, fire resistance and flexibility, possessing material regeneration by simple distillation, combustion, squeezing or combination of them.

Herein, a novel technique was developed to prepare multi-functional carbon fiber (MCF) aerogel using bamboo chocksticks as a carbon source. The morphology, thermal stability and surface properties of MCF aerogel were studied. The feasibility of using this material for the separation of oil-water mixture was investigated, and its advantages were demonstrated as well.

Experimental Section

Materials

Disposable bamboo chopsticks were used as raw material and collected from the local restaurant. Commercial available oil products (Gasoline, diesel, pump oil, colza oil) and crude oil (Karamay Oilfield, China) were used as probing liquids for oil-sorption-capacity studies. All other chemicals were analytical grade and used as received without further purification. De-ionized water (by a Millipore Water Purification System) was used in all experiments.

Preparation of MCF aerogel

1.0 g of shavings made from bamboo chopstick waste was put into a Teflon-lined stainless steel autoclave wherein a 70 mL of potassium hydroxide solution (KOH, 3 mol/L) was contained. The autoclave was sealed and placed in an oven at 150 °C for 6 h. The solid in the mixture was separated using a filter paper and then poured into water and stirred vigorously to form a uniform dispersion. The mixture was subjected to freeze-drying to form bamboo fiber aerogel. MCF aerogel was achieved by processing the fiber aerogel in a tubular furnace at 400 °C for 2 h in nitrogen atmosphere.

Adsorption behavior MCF aerogel and its regeneration

In a typical adsorption experiment, MCF aerogel was put into a specific liquid. Spontaneous adsorption occurred within a relatively short time, resulting in the immersion of sample in the liquid in a few seconds. The sample was suspended in the liquid for another 10 minutes to reach the adsorption equilibrium. The weight gain of aerogel was calculated using the weight change of material before and after adsorption. During the weight measurement, the sample was collected from the liquid using a tweezers and finely led on the edge of liquid container to

remove the excessive oil droplet. In order to avoid the evaporation of absorbed organic liquids, especially for those with low boiling points, the measurement was done as quickly as possible. All adsorption experiments were conducted three times and an average value of adsorption capacity was reported here. Various methods, including adsorption-distillation, adsorption-combustion, adsorption-squeezing, were applied to regenerate the aerogel after oil adsorption. In the adsorption-distillation way, the oil-adsorbed aerogel was heated at a temperature around the boiling point of adsorbate, and the dried aerogel was used for further cyclic operation. The adsorption-combustion method was accomplished by the direct combustion of aerogel in air. In the adsorption-squeezing operation, the oil-adsorbed aerogel was squeezed to recover the oil, and the material was used for next cycle.

Characterization

The morphology of aerogel was observed using a scanning electron microscopy (SEM, Zeiss Supra55VP). Functional groups in aerogel were determined using a Fourier-transform infrared spectrometer (FT-IR, FTS-165, BIO-RAD). Thermal stability of bamboo fiber aerogel were characterized on a thermogravimetric analyzer (TGA, STA 449F3, Netzsch) with a heating rate of 5 °C/min from 30 to 1000 °C in nitrogen atmosphere. Static contact angle of aerogel was measured on a goniometer (XG-CAMA, Shanghai Xuanyichuangxi) at ambient temperature. At least three measurements were made within 10 s of water droplet formation on the sample, and the average value was reported here. The nitrogen adsorption-desorption isotherm was obtained using a volumetric adsorption analyzer (SSA-4300). Raman spectra were conducted on a confocal laser micro Raman spectrometer (LABRAM-HR800, Horiba Jobin-Yvon). The X-ray photoelectron spectroscopy (XPS, Thermo Escalab 250xi, Perkin Elmer) was tested by using a Al K α radiation exciting source. The optical images and movies were obtained by digital camera

(600D, Canon). The compressive properties of aerogel were determined on a universal testing machine (C43-104, MTS) with a constant cross-head speed of 0.5 mm/min.

Results and discussion

Morphology and properties of aerogel

The fabrication process for MCF aerogel and the morphological changes of materials at different steps are illustrated in Figure 1. Typically, shavings made from recycled bamboo chopsticks (Figure 1a) were processed in an autoclave under the alkaline condition. During this process, the lignin parts between the neighboring fibers were gradually dissolved into KOH solution, producing cotton-like cellulose fibers (Figure 1b). After these fibers were re-dispersed in water and subjected to a following freeze-drying process, bamboo fiber aerogels were produced (Figure 1c). The white aerogel was converted to black MCF ones at the elevated temperature with the protection of nitrogen gas (Figure 1d). The macroscopic dimension of as-synthesized MCF aerogel can be controlled by using various shapes of containers. For example, a cylindrical piece of fiber sample with a diameter of 3.1 cm was converted to MCF aerogel with a shrunken diameter of 2.4 cm after thermal treatment. The calculated density of aerogel (based on the weight and dimension of sample) was 8.3 mg/cm^3 , belonged to the range of ultra-low density material²⁶. The result is comparable to that of spongy graphene ($\sim 12 \text{ mg/cm}^3$)¹⁵ and un-densified vertically aligned CNT arrays (10 mg/cm^3)²⁷, and much less than those of traditional carbon aerogels ($100\text{-}800 \text{ mg/cm}^3$)²⁸ and hydrophobic cellulose aerogels ($20\text{-}30 \text{ mg/cm}^3$)²⁹. As a result, a piece of MCF aerogel could effortlessly stand on the top of grass without deforming the supporter at all (Figure 1d).

The morphology of aerogel at different steps was characterized using SEM. The results showed that the bamboo fiber aerogel exhibits 3-D frameworks in microscale. The length of individual fibers was up to hundreds of micron or even longer, and porous structure can be observed among the neighboring fibers (Figure 1e). High-magnification SEM image indicated that the diameter of fibers ranged from 8 to 10 μm , and some fragments were attached on fiber surface (Figure 1f). After the pyrolysis, the porous structure of aerogel was remained (Figure 1g), and the fibers exhibited a smooth surface with a smaller diameter of 4-6 μm (Figure 1h). This was due to the thermal treatment of sample: under the high temperature in inert atmosphere, the organic compounds in bamboo fibers were removed, leading to the volumetric contraction of fibers. Type III nitrogen adsorption-desorption isotherm is observed in the MCF aerogel (Figure 1i), suggesting the formation of multilayer adsorption of nitrogen in the sample. Kindly note that by employing this isotherm, the surface area and pore size distribution of sample can be obtained using Brunauer-Emmett-Teller (BET) method. However, this theory is applicable for the monolayer adsorption, and favorable for the material with micropore (<2 nm) and mesopore (2-50 nm). Whereas the whole aerogel shows macroporous structure in micron scale (Figure 1g). Under such circumstances, the surface area and pore size distribution obtained from BET method are not reliable to be reported here.

The thermal stability of bamboo fiber aerogel was studied to provide reference on selecting suitable pyrolysis temperature with an effort to reduce the energy cost for the production of MCF aerogel. Figure 2 presents the TGA and corresponding derivative results (DTG) of fibers tested from room temperature to 1000 $^{\circ}\text{C}$ in nitrogen atmosphere. An obvious weight change was observed in the range of 300-400 $^{\circ}\text{C}$, and the peak from DTG curve suggested that the maximum decomposition of material occurred at 340 $^{\circ}\text{C}$. The significant weight loss of sample should be

attributed to the pyrolysis of cellulose in the fibers with byproducts of water vapor, CO₂, CO, H₂ and trace amounts of organic gases³⁰. Suitable temperature for the treatment of the precursor for MCF aerogel should be above 340 °C in an inert gas. Increasing the temperature can enhance the degree of carbonization for the bamboo fiber, however, it will bring negative effect on maintaining the structural stability of aerogel, especially when the material was subject to a mechanical load. For example, the bamboo fibers pyrolyzed at 800 °C became powders after they were compressed and released for two cycles. In addition, marginal decrease on the weight of fiber from 400 to 1000 °C was observed. Therefore, 400 °C was adopted as the optimized temperature to prepare MCF aerogel.

Besides the ultra-light and excellent thermal stability, the MCF aerogel exhibited outstanding hydrophobic property. Figure 3 shows the wettability of two aerogels probed by water and oil. A few drops of water stained with methylene blue (MB) were adsorbed quickly by the bamboo fiber aerogel, leaving a blue stain in the material (Figure 3a). In sharp contrast, the penetration of water into the MCF aerogel was inhibited greatly, and spherical droplets were formed on the surface of aerogel (Figure 3b), suggesting the conversion of fiber aerogel from the hydrophilic to the hydrophobic after thermal treatment. The hydrophobicity of MCF aerogel was further confirmed by immersing the material into water. A uniform mirror-reflection was observed on the surface of material (Figure 3c) due to the formation of an interface between the entrapped air in the aerogel and surrounding water. The measured contact angle between the MCF aerogel and water was $145 \pm 2^\circ$ (Figure 3d), whereas a drop of lubricating oil penetrated into the aerogel completely in one second, making it impossible to determine the static contact angle (Figure 3e). These results suggested that the as-prepared MCF aerogel exhibited good adsorption selectivity

for oil thanks to the hydrophobic nature of carbon-based materials and porous networks in the structure.

The mechanism behind the differences on the wettability of fiber and MCF aerogels was further justified by FT-IR (Figure 4a). Specifically, several peaks representing oxygen-contained groups, such as C=O ($1642, 2922\text{ cm}^{-1}$), C-O (1062 cm^{-1}) and -OH (3424 cm^{-1}), were observed in the fiber aerogel (Curve (1) in Figure 4a). However, these peaks became weak or disappeared once the bamboo fibers were processed to MCF aerogel (Curve (2) in Figure 4a), suggesting the removal of hydrophilic moieties in the sample during the thermal treatment. This assumption was partly confirmed by studying the Raman spectrum of samples. As shown in Figure 4b, the fiber aerogel exhibits a smooth line in the range of $500\text{--}2000\text{ cm}^{-1}$. In contrast, the MCF aerogel shows a strong peak at 1590 cm^{-1} along with a weak hump at around 1378 cm^{-1} , representing the vibration of hexagonal graphite and disorder-induced carbon structure in the sample, respectively. These results indicate the conversion of cellulose fibers in bamboo to graphitic carbon after pyrolysis treatment. It can be expected that the carbon structure in MCF aerogel is hydrophobic and oleophilic. The C1s XPS spectra of fiber and MCF aerogels were also used to confirm the deoxygenation process³¹, as shown in Figure 4c. The atomic percentages of C and O in bamboo fiber aerogel are 44.8% and 55.2%, respectively. The content of C increases to 83.9% with a simultaneous decrease on O to 16.1% in MCF aerogel. The higher C/O ratio (5.2 for MCF aerogel, 0.8 for bamboo fiber aerogel) provides direct evidence showing the enhanced carbonization in MCF aerogel. Furthermore, the removal of oxygen-contained groups in the sample was verified by a significant improvement on the intensity of sp^2 C-C bond at 284.6 eV and decreasing oxygen-contained carbon (C-O at 286.6 eV, -C=O at 287.6 eV, and -O-C=O at

289.0 eV), as shown in the deconvoluted C1s spectra of aerogels in Figure 4d. The results were in good agreement with the hydrophobic nature of MCF aerogel, as observed in Figure 3.

Interestingly, the MCF aerogel developed in this study displayed excellent flexibility, and it can bear a large volume reduction under the compression and excellent recovery once the mechanical load was released. Figure 5 shows the compressive stress-strain curves for the set strain (ϵ) maxima of 70% and 90%. Two distinct stages were observed when the material was subjected to the mechanical load, including a linear region with $\epsilon < 70\%$, and a following densification region. In the first region, the compressive stress increased linearly with increasing strain, representing the elastic deformation of aerogel. In the densification region ($\epsilon > 70\%$), the stress increased steeply with strain because the fibers in the aerogel were impinged upon each other. The curves under release gave a stress near zero until $\epsilon = 0\%$, suggesting the aerogel can be completely recovered to its original dimension. These robust mechanical properties were attributed to the constitute and structure of aerogel: i) the excellent mechanical properties of carbon fibers in aerogel offer the elasticity of material, and ii) the porous and 3-D networks make the material flexible, even with an extreme compressive strain of 90%.

Adsorption behavior of MCF aerogel for oils and organic solvents

The low density, flexible and hydrophobic properties of MCF aerogel make it a promising material to remove oils and organic solvents from water. This feasibility was demonstrated by studying the oil-spill cleanup capacity of aerogel, as shown in Figure 6a. Crude oil was suspended on water surface, forming a thin layer due to its low density and immiscibility with water. Once a piece of MCF aerogel contacted with the layer, the oil was adsorbed completely, leaving a transparent region on water surface (Movie S1 in the Supporting Information). The

aerogel saturated with oil can float on water surface due to its low density and hydrophobicity, offering convenience in collecting and recycling the aerogel in practical applications.

Furthermore, MCF aerogel can adsorb solvents with a higher density than water. For example, when the material was immersed into water and put in contact with chloroform (1.48 g/mL, stained with oil red), the water-immiscible droplet was sucked into the aerogel within a few seconds (Figure 6b, Movie S2 in the Supporting Information). No detectable water was found in the saturated aerogel, showing the excellent adsorption selectivity of material for oily liquids.

The versatility of MCF aerogel for the adsorption of organic compounds was also studied by employing a series of commonly used liquids, such as petroleum products (gasoline, diesel, pump oil, etc), water-immiscible solvents (hexane, decane, toluene, etc). The results showed that the MCF aerogel exhibited a very high adsorption capacity for these organic liquids ranging from 30 to 129 times of its original weight (Figure 6c). The adsorption capacity and estimated cost (With considerations on the availability of precursors, fabrication process, convenience and applicability of materials, and so on) of MCF aerogel were compared with other 3-D carbon-based materials (Figure 6d). It was obvious that the aerogel reported here exhibited much higher adsorption capacity than activated carbon and graphite-based materials, and comparable to the sponge/foam consisting of carbon nanotubes (CNTs) and graphene. While its adsorption performance was lower than some newly-developed ultra-flyweight aerogels, the method for the fabrication of MCF aerogel was much simpler and its precursor, i.e., bamboo chopsticks, is a low-cost and sustainable biomass. Therefore, the aerogel developed in our work was effective with a low-cost performance, environment-friendly and had a high scalability for the practical separation of oil-water mixture.

Besides the adsorption capability, the recyclability of adsorbent and recoverability of adsorbed liquids were key considerations when selecting suitable material for water-oil separation. Figure 7 shows the recyclability of MCF aerogel by studying the weight change of material after saturated adsorption of organic liquids with different boiling points. Various methods, including distillation, combustion and squeezing, were employed to recover the aerogel. For instance, for the removal of valuable pollutants or those with low boiling points (n-hexane as an example), distillation is a commonly used method. MCF aerogel was recovered by heating the sample at 70 °C (boiling point of n-hexane). The results showed that negligible changes on the adsorption capacity were found after six adsorption/distillation cycles (Figure 7a). The morphology of aerogel (Figure 7b) was pretty similar to those of original one, suggesting marginal damage to the MCF aerogel during the cyclic runs.

For the adsorption of oils with high boiling point, combustion was applied to recover the MCF aerogel. In such case, hexadecane (boiling point: 287 °C) was used as an adsorbate. The results showed that the material maintained 62.1% for its initial adsorption capacity under the second cyclic run, and this capability was nearly unaltered under the further cycles (Figure 7c). The decrease on the adsorption capacity was likely due to the deposition of combustion residues on fiber surface. This assumption was partly confirmed by examining the surface of individual fibers, as amorphous materials were observed on fiber and its diameter increased to 8-10 μm (Figure 7d). During the combustion, the aerogel adsorbed with hexadecane was burned quietly and stably, leaving an unchangeable shape of material after the burnt-out of oil (Figure 8a). This capability was attributed to: i) the excellent thermal stability and fire resistance of aerogel, and ii) the porous structure of material facilitated the transfer of heat generated during the combustion

As an alternative for aforementioned methods, squeezing was a simple and easy-applied one. The recycling of MCF aerogel by this method was also studied using gasoline as a probe liquid. The sorption capacity of MCF aerogel possessed 61.4% of the original value after six cycles (Figure 7e), and about 25.3% of adsorbed gasoline was left in the second cycle due to the incomplete compression of MCF aerogel. The porous structure of aerogel and the carbon fibers remained unchanged during the whole process (Figure 7f, Figure 8b). While the squeezing was less effective as compared with other two, the energy consumption and operation convenience of this method make it very competitive in practice.

Polymer foams/aerogels with novel structures/composition have been applied in recent years for oil-water separation, thanks to their large surface area, controllable porosity and hydrophobic properties^{41, 42}. The materials showed relatively low cost and high absorption ability for oil contaminants. However, polymers are limited for wide applications possibly due to their unclear impact to the environmental and ecological systems^{15, 33}. In addition, there is a concern that most polymers can be dissolved/swelled by the organic liquids, resulting in a negative effective on the structural stability of materials. In this context, sustainable materials originated from natural resources with excellent stability and recyclability, eco-friendliness, like the aerogel developed in this study, are a good candidate for practical applications. This field is in need of more participation from the material communities to achieve the high impact it deserves.

Conclusions

In conclusion, MCF aerogel was developed by a facile method using sustainable bamboo chopsticks as starting material. Compared with some newly-developed ultra-flyweight aerogels, the MCF aerogel possessed a moderate adsorption capacity for oils and organic liquids, however,

the material can be recycled easily using distillation, combustion and squeezing, mainly thanks to its porous structure and inherently hydrophobic nature, high thermal stability and excellent mechanical properties. The multi-functionality of aerogel integrated by the super hydrophobicity, mild adsorption capacity, stable recyclability and low-cost for production endowed the aerogel an ideal adsorbent for collecting oily compounds on water surface.

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Supporting Information.

Movie S1: The movie shows that the removal of crude oil on water surface using MCF aerogel.

Movie S2: The movie shows that the removal of chloroform (dyed by Sudan-Red) in water using MCF aerogel.

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Figure 1

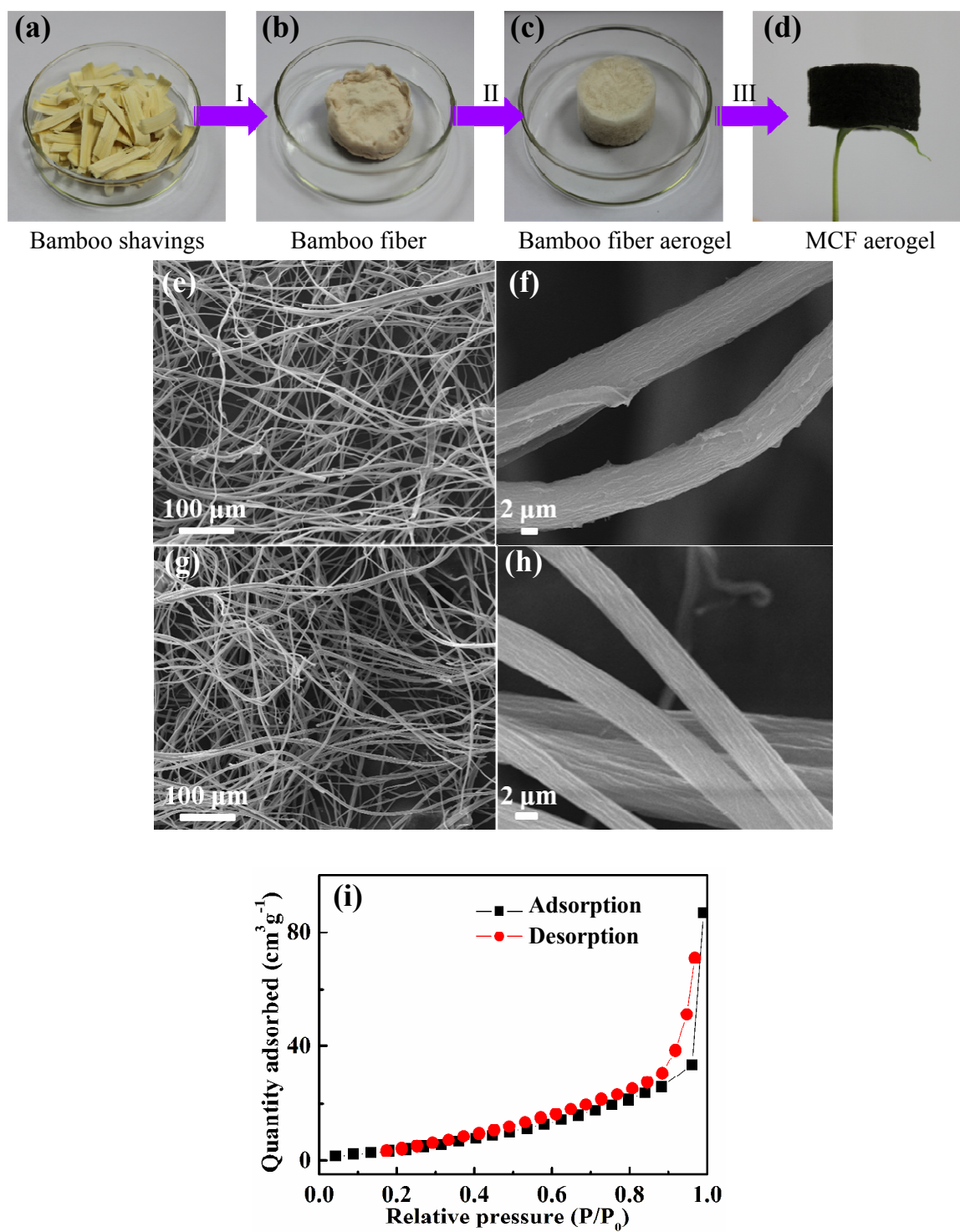


Figure 2

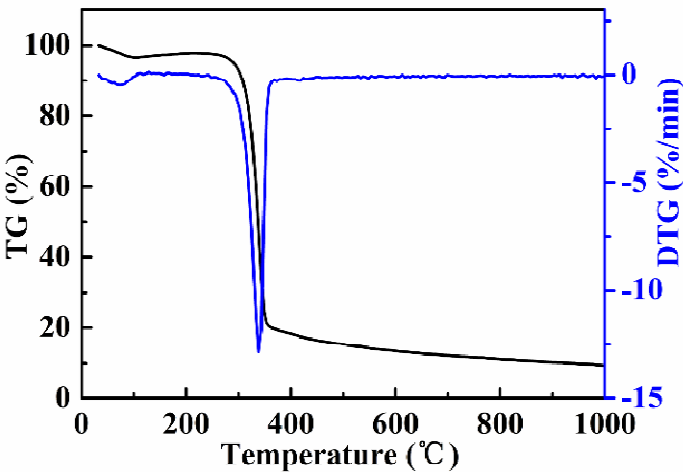


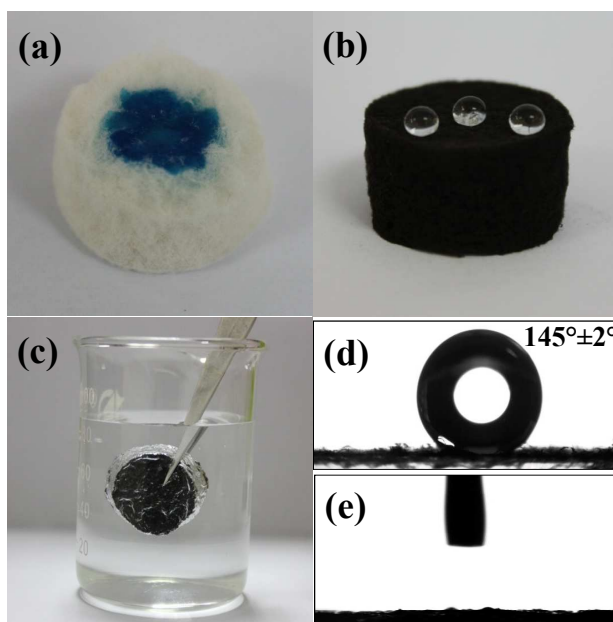
Figure 3

Figure 4

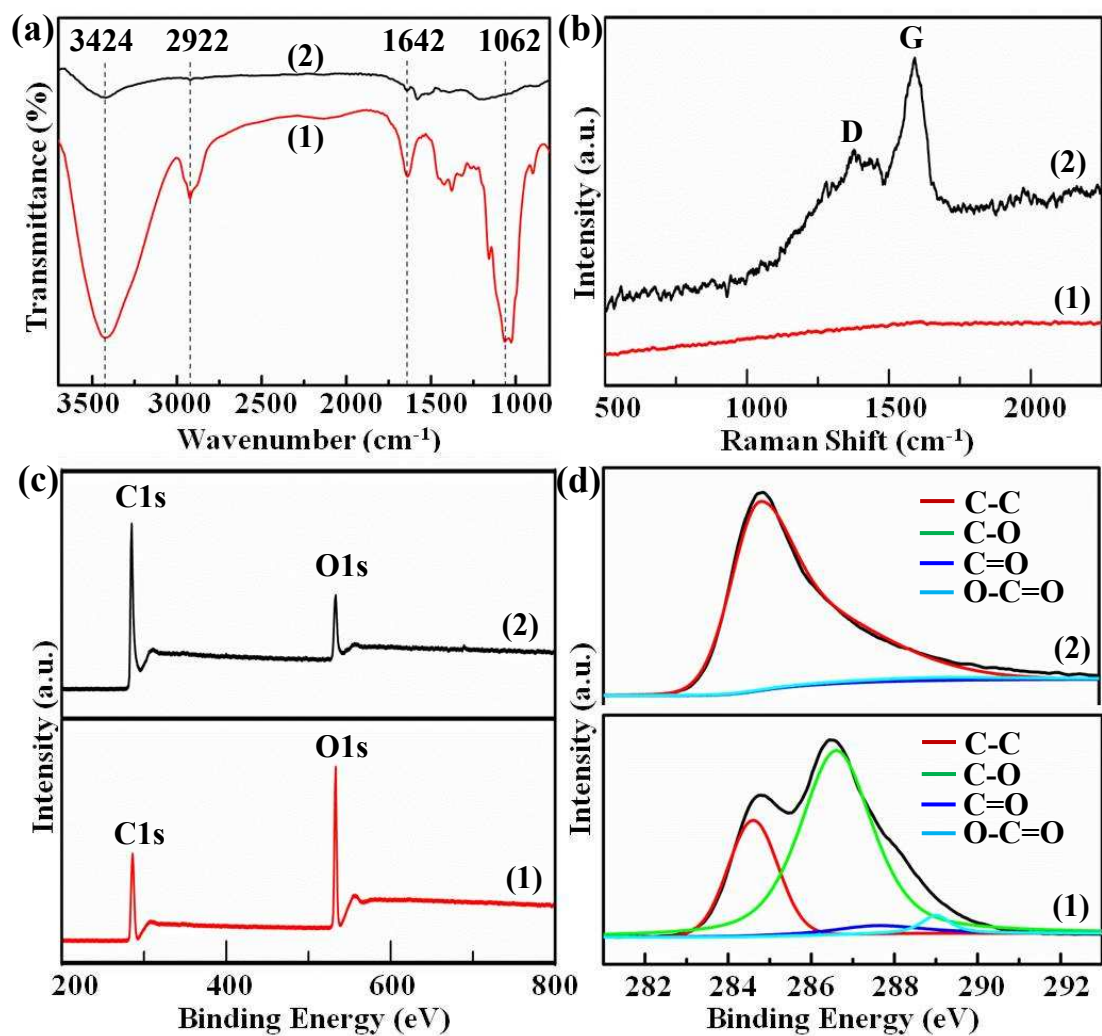


Figure 5

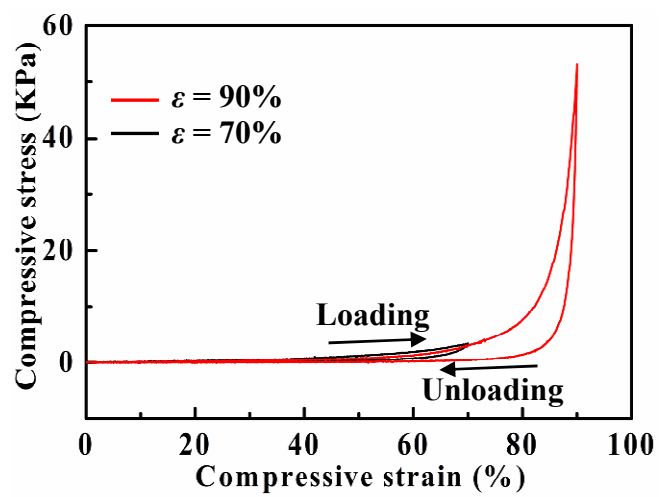


Figure 6

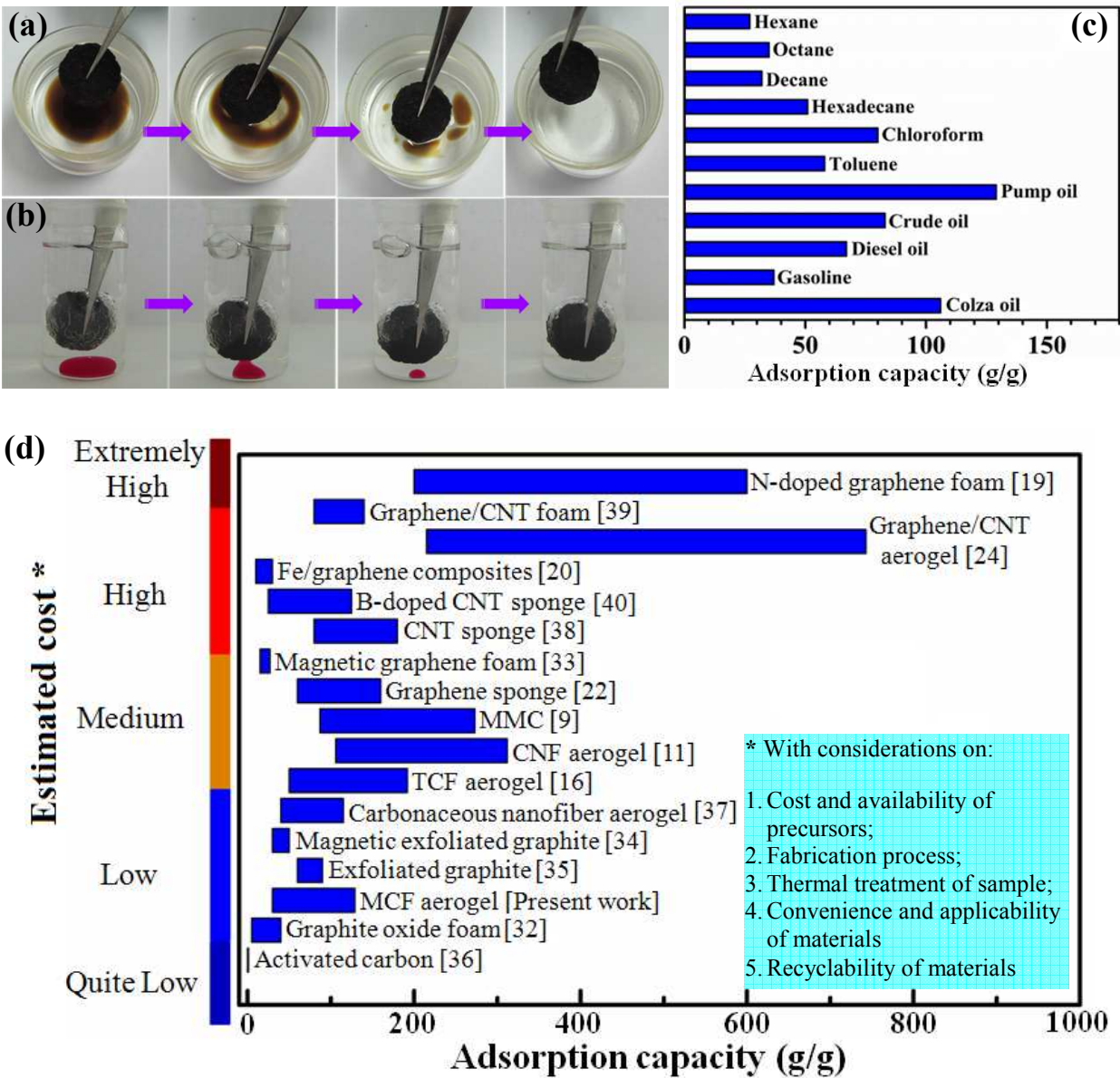


Figure 7

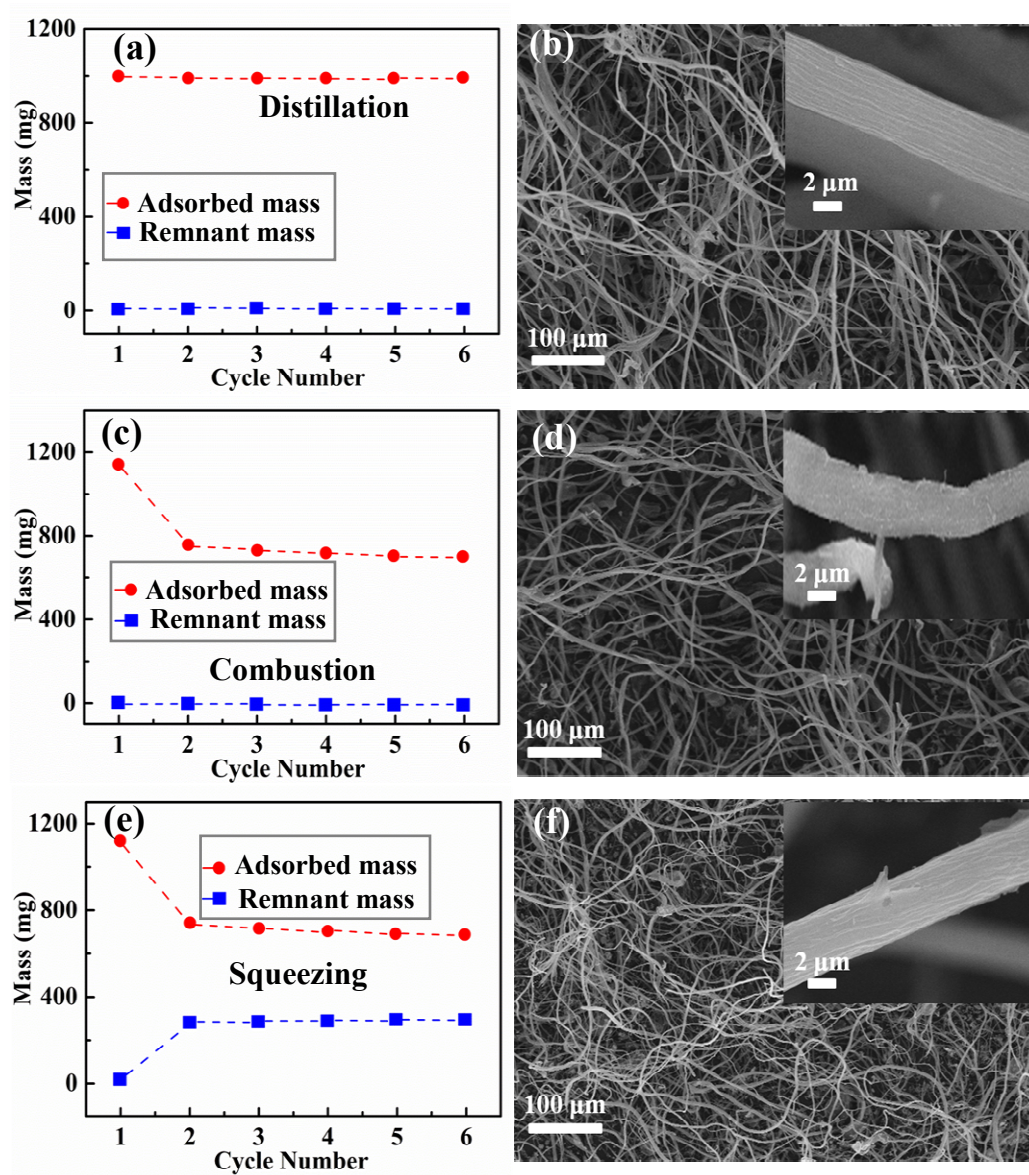


Figure 8

