Journal of the Taiwan Institute of Chemical Engineers 000 (2017) 1–8

Contents lists available at ScienceDirect

Journal of the Taiwan Institute of Chemical Engineers

journal homepage: www.elsevier.com/locate/jtice

Removal of organic solvents/oils using carbon aerogels derived from waste durian shell

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ARTICLE INFO

Article history:
Received 4 January 2017
Revised 3 June 2017
Accepted 19 June 2017
Available online xxx

Keywords:
Carbon aerogel
Durian shell
Absorption
Organic solvents
Oils

ABSTRACT

The water contamination caused by organic solvents and oils has become the focus of attention and research for every country in the world. This study was focused on the preparation method of the carbon aerogel using waste durian shell (DSCA) as the biomass precursor and its application in removal of organic pollutants. A variety of characterization techniques including the water contact angle (WCA), scanning electron microscopy (SEM), transmission electron microscopy (TEM), N_2 adsorption/desorption isotherms, Fourier transform infrared (FT-IR) spectroscopy, X-ray diffraction (XRD), and Raman spectroscopy had been exploited to investigate the synthesized materials. The results indicated that DSCA had an interconnected mesoporous structure mainly composed of the defective graphitic layers and its specific surface area was 734.96 m^2/g. Furthermore, the prepared carbon aerogels were applied to the adsorption of various organic liquids and oils in our daily life. The recyclability of DSCA was also tested in this work. DSCA, therefore, might be an alternative adsorbent for removal of organic solvents/oils in polluted waters.

1. Introduction

Among various apocalyptic warnings focused on environmental resources, the water crisis should be the most frightening and immediate problem looming on the horizon of the 21st century. It is generally known that more than 70% of the Earth's surface is covered by water, but there are still tens of thousands of thirsty humans who rely on finite supplies of freshwater to alive. Admittedly, a string of major chemical accidents in the past few decades had magnified the water scarcity issue, such as organics leak in Songhua River in China, the Gulf of Mexico oil spill and the gold event in Romania, etc. These catastrophic events usually have the characteristics of complex situation, long continuance, and great perniciousness, etc. Almost all kinds of creatures associated with the aquatic ecosystems are affected by these water pollutants owing to their inflammability, toxicity, carcinogenicity, and/or mutagenicity, especially some organic solvents and oils, such as aromatic compounds, ester, ketone, gasoline, diesel and kerosene, etc. Therefore, it is urgent to utilize effective techniques to dispose of such organic contaminants in water [1,2].

In recent years, primarily three kinds of methods have been implemented for such organics spill cleaning: chemical methods like dispersion, in-situ burning and solidification, bioremediation involving microorganisms to degrade hydrocarbon in organics, and physical methods using booms and skimmers to corral the oil. However, there are lots of disadvantages in most traditional ways, such as low purification efficiency, high energy consumption, equipment complexity, high operating costs, and restricting the scope of application of them, etc. Inversely, physical adsorption technique has attracted increasing attention because of its simple design, insensitivity and low cost of operation [3]. And it is undeniable that the adsorbent plays an important role in the process of adsorption. Therefore, it is significant to develop the novel absorbents with eminent absorption capacity, superior hydrophobicity, great recyclability and low cost [4].

Carbon aerogels are a class of important aerogels that are spongy three dimensional (3D) structured materials with millions of tiny open pores filled with air [5]. As carbon aerogels have multifunctional properties like high surface area, abundant porous structure, low mass density and excellent electrical conductivity, it is no surprise that they are considered as the ideal adsorbents for oil-spill cleanup and have triggered a number of scientific research activities [6-8]. A variety of graphene [9] and graphene oxide [10], carbon nanotube [7], cellulose [6,11] and their composite [12,13] carbon aerogels were successfully prepared and widely used in water treatment. It is noteworthy that a huge array of carbon aerogels, derived by some biomass such as watermelon [14], winter melon [15] and sugarcane [16], were directly synthesized

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http://dx.doi.org/10.1016/j.jtice.2017.06.037
1876-1070/

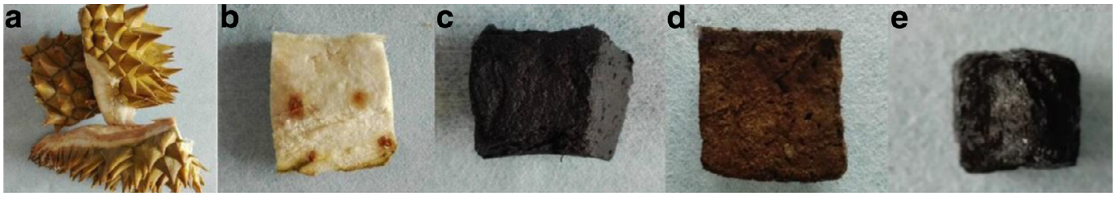

Fig. 1. The photographs of raw material (a and b), hydrogel (c), aerogel (d), and carbon aerogel (e) in the preparation process of DSCA.

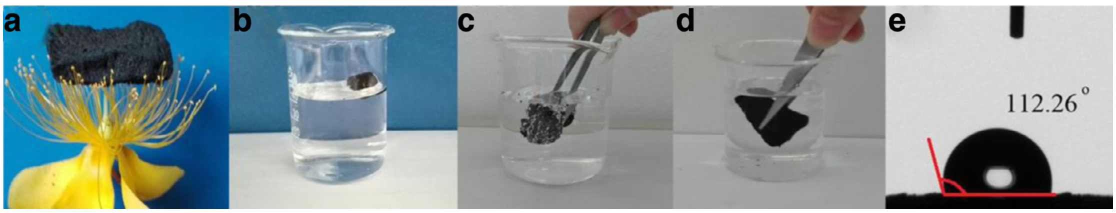

Fig. 2. Digital images of DSCA standing on a fragile soft flower (a); floating on the water surface (b); immersed in water (c) and DMF (d) by an external force. The water contact angle image of DSCA (e).

by a simple hydrothermal carbonization (HTC) process. However, the preparation of traditional carbon aerogels usually involves the harmful and costly precursors, complex process and complicated equipment [17]. While biomass-derived carbon aerogels are large class of cheap sustainable and non-toxic materials, so they have received more and more attention in recent year.

Durian, a kind of common tropical fruit, has been considered to be of high edible value and medicinal value due to its rich bioactive components. As by-products of the fruit, large amounts of durian shell are threw away after consumption, resulting in resources waste and environmental pollution. In fact, these biomass materials consisting of lignin, hemicellulose, phenolic compounds and flavonoids are some effective natural sorbents with unique chemical composition, availability in abundance, renewable nature and low cost [18]. The application examples of durian shell based materials, therefore, have been increasing in the recent years. For instance, Tham et al. [19] prepared a sort of activated carbon based on durian shell with different concentrations of H_3PO_4 and studied its adsorption capacity for toluene. The capability of durian shell waste biomass as a sorbent for Cr(VI) removal from synthetic wastewater has been explored by Kurniawan et al. [20]. And with simple physical treatments, adsorption potential of durian peel for the Brilliant Green from aqueous solution was studied by Dahri et al. [21]. Encouraged by these studies, there was a strong desire to carry out this study as the following.

Herein for the first time, durian shell as the biomass precursor that was spongy in nature and had interconnected macropore network, was used for the preparation of carbon aerogel via HTC, freeze-drying and pyrolysis process. The relevant physicochemical properties of the prepared materials, such as characteristics of the water contact angle, surface morphology, pore size, functional groups and crystal structure, were examined by utilizing a sequence of analytical techniques. The carbon aerogel derived from the durian shell (DSCA) was used as the inexpensive and environment-friendly adsorbent for organic solvents/oils removal. The absorption capacity and recyclability of DSCA for various organic solvents/oils were investigated and compared.

2. Materials and methods

2.1. Materials

Waste durian shell was collected from a fruit stall in Zhenjiang, China. Dimethylformamide (DMF), ethanol, acetone, para-xylene (PX), n-heptane, toluene, methanol, formic acid, 1-methyl-2-pyrrolidone (NMP), pump oil, castor oil, sunflower oil, soybean oil and Sudan red III were supplied by Sinopharm Chemical Reagent Co. Ltd., Shanghai, China. All reagents employed were of analytical grade and used as received without any further purification.

2.2. Preparation procedure of carbon aerogels

As shown in Fig. 1, prickly husk of durian shell was removed and the flesh of the durian shell was cut into small pieces with dimension of about 4 × 2.5 × 1 cm^3 at first. Then small pieces were put into a Teflon-lined stainless-steel autoclave (KH-200, China.) and hydrothermal treated at 180 °C for 10 h Successively, spongy durian shell hydrogels were taken out and washed repeatedly with hot water about 70 °C for two days to remove soluble impurities. After that, the brown monoliths were dried at −80 °C for 48 h using a vacuum freeze dryer (FD-1A-80, China). Finally, the black carbon aerogels were obtained by pyrolyzing durian shell hydrogels at 800 °C for 1 h in N_2 atmosphere using a tube furnace (OTF-1200X, China) [15,16].

2.3. Materials characterization

A contact angle system (KSV CM20, Finland) was used to investigate the water contact angle (WCA) of DSCA prepared in our work. In a static contact angle measurement, a water droplet about 0.5 µl is deposited on the sample surface via a syringe system of the tester. Besides, the water contact angle images of DSCA were captured by a high resolution video system and then were analyzed using image analysis software. And The WCA of DSCA was an average at three different positions [6]. The surface morphology of various materials were observed by Scanning electron microscopy (S-3400 N, Japan) at an accelerating voltage of 15 kV and the samples were coated with gold for 5 min before tests [22]. In addition, TEM images of them were obtained by Transmission electron microscope (JEM-2100HR, Japan) at an accelerating voltage of 200 kV. Before measurements, the samples were dispersed evenly in ethanol, and the obtained suspension dripped on the copper meshes [23]. The specific surface area and the pore size distribution of DSCA were obtained by utilizing the surface area analyzer (Quanta Chrome Corporation, USA) at 77 K [24]. To identify the chemical functional groups present on the prepared samples, the FTIR spectra were recorded on NEXUS 670 FTIR Spectrometer using the KBr method in a wave number range of 400–4000/cm. The samples and KBr pellets were mixed at a mass ratio