Removal of Crude Oil from Aqueous Medium by Sorption on Sterculia setigera

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Authors’ contributions

This work is carried out in collaboration between both authors. Author BJD carried out the laboratory work under the supervision of author SAO. The first draft was written by author BJD and cross check by SAO. Both authors read and approved the final manuscript.

Article Information

DOI: 10.9734/AJACR/2020/v5i30133
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Complete Peer review History: http://www.sdiarticle4.com/review-history/55040

ABSTRACT

This study will present a novel method for crude oil remediation in water. The research was carried out to explore the possible application of Sterculia setigera as a potential biodegradable sorbent for oil cleanup from water. The crude Sterculia setigera (CSS), retted Sterculia setigera (RSS) and bleached Sterculia setigera (PFSS) were subjected to sorption studies to optimize their sorption capacity. The results revealed that the efficiency of sorbent to remove crude oil from water is related to the sorbent weight, contact time, initial oil concentration and temperature of sorption. It was found that increase in sorbent weight led to increase in sorption capacity from 3.75 - 5.12 g/g, 4.72 - 6.41 g/g, and 4.61-6.18 g/g in CSS, RSS and PFSS respectively. Oil sorption capacity increases by 21-27% when oil concentration was varied from 5-20 g. Contact time played a role only at the beginning of oil sorption study and became less important near equilibrium. Sorption time was varied from 10-70 min and the highest sorption capacity was recorded at 30 min. then it gradually reduced and became steady. The effect of temperature was investigated from 30-60°C. A decreased of 34-37% in oil sorption capacity was observed with increased in temperature. RSS exhibit lower water sorption when compared to the other sorbents. The sorbents showed good reusability after 8 cycles, with less than 50% reduction in sorption capacity and good reusability. Sterculia setigera demonstrated good potentials for utilization as natural sorbent for oil cleanup.

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Keywords: Crude oil; sorption; sorbent; Sterculis setigera; remediation.

1. INTRODUCTION

The results of crude oil activities leave most water surfaces polluted with different pollutant ranging from dissolved and dispersed oils, posing threat to aquatic life and soils [1,2]. Oily waste water contains persistent organic pollutants that are harmful to human health and the environment and can be hazardous if not properly treated [3,4].

Oil are introduced into the water as a result of extraction, production, transportation or storage process [5]. If oil is spilled on water or land, the physical and chemical properties of oil change progressively. Spilled oil gives an undesirable taste and odour to drinking water and causes severe environmental damage. Contaminated water cannot be used as a municipal water supply, in industry, or for irrigation. Oil settles on beaches and kills organisms that lie there. Cleanup are necessary after a spill for the protection of the environment and human health [6,7].

The physical environment in Nigeria especially in the Niger delta region is affected by activities of oil companies [8]. Many researches have worked on the effect of spills on the ecosystem over the years [9,10,11] and different method of oil spill cleanup has been developed which include physical/mechanical, chemical, biological [12,13]. Oil sorbent are promising materials for oil spills cleanup because they have the ability to concentrate and transform liquid oil into a semi solid or solid phase which can be from water conveniently [14]. Waste water is widely purified using sorbent from synthetic and natural materials. The synthetic sorbents have high sorption properties but they are expensive and difficult to biodegrade hence the recent interest in natural sorbent [15].

Many natural and modified biomass materials are available and have been reported as sorbent for remediating oil contaminated water, this include walnut shell, cotton, bagasse, biochars, sugarcane bagasse [16,17,18,19]. The plant Sterculia setigera, which is also called karaya gum tree, belongs to the family Sterculiaceae. It is a deciduous savanna tree up to 15m high. Bark grey purple, slash red with paler streaks, exuding white gum and watery sap. Traditionally, the stem bark has been used for treatment of diseases and the seeds can be eaten and contain edible oil [20].

Raw natural sorbents have good adsorption capacity, biodegradability and non-toxicity when compared with synthetic sorbents, but their low hydrophobicity increases water uptake and lowers oil sorption capacity. This disadvantage can be reduced through pre-treatment [21]. Different pretreatment can be applied in fibrous materials to enhance their application, including physical (Milling and grinding), Chemical (acid, alkaline, oxidizing agents and organic solvent treatment); biological and combination of physical and chemical techniques can be used. Chemical pretreatment is one of the most efficient and cost effective method [22]. When chemicals are added to clean the sorbent, it improves adsorption capacity and stability of the biosorbent and makes it effective [23].

The focus of the present study is to assess the potential application of Sterculis setigera as natural sorbent for oil removal from water. The effect of variations in sorbent weight, contact time, initial oil concentration and temperature were studied. Water sorption capacity, sorbent reusability and oil retention were also investigated.

2. METHODOLOGY

2.1 Sample Collection and Sorbent Preparation

The fibrous plant Sterculia setigera was collected from a farmland located in Girei Local Government Area, Adamawa State, Nigeria and identified by a Botanist from Modibbo Adama University of Technology, Yola. The plant part obtained was cut from the stem with a knife, the bark removed and washed with distilled water. It was spread on a clean polythene and allowed to dry in the laboratory for one week.

2.2 Extraction of Fiber Procedure

The fiber was extracted from the fibrous plant stem using chemical retting extraction process, giving fiber of different lengths and diameters. The fibrous plant (Sample) was treated with 6% NaOH solution in accordance with work done by Cai et al. [24]. 15 g of the sample was submerged in 6% NaOH solution and heated at
100°C for 30 min in a water-bath. The fiber was rinsed in cold water to free fibers strands. It was neutralized with acetic acid and washed with distilled water repeatedly until all sodium hydroxide is eliminated. Finally, the fiber was dried at room temperature for 48 h.

2.3 Bleaching of Fibers

Retted fibers were scoured in 2% NaOH solution at 100°C for 30 min. Scouring of the fiber was carried out before bleaching. Dry scoured fibers were measured and submerged in a solution of 3% H₂O₂, with sodium pyrophosphate/sodium oxalate as buffering medium at 55°C for 30 min to remove any colouring matter and white fibers was obtained.

2.4 Characterization of Crude Oil Sample

The properties of crude oil sample (COS) was characterized according to the method described by Nwabueze et al. [25]. The density, viscosity, specific gravity and API gravity of the crude oil sample was investigated.

2.4.1 Density

The density of COS sample was taken by using a specific gravity bottle. The bottle was filled with oil and weighed at room temperature (28-30°C) and the density calculated from:

$$\text{Density} = \frac{(\text{MS} - \text{Mb})}{\text{Vb}}$$ (1)

Where MS = mass of oil plus bottle
Mb = mass of bottle
Vb = volume of bottle

The method was repeated in triplicate to obtain a mean value.

2.4.2 Viscosity

The viscosity for crude oil sample was determined using viscometer. The viscometer was cleansed with a non-toxic solvent and dried. A certain amount of crude oil sample was poured into a beaker, and then transferred to the viscometer. The viscometer was inserted into the water bath at the required temperature and the viscosity was recorded in poise and converted to centistokes. This was carried out in triplicate to obtain a mean value.

2.4.3 Specific gravity

The specific gravity of crude oil was determined from the results obtained for density. The specific gravity, being a more standard measurement was obtained by multiplying the density calculated with density of water 0.998 g/cm³.

2.4.4 American Petroleum Institute (API) Gravity

The API gravity was calculated using the formula:

$$\text{API} = (141/\text{s.g}) - 131.5$$ (2)

Where s.g = specific gravity of crude oil calculated.

2.5 Characterization of Crude Fibers

The physiochemical properties of the sorbents will be investigated according to the method described by Donatus et al. [26]. All the following physiochemical properties were determined: Moisture Content, Ash Content, Volatile Content, Fixed Carbon, Density, Specific Gravity and Swell ability.

2.6 Determination of the Amount of Water Sorption

The water content of the sorbent was determined in the laboratory using the method of centrifuge technique described by Al Zubaidy et al. [27]. The sorbent was subjected to pressing to desorb the crude oil. During the pressing stage, petroleum ether was added to help extract the oil in the sorbent; the extracted liquid was collected in a centrifuge tube and placed in a water bath to break emulsion present and then, centrifuge for 20 min. The amount of water sorbed was weighed and recorded.

2.7 Test for Oil Sorption Capacity by Sorbent

Factors that affect oil adsorption were investigated, namely the effect of variation in sorbent weight, contact time, oil concentration and temperature in water/oil medium and in oil medium. Tests were carried out at room temperature. The methods describe by Onwuka et al. [28] was adopted for the sorption studies. To simulate the situation of oil spill and minimize experimental variation, the crude oil sample was held in beakers for 1 day in open air to release volatile hydrocarbon contents. The crude, retted and pure fibers were subjected to sorption studies to optimize the sorption properties.
To 100 ml of distilled water in a 250-ml beaker, 10 g of crude oil was being added. A portion 0.10 g of the sorbent was added into the mixture in the beaker and left unperturbed for 30 min at constant temperature of 30°C. After 30 min, the sorbent was removed using a spatula and placed on sieving net and left to drain by hanging the net over a beaker for 10 min. The drained sample was weighed and recorded. This was repeated at different weights of 0.2, 0.3, 0.4, 0.5, 0.6 and 0.7 g and results recorded. This experiment was also conducted at different times of 10, 20, 30, 40, 50, 60 and 70 min at constant sorbent weight (0.7 g), oil concentration (10 g) and temperature (30°C) and results were recorded. The effect of Initial concentrations of crude oil was also studied from 5, 7.5, 10, 12.5, 15, 17.5 and 20 g/100 ml of water at constant sorbent weight (0.7 g) and time (30 min), temperature (30°C) and results recorded. The effect of temperature on sorption was also investigated at different temperature (30, 35, 40, 45, 50, 55 and 60°C) at constant weight of sorbent (0.7 g), time (30 min) and temperature (30°C). The sorption capacity of the sorbent samples was calculated using the expression:

\[ \text{Oil Sorption Capacity} = \frac{\text{New weight} - \text{Gain}}{\text{Original weight}} \]

and recorded as gram per gram of sorbent. The procedure was carried out in triplicates and the mean of the results reported.

2.8 Sorbent Reusability

The sorbent sample was used eight times and after each time the sorbent was pressed to squeeze the oil content from the sorbent and ready for further use. The sorption performance was recorded in g/g. Reusability of the sorbent sample was studied in oil medium. 8 cycles of sorption processes were performed. After each cycle, the sorbent was squeezed and reweighed. The difference between the weight of the wet material after drainage and the initial weight of the material gives its sorption ability.

2.9 Oil Retention

To determine the oil retention, a known weight of sorbent was placed in 20 ml of oil for 30 min. The sorbent was removed and vertically hung, where upon the adsorbed oil began to drip from the sorbent, the weight of the material was measured after 10, 20, 30, 40, 50, 60 and 70 min of draining. The amount of oil retained was determined as the difference between the weight of the wet material after drainage and the initial weight of the material [29].

3. RESULTS AND DISCUSSION

3.1 Effect of Sorbent Weight

As shown in Fig. 1, the sorption capacity efficiency was dependent on the weight of the sorbent in the entire sample studied. In CSS the sorption capacity increased from 3.75-5.12 g/g, RSS from 4.72-6.41 g/g and PFSS 4.61-6.18 g/g. These indicate that increasing the amount of sorbent led to increase in oil sorption capacity. With increased in sorbent quantity, the sorbent surface area and number of adsorption sites available for sorption increases as reported by many studies [30,31,32]. This phenomenon here is associated with an increase in available binding sites for sorption at higher sorbent dosage [33]. Availability of greater number of active sites on the surface of the sorbent led to a higher interaction between the oil molecules and the sorbent [34]. Sorption capacity in the RSS and PFSS samples were higher because pretreatment makes the spatial structure looser, increased its specific surface area thus led to formation of more number of active sites and storage space which increase the sorption capacity compared to the raw CSS sample [14].

Table 1. Physicochemical properties of crude oil sample (COS)

<table>
<thead>
<tr>
<th>Properties</th>
<th>Values – Mean and Standard Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm³)</td>
<td>0.8651 ± 0.01</td>
</tr>
<tr>
<td>Specific gravity (g/cm³)</td>
<td>0.8634 ± 0.01</td>
</tr>
<tr>
<td>API gravity (30°C)</td>
<td>32.4 ± 0.02</td>
</tr>
<tr>
<td>Viscosity, 30°C (Cst)</td>
<td>5.04 ± 0.02</td>
</tr>
</tbody>
</table>

3.2 Effect of Contact Time

The effects of contact time in relation to sorption capacity of sorbents were studied at different time 10, 20, 30, 40, 50, 60, 70 min with other experimental variables kept constant. Fig. 2 shows the effect of contact time on sorption capacity. The plot shows that sorption capacity increases with the contact time for the first 30 min. After which a slow oil removal from the solution was observed. The sorption process was rapid at the initial stages of the contact period and thereafter it became slower towards equilibrium. The sorption sites become less
available as the contact time increased, resulting in a slow sorption phase. This phenomenon may be attributed to the fact that a large number of vacant surface sites and microscopic voids were available for sorption during the initial stages and after a lapse of time, the remaining vacant surface sites were difficult to be occupied due to repulsive forces between the solute molecules on the solid and bulk phases [35,36]. During the slow sorption phase, the breakage of oil droplets increased the interfacial area available for sorption [37]. With increasing duration of reaction, many more adsorption sites and pores were formed, so the oil adsorption was enhanced. However, as time goes by; the sorbed oil residue starts to clog the pores near the outer surface so oil residue can no longer diffuse to the active sites deep within the interior surface hence reducing the sorption capacity. Contact time plays a role at the beginning of sorption but its significant decreases near equilibrium [38].

3.3 Effect of Initial Oil Concentration

The initial concentration study is very important because the initial concentration of the oil residue in solute can strongly affect the sorption kinetics and more specifically, the mechanism that controls the overall kinetic coefficient. Fig. 3 showed the sorption capacity of the sorbent at different initial oil concentrations ranging from 5, 7.5, 10, 12.5, 15, 17.5, 20 g. The results indicate that oil sorption capacity increased with increased in initial oil concentration until it reaches equilibrium. The sorption capacity of all the sorbent increased with 19-23%. The sorption capacity at initial oil concentration of 5 g was relatively low, as the oil molecules available for attachment on the sorbent surfaces were limited [39]. An increase in oil concentration enhanced oil sorption capacity because increasing the initial oil concentration led to increases in the number of collisions between oil and the adsorbent, which enhances the adsorption process [40]. At higher initial concentrations, the surface of the sorbent enhances oil residue diffusion through the film surrounding the materials and into the porous network of the sorbent [41]. It is also evident that the sorption capacities of retted Sterculia setigera (RSS) and bleached Sterculia setigera (PFSS) were higher than that of crude Sterculia setigera (CSS). The raw fiber (CSS) easily forms dispersions in the water making sorption of the oil difficult. The high sorption capacity of the pretreated fiber is due to the alkalization which makes the sorbent surface rougher and allows better wetting.

![Fig. 1. Effect of sorbent weight on oil sorption capacity](image-url)
Fig. 2. Effect of Contact time on Sorption Capacity

Fig. 3. Effect of initial oil concentration on sorption capacity

Table 2. Physicochemical properties of sorbent

<table>
<thead>
<tr>
<th>Properties</th>
<th>CSS</th>
<th>RSS</th>
<th>PFSS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture Contents (%)</td>
<td>11.77± 0.01</td>
<td>8.90± 0.02</td>
<td>4.98± 0.02</td>
</tr>
<tr>
<td>Ash Content (%)</td>
<td>7.02 ± 0.01</td>
<td>8.03± 0.01</td>
<td>5.05± 0.03</td>
</tr>
<tr>
<td>Volatile Content (%)</td>
<td>63.06± 0.02</td>
<td>76.46± 0.02</td>
<td>51.38± 0.02</td>
</tr>
<tr>
<td>Fixed Carbon Content</td>
<td>18.15± 0.01</td>
<td>6.61± 0.01</td>
<td>38.59± 0.01</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>0.985± 0.02</td>
<td>0.990± 0.01</td>
<td>0.9901± 0.01</td>
</tr>
<tr>
<td>Swell ability (%)</td>
<td>172.04± 0.01</td>
<td>146.34± 0.03</td>
<td>273.20± 0.02</td>
</tr>
</tbody>
</table>
3.4 Effect of Temperature

Temperature variation exists in different areas and seasons, so it needs to be studied in oil spill treatment. The effect of temperature on the sorption capacity of the sorbent is shown in Fig. 4. The sorption capacity decreased as the temperature increased from 30-60°C. Moderate viscosity facilitates the penetration of oil into the pores and is also good enough for retention of oil in the structure of the sorbents [42]. Temperature increased result in decreased in the sorption capacity by about 31%, 23% and 29% in CSS, RSS and PFSS respectively. It is clear that adsorption decreases gradually with temperature increase at above 30°C. This can be explained based on the change in oil viscosity with temperature, resulting in low adherence of oil to the pore walls giving rise to more oil drain during draining step. In general oil removal decrease with increase in temperature which may be due to reduction in viscosity then small amount of oil could be soluble in water. These results are typical of sorption processes in which higher kinetic energy of the oil at high temperature makes retention becomes difficult due to decreased in adsorptive forces between the oil molecules and the active sites of solid phases [43]. RSS and PFSS exhibit better sorption capacity and retained more oil than the raw fiber (CSS) because they possessed more energy required for adsorption [44].

3.5 Water Sorption Capacity

Fig. 5 showed the water sorption capacity of the sorbent in percentage at different sorbent weight. Water uptake and diffusion coefficient values increased as fiber content increased. High cellulose content causes the sorbent to take in water that penetrates through the interface. Water molecules travel freely through the micro voids and pores [45]. When the sorbent weight increased, the interface between oil and water nearly disappeared allowing the sorbent to absorb high values of water [46]. The water sorption capacity increased with increase in sorbent weight, with RSS having a lower value of 198%, which may be as a result of reduction in polar hydroxyl group in the fiber by replacement of OH group on the surface of the fibers by alkalization [47].

3.6 Effect of Reusability

Reusability is one of the major factors for selection of sorbent materials. Recyclability help in reducing cost, if the sorbent can be recycle and reused. The main criterion which can be used for judging reusability of a sorbent is the number of cycles it can tolerate without becoming unusable due to tearing, crushing, or other general deterioration [48]. The relationship between the number of times the sorbent was reused and its sorption capacity are shown in

Fig. 4. Effect of Temperature on oil sorption capacity
Fig. 6. The sorption capacity of all the sorbent decreased with repetitive use with 32%, 24% and 25% for CSS, RSS and PFSS respectively. This decline in oil sorption capacity can be as a result of some oil retained. The retained oil occupies some of the sorption sites taking some portion of the storage space [49]. Decrease in sorption capacity with reused can also be due to irreversible deformation. The continuous deformation may be due to tearing during squeezing, in addition, there was no apparent change showed in the bulk volume of the sorbent but some local deformations inside the sorbent can be formed once oil drained out. There might be reorientation in the sorbent particle arrangement, which can change the inner porous structure then caused reduction of subsequent oil sorption. However, all the sorbent exhibit good reusability since all maintains more than 60% of their sorption capacity.
3.7 Oil Retention

Oil retention is an important parameter because it prevents premature drainage of adsorbate. The oil retention capacity with time for CSS, RSS and PFSS sorbents were studied. The quantities of adsorbed oil as remains in sorbent were shown in Fig. 7. Though some of the oil was drained after some time (10-70 min, the draining of the oil in the extra lumen occurs when the capillary pressure is insufficient to hold the weight of the oil [17]. The inherent strength of the sorbent is susceptible to deformations easily due to moving and lifting of the material. Deformations results in pressures upon the internal pores forcing the adsorbed oil to be squeezed out. This phenomenon is observed with sorbent that are of organic materials. According to Darcy s law, smaller pore reduces the speed of oil drainage and may improve oil retention. The oil bridges formed among the bundles of the pretreated samples would have been difficult to destabilized [50], that is why the pretreated samples RSS and PFSS has higher retention value of RSS 68% and PFSS 76% compared to CSS with 64%.

4. CONCLUSION

The sorption capacity of *sterculia setigera* was investigated. It was found out that variation in sorbent weight, contact time, initial oil concentration and temperature all affect the sorption capacity of the sorbent. The experiments above showed that increased in sorbent weight and initial concentration led to better sorption capacity. Contact time of 30 min give higher sorption capacity and temperature of less than 35°C chosen for the sorption studies. CSS, RSS and PFSS sorbent exhibited good reusability and oil retention.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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