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REMOVAL OF OIL FROM CRUDE OIL POLLUTED WATER USING MANGO SEED BARK AS SORBENT IN A PACKED COLUMN

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Abstract: A column study was conducted to evaluate efficiency of mango seed bark as a sorbent material in the removal of crude oil from crude oil polluted water. The study was carried out in a packed continuous column with initial oil concentration of 200 mg/L. The result showed that increasing the bed height of the packing from 5 - 25 cm resulted to an increase in percentage removal of the oil from 47.69 - 95.38 % at a constant contact time of 30 min and flow rate of 26.52 cm^3/min and correlation coefficient of $R^2 = 0.92$. The study identifies mango seed bark as a suitable sorbent that can be used for continuous remediation of oil from crude oil polluted water. The maximum uptake of 190.77 mg/L oil was recorded at 25 cm bed height.

Keywords: Mango seed bark, oil pollution, packed column, remediation

Introduction

Since the advent of crude oil exploration, spillage from oil fields has created serious environmental concern due to its hazardous nature and its eventual contamination of both the aquatic and terrestrial ecosystems. Oil is a naturally occurring substance. The organic residue from the decay of plant and animals are converted by heat and pressure into petroleum, migrating upwards, sometimes over extensive areas, either to reach the surface or be occasionally trapped in to become oil reservoirs.

Oil is one of the most important sources of energy and is also used as raw material for synthetic polymers and chemicals worldwide. Oil has been a part of the natural environment for millions of years (Karan *et al*., 2010). The methods commonly used to remove oil involve oil booms, dispersants, skimmers, oil pumping, in-situ burning, bioremediation, solidifier and sorbents (Aliyu*et al*., 2015). The main limitations of some of these techniques are their high cost and inefficient trace level adsorption. Also most of the dispersants are often inflammable and cause health hazards to the operators and potential damage to fish, and marine mammals. They can also lead to fouling of shorelines and contamination of drinking water sources.

Removal of oil by sorption has been observed to be one of the most effective techniques for complete removal of spilled oil under ambient conditions. The use of sorbents is of great interest, as it allows the collection and complete removal of oil by achieving a change from liquid to semi-solid phase. The efficiency of an oil sorbent material is determined by properties, such as hydrophobicity, high uptake capacity, high rate of uptake, retention over time, oil recovery, reusability and biodegradability (Aliyu*et al*., 2015). The sorbent materials in use for oil spill clean-up include organic natural ones and agro wastes, such as egg shell (Aliyu*et al*., 2016), cassava peel activated carbon (Oghenejoboh*et al*., 2016), mango seed kernel powder (El-Nafaty*et al*., 2014), oil removal using banana peel (Aliyu*et al*., 2015), low grade raw cotton fibres (Hussein *et al*., 2011), coconut fibre carbon (Egwaikhide*et al*., 2007), hybrid peel waste of musabalbisiana and citrus sinensis in preliminary study (Abdullah *et al*., 2016) have been investigated. Since most oil products are biodegradable, oil could be disposed of for example by composting. A biodegradable material with excellent adsorption properties would be advantageous in this respect (Hussein *et al*., 2011). In practice, clean up oil spill is a difficult economic problem. It is uneconomical to store large quantities of sorbent materials that are used to clean up oil spills and their disposal. The use of sorbents made from organic materials does not cause additional problems in the disposal of the spilled oil (Karan *et al*., 2010). This consideration therefore underlines the significance of establishing sorbent properties of locally available agro waste.

Global production of mangoes is concentrated mainly in Asia and more precisely in India (number one producer in the world). Mangoes are grown in 85 countries and 63 countries provide more than 1000 metric tonnes a year. Total world production was 24,420,116 metric tonnes in 1999 with developing countries accounting for about 98% of total production (Yusuf &Salau, 2007). Despite lack of encouragement as to large scale production of tropical fruits in the country, Nigeria still occupies the 8th position in the world ranking of mango producing countries as at 2002. This is instructive as it suggests the potential of tropical fruit in Nigeria. The main producing states in the country include Benue, Jigawa, Plateau, Yobe, Kebbi, Niger, Kaduna, Kano, Bauchi, Sokoto, Adamawa, Taraba, and FCT (Yusuf &Salau, 2007). Therefore, there is need for an effective remediation process that will remove the oil spills from the water surface.

Materials and Methods

Reagents

All reagents used were of analar grade and distilled water was used throughout the work

Apparatus and equipment

Meter rule, Long glass column, pH meter (Jenway 3505, UK), Milling machine, Analytical balance (JD 400-3, UK), Hot oven (Memmert, Germany), Spectrophotometer (HACHDR/2400, USA), FTIR (SCHIMADZU8400S, Japan), BET (V-sorb 2800P, China).

Sample collection

The waste samples (mango seeds) were obtained in Basawa community, Zaria, Kaduna State. The crude oil was obtained from Kaduna refinery and petrochemicals, Kaduna State, Nigeria.

Sample preparation

Preparation of simulated crude oil polluted water

The simulated oil polluted water was prepared by pouring 5 g of the crude oil into 25L of distilled water in a 50L plastic gallon and was shaken vigorously and stored for the sorption experiment.

Preparation of sorbent material

The waste samples (mango seeds) barks were removed manually and the bark was oven dried at 105°C. It was then pulverized to finer particles, passed through a mesh of 300 μm particle size. The powdered bark was washed with distilled water severally to rid the sample of traces of impurities and dried in an oven at a temperature of 105°C until constant weight. The dried powdered then was stored in glass bottle labelled as mango seed bark (MSB) sample.

Characterization of sorbent material

The functional groups present at the surface of the mango seed bark (sorbent material) was determined using FTIR (SCHIMADZU8400S, Japan) ranging from 400 – 4000 cm^{-1}, and surface area of the sorbent was determined using BET (Brunaeur-Emmet-Teller) V-Sorb 2800P Surface Area and Porosimetry Analyzer China).

Sorption experiment

The glass column was clamped vertically and a meter rule was also clamped beside it as shown in Plate 1 below. The base of the meter rule and the column were at the same level. A perforated rubber cork was inserted at the bottom of the column. A plastic aspiration bottle containing the produced polluted water was placed above the column with a rubber tube connected to a valve and dropped into the column continuously. A beaker was placed below the column to collect the treated water. The column was packed with the sorbent material (mango seed bark) to a height of 5 cm. A control valve was slightly opened to allow the oil polluted water to flow into the bed and treated water was collected below the column using a beaker. The experiment was repeated for different bed heights of 10, 15, 20 and 25 cm, respectively at a constant flow that was determined during the experiment. Each of the samples with respect to their bed heights was collected separately in a plastic sample bottle and labelled appropriately. The concentration of polluted oil adsorbed was then determined using a spectrophotometer (HACHDR/2400, USA).

Plate 1: Column experimental set up

Results and Discussion

Characterization using FTIR

FTIR spectroscopy method was used to show the functional groups present on the surface of the sorbent material(Fig. 1).

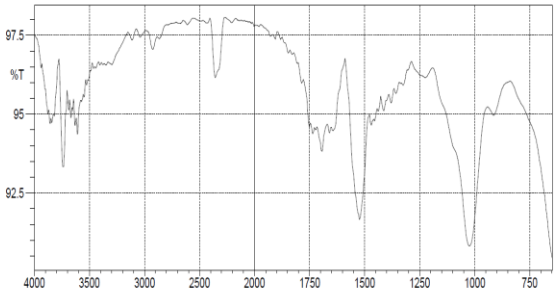

Fig. 1:FTIR spectrum of mango seed bark analysis

In Table 1, different functional groups were present on the material surface. The MSB was observed to have shift at 925 cm^{-1} which was attributed to aromatic C-H bend, at 1030cm^{-1} C-O stretch. At 1200 cm^{-1} the peak was assigned to C-O-C symmetric and asymmetric stretch, while at peak 1525 cm^{-1} was assigned to C=C-NO$_2$ symmetric. At 1625 cm^{-1} was assigned to aromatic C=C stretch and the mango seed bark was observed to have shift between 1700-1750 cm^{-1} which was attributed to C=O stretch, while at 2075 cm^{-1} was as a result of C≡C stretching.

Table 1: Functional groups and corresponding assignments present in the sorbent

Wavenumber (cm^{-1})	Functional group
925	C-H bend aromatic
1030	C-O stretch
1200	C-O-C symmetric & ass stretch
1525	C=C-NO$_2$ stretch
1625	C=C stretch aromatic
1700	C=O stretch
1707	C=O stretch
1725	C=O stretch
1750	C=O stretch
2075	C≡C stretch

Characterization using BET

The surface area of MSB was found to be 30.387848 m^2/g, surface area measurement by Langmuir method revealed the area to be 62.201848 m^2/g with pore volume 0.008853 cm^3/g and a pore size of 1.165314 nm which was better than the values reported for egg shell by (Muhammad *et al.*, 2015) reported surface are to be 0.0261 m^2/g with pore volume to be 0.00218 cm^3/g and pore size of 484.8494 nm.

Effect of bed height on percentage oil removal and service time

The graph of effect of bed height on removal and effect of service time against bed height were presented in Figs. 2 and 3, respectively

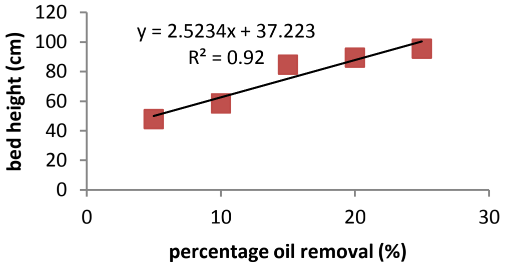

Fig. 2: Effect of bed height on oil removal

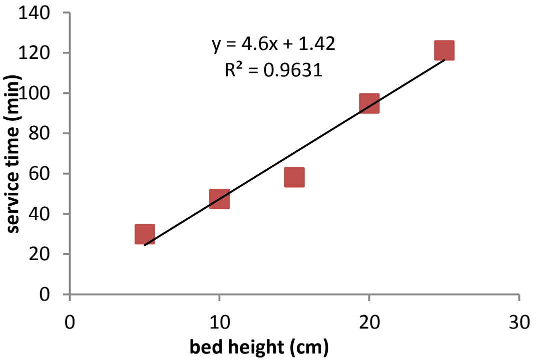

Fig. 3: Effect of service time against bed height

Figure 2, showed the breakthrough curves obtained for oil sorption on to adsorbent at different bed heights (5, 10, 15, 20 and 25 cm), at a constant flow rate of 26.52 cm^3/min and pH of 8.1. The results indicated that when bed height increased from 5 to 10 cm there was an increase in percentage removal of the adsorbent. The increase in the uptake of oil with the bed height was due to an increase in sorption sites for the oil removal. The optimum uptake which was 190.77 mg/L was found at the bed height of 25 cm. The plot of bed height against percentage oil removal is presented in Fig. 2 which shows there is direct relationship between bed height and percentage oil removal which also supported that as bed height increases, percentage oil removal also increases. The correlation coefficient ($R^2 = 0.92$) from the plot also indicated linearity which is approximated to unity 1.

Bed depth service time (BDST) models

The data collected during laboratory studies can be very well utilized for predicting and evaluating the performance of practical size columns by applying suitable mathematical models. Several models have been reported for predicting the breakthrough performance in fixed-bed sorption. BDST model was adopted in this study because of its simplicity for predicting relationship between bed height and service time. In this fixed-bed system, BDST is used to predict how long the adsorbent material will be able to sustain removing oil from water before regeneration is needed. Hutchins proposed a linear relationship between bed height and service time given by the equation below:

$$t = \frac{No}{Cou} - \frac{1}{KaCo}\left(\frac{Co-1}{Cb}\right) \text{----------} (1)$$

where: Co is the initial oil concentration (mg/L), Cb is the breakthrough oil concentration (mg/L), Ka is the rate constant in BDST model (L/mg/min), t is the time (min), u is the linear velocity (cm/min), No is the sorption capacity of bed (mg/L) and Z is the bed height (cm) of the column. Eq. (1) can be rewritten in the form of a straight line as presented in Eq. (2)

$$t = aZ - b \quad \text{-----------------} (2)$$

where: a = slope = $\frac{No}{Cou}$ and

b = intercept = $\frac{1}{KaCo}\ln\left(\frac{Co-1}{Cb}\right)$

The critical bed depth (Zo) is the theoretical depth of the sorbent sufficient to ensure that the outlet solute concentration does not exceed the breakthrough concentration, Cb, value (9.23mg/L in the present study) at time t=0 and solving Eq. (2) for Z yields Eq. (3)

$$Zo = \frac{u}{KaNo}\ln\left(\frac{Co-1}{Cb}\right) \text{-----} (3)$$

From the results obtained in Fig. 3, it was observed that as the bed height increases the bed service time increases. This was due to the availability of sorbent sites for sorption to occur. The plot of service time against bed height was used to calculate the values of sorption capacity of the bed No using equation (2) and bed depth service constant Ka was also calculated using equation (3). The values of No and Ka were found to be 920 mg/L and 0.0107 L/mg/min respectively. The critical bed depth Zo was found to be 0.3086 cm.

Conclusion

Mango seed bark was used to remediate crude oil polluted water in a continuous manner. Effect of bed height was investigated from 5 to 25 cm, which revealed that oil removal and service time were increased with increase in bed height at 30 min contact time and flow rate of 26.52 cm^3/min. he mango seed bark sorption capacity was found to be 190.77 mg/L at 25 cm bed height. BDST model was used to predict the relationship between bed height and service time, which is essential in column process design and experimental data gave fit to the BDST model.

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