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A Novel Method for Adsorption using Human Hair as a Natural Oil Spill Sorbent

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Abstract— Oil exploration has enormous economic benefits; however, these benefits are associated with environmental issues arising from oil spills. Numerous methods exist for cleaning up oil spills along with their after-effect as well as huge financial burden. Hence, the shift to methods that are environmentally friendly and cost effective is imperative. This paper investigates the efficiency and mechanics of using human hair to clean up crude oil contaminated water using various parameters such as contact time, recovery and reuse, adsorbent dosage, temperature, modification. Human hair has proven to be an efficient material in removing oil from water with a maximum adsorption capacity of 7470mg/g for crude oil as well as its recovery and reusability. Investigation reveals that African hair came first, followed by Asian hair and finally, European hair with an adsorption capacity of 7470, 6176 and 5246mg/g for crude oil respectively. The result obtained for the kinetics of adsorption revealed a good fit in pseudo-second order model, indicating that the mode of adsorption is exothermic which is controlled by a chemisorption process. The isotherm studies also revealed that the experimental data is better described with Freundlich isotherm model. A comparative analysis conducted using the maximum adsorption capacity of different adsorbents revealed that human hair performed better than organoclay, rice husks, reed bed canary grass, treated sludge, modified oil palm leaves, peat moss and activated carbon, but less than exfoliated graphytes, kapot and recycled wool based non-woven material. Hence, human hair can be modified into boom to clean-up oil spills. This is a promising area that researchers need to focus more on inorder to explore the huge benefits it presents.

Key words: Adsorption capacity, adsorption isotherm, clean-up, human hair, oil spill, sorption kinetics, sorbent

Introduction

Oil exploration, production and transportation are critical to the development of the economy of many nations across the world owing to the enormous benefits derived from it. Due to the essential nature of oil, it is very difficult for an individual or a country to complete the day's activities without directly or indirectly depending on petroleum products. However, these processes (petroleum exploration, production and transportation) over the years have impacted negatively on the environment as a legacy, ranging from oil spillages, release of dangerous substances into the air, water and land, species extinction etc [1]. Over the years there has been cases of oil spills disaster around the world giving rise to environmental degradation [2], species extinction to mention a few. Notably among them are: Torrey Canyon oil spill disaster 1969 [3], Sea Star – Gulf of Oman 1972 [4], Atlantic Express – West India 1979 [5], Exxon Valdez in 1989 [6], Dalian spill into Yellow Sea 2010 [2], Gulf of Mexico 2010 [7]. The consequence of oil spills has tremendous impact on the ecosystem e.g the Exxon Valdez resulted in the death of about 30,000 sea bird [8], 2000 sea otters, 250,000 seabirds, 302 harbor seals [9].The disaster and other incidents contributed to the reduction of pigeon guillemots from 15000 in the 70s, to 3000 in the 90s [10]. It also lead to the increase in the concentration of THC and TPAH in mussels to 62,000 ug/g wet wt and 8 ug/g dry wet respectively as against the background concentration of less than 60 ug/g THC and 0.5 ug/g TPAH [11]. In the case of the Gulf of Mexico oil spill, the ecological damage done to aquatic organisms are incalculable; however, the disaster affected about 20% of the national wetlands in the River Delta of Mississippi, which provides habitat for resting sea birds as well as resting migratory birds [7]. The oil spill covered about 88,000 square miles and also made its way to the beaches and estuaries thereby causing a great deal of damage to tourism, fishing industries, many marine animals and bird species inhabiting areas within and around the Gulf of Mexico [7].

The inevitable nature of oil spills has necessitated the need to seek for ways of mitigating its environmental impacts. There are several methods employed to clean up oil spills in water such as direct burning, use of dispersants, mechanical skimmers, use of booms and sorbents [1], [12]. During the famous Gulf of Mexico oil spill in 2010 about 700 km of boom were deployed to clean up the spill, the application of skimmers recovered about 27 million gallons of oily water and more than 1.5 million gallons of dispersants were used as at the first of July, 2010 [13]. The use of chemicals and mechanical recovery equipment may be time consuming, increase the cost of clean-up and may also require a lot of personnel as well as equipment, hence, the need to explore other environmentally friendly and low cost alternatives [14]. There are materials with low sorption capacity, non-biodegradable and usually expensive, but possess high hydrophobic and oleophilic properties that are available commercially which are made from polyeruthene, polyethylene and many cross-linked polymeric sorbents [15], [16]. The use of natural absorbents for the removal of oil spills has attracted attention and is considered among some of the most attractive options as a result of their effectiveness, reusability, low cost, environmentally friendly as well high sorption capacity [1]. There are available literatures showing that natural biosorbents have been used for oil spill removal in water. Notable among others are activated carbon [17], coconut husk, feathers [18], oil palm fibre [19], garlic peel [20], barley straw, sawdust, wool fibre, rice husk, cotton grass, sugar cane bagasse, walnut shell [21], [22], [23], [24], [25]. The idea of using human hair to clean up oil spill was initiated by Phil McCory, an Alabama hair stylist in 1989 during the Exxon Valdez disaster [26]. However, not much has been done in the area of exploring the efficiency of using human hair to clean up oil contaminated water [27]. Hence, having identified the existing research gap, it becomes imperative to investigate the capacity of human hair in adsorbing oil in water as well as the mechanics behind the process, which is the cardinal point of this research.

Human hair (50-100 um in diameter) is a natural biosorbents consisting of dead cells made up of the cuticle, water, lipids, trace elements and 65-95% proteins, mainly polymers of amino acids such as keratin and cysteine, medulla and cortex. The cuticle is highly hydrophobic, which makes it water repellent. It also contains numerous peptide bonds and CO- as well as NH- group which forms hydrogen bonds between neighbouring molecules on the human organic follicle surface and has a highly porous cortex [1], [28], [29].

Method

All the reagents used for the experiments were of analytical grade procured from accredited dealer in England, UK. The oils used in the sorption examination were crude oil (Bonny Light, Nigeria), vegetable oil (Sainsbury, UK) and diesel fuel (Esso, UK). Table 1 shows the characteristics of the different oils above. The human har used for the experiment was collected from local hairdressing salon and separated in line with their respective ethnic groups. Sorbent X (African Hair), Y (Asian Hair) and Z (European Hair).

Table 1

Physical characteristics of studied oils at room temperature.

Oil type	Density (g/cm)	Viscosity (Pa s)
Vegetable oil	0.910	0.061
Crude oil	0.849	0.024
Diesel fuel	0.840	0.005

Preparation of Adsorbent

The properly separated human hair(s) were soaked in hot water using detergent to ensure it is free from contamination. Then, they were rinsed with hot water and dried under natural sunlight for 48hrs.

Adsorption kinetics experiment

Adsorption experiments were conducted in artificial seawater, which was prepared artificially as has been previously described by Kester et al [30]. Table Table 2 shows the composition of the artificial sea water. 20 g of crude oil, vegetable oil and diesel fuel, were poured into separate 500 ml conical flask with 200 ml of seawater. Varying amounts of the sorbents (0–8 g) were added with a 60 min contact time following the ASTM F 726-99 standard method for testing oil spill sorbents [31]. Oil concentrations were measured based on US-EPA method 1664 [32]. Oil adsorption capacities (mg/g) were obtained from the following equation:

Sorption capacity = $\frac{Xo-Xs}{Xs}$ -- [1]

where X_O (g) is the total mass of wet sorbent after oil adsorption and X_S (g) is the mass of the sorbent before adsorption. All experiments were conducted at room temperature (25^0C+2^0C) and were performed in triplicate with the average value and standard deviation (SD) calculated. Sample data with SD greater than 10% were rejected with a re-run of the test carried out. A dosage of 4g was used for all subsequent experiments.

Table 2
Composition of salts in artificial seawater

Salts	Molecular Weight	g/kg solution
Sodium chloride (NaCl)	58.44	23.926
Sodium sulphate (Na_2SO_4)	142.04	4.008
Potassium chloride (KCl)	74.56	0.667
Sodium bicarbonate ($NaHCO_3$)	84.00	0.196
Potassium bromide (KBr)	119.01	0.098
Boric acid (H_3BO_3)	61.83	0.026
Sodium flouride (NaF)	41.99	0.003

Sorption kinetic and isotherm study

Batch adsorption isotherm studies were carried out using Type X human hair in all the three oil types. Experiments were performed in triplicate with samples withdrawn at intervals ranging from 5 to 70 min. The experimental data obtained were analyzed using pseudo-first-order and second-order kinetic models [33], the Langmuir [34] and Freundlich [35] adsorption isotherm models. The experiments were conducted with varying weights of the adsorbents to determine the adsorption capacities. The kinetics and adsorption isotherm were also investigated.

Reusability test

There are different methods in which the adsorbent can be recovered and reused, e.g. compression, centrifugation, solvent extraction [14]. The solvent extraction method was employed in this research. In evaluating the reusability of human hair for oil sorption, the adsorbent was first washed with hot and cold water. All remaining oil was then extracted using n-hexane. The recovered adsorbent was then dried and reused for the batch adsorption experiments in four continuous cycles after each desorption test.

Results and discussion

Oil recovery

To determine the efficiency of the experimental processes leading to all the results obtained, it was necessary to evaluate its applicability. Under normal circumstance, one would expect 100 percentage recovery of oil since there was no contact between the adsorbent and the adsorbate. However, this was not the case in this experiment as the percentage recovery achieved for vegetable oil, diesel and crude oil were 96.20%, 94.68% and 99.41% respectively (Figure 1). In a similar experiment conducted by Nguyen [36] the result shows a percentage recovery of 91.63% for crude oil, 97.27% for diesel and 99.87 for vegetable respectively. This indicates that the process is not 100% efficient, although crude oil showed a very high percentage recovery as such each of them will serve as the quality control throughout this research.

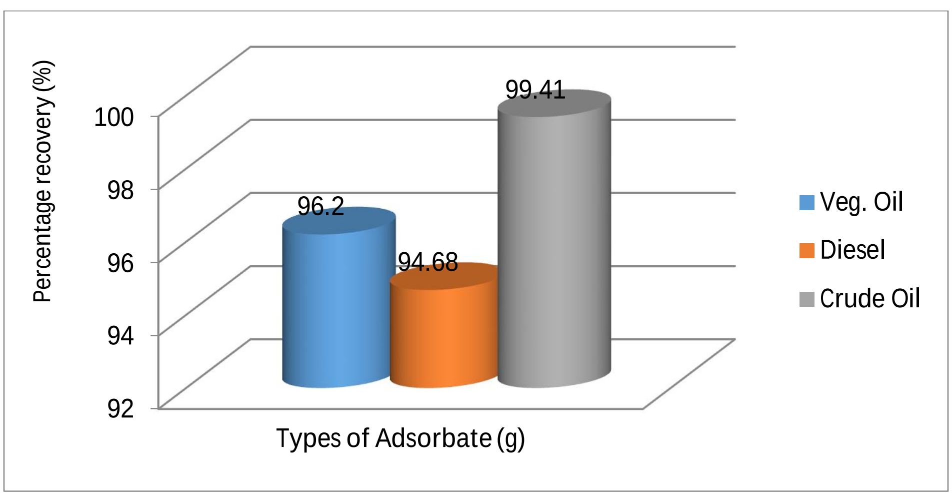

Fig 1. Oil recovery experiment for vegetable oil, crude oil and diesel fuel

Effect of contact time on adsorption

The effect of contact time in relation to the adsorption capacity of human hair were studied using batch adsorption test at different times of 5, 10, 20, 30, 40, 50, 60 and 70 minutes with other experimental variables kept at constant. Fig 2. Indicates a graphical representation of the data on the effect of contact time. It is obvious from the graph that in the first five minutes of the experiment there was rapid increase in the uptake of the different oils, thereafter the uptake capacity started reducing with time until equilibrium was reached at about the 60th minute. The initial high rate of oil uptake may be due to the availability of bare active sites on the surfaces of human hair [1], [36]. The gradual drop in the uptake capacity may be attributed to saturation of the active sites for adsorption of oil until equilibrium was attained. In a related research conducted using modified oil palm leaves, sludge, onion and garlic peel, the uptake capacity followed a similar pattern until equilibrium was reached [Sidik et al 2012]. However, for diesel the concentration adsorbed tend to follow an irregular pattern, the value decreased from 61.38mg/ml to 53.70mg/ml with an increase in contact time from 5mins to 10mins, then it increased to 63.92mg/ml in 20mins and finally to 64.69mg/ml in 60mins. This may be due to either human or equipment error during the experiment. This experiment gives a clear picture of the relationship between field application, duration and the effectiveness of the adsorbent.

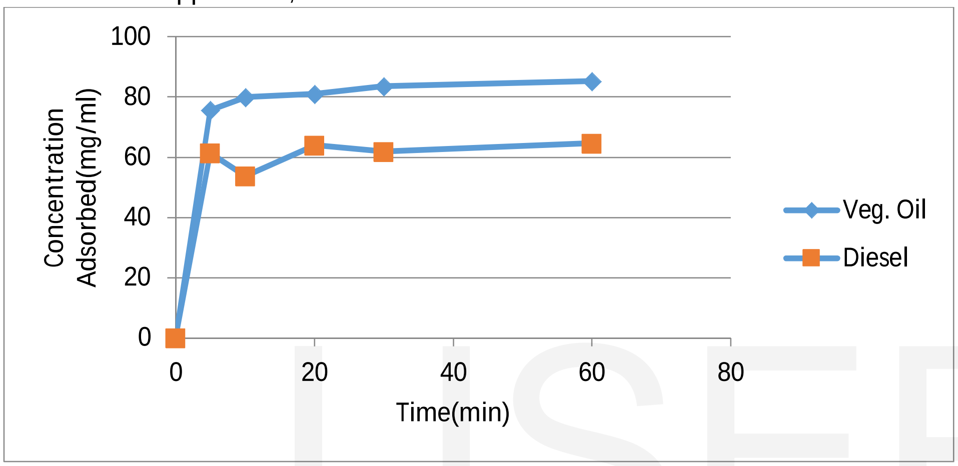

Fig 2. Sorption rate of Type Y human hair in vegetable oil and diesel at temperature (26^{0}C).

Effects of temperature on the adsorption capacity

The effect of temperature on the adsorption capacity of Type Y human hair using crude oil and vegetable oil were studied at different temperatures of 18, 25, 32 and 40. Fig 3 shows the results of the investigation. It indicates a two way pattern, with the experiment involving vegetable oil showing that the adsorption capacity of human hair decreases with increasing temperature, while that of crude oil indicates that the adsorption capacity of the adsorbent increases slightly with increasing temperature maybe as a result of differential viscocity between vegetable oil and crude oil. In a similar experiment conducted by Rajakovic-Ognjanovic et al [37] using natural wool fibres and recycled non-woven material while investigating the relationship between adsorption capacity and temperature, the result demonstrates that the adsorption capacity decreases with increasing temperature. Also, the adsorption capacity obtained by Vlaev et al [22] using rice husks, [38] using kapot an agricultural waste material, Sidik et al [39] using lauric and modified palm leaves reported a similar behaviour for the adsorption of vegetable oil. Ifelebuegu et al [1] reported a decrease in adsorption capacity with increasing temperature using crude oil. Hence, challenging the result using crude oil in this experiment which is the opposite direction.

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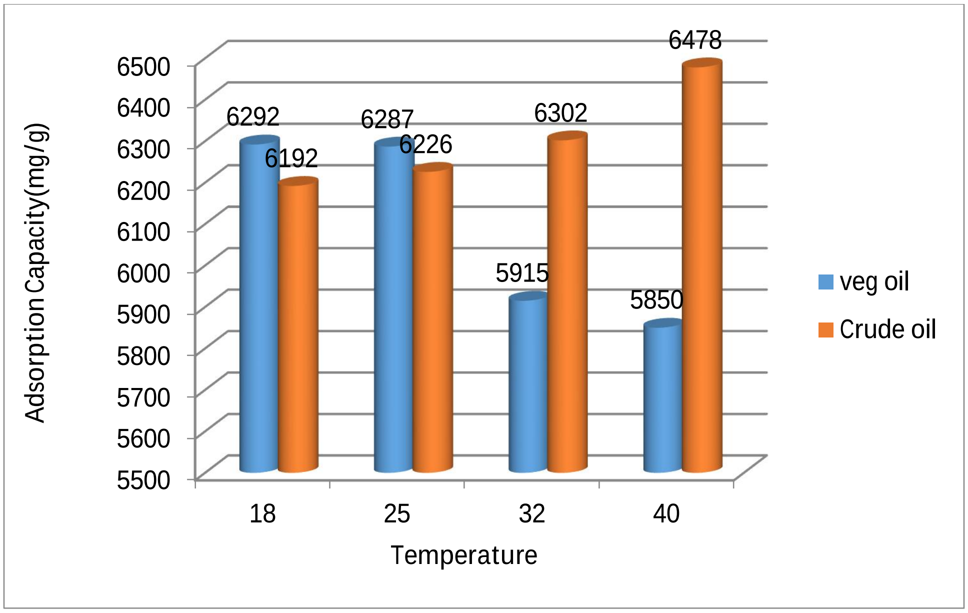

Fig. 3. Effect of temperature on the sorption capacity of Type Z human hair.

Effect of human hair dose and type

The effect of adsorbent dosage on the removal efficiency of vegetable oil, crude oil and diesel were studied by the application of different dosage between 2 to 6g into the different oily water samples. The concentration of oil adsorbed shows a rapid increase at the initial stage of adsorption (2g dose) and then increase gradually until equilibrium was reached at 87.96mg/ml for vegetable oil, 74.78mg/ml for diesel and 103.12mg/ml for crude oil respectively (Fig 4). The results demonstrate that the percentage uptake of human hair increases with increasing adsorbent dosage for vegetable oil, diesel fuel and crude oil. This is similar to the results obtained while studying the relationship between adsorbent dosage and the concentration of oil removed using peat-based sorbents, hydrophobic silica aerogels, sludge, onion and garlic peel and human hair [1], [39], [40], [41], [42]. The uptake efficiency for crude oil is encouraging over others.

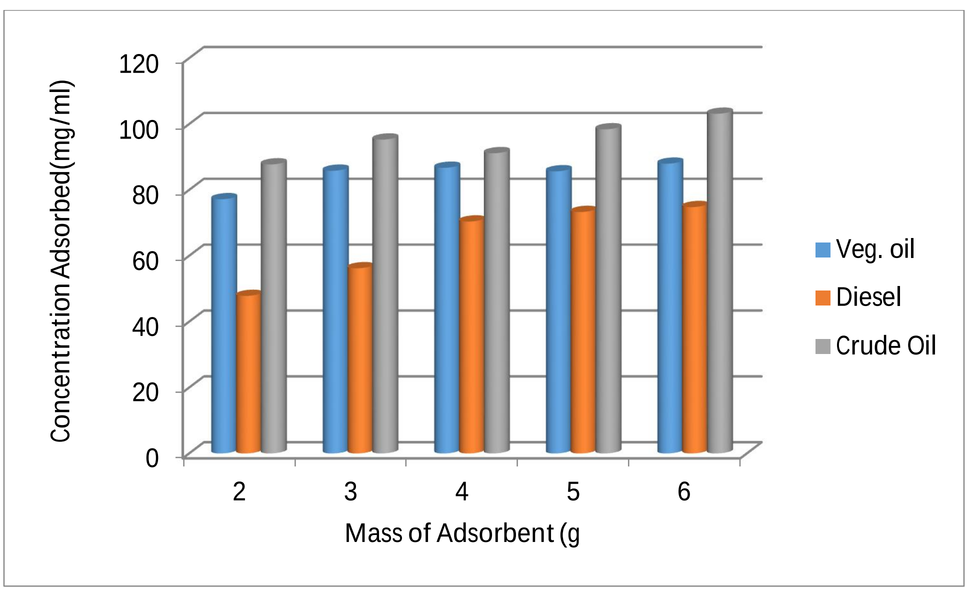

Fig 4. Effect of adsorbent dose on the adsorption capacity of human hair

Types of human hair and their adsorption capacities

The adsorption capacities of three different types of human hair were examined using crude oil. Fig. 5 shows the adsorption capacities of the adsorbent which indicates that type X came first with an adsorption capacity of 7470mg/g about eight times the weight of the adsorbent, followed by type Y with the adsorption capacity of 6176mg/g about seven times its weight and finally, type Z with adsorption capacity of 5246mg/g about six times it's weight of the adsorbent for crude oil. The superior performance of type X adsorbent can be attributed to its tight, coarse and spongy nature. According a study conducted by Ifelebuegu et al [1] to identify features that may be responsible for the dominance of type X adsorbent over others. The different adsorbents were examined under a scanning electron microscope; type X adsorbent has a rougher cuticles formed by many layers of hair which looks like a scalp. It is more hydrophobic as water tends to bead on the hair strands. Also, there are more macropores, which play a significant role in the sorption of oil by human hair due to capillary action of the oil at the hair/oil interface [28]. On the other hand, type Y and Z are les tightly packed when compared to type X, as such this features may have accounted for their performance.

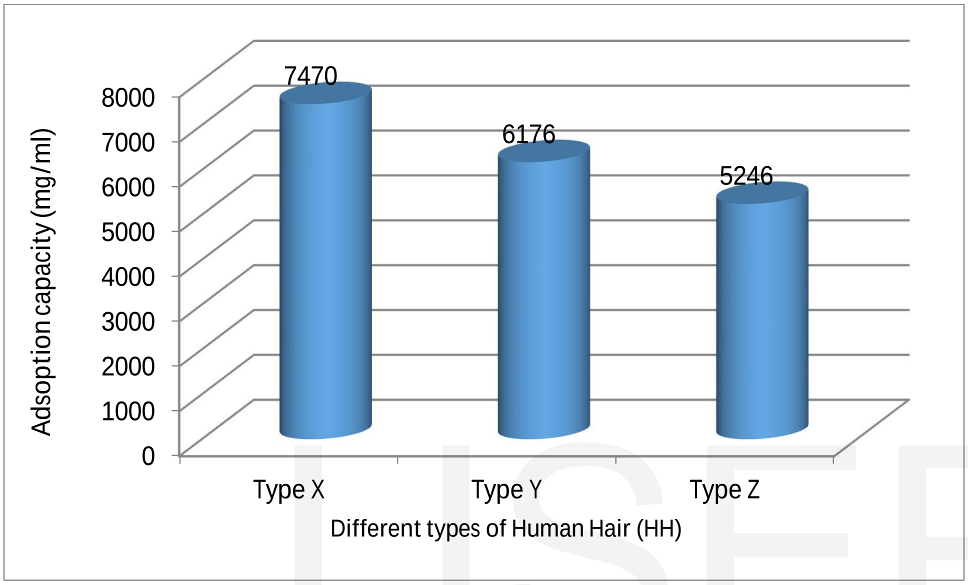

Fig 5. Sorption capacities of Type X (African Hair), Y (Asian Hair) and Z (European Hair)

Reusability of human hair (HH)

The suitability of human hair to undergo recovery and reuse was examined. Different methods exist in which adsorbents like HH can be recovered and reused e.g solvent extraction, compression and centrifugation. However, for the purpose of this research, solvent extraction was used to determine the potential of reusing synthetic hair without altering significantly its performance over time. The result as shown in Fig 6 indicates that there is no significant reduction in the adsorption capacity of the adsorbent after subjecting it to four cycles of recovery and reuse. This result seem to agree with the work of Liu et al [43] using kapot which showed no significant loss in its adsorption capacity after it was subjected to the process of recovery and reuse five consecutive times.

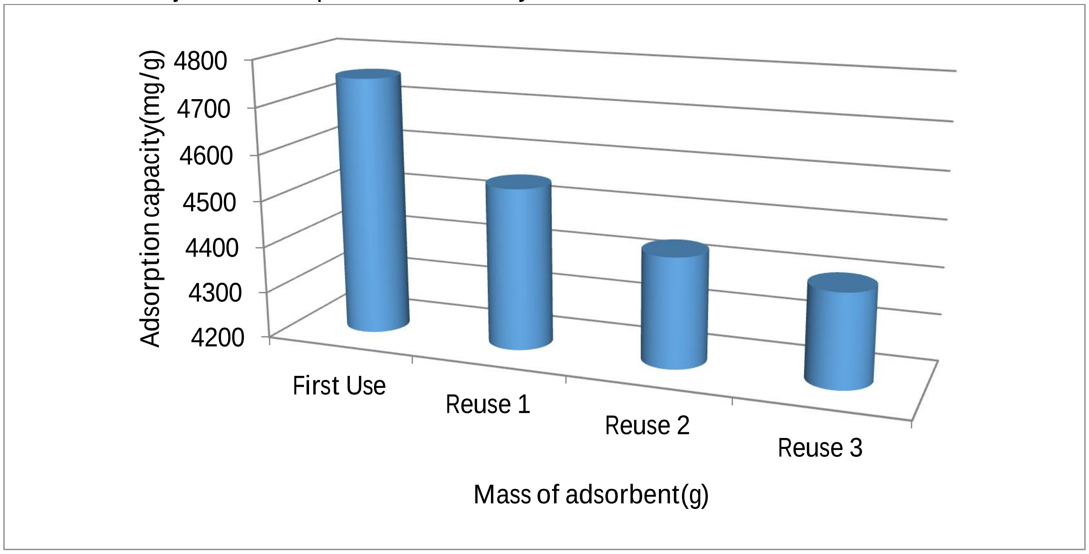

Fig 6. Sorption capacities of human hair during recovery and reuse

Adsorption isotherm and kinetics

Adsorption isotherm describes how solutes interact with adsorbents as well as its relevance in optimizing the use of adsorbents. The Langmuir and Freundlich adsorption isotherm models were used to analyse the data obtained inorder to attain equilibrium in this study.

The linear form of the Langmuir isotherm is expressed as:

$$\frac{1}{X/M} = \frac{1}{b} + \frac{1}{ab} \times \frac{1}{Ce} \quad \text{------(2)}$$

where X – the mass of adsorbate (mg), M – the mass of adsorbent (mg), Ce – the concentration of solute remaining at equilibrium in mg/L, a, b – the constants. a is the coefficient, and b is the amount of adsorbate needed to form a complete monolayer on the adsorbent surface and so increases with molecular size. A plot of (1/(X/M)) against (1/Ce) gave a straight line.

Also, the linearized form of the Freundlich isotherm is expressed as:

$$\text{Log}\frac{X}{M} = \text{Log}\, k_f + \frac{1}{n}\text{Log}\, Ce \quad \text{------(3)}$$

where X – the mass of adsorbate (mg), M – the mass of adsorbent (mg), Ce – the concentration of solute remaining at equilibrium in mg/L, kf and n – constants derived from the adsorption isotherm by plotting (X/M) against Ce on log–log paper which produces a straight line with a slope 1/n while the y-intercept is kf.

Table 3 presents the correlation coefficient as well as the Langmuir and Freundlich coefficients for single solute adsorption isotherms. The experimental data obtained indicates a good fit for Freundlich model which placed side by side with Langmuir. Sidik et al [2012] established that Freundlich isotherm model better fit the adsorption process while using oil palm modified leaves to adsorb crude oil. They however, suggested that having the value of n greater than unity (n = 1.39) indicates that the oil is favourably adsorbed on the adsorbent used.

The prediction of the rate of adsorption is determined using adsorption kinetics and it also helps in selecting the optimum operating conditions in designing and optimizing full-scale application [1]. The pseudo first and second order were used in this experiment to determine the adsorption kinetics of type X human hair for vegetable oil and diesel.

Below is the pseudo first order equation;

$$\text{Log}(C_e - C_t) = \text{Log}\, C_e - \frac{K1t}{2.303} \quad \text{------(4)}$$

Also, pseudo second order equation;

$$\frac{t}{Ct} = \frac{1}{K2Ce2} + \frac{t}{Ce} \quad \text{------(5)}$$

where C_e and C_t are the liquid-phase concentrations of oil in mg/l at equilibrium and time t, respectively. k_1 and k_2 are the first order and second order rate constants, respectively. The pseudo first order plot of Log (C_e_C_t) against t should give a linear relationship from which k_1 in (min^{-1}) can be calculated from the slope of the graph. A plot of t/C_t will give a rate constant k_2 (mg g^{-1} min^{-1}) for pseudo-second order kinetics. The constant values as well as the correlation coefficient R^2 of pseudo-first order and pseudo-second order kinetic models for adsorption of vegetable oil and diesel is stated in table 4. The kinetic plot for pseudo-second-order model, there was a good agreement between the experimental and the calculated values.

Table 3

Constant and coefficient of determination for Langmuir model and Freundlich for Type X human hair

	Langmuir			Freundlich		
	a	b	R^2	n	K_f(g/g)(L/g)$^{1/n}$	R^2
Crude oil	1.2550	4.215	0.3419	2.3790	1.4761	0.9635
Diesel fuel	0.0104	20.00	0.8832	2.3790	1.0080	0.9760
Vegetable oil	0.1315	15.28	0.5820	1.8635	1.0730	0.9640

Table 4

Pseudo first and second rate constants for Type X human hair

Adsorbate	Pseudo-first order kinetics		Pseudo-second order kinetics	
	k_1 (min^{-1})	R^2	k_2(mg/$g^{-1}$$min^{-1}$)	R^2
Diesel fuel	0.0200	0.214	0.0380	0.998
Vegetable oil	0.030	0.209	0.0200	0.990

Improving the adsorption capacity of human hair (HH)

The effect of modifying human hair using polymeric material was investigated to determine its sorption capacity and field applicability. The result indicates an improvement in its sorption capacity up-to 4606mg/g for vegetable oil (see Fig 7). The modification is aimed at giving it a similitude of boom owing to the fact that the adsorbent will need to be in such form before field application. Comparatively, HH modification performed better than the work of Sidik et al [39] while testing the adsorption capacity of unmodified and modified oil palm leaves with the resultant adsorption capacity of 92.32 mg/g for unmodified and 645.73 mg/g for modified respectively. However, Toyoda and Inagki [44] studied the adsorption capacity of exfoliated graphites with different bulk densities and obtained a maximum adsorption capacity of 86000 mg/g which is an impressive result.

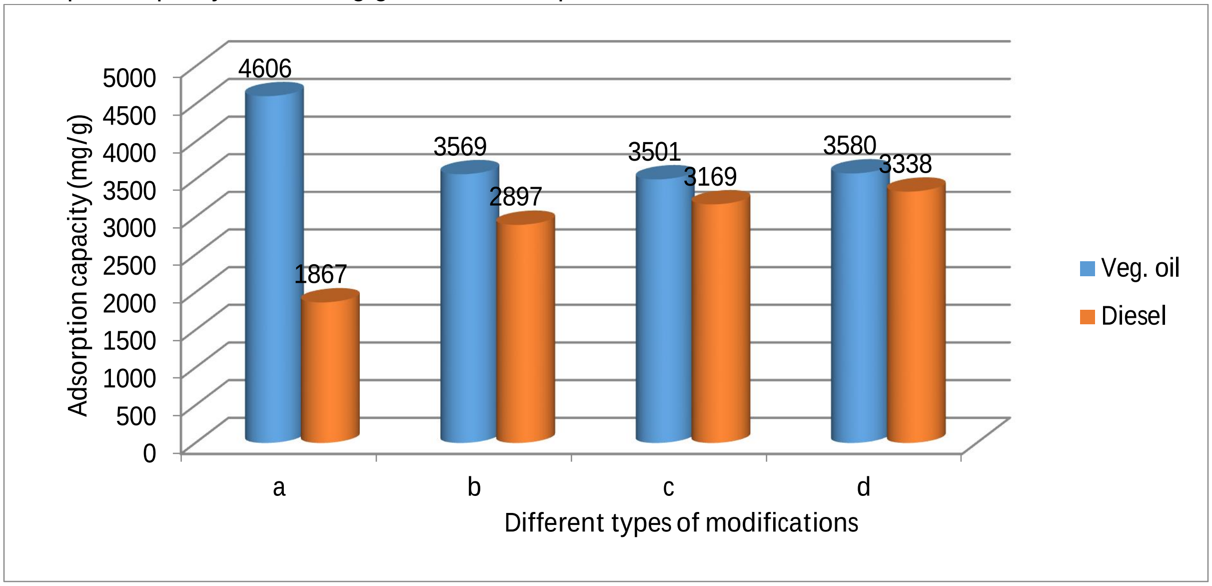

Fig 7. Sorption capacity for different modifications of human hair (HH). Key: a – d typify the following (a) polymeric material only, (b) polymeric material tightly packed with HH, (c) polymeric material moderately packed with HH and (d) polymeric material loosely packed with HH

Comparative analysis of different types of adsorbents

Table 5. Indicates different types of adsorbents cited in the literature including human hair. Human hair performed better than organoclay, rice husk, reed canary grass, treated sludge, modified oil palm leaves, peat moss and activated carbon. However, exfoliated graphytes, kapot and recycled wool based non-woven material top the table followed by human hair and others. The overall effect is that, human hair when given the right type of modification can compete favourably with other adsorbents present in the market in the field of oil spill remediation.

Adsorbent	Adsorbate	Maximum Adsorption Capacity (mg/g)	Reference
Exfoliated graphytes	Crude oil	76000	[44]
kapot	Engine oil	45000	[38]
Recycled wool based non-woven material	Vegetable oil	15800	[45]
Human hair	Vegetable oil	7470	
Organoclay	Diesel	7200	[46]
Rice husk	Crude oil	6220	[22]
Reed canary grass	Oily wastewater	3300	[47]
Treated sludge	Crude oil	2000	[42]
Modified oil palm leaves	Crude oil	645.73	[39]
Peat moss	Oily waste water	189	[48]
Activated carbon	Vegetable oil	47.8	[49]

Conclusion

The capacity of human hair to adsorb crude oil, vegetable oil and diesel fuel was investigated with the aim of establishing its potential for oil spill clean-up. This is timely not just because of the need to remove contaminants from the environment without due diligence to its impact, but because it is environmentally friendly and also encourage the principle of converting waste to wealth. The result obtained indicates that there was rapid increase in the sorption of the various oils in the first 5 minutes after which it proceed at a slower rate until equilibrium at 60 minutes. The relationship between sorption capacity and temperature was also examined and the result indicates that an increase in temperature reduces the sorption capacity of human hair, except for crude which shows a slight increase. Similarly, investigation on Type X,Y and Z human hair indicates that Type X performed impressively well with an adsorption capacity of 7470mg/g and 6176mg/g, 5246mg/g for Type Y and Z respectively. The kinetics of adsorption was found to fit pseudo second order than first order model. The adsorption isotherm experiment established a relationship between an increase in the dosage of human hair with the uptake efficiency as well as the concentration of oil adsorbed. All the experimental data fit well with Freundlich isotherm model indicating elements of chemisorption and oil retention occurring on heterogeneous sites with a non-uniform distribution of energy. The efficiency of human hair when subjected to the process of recovery and reuse was also examined and found to be effective after four consecutive times with little variation from the first to the fourth in its sorption capacity. Hence, this is an indication that human hair can be modified into boom and used as low-cost, environmentally friendly adsorbent for cleaning up oil spill, especially considering its potential for reusability without significantly altering the sorption features.

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