



Separation of dissolved oil from aqueous solution by sorption onto acetylated lignocellulosic biomass—equilibrium, kinetics and mechanism studies



Siew Chin Cheu^a, Helen Kong^a, Shioh Tien Song^a, Khairiraihanna Johari^b,
Norasikin Saman^a, Mohd Azizi Che Yunus^c, Hanapi Mat^{a,d,*}

^a Advanced Materials and Process Engineering Laboratory, Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor, Malaysia

^b Department of Chemical Engineering, Faculty of Engineering, Universiti Teknologi PETRONAS, 32610, Bandar Seri Iskandar, Perak, Malaysia

^c Centre of Lipid Engineering and Applied Research (CLEAR), Ibnu Sina Institute for Scientific & Industrial Research (Ibnu Sina ISIR), Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor, Malaysia

^d Advanced Material and Separation Technologies (AMSET) Research Group, Health and Wellness Research Alliance, Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor, Malaysia

ARTICLE INFO

Article history:

Received 27 September 2015

Received in revised form 11 December 2015

Accepted 27 December 2015

Available online 31 December 2015

Keywords:

Lignocellulosic biomass

Pineapple leaves

Acetylation

Oil-in-water solution

Adsorption

ABSTRACT

The potential use of lignocellulosic biomass as oil sorbents was demonstrated through acetylation of mercerized pineapple leaves (M-PAL) using acetic anhydride with catalysts (*N*-bromosuccinimide and pyridine) in order to enhance its surface hydrophobicity and thus oil sorption capacity for treatment of dissolved oil contaminated wastewaters. The raw and acetylated PAL sorbents, before and after oil sorption, were characterized by a scanning electron microscope (SEM), transmission electron microscopy (TEM), Fourier transform infrared (FTIR) spectroscopy and X-ray photoelectron spectroscopy (XPS). The oil sorption experiment was evaluated in batch sorption mode using dissolved o/w solution prepared by mixing crude oil and double-distilled water. It was found that the M-NBS-PAL sorbent yielded better oil sorption performance than R-PAL sorbent due to the replacement of hydroxyl group which has more hydrophobic acetyl group on the PAL surfaces, with strong affinity towards oil. The batch equilibrium data were fitted well by Langmuir isotherm model with the maximum sorption capacity (q_m) of 37.45 and 90.91 mg/g for the R-PAL and M-NBS-PAL sorbents, respectively. The oil sorption process was thermodynamically feasible and exothermic, while the nature of interaction could be described via the physisorption mechanism. The kinetic sorption data were found to be fitted well into the pseudo-second order kinetic model. The sorption–desorption cycle was repeated four times whereby isopropanol was used as a desorbing agent and the results were comparable with freshly prepared sorbent. Finally, the present findings indicate that the lignocellulosic biomass could be a potential alternative as sorbent precursors for oil removal from oil contaminated wastewaters.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

Oil is one of the major contaminants in water that exists in the forms of either heterogeneous or homogeneous oil-in-water (o/w) system, and/or a combination of both systems. The heterogeneous o/w normally contains free and dispersed oil with bigger oil droplet size (>5 microns) which is able to float on the water surface, while the homogeneous o/w is due to the dissolved oils such as aromatic and phenolic compounds [1]. Oil spill is the main

source of oil contamination in water which is a result of unintentional introduction of oil into the environment during the extraction, production, transportation or storage processes [2]. The oily wastewaters and produced waters generated from oil and gas exploration activities, crude oil refinery, petrochemical and petroleum refining plants, steel manufacturing and metal working, vehicle repair, and various types of manufacturing plants are other sources of water contamination by oil [3].

There are many techniques available for o/w separation regardless of being physical, biological or chemical such as gravity separation, chemical treatment methods, flotation system, coagulation, filtration, hydrocyclone, electrical process, reverse osmosis and membrane reactor which offer advantages and drawbacks

* Corresponding author: Fax: +607 558146.

E-mail address: hmat@cheme.utm.my (H. Mat).

over others [4–6]. Some existing methods are expensive, and sometimes some supplementary treatments are needed in order to achieve the stipulated environmental standards. Among these methods, sorption process has emerged as a highly efficient treatment method due to its simplicity of the design, ease in operation and no reaction with toxic substances in removing dissolved organic components from water [7–9]. The high commercial sorbent costs and stringent environmental regulations have led to the needs to develop low-cost sorbents derived from lignocellulosic materials, such as wheat straw, sawdust, rice residues, corncob, coconut husk, kenaf, kapok fibers, cotton, wool and wood [10]. The advantages of these materials include being low cost, biodegradable and non-toxic [11] as compared to the commercial sorbents; however, appropriate modifications are required in order to achieve acceptable sorbent technical specifications [12].

Mostly, the pristine lignocellulosic materials exhibit lower oil sorption capacity than chemically modified ones due to the presence of hydroxyl group (OH) on their surfaces that tends to adsorb water more than oil. Several modification methods have been introduced for utilizing lignocellulosic materials, including acid treatment, alkaline treatment, etherification, esterification, polymer grafting and carbonization [13–20]. The performance of these methods of modification also depends on sorbent precursors and therefore the use of published data do not to some extent allow for evaluation of the performance of one method to another due to the different lignocellulosic biomass used in the studies. For instance, acetylation is one of the modification methods and there were some previous studies on acetylation of lignocellulosic biomass using acetic anhydride using various types of catalysts on different lignocellulosic biomass. Furthermore, the application of this modified lignocellulosic biomass has widely been reported predominantly for the removal of oil from the dispersed o/w solutions [11,21–34]. There is no report on the oil sorption performance of lignocellulosic biomass from dissolved o/w solution and its comparison with the dispersed o/w solution.

The present study is mainly focused on the separation of dissolved oil by lignocellulosic biomass, pineapple leaves (PAL). The PAL has a high cellulose content (74.33% wt.) which could ease modification processes due to the presence of high content of hydroxyl functional groups on the surfaces. It was modified by acetylation and used as a sorbent to remove oil from dissolved o/w solution (or homogeneous o/w solution) which has been less studied compared to the dispersed o/w solution (or heterogeneous o/w solution). The synthesized PAL sorbents were characterized by using the Fourier transform infrared (FTIR), scanning electron microscope (SEM) and elemental analysis to study the morphological properties, surface functional groups and elemental composition of the PAL sorbents. Further analyses using the X-ray photoelectron spectroscopy (XPS) and transmission electron microscopy (TEM) analysis were also conducted. The oil sorption of raw PAL (R-PAL) and mercerized–acetylated PAL (M-NBS-PAL and M-P-PAL) sorbents from dissolved o/w solution (or homogeneous o/w solution) was experimentally investigated followed by equilibrium and kinetic model analyses. The oil sorption of PAL sorbents from a dispersed o/w solution was also carried out as a comparison to that of the dissolved o/w solution. Finally, the potential use of the PAL and thus lignocellulosic biomass as oil sorbent precursors was demonstrated by comparing with the published data.

2. Materials and methods

2.1. Materials

Lignocellulosic biomass of pineapple leave (PAL) was collected from Pekan Nenas, Johor, Malaysia. Acetic anhydride, *N*-

bromosuccinimide (NBS), pyridine, methanol, acetone, ethanol, sodium hydroxide (NaOH) and hydrochloric acid (HCl) were purchased from Sigma–Aldrich (USA) and Merck (Germany). The crude oil used as model oil for the whole experiment was obtained from a local petroleum company. Double-distilled water was used for solution preparation throughout the experiments.

2.2. Sorbents preparation

The PAL were cut into small pieces and dried under sunlight for several days. The dried PAL were then ground into small size and sieved to obtain targeted sample size of 75–150 μm . The sieved PAL sample was soaked in ethanol and water mixture, followed by agitation and centrifugation to remove dirt and impurities. Then, the washed PAL sample was oven dried for 24 h at 333 K and stored in a desiccator for further characterization and modifications. This PAL sample was referred to as a raw PAL (R-PAL) sample.

Alkaline pretreatment of R-PAL by mercerization was performed by treating 15 g of R-PAL with 5% aqueous NaOH solution under constant stirring at room temperature (303 K) for 18 h. The fibers were then washed with double-distilled water for several times to remove any traces of NaOH sticking on the fiber surface and then neutralized with acetic acid. The mercerized samples (M-PAL) were washed again with double-distilled water until pH 7 before oven dried at 333 K for 24 h.

The acetylation of the M-PAL sample in solvent free system was performed according to Teli et al [21]. 5 g of the R-PAL sample was placed in a 500 mL round bottom flask, which contained 150 mL of acetic anhydride and 2% NBS as catalyst. The flask was then placed in an oil bath at 403–413 K and refluxed for 2 h. After that, the acetylated PAL sample was washed thoroughly using ethanol and acetone to remove the excess acetic anhydride and acetic acid byproducts. The acetylated PAL (M-NBS-PAL) sample was then oven-dried for 24 h at 333 K. As comparison, pyridine was also used as a catalyst to synthesize the acetylated PAL (M-P-PAL). The R-PAL, M-NBS-PAL and M-P-PAL samples were used as sorbents in oil sorption experiments.

2.3. Morphological and functional groups determination

The total content of carbon, hydrogen, oxygen and nitrogen in the samples was analyzed by using CHNS/O analyzer (CHNS/O Analyzer 2400, PerkinElmer, USA). The surface morphology of the PAL before and after acetylation was determined using a scanning electron microscope (SEM) model JEOL JSM–7500 F (Japan) and transmission electron microscope (TEM) model H-7000 FA electron microscope (Hitachi, Tokyo, Japan) with 75 kV of acceleration voltage. The FTIR spectra of R-PAL and mercerized–acetylated PAL were acquired using FTIR spectrometer model Thermo Scientific Nicolet iS5 (USA) via the ATR sampling technique. The spectra were obtained by scanning the samples in the range of 4000 to 600 cm^{-1} at a resolution of 4 cm^{-1} to identify the surface functional groups responsible for the oil sorption. The X-ray photoelectron spectroscopy measurement of the samples was carried out using a X-ray photoelectron spectrometer (XPS) model Shimadzu KRATOS AMICUS (Japan), equipped with unmonochromated aluminum Al $K\alpha$ source (1486.8 eV) with an operating pressure of 10^{-9} mbar and 10 mA of current. The XPS spectra were taken at 150 eV of constant analyzer energy (CAE) mode with 1 eV of energy step size, while high-resolution XPS spectra of O_{1s} , N_{1s} and C_{1s} were taken at 25 eV of CAE and 0.05 eV of energy step size. The specific surface area was measured by a BET surface area analyzer model AutoSorb-1 (Quantachrome, USA). The pore size distribution, average pore size and pore volume of sorbents were determined using the Barrett, Johner and Halenda (BJH) method.

Download English Version:

<https://daneshyari.com/en/article/221938>

Download Persian Version:

<https://daneshyari.com/article/221938>

[Daneshyari.com](https://daneshyari.com)