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## Using of agriculture residue in removing of oil spill

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### ABSTRACT

Nowadays, the concern about the environment contamination possibility of petroleum oil extraction to its processing, transportation and storage has been increased. The petroleum industries are undoubtedly great responsible of environmental impacts. On the other hand, the importance of the petroleum in our society is widespread and fundamental, because besides being one of the main energy sources used to days, its derivatives are raw materials for the consumption of countless consumer goods. Many efforts have being accomplished to develop new alternatives to remediation petroleum contaminated sites. A number of agriculture residues are utilized as adsorbents. They are insoluble in water, have a good chemical stability and high mechanical strength, have possesses a granular structure, making them a good adsorbent material. The use of biomass sorbents, which are less expensive and more biodegradable than synthetic sorbents, in such a way is one of the primary research directions in the field of the environmental protection. When oil pollution does occur, the issue is not only the cleaning of the environment but also recovery of this precious commodity. Hence any oil absorbing material used must also be able to release the oil. © 2011 Trade Science Inc. - INDIA

### KEYWORDS

Agriculture residue;  
Sorption;  
Desorption;  
Remediation;

### INTRODUCTION

The chemical contamination of water with a wide range of toxic products such as metal ions, aromatic molecules, dyes and so on, is a serious of environmental problem. The industrial activity accelerates the process of environment degradation, through uncontrolled gaseous emissions, inadequate disposal of the industrial wastes and other several forms that cause the contamination of the environment. Environmental concerns, demonstrated as being of vital importance as long as more and more environmental accidents started to happen, stimulating the research in a way to repair or to minimize,

those environmental damages. Besides, the pressure from the governments and public opinion has been forcing that environmentally correct posture of the industries, establishing laws that regulate the waste management<sup>[1]</sup>.

The petroleum industries are undoubtedly great responsible of environmental impacts. On the other hand, the importance of the petroleum in our society is widespread and fundamental, because besides being one of the main energy sources used nowadays, its derivatives are raw materials for the consumption of countless consumer goods. Whenever oil is explored, transported and stored and its derivatives are used, there is risk of spillage with the potential to cause significant environ-

mental impact. Pollution by petroleum oils affects sea life, economy, tourism and leisure activities because of the coating properties of these materials. Oil spills harm the beauty of polluted sites; the strong odor can be felt miles away and the excessive growth of green algae alters sea color and the landscape [2].

Each year, an average of 14 million gallons of oil from more than 10,000 accidental spills, are discharged into fresh water and salt water environments. These spills occur as a result of under water well blowouts, tanker accidents, storage tank failures, production platform blowouts, intensified petroleum exploration on the continental shelf, transfer operations between ships and shores, economic sabotage and youth restiveness [3]. Immediate response in the cleaning up of the immiscible oil does much good in reducing the environmental hazards, the use of booms to contain the oil, as well as other methods have proved very useful cleaning method, nevertheless, there is the problem of dissolved fractions of the oil in the water bodies, these, in fact cause more harm than the bulk of the oil that floats on the top of the water since it interferes with the ecosystem of the water bodies. The dissolved fraction of the oil are made up of hazardous organic substances such as benzene, xylene, toluene, that are capable of causing diseases such as carcinogenesis and mutagenesis; they can also lead to death of aquatic lives [4].

Efforts have being accomplished to develop new alternatives to remediation petroleum contaminated sites. Organic contaminated soils may demand the utilization of a biological, physical and chemical combination of technologies to reduce the contamination up to a safe and acceptable level [2]. Mechanical recovery is the transfer of oil from the spilled area to some transportable form of temporary storage by the help of oil sorbents or skimmers [5]. The sorbents in use can be classified as polymers, natural materials or treated cellulosic materials [6]. The most commonly used commercial sorbents are synthetic sorbents made of polypropylene or polyurethane [7]. They have good hydrophobic and oleophilic properties, but their non-biodegradability is a major disadvantage [5]. Since most oil products are biodegradable, oil could be disposed of for example by composting. A biodegradable material with excellent absorption properties would be advantageous in this respect. A number of natural sorbents have been studied for use in oil-spill cleanup, e.g. cotton [8,9] wool [10], bark [11-12] and rice straw [13].

A wide variety of natural sorbents such as rice straw,

corn corb, peat moss wood, cotton, milkweed floss, kapok, kenaf and wool fibers have been employed as sorbents in oil spill cleanup. These natural sorbents have the advantages of economy and biodegradability but have also been observed to have the disadvantages of poor buoyancy characteristics, relatively low oil sorption capacity and low hydrophobicity. However, it has also been shown that it is possible for some natural sorbents to sorb significantly more oil than even polypropylene materials that are normally used commercially [14]. For example, Kobayashi et al. [15] previously reported that the oil sorption of cellulosic kapok fiber used in a mat, block, band or screen was approximately 1.5–2.0 times greater than that of polypropylene mat which was observed to sorb 11.1 g heavy oil and 7.8 g machine oil in water. It has also been reported [15,8] that milkweed and cotton fibers sorbed significantly higher amounts of crude oil than polypropylene fiber and polypropylene web from the surface of an artificial sea water bath containing crude oil and from a crude oil bath.

Agricultural by-products have been widely reported for effective removal of metal ions from aqueous solution. There are few reports on their capacity to remove dissolved organic compounds from aqueous solution partly because the binding sites in agricultural by-products are considered to be ineffective in the removal of many polluted molecules. These agricultural products and residues are locally inexpensive and available. Some are waste materials and hence their reuses will in savings in disposal fee. The cellulosic products which exist in fibrous form can be easily formed into mats, pads and non-woven sheets for convenient applications [16]. They are insoluble in water, have a good chemical stability and high mechanical strength, have possesses a granular structure, making them a good adsorbent material.

### Non-treated agriculture residue

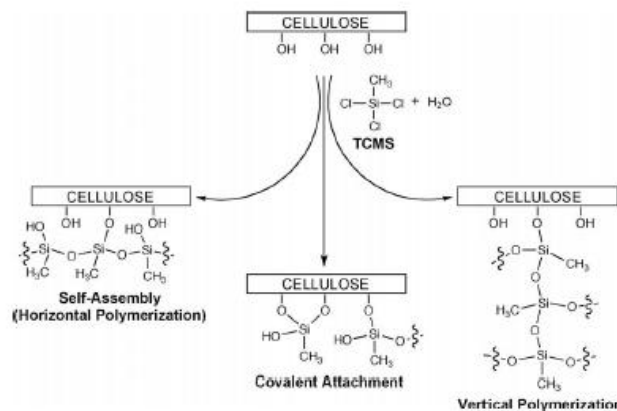
The importance of systematic utilization of bagasse (sugar cane cellulosic residues) has been noted in the past decade. Environmental concerns have fueled this focus not only because of the quantity of bagasse produced annually but also because of the nature of the material [17-18]. The sugar product from this plant represents only thirteen percent of the biomass. Bagasse from sugar production is twenty eight percent of the biomass [19]. The study of bagasse as an absorbent of environmental pollutants has received attention recently [20-22]. The utilization of bagasse as raw materials would access to cheaper material for adsorption in water pollut-

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ants control system.

Plant materials have long been used as sorbents for oil spill clean up in which oil concentration is relatively high<sup>[6,23]</sup>. However, the oil concentration in storm water runoff was relatively low<sup>[24]</sup> Moreover, a limited number of studies on the application of plant biomass to remove oil from stormwater runoff have been conducted<sup>[25]</sup>. A recent study investigates the feasibility of using biomass sorbents to remove oil from water runoff. Seven plant biomass; kapok fiber, cattail fiber, *Salvinia* sp., wood chip, rice husk, coconut husk, and bagasse were examined for oil sorption and desorption<sup>[3]</sup>. Polyester fiber, a commercial synthetic sorbent, was also experimented for comparison purpose. The sorbents were tested in the simulated conditions that would occur in the field. More than 70% of oil mass removal was achieved with all sorbents, except coconut husk and bagasse, which sorbed only 32% and 20% of oil mass. The sorbents that are hydrophobic such as kapok fiber, cattail fiber, *Salvinia* sp., and polyester fiber, could remove more oil than the other sorbents. The loose-fiber structures of these four sorbents, containing ample void spaces, also facilitated oil sorption. Polyester and cattail fibers provided the least average effluent oil concentrations. The oil sorption ability of some biomass sorbents (kapok fiber, cattail fiber, and *Salvinia* sp.) was not much different from the commercial sorbent (polyester fiber). During the desorption tests, less than 1.5% of the oil sorbed leached out of each of the sorbents tested except for polyester fiber that released the highest amount of oil of 4% of total oil sorbed. The use of polyester fiber as an oil sorbent must be practiced with care; frequent inspection to avoid the oil saturation is needed<sup>[26]</sup>.

Ch. Teas et al<sup>[7]</sup> examined the oil sorption capacities of five sorbents a commercial cellulosic material from processed wood, a commercial synthetic organic fiber from polypropylene and three commercial types of local expanded perlite from the island of Milos in three types of oil, namely Light cycle oil, Iranian heavy, and Light gas oil, to determine the potential use in oil spill clean-up, with and without the presence of seawater. They found that; in wet environments, the best performing materials were polypropylene and cellulosic fiber, while in the presence of water and Light cycle oil, one type of hydrophobic perlite had comparable absorption efficiencies with polypropylene and cellulosic material. In a dry environment, polypropylene exhibited the best absorption characteristics, whereas all expanded perlites had similar absorbing efficiencies with



**Figure 1: Schematic representation of the possible products of the reaction of TCMS with a cellulose substrate in the presence of controlled humidity (adapted from Ref. <sup>[41]</sup>).**

the cellulosic material for all oils used. The results suggest that substitution of commercial synthetic oil sorbents in oil spill clean-up and wastewater purification from organic molecules is possible by naturally occurring minerals with regional interest, given their friendliness to the environment and their local abundance.

### Treated agriculture residue

The hydrophobization, and especially the superhydrophobization, of materials which are not naturally hydrophobic has been a topic of chemical research for at least 60 years<sup>[27]</sup>. In the last decade that interest in this topic has grown spectacularly, as witnessed by the exponentially increasing number of studies<sup>[28-31]</sup>.

The hydrophobicity of a material may be assessed by the contact angle of a water droplet deposited onto its surface. In general, hydrophobicity is achieved, on the one hand, by lowering the surface energy, and more specifically the polar contribution to it, and, on the other, by creating adequate surface micro- and/or nanomorphologies, which hinder water spreading. When the water contact angle is higher than 90°, the corresponding material is conventionally defined as hydrophobic<sup>[32]</sup>.

Nonhydrophobic materials may be rendered hydrophobic by appropriate surface modifications involving both physical treatment and/or chemical reactions<sup>[33]</sup> e.g., by appending apolar moieties, such as alkyl or perfluoro groups, to the fiber surface<sup>[27]</sup>

Chlorosilanes are a good example of molecules extensively studied for this purpose, since they readily couple with OH bearing substrates through a condensation reaction that releases HCl<sup>[34]</sup>

Organic chlorosilanes are commonly used as coat-

ings for silicon [35] and glass surfaces, as well as in the production of silicone polymers (polysiloxanes) [27,36-38] specifically, methylchlorosilanes are an important and widely studied family of chlorosilanes [39,40], which find different applications depending on the number of methyl groups, varying between one and three. Trichloromethylsilane (TCMS) has already been employed to generate branched structures onto the surface of several OH-bearing substrates in order to increase their surface hydrophobicity.

The hydrophobization and lipophobization of cellulose fibers through their reaction with gaseous trichloromethylsilane (TCMS) were processed by Ana et al [27].

A simple system was used for the silanization of the filter paper with TCMS as shown in Figure 1.

Abdelmouleh et al [42] modified the surface of cellulosic fibres using organofunctional silane coupling agents in an ethanol/water medium. They found that; silane coupling agents adsorbed from diluted solution on cellulose fiber surfaces, followed by heat treatment, were shown to condense both among themselves and with the OH groups of the substrate to give Si-O-Si and Si-O-C couplings, respectively. These reactions ensured efficient and irreversible chemical bonding of the silane onto the cellulose surface. Contact angles and Inverse gas chromatographic measurements revealed that the hydrophilic character of the cellulose fibres can be strongly decreased after the silane treatment. A particularly hydrophobic surface could thus be obtained after treatment with Hexadecyltrimethoxysilane or  $\gamma$ -Methacryloxypropyltrimethoxysilane in the presence of triethylamine. Moreover, polymerisation tests, carried out in the presence of different monomers, showed that the presence of adequate functional groups on the silane molecule enable them to participate in the polymer chain growth and give rise to chemical grafting of the fibers. The chemical modification of natural cellulose fibers has been the object of much attention [43-45]. Esterification reactions can be carried out to modify the fiber surface and decrease the natural hydrophilic character of cellulose Fatty acids are carboxylic acids, often with along aliphatic tail that may be either saturated or unsaturated. They are found in animal and vegetable fats and oils and have the general formula  $C_nH_{2n-1}COOH$ . Cellulose esters of short chain fatty acids (length of four carbons or less) are commercially synthesized using acid anhydrides and a sulfuric acid catalyst in a heterogeneous phase reaction. Cellulose

esters of higher acid chain lengths have been prepared using acid chlorides with pyridine, acid chlorides under vacuum [46] and aliphatic acids with trifluoroacetic acid.

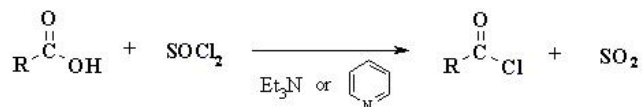


Figure 2: Acyl chloride preparation

Wang and Tao [44] reported the synthesis of fatty acid cellulose esters using hydrolyzed soybean oil and thus soybean fatty acids (mainly oleic, linoleic and linolenic acid). They prepared the fatty acid chlorides with

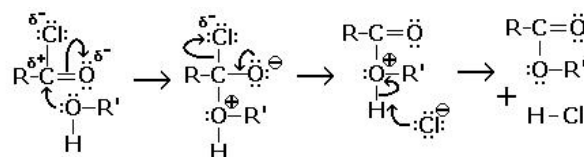


Figure 3: Esterification- Mechanism of Reaction

phosphorous pentachloride in benzene and reacted these with activated (by mercerization)  $\alpha$ -cellulose. The esterification reaction was performed in DMF and pyridine was also added to chelate the condensation reaction by-product HCl.

Kwatra et al studied the synthesis of long chain fatty acid cellulose esters by the vacuum acyl chloride method [46]. In this process, the HCl is removed by vacuum distillation, [47] thus eliminating the need for additional solvents and producing less complex solvent waste streams. The acyl chloride is prepared by treating the carboxylic acid with thionyl chloride ( $SOCl_2$ ) in the presence of a base [46].

Acyl chlorides are the most reactive of the carboxylic acid derivatives and therefore can be readily converted into other carboxylic acid derivatives. They are sufficiently reactive that they react quite readily with cold water and hydrolyze to the carboxylic acid.

The esterification reaction occurs by nucleophilic addition/elimination. The acyl chloride reacts with the alcohol to produce the ester and HCl. The reaction mechanism is as follows:

The acyl chloride carbonyl is highly polarized and the positive carbon is attacked by the nucleophilic alcohol. The alcohol adds to form a highly unstable ionic intermediate via a C-O bond and the  $\pi$  electron pair of the C=O double bond moves onto the oxygen atom to give it a full negative charge. The C-Cl bond electron pair then moves onto the chlorine atom which leaves as a chloride ion. Parallely, one of the lone pairs of elec-

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trons from the negative oxygen atom shifts to reform the C=O carbonyl bond. Finally, the previously formed chloride ion abstracts a proton to form the oxonium ion and the ester product<sup>[47]</sup>.

The modification of bagasse by acylation grafting with a fatty Acid, stearic acid, produced a material significantly more hydrophobic than the bagasse starting material. This grafted bagasse had little affinity for water and good affinity for oil. Raw bagasse had been shown to absorb oil better than the other materials in this study. However, raw bagasse also absorbed water. Hence the grafted raw bagasse would be most suitable for applications where oil is to be removed from an aqueous environment. For oil absorbing applications in the absence of water, the raw bagasse was found to be an excellent material. It can be prepared for this application by simply cleaning to remove residual sugar and drying<sup>[48]</sup>.

Cellulose fibers were grafted with aliphatic anhydrides having C6, C8, C12 and C16 chain length using a heterogeneous solvent exchange acylation procedure. The prepared modified fibers appeared to be efficient to trap different organic molecules dissolved in water. Recycling tests revealed that the saturated substrates could be regenerated tens of times without losing their capacity of absorption of organic contaminants<sup>[49]</sup>. The surface of cellulose fibers, Avicell (AV), as well as that of Whatman paper (WP) was chemically modified with two fluorine-bearing alkoxy silane coupling agents, namely: 3,3,3-trifluoropropyl trimethoxysilane and 1H,1H,2H,2H,perfluorooctyl trimethoxysilane. The contact angle measurements showed that, after grafting, the surface of AV and WP samples became totally highly hydrophobic with a contact angle of 140°. Thus, the polar contribution to the surface energy of the modified substrates was found to be close to zero. These modified substrate could be interesting for application such as self-cleaning surface, wipes paper, grease barrier paper or for biocomposite with a polar matrix<sup>[50]</sup>.

### Using Surfactant

The adsorption of surfactants at the solid-liquid interface has been a subject of considerable interest and lays an important role in controlling a variety of interfacial processes such as flotation, paper manufacturing and recycling, flocculation and dispersion, soil remediation, and adsorption of contaminants in water treatment<sup>[51,52]</sup>.

The aggregation of surfactant molecules at the solid-liquid interface generates hydrophobic cores formed by the associated surfactant tails on which organic com-

pounds can be trapped. This process, termed adsolubilization, stimulated much research and has found many applications in different domains, such as surface modification using admicellar polymerization<sup>[53,54]</sup>, chromatographic separation, wastewater treatment and drug carrier targeting in pharmacology<sup>[55]</sup>.

Fadhel Aloulou et al found that surfactant-modified cellulose fibers namely octadecyltrimethylammonium bromide (C18), hexadecylpyridinium chloride (C16), tetradecyltrimethylammonium bromide (C14) and dodecylpyridinium chloride (C12) appeared to be effective sorbents for the removal of organic compounds (Benzene, Chlorobenzene, 1,3,5-Trichlorobenzene, Nitrobenzene, 2-Naphthol, Naphtalene, 2-Chlorophenol, and Quinoline) dissolved in aqueous media<sup>[56]</sup>.

### Activated carbon

Activated carbon, with a good adsorption property, has been widely applied as an effective adsorbent to organic contaminants in water and wastewater treatment. Adsorption by activated carbon is the most commonly used method in removing organic pollutants from water<sup>[57-59]</sup>. Nevertheless, the adsorption of organic pollutants on activated carbon is only a superficial process, and with poor selectivity to organic pollutants and saturation-prone property, activated carbon needs to be regenerated frequently during the adsorption process. So it is dissatisfactory to use activated carbon to remove hydrophobic persistent organic pollutants in trace or ultratrace amount from water.

Jia et al<sup>[60]</sup> prepared a composite adsorbent which composed of the supporting activated carbon and the surrounding triolein-embedded cellulose acetate membrane. The adsorbent was stable in water, for no triolein leakage was detected after soaking the adsorbent for five weeks. The adsorbent had good adsorption capability to dieldrin, and the removal efficiency of the composite adsorbent was higher than the traditional activated carbon adsorbent.

Pyrolysis of rice husks at temperature of 450 °C for 3 hrs formed a composite comprised of hydrocarbon component containing over 98% SiO<sub>2</sub>. This treatment leads to obtaining of an adsorbent material with 2 to 3.5 times higher sorption capacity for the whole range of petroleum products – petrol, diesel fuel, motor oil, light and heavy crude oil<sup>[61]</sup>

Activated carbon adsorbents from corncobs were produced by Diya'uddeen et al at various conditions and successfully employed in the removal of crude oil

from water surface. The optimum activation temperature was 700°C and the temperature had no definite effect on both the apparent and solid densities of the corncob activated carbons. The result of the analysis indicates the dependence of the adsorption of the crude oil on the surface textural properties of the adsorbents. Values of purity indices of the crude oil showed that the process is a physical adsorption<sup>[62]</sup>.

An attempt by Hussein et al<sup>[63]</sup> was made to provide an efficient, easily deployable method of cleaning up oil spills and recovering of the oil. Carbonized pith bagasse, a relatively abundant and inexpensive material is currently being investigated as an adsorbent to remove oil contaminants from water. Fibers extracted from bagasse and carbonized at 300 °C were found to have a high performance for sorption and recovery of light, heavy oils and even the viscous ones. The carbonized pith bagasse was packed into a polypropylene bag. A comparison was made between the prepared pad and the commercial sorbents show that the pad containing carbonized pith bagasse has higher sorption capacity in comparison to the commercial sorbents. The pad exhibited high oil retention ability and a high selectivity for the oils over the water. The pad showed a possibility of reuse for eight times. The sorption capacity of the pads containing carbonized pith bagasse was found to increase with increasing the time of sorption till it reaches the maximum value at the time of sorption equal to 60 min.

## CONCLUSIONS

This review provides a general overview of the wide variety of sorbent materials that have been investigated for removal of oil spill with natural sorbents. The review shows that various workers have successfully prepared hydrophobic lignocellulosic materials by employing various modification procedures to incorporate chemical functionality. It is suggested that esterification or carbonization of rice straws and other biodegradable lignocellulosic vegetable products such as cotton wool, sugar cane, paper, wood, etc may prove very economical, technically feasible and environmentally acceptable for application in oil spill cleanup technology.

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