



Research article

Coconut endocarp and mesocarp as both biosorbents of dissolved hydrocarbons in fuel spills and as a power source when exhausted



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ABSTRACT

Health and environmental problems associated with the presence of toxic aromatic compounds in water from oil spills have motivated research to develop effective and economically viable strategies to remove these pollutants. In this work, coconut shell (endocarp), coconut fiber (mesocarp) and coconut shell with fiber (endocarp and mesocarp) obtained from coconut (*Cocos nucifera*) waste were evaluated as biosorbents of benzene, toluene and naphthalene from water, considering the effect of the solution pH (6–9) and the presence of dissolved organic matter (DOM) in natural water (14 mg/L). In addition, the heat capacity of saturated biosorbents was determined to evaluate their potential as an alternative power source to conventional fossil fuels. Tests of N₂ physisorption, SEM, elemental and fiber analysis, ATR-FTIR and acid-based titrations were performed in order to understand the materials' characteristics, and to elucidate the biosorbents' hydrocarbon adsorption mechanism. Coconut fiber showed the highest adsorption capacities (222, 96 and 5.85 mg/g for benzene, toluene and naphthalene, respectively), which was attributed to its morphologic characteristics and to its high concentration of phenolic groups, associated with the lignin structure. The pH of the solution did not have a significant influence on the removal of the contaminants, and the presence of DOM improved the adsorption capacities of aromatic hydrocarbons. The adsorption studies showed biphasic isotherms, which highlighted the strong affinity between the molecules adsorbed on the biosorbents and the aromatic compounds remaining in the solution. Finally, combustion heat analysis of coconut waste saturated with soluble hydrocarbons showed that the heat capacity increased from 4407.79 cal/g to 5064.43 ± 11.6 cal/g, which is comparable with that of woody biomass (3400–4000 cal/g): this waste biomass with added value could be a promising biofuel.

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1. Introduction

Aromatic hydrocarbons enter the environment through anthropogenic activities such as crude oil spills, burning fossil fuels and fuel leakages (Foght, 2008). These hydrocarbons are comprised of simple aromatic compounds (monocyclic) such as benzene and toluene, polycyclic aromatic hydrocarbons (PAHs) such as naphthalene and pyrene, and isomers with countless alkyl substituents (Flores-chaparro et al., 2016; Foght, 2008). Annually, aromatic compounds impact environments such as groundwater, surface freshwater bodies, soils and terrestrial and marine sediments (Ali and Aboul-Enein, 2004; Foght, 2008), causing damage to flora,

fauna and humans. For example, the consumption of water containing aromatic compounds may cause in the human population: anemia, problems in the nervous system, kidney or liver, as well as increased risk of cancer (WHO, 2011). Therefore, it is important to contribute to the development and understanding of the techniques that are the most appropriate to remove aromatic compounds from water. The adsorption process is a feasible technique for the removal of several pollutants (Ali and Gupta, 2006). A wide variety of adsorbent materials have been used to remove organic pollutants from water, including activated carbon, zeolites, activated alumina and polymeric resins (Ali et al., 2012; Flores-chaparro et al., 2016). In recent years, the use of low-cost materials, among them biomass, is considered a promising alternative for replacing conventional adsorbents to remove organic compounds from water (Abdolali et al., 2014; Fomina and Gadd, 2014; Michalak et al., 2013; Tran et al., 2015). Biosorption is the

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property of certain types of biomass to concentrate on its surface certain molecules or ions from aqueous solutions (Volesky, 2007); basically, a low-cost biosorbent requires little processing, is abundant in nature or is a byproduct or waste from any industry or process (Fomina and Gadd, 2014). Additionally, the disposal of exhausted biomass is usually a problem; hence, its reuse is crucial for a cost-effective process. Its combustion could be a viable alternative to conventional fossil fuels (Almendros et al., 2015; Ronda et al., 2016), moreover, it is known that biomass can become one of the most valuable sources to reduce pollution emissions from energy production processes (Ronda et al., 2016).

The use of agricultural waste from the coconut (*Cocos nucifera*) has received attention as an effective adsorbent due to its low cost, high potential to remove several water contaminants and as an energy resource due to its high carbon content and fuel value (Yerizam et al., 2013). Moreover, the physical characteristics of the pristine material allow it to be used directly in adsorption processes in packed-bed columns on an industrial scale, or in reactive permeable barrier (PRBs) technologies for sustainable in situ remediation of contaminated groundwater. Coconut-based adsorbents have been applied mainly in the removal of metals (Conrad and Bruun, 2007; de Sousa et al., 2010; Mohan et al., 2006; Parab et al., 2008; Selvi, 2001), dyes (Gupta et al., 2010; Hameed et al., 2008a; Kavitha and Namasivayam, 2008; Sureshkumar and Namasivayam, 2008; Vieira et al., 2009), phenol compounds (Hameed et al., 2008b; Sathishkumar et al., 2007; Tan et al., 2008), inorganic anions (Namasivayam and Sangeetha, 2008, 2006; Namasivayam and Sureshkumar, 2007) and radionuclides (Anirudhan et al., 2009; Bhatnagar et al., 2010; Parab and Sudersanan, 2010). In addition, since the coconut waste is produced in large quantities in several countries, e.g., Indonesia, the Philippines, India, Brazil, Mexico, among others (CONACOCO, 2012), these are widely available and therefore it is important to analyze strategies to valorize them as biosorbents and also the exhausted biosorbents, as a power source such as an alternative to fossil fuels.

Reports regarding the removal of aromatic hydrocarbons from aqueous solutions using biosorbents are scarce (Agarry et al., 2013; El-gendy and Nassar, 2016; Nduka et al., 2008; Sanchez-Galvan et al., 2013; Wahi et al., 2013; Younis et al., 2015; Zhang et al., 2014). Owabor and Agarry (2014) investigated the use of coconut shell for the removal of naphthalene and pyrene from synthetic wastewater considering the variation of adsorbates, dose of adsorbent, the particle size and the stirring time. The maximum adsorption capacity for naphthalene obtained by the Langmuir model was 24.3 mg/g. The authors did not show data on the characterization of the material, neither was an adsorption mechanism proposed. To the best of our knowledge, there are no studies reporting either the removal of benzene and toluene from aqueous solutions by coconut waste, or the assessment of the exhausted biosorbents to be used as a power source.

According to the above, the main objective of this study was to assess, in batch studies, the removal efficiency of benzene, toluene and naphthalene from water, by using the mesocarp and endocarp of coconut waste as biosorbents, and to determine the suitability of hydrocarbon-loaded coconut biosorbents for energy production by combustion. For this purpose, the biosorbents were physically and chemically characterized, the enthalpy of adsorption was experimentally determined and the heat of combustion of exhausted materials was acquired. Since the pH of natural waters should be between 6.5 and 8.5, and the fate of organic contaminants in aquatic environments can be altered by their interactions with complex mixtures such as dissolved organic matter (Xiao et al., 2014), the effect of pH and dissolved organic matter on the removal of aromatic compounds was also evaluated.

2. Materials and methods

2.1. Materials

Coconut residues were obtained from a local seller in San Luis Potosi, Mexico, and their fractions were divided into: the hard part (endocarp-coconut shell), the fibrous part (mesocarp-coconut fiber) and a combination of both (coconut shell with fiber), which were designed as CsF, F and CcF, respectively. The coconut shell was ground and sieved to get a particle size between 0.5 and 1.7 mm and the fibers were standardized to a size of approximately 1 cm. These materials were washed with deionized water until colorless water was obtained. Finally, the biosorbents were dried in an oven at 50 °C for 48 h, and were stored in polyethylene bags until the experiments were carried out.

2.2. Biosorbent characterization

The surface area and pore size distribution of the biosorbents were obtained by N₂ physisorption employing a surface analyzer (Micromeritics ASAP 2020). The samples were outgassed at 40 °C for 12 h. The morphology of the endocarp and mesocarp was observed by a field emission scanning electron microscope (FEI Quanta 200 Environmental SEM) equipped with a BSD detector, operated at 20–25 kV and a working distance of 10 mm; the samples were coated with a layer of gold before the microscopic examination. The elemental analysis was done in an Elemental Combustion System (COSTECH Model 4010), using 0.5 mg of each biosorbent. The main lignocellulosic fractions in the coconut waste were quantified by using a fiber analyzer (ANKOM 200). The procedure was based on the sequential extractions method proposed by (Van Soest, 1963). The point of zero charge (pH_{PZC}) and the quantification of functional groups were obtained through potentiometric titrations (Velazquez-Jimenez et al., 2013) by using NaCl 0.01 M as a supporting electrolyte; the data were analyzed by using SAIEUS program. Finally, ATR-FTIR spectroscopy (Thermo Nicolet Nexus 470 FT-IR) was performed to identify the functional groups present in the biosorbents, the spectra were obtained in the range between 500 and 4000 cm⁻¹ with 4 cm⁻¹ of resolution and 64 scans averaged.

2.3. Aromatic hydrocarbons adsorption: equilibrium and kinetics experiments

The hydrocarbon solutions used for equilibrium and kinetic experiments were prepared, based on the solubility reported (Flores-chaparro et al., 2016), within the range of 3.6–30 mg/L, 60–500 mg/L and 213–1780 mg/L for naphthalene, toluene and benzene (Sigma Aldrich, 99% purity), respectively. 0.05 g of CsF, CcF (10% fiber weight), or 0.015 g of F were contacted with 0.025 L of solution in amber glass containers and then closed with Teflon caps, with no free headspace to avoid volatilization. The bottles were kept at 25 °C and 110–120 rev/min for two days. The influence of initial pH in the adsorption process was investigated within the pH range of 6–9 for each chemical compound. All tests were done in duplicate. Moreover, adsorption kinetics were performed using a total of 15 bottles for every solution, and at given time intervals a bottle was sampled. The equimolar kinetics were performed using a stock solution with a concentration of 195 μM of each studied compound. The effect of dissolved organic matter in the adsorption process was performed by using natural water from a reservoir located in San Luis Potosi, Mexico. The adsorption capacity of the biosorbents (q_e, mg/g) was calculated as follows:

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