Acta Scientific Agriculture (ISSN: 2581-365X)

Volume 2 Issue 6 June 2018

Research Article

New Approach for Adsorptive Removal of Oil in Wastewater using Textile Fibers as Alternative Adsorbent

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Received: April 06, 2018; **Published**: May 21, 2018

Abstract

Nowadays, removing of petroleum and nonpetroleum spills using low cost adsorbent materials is one of the important challenge all over the world. In this work, recycled textile fiber (TF) obtained from used tires has been used as a low cost static adsorbent for the treatment of water pollution by petroleum and non-petroleum oil. The effect of important parameters which are affecting on the removal efficiency these pollution from waste water including fiber mass, removal time, and temperature were investigated. The maximum yield removal of water pollutions by TF at room temperature were found to be 95.2% for fatty oil and 91.0% for crude oil. While, the maximum capacity and removal percentage yield removal of water pollutions by TF at room temperature were found to be 4.40 g/g and 95.2% for cooking oil and 4.20 g/g and 91.0% for crude oil respectively. The evaluation of adsorbent efficiency for only oil uptake which named "dry system" was also studied. The maximum adsorption capacity was found to be about 17 g/g for used cooking oil, and 14.4 g/g for crude oil which reached within 24 hr. Influence of compressed textile fiber from waste tires for the removal efficiency of oil from water-oil system was also studied. The compressed TF showed lower adsorption capacity than normal fibers (uncompressed) which found to be about 1.04 g/g. But the obtained results showed that the compressed TF can be successfully used for the storage and transport of recovered oil and the adsorbed oil not leaches out at room temperature for long time and it's easier to handle.

Keywords: Adsorption; Crude Oil; Cooking Oil; Oil Spill; Static System; Textile Fiber; Water Pollution

Introduction

Today, one of the important tasks and challenges worldwide is oil spill pollution, because oil is considered one of the most dangerous pollutants, due to its toxicity to many life forms. Oil spill also has a great negative impact on the ecosystem by putting the marine lives at high risk [1]. Industrial development and human activities have increased oily wastewater discharge into the natural water streams. There are many sources of oily wastewater, including crude oil spill, power plant, stations, petrochemical plant, and household waste such as used cooking oil and lubricant from vehicle spills. Transportation of crude oil also one more source of oil spills on the surface water [2-4].

Besides, corrosion of oil pipes, poor maintenance of infrastructure, spills or leaks during processing at refineries, and accidental discharge from tankers or vessels and less due to sabotage, vandalisation of the oil infrastructure, and theft of oil [5]. It was reported that up to 20 million tons of crude oil is annually spilled on the surface of the ocean [6]. Reports in Niger indicate that the Niger Delta area of Nigeria, about 7 to 10 million barrels of oil have been spilled both onshore and offshore in recent year [5]. Regarding to household waste oil [7], it was reported that many consumers and small-scale reuse their cooking oil many times "repeatedly" which is simply called waste oil. This waste has a great impact on the environment and human health [3,4,7]. However, environmentally very important to remove oil from oily wastewater before discharging it into the environment due to its toxicity. Synthetic polymeric materials such as polypropylene (PP) and polyurethane foam (PUF), which are the most widely used commercial adsorbents in oil spill cleanup due to good mechanical properties. For example, "open-cell polyurethane foams capable of absorbing about 100 times their own weight of oil from oil-water mixtures were previously developed through chemical modification of the matrix, and by specific trimming of the foam structure" [8,9]. Thus, cleanup of petroleum and nonpetroleum spills from the water surface is an important task. Different methods has been used for the removal of oil from the water surface, for example, thermal, biological, mechanical and physicochemical (using coagulants and adsorbent materials) techniques [6-8]. Currently, removing of petroleum spills by adsorbent materials is the most safety and effective methods [6].

In recent year, there are number of works have been conducted on using industrial solid wastes as an adsorbent materials in the removal of oil from water-oil system such as, crumb rubber, polyurethane foam, polypropylene fiber, wastes of wool and cotton, and others [4,8-12]. Therefore, researches and scientists all over the world are looking for different alternatives for conventional expensive adsorbents. For instance, Chitsan and his coworkers [11] have investigated crumb rubber (CR) powder, polypropylene (PP) fibers, CR-PP composite as adsorbents for oily wastewater treatment. According to the data obtained in their work the following points were reported 1) crumb rubber has an average oil adsorption of 2.8g/g, with good elasticity to maintain its oil sorption capability after 100 reused. 2) Results on PP fibers showed that its oil adsorption average was 48.8 g/g. But this value of oil adsorption capability could rapidly decrease to about 50% after 8 reuses. 3) When CR powder is mixed with PP fibers to create a composite material, the oil adsorbent capacity was increased and reported as 85.5 g/g. Besides, the authors observed that CR powder and composite material of PP fiber and CR powder can be used at least 100 times without a reduction in oil adsorption capability [11].

Textile fiber is one of the materials that have been used for creating a new tires. The main compositions of recycled textile fiber derived from scrap tires in weight percent are 77.6% polyethylene-terephthalate (PET), 18.7% polyamide (PA), and 3.7% polypropylene (pp) [13]. Unfortunately, recycled textile fiber presented remain unused material and lack technical applications as far as we know. Therefore, the current research study is intended to investigate the utilization of TF as a solution for sustainable development by using this material as an adsorbent for oily wastewater treatment. This would subsequently raise the confidence to reduce the landfill disposal of such waste and thus afford a valuable solution to this global environmental pollution. However, the utilization of TF obtained from scrap tires without chemical treatment may possibly be named novelty as no previous works are applied in this direction to the best of our knowledge. Another benefit of the study is that, TF has a possibility of collection and complete removal of the oil from the oil spill site, particularly from the surface water. Additional advantages of TF is that inexpensive material, and can be used with or without a small amount of processing for oil spill cleanup.

Experimental Section

Materials

In this study textile fibers were obtained from the recycling process of used tires and supplied by Orzel Company from Poland. Length of fiber was about (2.5 - 5 mm) and used without any treatment as shown later in section (3.2.3); (Figure 7). Crude oil was delivered by Lotos Company from Poland. Used corn oil was collected from the standard fast food restaurant. Both oils were used without any purification or treatment. The recycled fibers were washed completely with tap water to remove impurities, subsequently was washed few times with distilled water, and dried at ambient conditions for few days and the other in an oven until constant weight was recorded.

Removal experiments

A removal process using textile fibers technique was carried out using static process. However, static experiments have been done regarding to the previous study [4] as follows: A specific amount of crude oil 5 ml (4.62g) was firstly poured into glass beakers with a capacity 150 ml, then 50 ml of distilled water was added to each beaker and kept for few minutes to form an oil layer. After that, 0.1g of recycled fibers was added into each of the beaker. Fibers was carefully removed from the sample at distinct time intervals of 10, 20, 30, and 40 min as presented in table 1, and dried at room temperature for 72 hr. The samples of fibers were also dried using an oven at temperature of 70°C until constant weight. The same procedure was repeated with waste cooking oil. Natural pH solution (7) was used during the whole process.

Independent variable	Unit	Sample symbol	Levels -1	0	1	2
Removal time	min	A	10	20	30	40
Adsorbent doses	g	B	0.1	0.3	0.5	1.0
Removal temperature	°C	C	5.0	25	35	45
Initial oil concentration in water	g/ml	D	4.62g (5 ml)			
pH	--	E	Natural, 7			

Table 1: Experimental range and levels of independent process variables.

In the case of "dry system", 100 ml of crude oil and used cooking oil was poured into beakers with a capacity of 250 ml, after that 1.0g of fiber was put into a beaker and kept without agitation for 24 hr. Every 6 hr, the fiber was removed with a net, which was hung over the beakers for 1 minute to allow the oil that was not adsorbed to separate as shown in figure 1. A similar procedure was used in the works [4,14]. The effect of removal time, temperature, and amount of adsorbent on effectiveness of oil adsorption for all systems were studied. The percentage removal of oil and adsorption capacity of fibers were calculated using Equations 1 and 2, respectively [4]. Cold system "fridge", was used for the effect of low temperature (5°C), while water bath system was applied for the effect of high temperature on the rate of adsorptive removal of oil by textile fiber.

$R = (w_1/w_2) \times 100\%$ ....(1)

where R is the oil removal (wt%), w_2 is the initial weight of oil in the system (g), and w_1 is the weight of oil adsorbed by the fiber.

$Q = (C_1 - C_2) / M$ ....(2)

where Q is the fiber capacity (g/g); C_1 and C_2 are the initial and final amount of oil (g).

Equation 2 can be given as:

$Q/M = (C_1 - C_2)$ .... (3)

Figure 1: Static system used for adsorption of oil: layer of oil (I), layer oil with textile fiber (II) and treated water (III).

Heat compression technique of textile fiber

From the storage and transport point of view, for concentration of fibers a method using compression molding at temperature of 180°C for 15 minutes has been applied to made a layer with more concentrated fibers for adsorbing oil from water as shown in figure 2 and 3.

Figure 2: Compression molding process of TF derived from scrap tires (A), and layer oil with TF (B).

Figure 3: TF after the concentration in compression molding.

Results and Discussion

Characterization of fiber

Elemental analysis of textile fiber

The elemental analysis of textile fiber was done using FLASH 2000 CHNO Analyzed, USA. The amount of carbon (C), hydrogen (H), nitrogen (N), and oxygen (O) were found as follows: 64.6 wt%, 6.6 wt%, 4.4 wt%, and 17.0 wt%. High content of carbon was observed for textile fiber. Another, elements were conducted by Wavelength Dispersive X-ray Fluorescence (WDXRF) Spectrometer and the values are presented in table 2.

Textile fiber derived from used tires			
Element	**Amount, wt%**	**Element**	**Amount, wt%**
Zn	0.76	Cl	0.04
Fe	0.62	Mg	0.03
S	0.50	Br	0.01
Si	0.27	Ti	0.0089
Ca	0.18	P	0.0078
Al	0.07	Mn	0.0041
Cu	0.040	Co	0.0035

Table 2: Almost elements found as constituents of textile fiber derived from scrap tires.

FTIR- analysis

Tensor 27 Bruker model of Fourier transform infrared (FTIR-ATR) Spectroscopy, was used in this study to obtain the IR spectra of the fiber material. Measurements were done using the ATR technique (Attenuated Total Reflection) applying the zinc selenide and diamond base optics. Samples were analyzed in spectral range of 550 - 4000 cm^{-1}. Figure 4 shows the spectra of TF involved in this study as oil adsorbent. Absorption band at 2848 and 2916 cm-1 are related to groups -CH2- and -CH3- in aromatics polymer present in TF. Bands at 1450, 1500, and 1637 cm^{-1} may be associated with C=C of aromatics in TF. A band at 1712 cm^{-1} is associated with C=O of carbonyl group. Finally, broad peaks at 3297 cm^{-1} is attributed to N-H functional groups in the TF. According to the chemical nature of textile fiber, band at 1712.87 are associated to (C=O), related to mixture of both polyester (PET) and polyamide because are the main materials which were used to produce the textile cord for tires. It was reported that the important functional groups responsible for oil uptake are O-H, N-H, C-O and C=O groups [15,16], which are presented in textile fibers used in this study.

Figure 4: The spectra of textile fiber from used tires involved in this study as oil adsorbent.

Scanning electron microscope (SEM) of textile fiber

It is well known that natural and synthetic polymeric materials such as cotton, polyester, nylon, polypropylene, among others are involved in the composition of most of the fibrous waste [17]. The use of Scanning Electron Microscope (a Zeiss EVO 40, SEM) showed that textile fiber derived from scrap tires was not a pure textile material; it was a mixture of rubber and textile (Figure 5A). It was believed that TF was still containing a small amount of rubber material and a small portion of steel, thus the characteristics were different from the synthetic textile material, which was usually free of any impurities. As a result, the next step has implied the removal of these CR within TF by simply stroking the TF mechanically, providing almost a clean TF as presented in figure 5B.

Figure 5: SEM of TF with CR (A) and TF after removal of CR particles (B).

Affecting factors on the adsorption system

The effect of fiber mass

The influence of fiber mass on the adsorptive removal of oil was studied by adding 0.1g of fiber in 50 ml distilled water containing 5 ml (4.62g) of crude oil at natural pH 7, room temperature for 30 minutes without any agitation. The step was then repeated using different amounts of the fiber 0.3, 0.5, and 1.0 g, respectively under the same conditions. The same procedure was applied in the case of waste cooking oil. Finally, the optimum amount of fiber for both adsorption process (using used cooking oil and crude oil) was selected for use in the second step. Table 3 shows the effect of adsorbent mass on the percentage removal of oil, as well as the adsorption capacity of fibers. From the table, it can be concluded that the trend revealed a progressive increases in the removal of both oil as adsorbent mass increased from 0.1g to 1.0g. The percentage of oil removal increases from 64.5 to 84.9% for crude oil and from 69.3 to 93.3% for waste cooking oil respectively, and then the value of percentage removal were slowly increased until the equilibrium was reached at 1.0g of adsorbent mass, with percentage removal of 91 and 95.2% for crude oil and cooking oil respectively. However, a higher dose of adsorbent permanently binds the oil, which facilitates the storage and transport of waste material [4,18]. So, 1.0g of textile fiber was selected as optimization of adsorbent mass. For the adsorbent capacity which identified by using Equation 2 revealed that the amount of oil (g) adsorbed per unit mass of adsorbent (q) increased from 2.98 g/g to 4.2 g/g for crude oil and from 3.2g/g to 4.4 g/g for cooking oil with the increase in adsorbent dose from 0.1g to 1.0g as shown in table 3.

Adsorbent dose, g	Removal percentage, %		Adsorbent capacity, g/g	
	Crude oil	Waste cooking oil	Crude oil	Waste cooking oil
0.1	64.5	69.3	2.98	3.2
0.3	75.8	86.6	3.5	4.0
0.5	84.9	93.3	3.9	4.3
1.0	91.0	95.2	4.2	4.4

Table 3: Relationships between adsorbent dose and the removal percentage and adsorption capacity for crude oil and waste cooking oil by textile fiber.

As the amount of adsorbent (textile fiber) increases, this definitely enhances the increase in the surface area, which in turn would result in a higher oil molecules removal efficiency in the system. Besides, the increase in the amount of fibers leads to a decrease in oil solution concentration in the system at the same time [19]. A similar phenomenon was observed by the works [4,18,20]. For instance. Sulyman., *et al.* [4] reported that the percentage removal of used cooking oil by wool fibers was increased from 69% to 98% when the wool fiber increased from 0.2g to 1.0g.

The effect of removal time

The influence of removal time was studied by adding 1.0g of textile cord fiber which has been selected as optimum fiber mass into beakers containing 50 ml of 4.62g crude oil at pH 7, room temperature, for time periods of 10, 20, 30, and 40 minutes. The same procedure was applied in the case of waste cooking oil. Finally, the optimum removal time was selected for the next step. The removal percentage and capacity of adsorbent obtained during the experiments were presented in table 4. From the table, it can be seen that the percentage removal rapidly increased with the increase of sorption time at the first stage (20 minutes for waste cooking oil and 30 minutes for crude oil) and reached equilibrium in 40 minutes with oil removal efficiency of 92.4% for crude oil and 95% for cooking oil. Rapid increase in the retention of used cooking oil and crude oil in the first 20 minutes, as well as the rate of removal was higher because of the large amount of oil molecules attached to the fibers within the first period time of adsorption [19] while, the slowly and finally reached equilibrium due to the saturation of the surfaces of textile fibers [15]. The adsorption capacity was also reached equilibrium between 35 minutes and 40 minutes and found to be 4.27 g/g 4.39 g/g for crude oil and cooking oil respectively at removal time of 40minutes. The equilibrium time for both oil by textile fiber was between 35 minutes and 40 minutes at the operating conditions of room temperature, pH 7, initial oil concentration of 4.62g/50ml. A similar trend was observed by literature [4,6]. In the case of dry system, the maximum removal capacity of textile fiber for crude oil and used cooking oil was found to be14.4 g/g and 17.0 g/g respectively which reached within 24 hr. This behavior is agreement with related study [4] for the removal of cooking oil by wool fiber at equilibrium time of 30 minutes and [15] for the adsorptive of vegetable oil and diesel fuel by coconut husk with equilibrium time at about 60 minutes.

Removal time, minutes	Removal percentage, %		Adsorbent capacity, g/g	
	Crude oil	Waste cooking oil	Crude oil	Waste cooking oil
10	71.4	90.9	3.30	4.20
20	80.1	92.4	3.71	4.27
30	90.1	94.2	4.20	4.35
40	92.4	95.0	4.27	4.39

Table 4: Relationships between contact time and the removal percentage and adsorption capacity for crude oil and waste cooking oil by textile fiber.

The effect of temperature

Temperature is a very important parameter for the study of oily wastewater [1]. Since there are different temperature of effluent oily wastewater particularly that coming from the industrial wastewater. From the viscosity point of view, crude oil, as well as used cooking oil are very sensitive for temperature. However, the effect of temperature on the removal rate was investigated regarding to the above steps and obtained result are presented in table 5 and figure 6. In general, the percentage of oil removal decrease with an increase in temperature, which may be due to the reduce in viscosity of oil and then a small proportion of oil could be soluble in water. However, the adsorption capacity has also been decreased as the temperature increases. For instance, there were decreases from 4.49 g/g and 4.57 g/g at temperature of 5°C to 3.3 g/g and 3.5 g/g at temperature of 50°C for crude oil and cooking oil respectively. Wile, removal percentage was decreased from 97.2% and 98.9% at temperature of 5°C to 71.4% and 75.8% at temperature of 50°C for crude oil and cooking oil respectively. At a low temperature, the solubility of oil in water is relatively lowered [1]. However, the percentage removal, as well as adsorptive capacity were higher at low temperature. Similar results have been observed for adsorptive removal of vegetable oil and diesel fuel by coconut husk [15]. From the above details it can be conclude that these results are typical of sorption processes, in which the higher kinetic energy of the oil at higher temperature makes retention or adsorption of oil by the textile fiber more difficult. This is in agreement with similar work done by Aisien and Aisien (2012) [5], when the authors have investigated the effect of temperature which was one of the affecting parameters that they studied using activated recycled rubber from used tires in oil spill cleanup. On the other hand, decreased in adsorption capacity with increasing temperature of the liquid media can also be explained as decreasing in the adsorptive forces between the oil molecules and the active sites of solid phase (fiber). Raw TF derived from scrap tires involved in this study as shown in figure 7A, while TF loaded oil after tried at 60oC for few hours to remove any absorbed water has been shown in figure 7B.

Temperature, °C	Removal percentage, %		Adsorbent capacity, g/g	
	Crude oil	Waste cooking oil	Crude oil	Waste cooking oil
5	97.2	98.9	4.49	4.57
25	92.0	95.0	4.2	4.4
35	80.1	84.4	3.7	3.9
50	71.4	75.8	3.3	3.5

Table 5: Relationships between temperature and the removal percentage and adsorption capacity for crude oil and waste cooking oil by textile fiber.

Figure 6: Relationships between temperature and the adsorption capacity (A) and removal percentage (B) for crude oil and waste cooking oil by textile fibers

Figure 7: Recycled TF before adsorption of oil (A), and after adsorption of oil from oil-water system (B).

Results of recovered oil onto textile fiber after storage

In this study, storage of recovered oil by TF has been studies as follows: 1.0g of TF loaded oil was put on glass, then dried and stored for one year into save place at room temperature. The amount of adsorbed oil was 4.4g oil/g fiber, after one year the same material loaded oil was firstly dried at 60oC for few hours to remove any humidity, and then carefully weighted using electronic weight balance to know the amount of adsorbed oil onto the adsorbent. According to the data obtained, there is a very small reduction in weight of oil, which estimated by (about 6%), this reduction may be due to the leached out of oil during the long time of storage. On the other hand, the compressed TF using the compression molding, showed lower sorption capacity about 1.04 g/g. But at the same time indicates that such material can be successfully used for the storage and transport of recovered oil without or with a very small leeches out of the oil, as well as easy to handle.

Conclusion

The present study concerns the use of recycled textile fiber obtained from scrap tires, for the removal of Crude oil and waste cooking oil from the water. In this study, experiments were carried out under small scale conditions using the static system. According to the obtained data the following conclusion can be drawn:

1. Recycling of textile fiber as adsorbent material for oily wastewater treatment is possible and showed a good removal efficiency of oil from oil-water system.

2. The removal of both oils from waste water was found dependent on fiber mass, removal time, and temperature of media.

3. Data obtained in this study showed that recycled textile fibers can adsorb 4.2 - 4.4 times its weight from oily wastewater at room temperature. While, 14 - 17 times its weight in the case of dry system.
4. According to the experimental work, TF has a possibility of collection and complete removal of oil from the environment.
5. Adsorption process for both oil at low temperature showed better removal efficiency than high temperature, which is reported as 97.2% and 98.9% for crude oil and cooking oil respectively at 5°C.
6. From the storage and transport point of view, TF showed a good example for recovery, storage, and transport of petroleum and nonpetroleum oil.
7. Initial concentration of oil, as well as pH were fixed constant during the whole process. Further work for such parameters are necessary for support this study. The use of continue process using packed bed column for the same purpose is also recommended and will be the future work.

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