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Research Article

Modified methods of oil cleanup with cellulose–based adsorbents: a review

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Abstract: Oil spills and contaminated water sources are responsible for polluting marine environments, which in turn has adverse effects on marine ecosystems and public health. Among various oil removal methods, adsorption is the preferred technique due to its speed, simplicity, low cost, and eco–friendliness. In this paper, we will review methods for modifying the oil adsorption properties of cellulose to enhance its adsorption capacity. We have reviewed 287 relevant worldwide documents in recent years and selected 142 documents for use in this study. The review results of the number of documents show that there are three main methods for transforming cellulose–based adsorbents, including (1) Physical transformation methods such as mechanical crushing, or pressing, heat treatment, and the plasma technique; (2) Chemical modification methods such as mercerization, acetylation, grafting, acidification, aerogel modification, cationic surfactant; and (3) Bioremediation (immobilization of microorganisms). Among these modification methods, cellulose–based aerogels have shown remarkable oil absorption capabilities of up to 170.0 g/g, superhydrophobicity (with a water contact angle of 156.7°), and the ability to be reused up to 80 times. Cellulase denaturation and microbial immobilization are eco–friendly techniques that have potential to replace non–biodegradable oil adsorbents. Furthermore, utilizing agricultural by–products to produce high–capacity absorbent materials is a promising solution that benefits both the economy and the environment.

Keywords: Adsorption; Cellulose; Modified methods; Natural adsorbents; Oil cleanup.

1. Introduction

The worldwide deterioration of water quality and negative impact on the underwater ecosystem is caused by oil pollution resulting from oil spills and leaks during various activities such as transportation, shipping, exploration and production, oil refining, and disposal [1]. This has led to severe health consequences [2]. The majority of oil spill volumes in water originate from oil refinery ports (25%), ships (25%), oil tankers (20%), barge tankers (15%), and oil rigs (15%). The oil spills consist of various types of oil, with crude oil being the most common (35%), followed by diesel oil (20%), marine oil (10%), gasoline (8%), and the lowest is bunker oil (3%) [3].

There are various methods available for removing oil from water, including in–situ burning, chemical techniques such as solidification and dispersion, biological methods, and physical methods such as skimming and oil sorbents [4]. Nevertheless, oil absorbent materials are the most favored method due to their quickness [5], convenience, affordability [6], and eco–friendliness [7]. There are about 200 different sorbent materials available in the market, and the choice of sorbent material depends on factors such as availability, cost, and safe usage [8]. The three main categories of oil sorbent materials are inorganic mineral products, synthetic organic products, and natural organic products [9]. Inorganic products like perlite, vermiculite, and diatomite are mostly buoyant and have low oil absorption capacity [10], while organic polymer products such as polypropylene, polyethylene, and polyurethane have a major drawback of being difficult to biodegrade [11]. However, lignocellulose sorbent materials have significant advantages over other materials, especially in terms of their environmental friendliness and lightweight nature, making them easily recoverable and reusable [12].

Natural adsorbents from agricultural waste and byproducts, including rice straw [13], sawdust [12], onion skin [14], garlic skin [14], walnut shell [15], bagasse [16], barley straw [17], banana stem fibers [18], banana stem [18], peanut shell [19], rice husk [20], corn stalk [21], corn cob [22], pomelo peel [23], wheat straw [24], mango seed coat [25], banana peel [4], rice bran [26], Cha La seeds [27], flax fiber [28], durian peel [29], and coconut coir [30], have gained significant attention from researchers in recent years for their effectiveness in removing oil from water.

Natural absorbent materials are an appealing choice for cleaning up oil spills owing to their cost–effectiveness, widespread availability, biodegradability, and non–toxic nature. Nevertheless, they exhibit limitations such as low adsorption capacity, limited floatability, and hydrophilicity [31]. Researchers have reported different conversion methods in literature to enhance the properties of natural absorbent materials, such as their adsorption capacity and buoyancy, while maintaining their original properties. Ideally, these modifications should be minimal and should not affect the biocompatibility of the material [32].

To improve these properties, adsorbent materials can be modified by physical methods: mechanical [33], thermal [34–35]; chemical methods: alkaline treatment [36], acetylation [37], benzoylation and grafting [38–40], and biological methods [41], which can significantly enhance the adsorption properties of the natural base material.

This study will delve into investigating and evaluating methods to improve the oil adsorption capability of natural organic sorbents derived from environmentally friendly plant sources.

2. Cellulose–based adsorbents

Lignocellulosic, which refers to wood and plant materials, is a type of composite material that consists of three different polymers, namely cellulose, hemicelluloses, and lignin [42]. Cellulose is made up of straight crystalline chains of glucose units that are joined together by 1,4–β–glycosidic bonds, and the length of the polymer chain can be as long as 15,000 units [43]. One of the remarkable characteristics of cellulose is its ability to biodegrade and regenerate. Additionally, cellulose exhibits high strength and stiffness, low density, and excellent biocompatibility [44].

Natural organic sorbents are composed of cellulose, hemicellulose, and lignocellulose, which mainly composed of carbon (C), hydrogen (H), oxygen (O), and nitrogen (N), as noted by [45]. As pointed out by [46], high levels of carbon or oxygen content are important for improving the oil recovery capacity of sorbent materials in water. Although cellulose can serve as an efficient oil sorbent, its hydrophilic characteristics due to the hydroxyl (–OH) groups on the material’s surface can impede oil clean–up efforts in water. One solution is to substitute the hydroxyl groups to modify the sorbent's surface and increase its hydrophobicity [47]. This is also the rationale behind numerous modification methods used to substitute the hydroxyl groups.

The structure of the sorbent is a significant factor, including its hydrophobicity, porosity, suitable pore size, and surface area [46]. An oil absorbent that is considered ideal should exhibit high porosity and surface area while also being able to selectively absorb oil. To be useful for oil spill treatment purposes, the sorbent must also be stable chemically and mechanically, environmentally friendly, buoyant, and possess low density, in addition to being recyclable [48].

The next section will analyze methods of modifying sorbents to improve their hydrophobicity and oil sorption capacity of natural adsorbents.

3. Adsorbent modification method

Natural sorbents are typically only effective for oil absorption in non–aqueous environments [49]. To improve their performance, pretreatment methods are used to enhance functional groups, such as hydrophobic and ankylic groups, and increase pore numbers. Sorbent modification methods include physical modification, chemical modification (such as hydrolysis, acetylation, benzylation, grafting, and other methods), and biological modification [33].

3.1. Physical modification methods

3.1.1. Mechanical crushing or pressing

According to [33], mechanical crushing or pressing does not affect the hydrophobicity of natural adsorbents, but it can affect their effectiveness in removing oil. Crushing or pressing the absorbent can increase its oil absorption capacity by increasing the contact surface area between the absorbent and oil. However, if the absorbent is highly compressed, its oil absorption capacity may decrease because the oil has difficulty infiltrating the material [17, 50].

3.1.2. Heat treatment

Natural adsorbent modification by high–temperature treatment methods includes drying [35], hydrothermal treatment [33], and pyrolysis [34].

By drying, the material's porosity can be augmented, leading to an increase in the surface area of the adsorbent and an improvement in oil absorption capacity. However, conventional drying methods can cause a substantial reduction in the porous structure of the natural adsorbent, thereby decreasing its ability to adsorb and be reused [51]. Additionally, the drying process does not enhance the oil absorption capacity and water repellency of the organic natural adsorbent in comparison to raw fibers [35].

According to [52], hydrothermal treatment is an economical and eco–friendly technique that can eliminate cellulose components that are hydrophilic. When adsorbent materials are subjected to hot water treatment, it can help eliminate impurities, wax coatings, and volatile compounds from cellulose fibers, increasing their contact with the adsorbing environment [53]. Dried moss fibers were soaked in deionized water at temperatures of 80°C and 100°C for an hour, and the results showed that the adsorbent material treated with hot water at 80°C had a 12.4% increase in diesel oil adsorption capability, while treatment at 100°C showed a 6.7% increase when compared to untreated materials [33]. In another study, [54] utilized superheated steam treatment to remove hemicellulose from coir fibers. The findings of [33] indicated that hot water modification of the fiber surface resulted in a greater contact surface area, which improved the oil adhesion of the modified fiber surface and removed the wax coating and some volatile components. In addition, the heat treatment will increase the oil absorption capacity, enhancing the hydrophobicity and oil affinity of the fibers [55].

Pyrolysis is an alternative heat treatment method that can increase the hydrophobicity and oil absorption capacity of materials through carbonization. However, this process is expensive and time–consuming [56]. Carbonized rice husk has been found to be a superior natural adsorbent due to its high content of oil–affinity SiO_2 group, which enhances its oil absorption capacity and hydrophobicity. However, the efficiency of pyrolysis depends on the number of natural fibers in the plant material being used [34]. Rice husk waste is rich in silica, and pyrolyzing the husk leads to the decomposition of organic matter and the breakdown of bonds between silica and organic matter. This increases the hydrophobicity of the husk, allowing it to selectively absorb crude oil [20]. Table 1 presents a summary of the synthesis of natural adsorbents using various heat treatment methods such as drying, hydrothermal treatment, and pyrolysis.

Table 1. Synthesis of natural adsorbents treated by heat methods.

Material	**Method**	**Oils**	**Sorption capacity/ Removal**	**Ref.**
Barley straw	Drying	Gas oil	7 – 8.5 g/g	[57]
	Carbonization, 400°C in 0.5 to 3 hours	Gas oil	8.5 – 9 g/g	[57]
Calotropis procera fiber	Thermally treated, 150°C	Crude oil	76.32 g/g	[58]
	Thermally treated, 200°C	Crude oil	94.31 g/g	[58]
	Hot water treatment at 80°C in 1 hour	Crude oil	99.2 g/g	[36]
	Oven drying, 60°C, 24 hours	Engine oil	1.107 g/g	[59]
Coconut husk	Oven drying, 60°C, 24 hours	Engine oil	0.058 g/g	[59]
Kapok fiber	Oven drying, 60°C, 24 hours	Engine oil	0.827 g/g	[59]
	Sun–drying	Diesel oil	19.35 g/g	[60]
	Sun–drying	Engine oil	60.51 g/g	[60]
	Sun–drying	Used engine oil	49.94 g/g	[60]
Luffa (an agricultural waste)	Cutting, Sieving, washing, and drying (105°C)	Diesel oil	> 85 %	[60]
Moss	Hot water treatment, 100°C	Diesel oil	7.85 ± 0.09 g/g	[33]
	Hot water treatment, 80°C	Diesel oil	8.27 ± 0.06 g/g	[33]
Potato peel	Drying, 70°C and crushing	Waste lubricating oil	2.15 g/g	[23]
Rice husks	Oven drying, 60°C, 24 hours	Engine oil	0.298 g/g	[59]
	Sun–drying	Diesel oil	2.60 g/g	[60]
	Sun–drying	Engine oil	9.26 g/g	[60]
	Pyrolysis	Diesel oil	5.02 g/g	[20]
	Pyrolysis (N_2 or inert atmosphere)	Diesel oil	2.78 g/g	[20]
	Pyrolysis (Carbonized), 480°C	Diesel oil	5.5 kg/kg	[34]
	Pyrolysis (Carbonized), 480°C	Motor oil	7.5 kg/kg	[34]
Salvinia cucullata Roxb	Oven drying, 60°C, 24 hours	Engine oil	0.944 g/g	[59]
Silkworm cocoon waste	Oven drying, 110°C reduced to 60 °C, cutting and milling	Motor oil	42 – 52 g/g	[62]
Sugarcane bagasse	Oven drying, 60°C, 24 hours	Engine oil	0.019 g/g	[59]
	Sun–drying	Diesel oil	10.51 g/g	[60]
	Sun–drying	Engine oil	19.95 g/g	[60]
	Sun–drying	Used engine oil	18.01 g/g	[60]
	Oven drying	Engine oil	3.2 – 5.3 g/g	[61]
Wood chips	Oven drying, 60°C, 24 hours	Engine oil	0.343 g/g	[59]

Table 1 provides information on the oil adsorption capacities of naturally derived organic adsorbents that have undergone thermal treatment. Calotropis procera fiber and Kapok fiber are examples of natural fiber adsorbents with excellent oil adsorption capabilities. Heat treatment of Calotropis procera fiber at 150°C and 200°C for 1 hour resulted in the reduction of functional groups, such as C–H (2920 cm^{-1}), C=O (1734, 1368, and 1244 cm^{-1}), and C–O (1032 cm^{-1}), and the disappearance of lignin (1505 and 1597 cm^{-1}) and hemicellulose (1737 and 1248 cm^{-1}) peaks. Furthermore, the heat treatment helped eliminate the water–repellent waxes from the fiber surface, thereby increasing its oil adsorption capacity. The *Calotropis procera* fiber with a large lumen, has a high oil adsorption capacity of up to 74.04 g/g. Raising the temperature from 150°C to 200°C results in an increase in the maximum adsorption capacity from 94.31 g/g to 124.60 g/g [58]. Similarly, Kapok fiber has oil adsorption capacities of 19.35 g/g, 49.94 g/g, and 60.51 g/g for diesel oil, used engine oil, and engine oil, respectively, after being sun–dried [60].

3.1.3. The plasma technique

The plasma method is a physical process that transforms the surface of fibers by forming strong bonds between the fiber matrix and the new functional groups, leading to improved mechanical properties of natural fibers [63–64]. Low–pressure flo plasma treatment has been used to enhance the surface hydrophilicity of cellulose fibers towards hydrocarbons that are dispersed in water, which improves their ultra–hydrophobic adsorption properties. The cellulose fibers were obtained from various vegetables, treated by NaOH (0.5% w/w) at 353 K for 20 minutes, and then treated with low–pressure plasma in the laboratory. The results of the kinetic analysis demonstrated that the fibers treated with plasma achieved a removal efficiency ranging from 80% to 90% after only one minute of exposure, which depended on the initial weight ratio of hydrocarbon to fiber (ranging from 20 mg/g to 240 mg/g). Moreover, the maximum adsorption capacity of the treated fibers exceeded 270 mg/g, and the adsorption process adhered to the Langmuir adsorption isotherm, as noted by [65].

3.2. Chemical modification method

The chemical treatment of adsorbents is a technique that can be used to cleanse the surface of fibers, alter the surface chemically, and increase the roughness of the surface [66]. This modification can lead to a reduction in water adsorption and an improvement in oil adsorption as compared to unmodified adsorbents [33]. Many techniques have been studied to improve the ability of adsorbents to repel water (hydrophobic) and attract oil (lipophilic), including alkalization, acetylation, benzoylation, crosslinking agents (with or without heat), grafting, and other techniques [67]. In order to attach different chemical groups to the hydroxyl (–OH) group on the cellulose chain, a variety of reactions are used. These reactions typically involve chemicals such as alkali, acetic anhydride, coupling agents, peroxide, stearic acid, fatty acid derivatives (oleoyl chloride), and others [68–69].

3.2.1. Mercerization

Alkali treatment is defined by the ASTM D1965 as a method in which strong bases are used to treat plant fibers, causing significant swelling and changing the fibers’ fine structure, size, shape, and mechanical properties [70]. Mercerization, a process that breaks down fiber bundles into smaller ones by treating them with alkali, is widely used for modifying organic adsorbents because it is simple and effective [47, 71]. The initial alkali treatment is used to remove non–cellulose components like lignin and pectin, which exposes the inner surface and creates a rougher surface structure for natural fibers [56]. This increases surface roughness and promotes mechanical interlocking and better contact of cellulose on the fiber surface, which increases the number of reactive sites [72]. Hasim has shown that the change in natural crystalline structure depends on the type and concentration of alkali used [73]. Increasing the NaOH concentration in the alkali treatment increases water repellency, leading to a reduced water adsorption capacity of single abaca fiber [74]. Table 2 summarizes the literature on the parameters of the alkali treatment process, focusing on NaOH concentration, treatment temperature, and soaking time.

Table 2. Mercerization conditions of natural cellulose fibers.

Material	NaOH treatment parameters	Oils	Effect/Comments	Sorption capacity	Ref.
Calotropis procera fiber	Agitate NaOH 0.1 M for 1 hour	Crude oil	The functional groups' intensity was reduced, and the surface became slick with a waxy hydrophobic coating on the inner surface of the hollow structure. The fiber diameter increased from an average of 23.84 ± 4.44 µm to 37.47 ± 3.80 µm, and the specific surface area improved from 146.6 to 390.8 m²/kg. Furthermore, 93% of the structure is made up of voids.	103.9 g/g	[36]
Hybrid peel waste (banana skins + orange peel)	Hybridization of peels with NaOH 1 M, stirred at 350 rpm for 24 hours and drying at 70°C for 24 hours	Lubricant oil	The process of hydrolyzing the adsorption band of $-COOCH_3$ on the surface of fruit peels results in an augmentation of carboxylic acid amount due to the formation of $-COO-$ groups.	38.12%	[75]
Moss	NaOH solvent (5%) at 25°C for 48 hours	Diesel oil	The treatment of moss fibers with mercerization resulted in an enhancement of surface roughness and area, leading to swelling and an increase in diesel sorption capacity by 22%.	8.99 ± 0.08 g/g	[33]
Plant fibers (soybean)	The fibers were subjected to alkali treatment by immersing and shaking them in a 5% NaOH solution for two different periods: 48 hours at 25°C and 1 hour at 300°C. Following the treatment, the fibers were drained and dried at 105°C for 24 hours	Crude oil		5 g/g	[76]
Rice Husks	NaOH 4 M at 90°C	Marine diesel	The use of low NaOH concentrations (0.1 M) can effectively eliminate surface fat, wax, and hemicellulose, whereas comparable oil uptake outcomes are seen for samples treated with NaOH concentrations ranging from 2–6 M.	20 g/g	[77]

Table 2 shows that *Calotropis procera* fibers treated with 0.1 M NaOH for 1 hour resulted in a reduction in functional groups' intensity, such as CH (2920 cm^{-1}), C=O (1734, 1368, and 1244 cm^{-1}), and CO (1032 cm^{-1}). This is because the treatment removed impurities, wax and pectin from the surface of the fibers. The contact angle θ of the hydrophobic surface was measured at 119° and 0° for diesel. Mercerisation considerably increased the inner diameter and surface area of *Calotropis procera* lumens, leading to an increased absorption capacity of up to 103.9 g/g [36]. The mercerization process using NaOH concentrations ranging from 0.1 M to 6 M on rice husks resulted in broken cellulose hydrogens and increased surface roughness. Wax and oil coatings on the outer layer of fiber cells were eliminated, leading to an improved absorption capacity of up to 20 g/g for marine fuel RMG380 [77]. Anuzyte discovered that the alkaline hydrolysis process using 5% NaOH induced swelling and increased surface area and roughness of moss fibers, resulting in an additional 22% increase in diesel absorption capacity [33]. Additionally, alkali treatment serves as a pretreatment method for other adsorbent transformation techniques. In the study by [24], wheat straw underwent a pretreatment process by extraction with a toluene–ethanol solvent system (2:1, v/v) to remove the wax layer, followed by treatment with 1.3% $NaClO_2$ solution and 10% NaOH solution at 20°C for 10 hours to eliminate hemicellulose and expand the cellulose before acetylation reactions.

3.2.2. Acetylation

The process of acetylation is commonly used to modify cellulose under appropriate reaction conditions, either in industrial production of cellulose acetate or in laboratory synthesis. During acetylation, the –OH bonds in cellulose are replaced by ester [–COO] or acetyl (CO) bonds, resulting in increased hydrophobicity compared to the original hydroxyl groups, which can be hydrolyzed with water [52]. The acetylation process replaces the hydroxyl groups with hydrophobic acetyl groups, which enhances the oil adsorption capacity of cellulose adsorbents, making them easier to recover [78]. Table 3 summarizes the oil adsorption capacity and agents used for some natural adsorbents that have been modified through the acetylation reaction.

According to Table 3, the oil adsorption capacity of the adsorbent significantly increased after acetylation, showing an increase from 67.51% to 195.60% compared to before the acetylation process. Notably, a study by [79] found that the acetylation of corn cobs with acetic anhydride in a solvent–free system in the presence of iodine under mild reaction conditions resulted in excellent oil adsorption capacity both before and after modification, with values of 28.2 g/g and 68.8 g/g, respectively. Similarly, [24] reported that the acetylation process modified the surface properties of wheat straw of wheat straw, creating new sites for oil adsorption and storage. This resulted in an increase in oil adsorption capacity by 177.46% to 195.60% compared to the original values. Corn silk was also acetylated by [49] to improve its oil adsorption capacity using acetic anhydride and N–bromosuccinimide as a catalyst, resulting in a maximum weight percentage gain of 11.45% and an increase in oil adsorption capacity from 72.02% to 77.45%. Acetylated flax fibers were synthesized by [28] using a mixture of liquid acetylation agents and observed an increase in oil adsorption capacity from 13.25 g/g to 17.42 g/g and 24.54 g/g after microwave treatment and acetylation modification, respectively. Finally, [37] found that acetylated sugarcane bagasse had a higher affinity for oil than raw sugarcane bagasse and that acetylation could increase the oil adsorption capacity in salty water by approximately 88.33%.

The disadvantage of acetylation method by acetic anhydride that it produces acetic acid as a byproduct. Acetic acid causes residual odor, loss of material durability due to acid hydrolysis of hemicellulose, and metal corrosion [13].

Table 3. The agents used and oil adsorption capacity of some natural adsorbents modified by acetylation reaction.

Adsorbent	Agent	Oils	Adsorption capacity before modification	Adsorption capacity after modification	The adsorption efficiency increased	Ref.
Bagasse	The process of acetylation with acetic anhydride as a	Crude oil	6 g/g	11.3 g/g	88.33 %	[37]

Adsorbent	**Agent**	**Oils**	**Adsorption capacity before modification**	**Adsorption capacity after modification**	**The adsorption efficiency increased**	**Ref.**
	reactant and NBS as a catalyst is carried out under mild conditions					
		Crude oil	5.4 g/g	9.1 g/g	68.52 %	[37]
Cocoa pods (natural fiber)	Acetylation with acetic anhydride and (1 % of the solvent) N–bromosuccinimide (NBS) as a catalyst	Crude oil	3.97 g/g	6.65 g/g	67.51 %	[80]
Corn Cobs	Acetylation with iodine and acetic anhydride	Oil spill	28.2 g/g	68.8 g/g	143.97 %	[79]
Corn silk fiber	The catalyst used for the acetylation reaction was NBS, which was added at a concentration of 1–3% by weight of the sorbent. Acetic anhydride was used in a solid–to–liquid ratio of 1 g/30 mL.	Tapis crude oil	8.15 g/g	14.02 g/g	72.02 %	[81]
		Arabian crude oil	9.4 g/g	16.68 g/g	77.45 %	[49]
Flax fiber	A blend of 200 mL methylbenzene, 100 mL ethanoic anhydride, and 3 mL perchloric acid as a catalyst for acetylation	Motor oil	13.25 g/g	24.54 g/g	85.21 %	[28]
Oil palm empty fruit bunch (natural fiber)	Acetylation with acetic anhydride and (1 % of the solvent) N–bromosuccinimide (NBS) as a catalyst	Crude oil	3.04 g/g	6.48 g/g	113.16 %	[80]
Peat moss	Acetylation with acetic acid	Oil spill	–	7.6 – 8 g/g	–	[82]
Rice Husks	Acetylation in a solvent free system using NBS	Crude oil	–	10.31 g/g	–	[83]
Sugarcane bagasse	Under mild conditions and without the use of a solvent, an acetylation reaction be carried out with acetic anhydride as the reactant and N–bromosuccinimide as the catalyst	Machine oil	–	18.8 g/g	–	[84]
Wheat straw	Acetic anhydride as an acetylation reagent and NBS as a catalyst	Diesel oil	8.19 ± 0.47 g/g	24.21± 0.76 g/g	195.60 %	[24]
		Diesel oil slick	7.83 ± 1.14 g/g	22.39 ± 0.77 g/g	185.95 %	[24]
		Corn oil	9.23 ± 1.34 g/g	25.61 ± 2.13 g/g	177.46 %	[24]
		Corn oil slick	8.68 ± 1.23 g/g	24.73 ± 1.19 g/g	184.91 %	[24]

3.2.3. Grafting

The process of grafting is a straightforward and efficient approach to modify natural fibers for better oil adsorption by introducing oil–friendly groups onto the fibers, which increases their hydrophobicity [52]. Various reactions, including silylation, etherification, and esterification, can be used for grafting [38]. Esterification grafting, which involves attaching fatty acids onto the adsorbent material's surface, is commonly employed to modify natural oil adsorbents. This section aims to provide a comprehensive description of the esterification grafting method.

a) Esterification

Cellulose esterification is a process of acylation that involves using carboxylic acids as acylating agents, catalyzed by strong acids or activated derivatives like anhydrides or acid chlorides with a base or Lewis acid [85]. The two types of cellulose esterification are chemical esterification [86] and mechanochemical esterification [39]. Chemical esterification of cellulose is often performed using pyridine–acyl chloride or anhydride reactions, which generate hydrochloric acid as a by–product, leading to cellulose degradation and environmental harm [87–88].

According to [89], ionic liquids like EmimOAc could serve as catalysts and reaction media for cellulose hydroxy group esterification. The researchers performed cellulose modification using oleic acid in the TsCl/BmimOAc system through mechanochemical esterification, and a new absorption peak was observed at 1731 cm^{-1} in the Fourier transform infrared spectra, indicating the formation of C=O groups [39]. In another mechanochemical esterification method conducted by [90], cellulose fibers were ground to create microcrystals, and these microcrystalline cellulose particles were then reacted with asymmetric mixed anhydrides (formed from acetic anhydride and oleic acid) in a ball mill. The study found that the ball–milling method was an effective way to modify cellulose powder surfaces with acetic–oleic acid mixtures, and the modified cellulose esters demonstrated improved hydrophobicity and thermal stability when compared to untreated cellulose powder.

Typical fatty acids, including oleic acid, stearic acid, lauric acid, decanoic acid, fatty acid chlorides (oleoyl chloride and octanoate chloride), and N–bromosuccinimide (NBS), can be used as agents for cellulose esterification reactions to enhance oil absorption and hydrophobicity of natural adsorbents [12, 29, 86, 91–94]. Table 4 presents the agents used for cellulose esterification reactions and the oil absorption capacity of the modified adsorbents by esterification reactions.

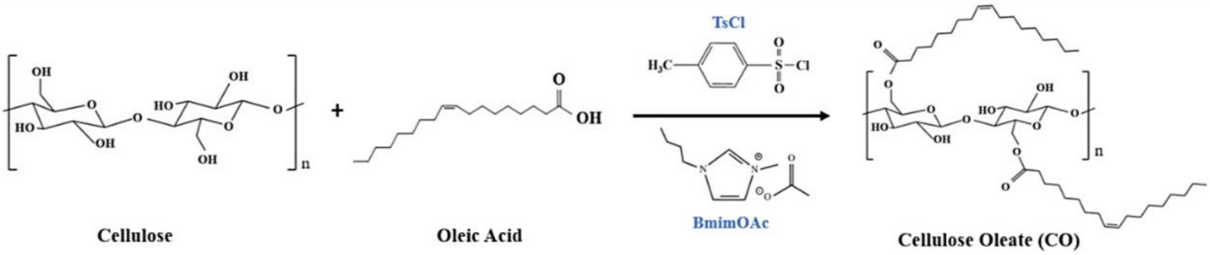

Figure 1. Diagrammatic illustration of the esterification process of cellulose in BmimOAc with oleic acid and TsCl as an activating agent [39].

Table 4. The agents and oil adsorption capacity of the modified adsorbents through esterification reaction.

Adsorbent	Esterifiers	Solvent	Catalyst	Oils	Sorption capacity/ Removal	Ref.
Coconut coir	Fatty acid chloride (oleoyl chloride)	DMaC/LiCl	N–bromosuccinimide (NBS) 1% (v/v)	Engine oil	15.31 g/g	[94]
	Fatty acid chloride (octanoate chloride)	DMaC/LiCl	N–bromosuccinimide (NBS) 1% (v/v)	Engine oil	11.96 g/g	[94]
Coconut husk	Stearic acid	n–hexane	H_2SO_4			[86]

Adsorbent	Esterifiers	Solvent	Catalyst	Oils	Sorption capacity/ Removal	Ref.
Cotton fiber	Stearic acid (C18 fatty acid)	Solvent free system	5.0 g of p–TsCl/ 100 mL of pyridine	Crude oil	35.58 g/g (49 % raw cotton fiber)	[95]
Durial Peel	Stearic acid	n–hexane	H_2SO_4	Diesel oil	0.3780	[29]
Microcrystalline cellulose [MCC] powder	Oleic acid	Solvent free system	TsCl/BmimOAc	–	–	[39]
Pineapple Leaf	Stearic acid	Methanol	Pyridine/p–toluenesulfonyl clorua	Crude oil (100 ppm)	0.108 ± 0.004 g/g	[93]
	Lauric acid	Methanol	Pyridine/p–toluenesulfonyl clorua	Crude oil (100 ppm)	0.138 ± 0.002 g/g	[93]
Sago bark	Stearic acid	Ethyl acetate	CaO	Used engine oil	2.14–2.30 g/g	[96]
Sawdust	Oleic acid	n–hexane	H_2SO_4	Crude oil	6.40	[12]
	Stearic acid	n–hexane	H_2SO_4	Crude oil	5.23	[12]
	Decanoic acid	n–hexane	H_2SO_4	Crude oil	4.23	[12]
Sugarcane bagasse	N–bromosuccinimide used as a catalyst in the acetylation reaction with acetic anhydride	Solvent free system	N–bromosuccinimide (NBS) 1% (v/v)	Machine oil	20.2 g/g	[91]
	Esterification with stearic acid			Engine oil	1.3 – 3.2 g/g	[61]
Grapefruit peel	Stearic acid	n–hexane	H_2SO_4	Diesel oil Lubricating oil	10.93 g/g 10.53 g/g	[92]
	Oleic acid	n–hexane	H_2SO_4	Diesel oil Lubricating oil	11.39 g/g 13.35 g/g	[92]

According to studies by [65, 90], oleic acid, which has a long carbon chain and is hydrophobic, is considered the most suitable agent for cellulose esterification. It has advantages such as flexibility, a wide thermal range, low melting point, easy processing, renewability, and does not produce harmful by–products. Fatty acids with longer carbon chains have better oil absorption capacity. Table 4 shows that [12] found that sawdust modified with oleic acid (6.40 g/g) had higher crude oil absorption capacity than that treated with stearic acid (5.23 g/g) or decanoic acid (4.23 g/g). Similarly, grapefruit peel modified with oleic acid had better absorption capacity for diesel and lubricating oils than that treated with stearic acid [92]. The higher solubility of oleic acid in n–hexane than stearic acid can be a possible explanation for these results. Pineapple leaves modified with stearic acid showed higher oil absorption capacity than lauric acid, which has a shorter carbon chain [93]. In summary, the oil absorption capacity of cellulose modified with fatty acids can be ranked as follows: oleic acid > stearic acid > lauric acid > decanoic acid.

In esterification reactions, powerful acids are typically used as catalysts, with concentrated sulfuric acid being the most used [86, 92]. Other catalysts used include TsCl [96], CaO [96], and N–bromosuccinimide (NBS) 1% (v/v) [94]. The solvent plays a crucial role in dissolving and transforming fatty acids into absorbent materials during esterification reactions. Some of the catalyst/solvent systems used include sulfuric acid/n–hexane [29, 86, 92], TsCl/ethanol [95], CaO/ethyl acetate [96], and N–bromosuccinimide (NBS) 1% (v/v)/DMaC/LiCl [94].

Once the dry adsorbent has undergone pre–treatment, the esterification process can take place. For example, esterifying the –OH group of sawdust with a fatty acid, such as oleic acid, stearic acid, or decanoic acid, or with vegetable oil, such as castor oil or mustard oil. To carry out this process, 1 gram of dry sawdust is mixed with 0.2 grams of the fatty acid or 0.5 grams of vegetable oil in 100 mL of n–hexane, which contains a drop of concentrated H_2SO_4 as a catalyst. The mixture is then refluxed in a Dean–Stark system at a temperature of 65 ± 2°C for 6 hours. After the reaction has occurred, the product is washed with n–hexane multiple times, dried in an oven at 80°C for 24 hours, and then stored for later use. According to equation [1,12], this process will result in the desired reaction.

$$\text{SD-OH} + \text{C}_{17}\text{H}_{33}\text{-COOH} \underset{\text{H}_2\text{O}}{\overset{\text{H}^+}{\Leftrightarrow}} \text{Cell-OCOC}_{17}\text{H}_{33} \qquad (1)$$

The process of esterification can be integrated with the carbonization process. Example carbonizing ogbono shell at 600°C for 4 hours [97], coconut husk at 800°C for 6 hours [86], and rice husk and sawdust at 600°C for 8 hours, followed by esterification with stearic acid [98].

Moreover, the esterification process can be accomplished by initially modifying the material with oleic acid, followed by a second modification using leaving group chemistry with a different fatty acid. This process entails replacing the surface hydroxyl group with a p–toluenesulfonyl group, and then covalently bonding a fatty acid on the sawdust surface through high–temperature (55°C) esterification process (Figure 2).

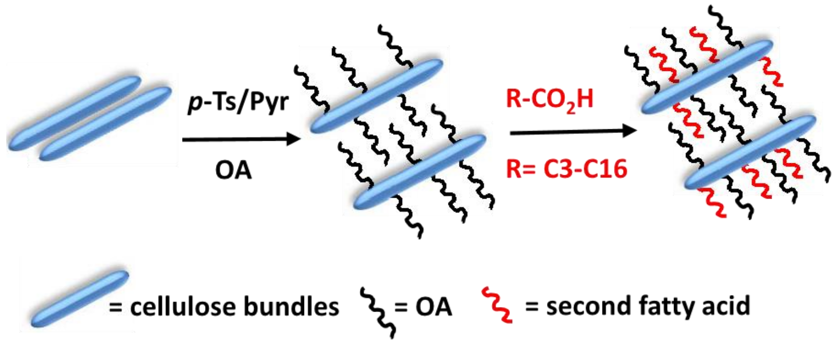

Figure 2. An illustration depicting the hydrophobic modification of cellulose networks in sawdust [99].

Shin prepared a reaction mixture containing p–TsCl and sawdust in pyridine, and added oleic acid while stirring for 6–10 hours at 55–58°C. The resulting material was purified by Soxhlet extraction and vacuum–dried, then modified with different fatty acids at 55°C for 8 hours. Two basic materials (pine/OA–106 and pine/OA–124) were used in the study [99].

b) Other grafting reactions

In the study of [38], they altered cotton fibers by attaching alkyl groups to them. Specifically, the cotton fibers were grafted with silyl ether substituents through solvent–free silylation reactions, replacing dialkyl groups. This conversion transformed the hydroxyl groups on the surface of the cotton into hydrophobic alkylsilyl ether chains. The resulting oil sorption capacity was about 18 g g/g, which was 5 times greater than that of unmodified cotton fibers [38]. In a similar vein, [100], the hydrophobicity of cellulose fibers was increased by grafting them with epoxidized soybean oil through an open–ring polymerization reaction. The modified cellulose film that contained nano ESO polymer particles demonstrated increased smoothness and improved water repellency properties [100]. Meanwhile, [40] utilized corn stalk pith to create a biosorbent with excellent oil sorption and water–repelling properties. They modified the biosorbent by attaching octadecylamine to its structure through a laccase/TEMPO–mediated Schiff–Base reaction (Figure 3). This method resulted in an increase in oil adsorption capacity from 13.24 g/g to 44.25 g/g, accompanied by a decrease in water adsorption capacity from 15.52 g/g to 2.76 g/g.

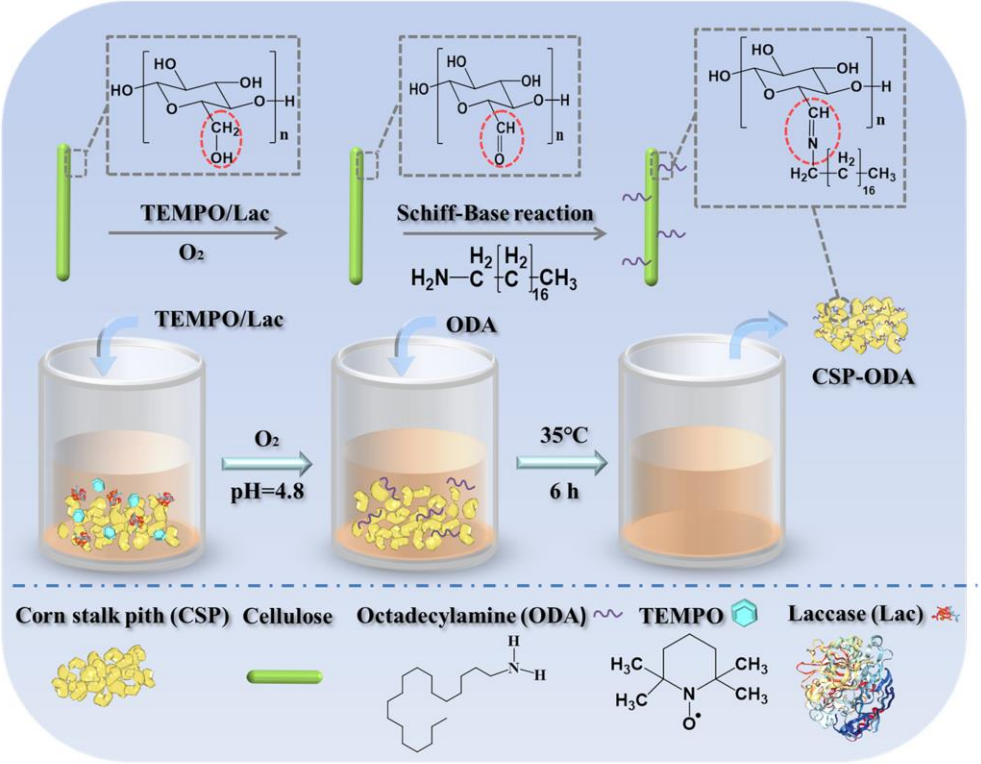

Figure 3. The diagrammatic representation of the steps involved in the surface modification of corn stalk pith [40].

3.2.4. Other chemical modification methods

a) Acidification

Acid treatment is a common form of wet oxidation process used for chemical modification [101]. Typically, acid treatment is used as a preliminary reaction to adjust the processed adsorbent [102], and it involves the use of inorganic acids and other oxidizing agents. The primary goal of acid treatment is to eliminate wax content, increase cellulose hydroxyl groups, and prevent exposure on the surface of the adsorbent [56]. Several chemicals can be used for acid treatment, including dodecylbenzene sulfonic acid [14], and H_2SO_4 [103], which is popularly used to remove wax content and increase the corresponding cellulose hydroxyl groups. Acid treatment has been shown to affect the surface area, surface charge, oil and water affinity, oxygen functional group content, and pore volume, which make it useful for adsorption in wastewater treatment [101].

In study of [4], sawdust was treated with a solution of 0.5 wt.% NaOH and 30% H_2O_2 for 13 hours at room temperature. The pH was then adjusted to 6.5–7.5 by adding drops of 6 mol/L HCl. Acid hydrolysis was employed to decrease the total amount of hydroxyl (–OH) groups in the sawdust, indicating a successful reduction of the alcohol content. On the other hand, [103] performed acid hydrolysis on wheat straw using a 0.045 N H_2SO_4 catalyst with a liquid: solid ratio of 20:l at temperatures ranging from 160–220°C for 0–50 minutes. The process was conducted in a heated reactor with continuous stirring.

b) Aerogel modification

Aerogel is a highly porous solid material with a porosity of up to 99% and a large surface area of over 100 m^2/g that is well–suited for absorbing oil spills. It can be produced from either inorganic or bio–based materials [104]. Using cellulose as a raw material for aerogel production has several advantages over chemically sourced aerogels, including biodegradability and lower cost. Recently, [105] developed cellulose–based aerogels with high porosity and low density by using *Eichhornia crassipes* as the cellulose source and polyvinyl alcohol as the crosslinking agent. These aerogels had oil/solvent uptake capacity ranging from 60.33 to 152.21 g/g, superhydrophobicity (water contact angle of 156.7°), and reusability up to 16 times. Sugarcane bagasse aerogels were also produced by using polyvinyl alcohol as the binder and freeze–drying at –70°C. These aerogels had a very low density (0.016–0.112 g/cm^3), high porosity (91.9–98.9%), and low thermal conductivity. They were superhydrophobic and had a high oil absorption capacity after being coated with methyltrimethoxysilane [106]. Pyrolyzed twisted carbon fiber aerogels developed by [107] showed exceptional oil uptake capacity up to 192 times their own weight and could be reused multiple times without a reduction in absorption capacity.

Nanocellulose–based aerogels are highly porous, lightweight materials that have excellent absorption capacity [108]. Nanocellulose is composed of cellulose nanofibers, nanocrystals, and nanoparticles, which can be isolated using various techniques [109]. The properties of nanocellulose depend on the isolation technique, precursor material, and surface modifications [110]. Researchers often modify the surface of nanocellulose to enhance its properties [111]. In one study, researchers developed aerogels using modified nano–crystalline cellulose (CNC) with 3–triethoxysilyl propyl isocyanate (TEPIC) for oil absorption. The aerogels had a porous structure, low density, small pore diameter, and high specific surface area. All TEPIC–modified CNC aerogels exhibited hydrophobicity, with a water contact angle greater than 130°. The sample modified with 3% TEPIC had the highest absorption capacity for motor oil at 130 ± 7.22 g/g [112]. Another study created a hydrophobic/hydrophilic nanostructured aerogel absorbent by functionalizing cellulose fibers with low surface energy moieties, dissolving and cross–linking the product in an organic solvent, and freeze–drying it. The resulting aerogel had a high oil absorption capacity, comparable to most synthetic oil sorbents [113]. The aerogels had ultralow density, high porosity, and extremely good absorption capabilities for numerous oils and solvents, with selectivity in absorbing marine diesel oil. They were much more effective than commercial absorbent materials and maintained a high absorption capacity for at least 30 cycles, indicating their potential for remediation of oil and chemical spills [114]. A recent study on modification of cellulose and nanocellulose–based aerogels for oil treatment is presented in Table 5.

Table 5. The synthesis of the oil adsorption results of some nanocellulose–based aerogel modifications.

Aerogel composites	Preparation Method	Density	Porosity	Cycle	Water Contact Angle	Absorption Capacity	Ref.
Cellulose–based aerogels from waste biomass (*Eichhornia crassipes*)	Freeze–drying and crosslinking	6.5 mg/cm^3	99.56 %	16	156.7°	60.33 – 152.21 g/g	[105]
Sugarcane bagasse aerogels	Freeze–drying at –70 °C	16 – 112 mg/cm^3	91.9–98.9 %	–	142.2 – 150.05°	25 g/g (Crude oil)	[106]
Cellulose nanofiber (CNF)	Freeze drying and modified via chemical vapor deposition (CVD) of hexadecyltrimethoxylan (HDTMS)	11 – 17.5 mg/cm^3	98.8 – 99.3 %	–	> 90°	78.8 g/g (Motor oil)	[115]
Macroporous cellulose aerogel via dissolving–	Sol–gel and freeze–drying techniques	–	–	80 cycles	154°	~ 25 g/g (Diesel oil)	[116]

Aerogel composites	**Preparation Method**	**Density**	**Porosity**	**Cycle**	**Water Contact Angle**	**Absorption Capacity**	**Ref.**
regenerating raw cotton fiber							
Hybrid coffee–cellulose aerogels from spent coffee grounds	Freeze drying and surface modification	45.8 – 79.7 mg/cm^3	92.1 – 95.5 %	–	124.7 – 139.1°	16 g/g (Motor oil)	[117]
Nano–crystalline cellulose (CNC)		–	–	–	> 130°	130 ± 7.22 g/g (Motor oil)	[112]
Cotton cellulose–based aerogel	Dissolution, cross–linking and freeze–drying	–	–	–	134.7 – 138.7°	40.7 g/g (Crude oil – Balal field oil) 57.0 g/g (Crude oil – Soroush field oil) 47.3 g/g (Crude oil – Azadegan field oil)	[113]
Waste cellulose fibers (Recycled box board and recycled milk–container board)	Freeze–drying	2.9 mg/cm^3	99.81 %	Adsorpticapacity about 71.4 – 81.0 % after 30 cycles	159.0°	42.9 g/g (Marine diesel oil)	[114]
Wool waste fiber aerogel	Direct freeze–drying	4 – 23 mg/cm^3	97.73 – 99.63 %		138.0°	136.2 g/g	[118]
Kapok/microfibrillated cellulose (MFC) aerogels	Simple vacuum freeze–drying and surface modification	5.1 mg/cm^3	99.58 %	–	140.1°	104 – 190.1 g/g	[119]
Kapok/microfibrillated cellulose (MFC) aerogels	Hydrophobic modification and freeze–drying	4.9 mg/cm^3	–	–	147.6°	141.9 g/g [Crude oil]	[120]
Natural cellulose aerogel from rice straw	Freeze–drying	2.2 – 24 mg/cm^3	98.4 – 99.8 %	–	151 ± 7°	170.0 g/g [Crude oil]	[121]

Table 5 displays various cellulose or nanocellulose–based aerogel absorbents derived from natural materials like sugarcane bagasse, cotton fiber, coffee grounds, kapok fiber, and rice straw. Aerogels are modified using techniques like freeze–drying, cross–linking, sol–gel, surface modification, and hydrophobic modification. The aerogels typically have low densities, ranging from 2.2 to 79.7 mg/cm^3, with some examples including natural cellulose aerogel from rice straw (2.2 mg/m^3) [121], waste cellulose fibers from recycled box board and recycled milk–container board (2.9 mg/cm^3) [114], cellulose–based aerogels from waste biomass (*Eichhornia crassipes*) (6.5 mg/cm^3) [105], and kapok/microfibrillated cellulose (MFC) aerogels (4.9–5.1 mg/cm^3) [119–120]. The porosity of modified aerogels ranges from 91.9–99.81 %, with some of the highest porosity aerogels listed as cellulose–based aerogels from waste biomass (*Eichhornia crassipes*) (99.56%) [105], cellulose nanofiber (CNF) (99.3 %) [115], wool waste fiber aerogel (99.63%) [118] and natural cellulose aerogel from rice straw (99.8 %) [121]. Additionally, modified aerogels have high water repellency, with water contact angles ranging from 130° to 159°, and excellent oil absorption capabilities, with the highest oil absorption capacity up to 170.0 g/g [121] and the average absorption capacity of aerogels listed in Table 5 being about 76.8–80.5 g/g.

c) Cationic surfactant

The natural materials are treated with surfactants to remove oil from water, such as wheat straw [123] and barley straw [122]. The addition of cationic surfactants results in an attraction between the surfactant and the positively charged adsorption sites on the adsorbent's surface [124]. One such surfactant, hexadecylpyridinium chloride monohydrate (CPC), has a 16–carbon chain tail attached to a pyridine head group with a permanent +1 charge. Barley straw was treated with sodium hydroxide solution to enhance the formation of carboxyl groups responsible for binding activities before being modified with CPC to enhance its oil adsorption capacity [125]. The Langmuir isotherm showed that the maximum adsorption capacity for standard mineral oil at 25°C was 584.2 mg/g. Desorption experiments in aqueous solution demonstrated that the adsorbent had a strong oil binding affinity, with only 1–2% desorption in 24 hours [122].

3.3. Bioremediation

The immobilization of microorganisms is a commonly used and effective method for biological treatment [126–127] due to its many advantages, including its low cost, simplicity, and minimal effect on microbial activity [128–129]. Compared to free–floating bacteria, immobilized bacteria can avoid unfavorable conditions in the environment, such as predators, native microorganisms, and toxic compounds [130]. Moreover, in an open water system, immobilization also helps to prevent the loss of bacteria due to being washed away [131].

In study of [41], they immobilized bacterial cells from the Vibrio and Acinetobacter genera on cotton fiber surfaces. Both immobilized and planktonic bacteria exhibited a degradation ability of over 60% for saturated hydrocarbons in crude oil, over a pH range of 5.6 to 8.6 and a NaCl concentration of up to 70 g/L. The degradation efficiency of immobilized bacterial cells was found to be approximately 30% higher than that of the planktonic cells. The effectiveness of the MPD–M bacterial group immobilized on polypropylene fibers in treating crude oil at different salinity levels was also demonstrated by [132]. In an experiment of [133], corn stalk pads were supplemented with different amounts of bacteria (1, 3, and 5 mL) and exposed to crude oil for 4 hours at 25 ± 1^{o}C, resulting in higher oil adsorption efficiency compared to a control sample without bacteria. The chemical properties of the hydrocarbon compounds, environmental conditions, and bacterial species influence the biodegradation of oil components, while oxygen molecules are essential for the process. In addition, [134] suggests that providing fertilization with nitrogen and organic phosphorus can significantly boost the growth of hydrocarbon–degrading bacteria. Fertilization with nitrogen and organic phosphorus can significantly enhance the growth of hydrocarbon–degrading bacteria. *Pseudomonas*, *Achromobacter*, *Arthrobacter*, *Micrococcus*, *Nocardia*, *Vibrio*, *Acinetobacter*, *Brevibacterium*, *Corynebacterium*, *Flavobacterium*, *Candida*, *Rhodotorula*, and *Sporobolomyces* are among the important bacterial communities involved in crude oil biodegradation in marine environments [135].

Additionally, it is possible to modify an oil sorbent material by treating it with cellulase solution. To create an oil sorbent material from corn stalk, the raw material is treated in cellulase solution at various temperatures (40 – 60^{o}C) and enzyme loadings (50–200 U/g) for 6 hours. This treatment method reduces the amount of hydrophilic cellulose in the corn stalk and increases its specific surface area. Compared to chemical methods, using cellulase to modify corn stalk is more effective, uses fewer hazardous chemicals, and has the potential to replace non–biodegradable oil sorbents. In systems with only oil, the cellulase–treated corn stalk has sorption capacities of 18.47 g/g for vegetable oil, 16.15 g/g for diesel oil, and 27.23 g/g for crude oil. In systems with crude oil and water, the sorption capacity is 24.98 g/g [21].

The advantages, disadvantages, and limitations of 03 methods in the article are summarized in Table 6.

Table 6. The advantages, disadvantages, and limitations of 03 methods in the article: Chemical modification method, Physical modification methods and Bioremediation.

Method	Advantages	Disadvantages and limitations
Chemical modification method	– High modification efficiency	– May require toxic or hazardous reagents that can cause environmental problems
	– Chemical modification can alter surface properties	– Limited stability in harsh environments, making it difficult to use for long–term remediation
	– Can modify a wide range of materials	– Expensive and time–consuming
Physical modification methods	– Simple and cost–effective	– Limited effectiveness for removing complex pollutants
	– Can use natural or recycled materials	– Modification process may be slow and require long–term maintenance
	– Can modify the physical and chemical properties of adsorbents	– Limited modification capacity for some materials
Bioremediation	– Can use natural, eco–friendly methods	– May require specialized expertise and equipment
	– Can be used for in situ remediation	– Limited effectiveness for removing persistent pollutants
	– Can potentially degrade pollutants into harmless byproducts	– May take a long time to achieve desired results

4. Conclusions

4.1. Conclusions

Lignocellulosic biomass is attracting the attention of researchers because it is a renewable and environmentally friendly natural source. However, the natural material has limitations such as buoyancy and hydrophilic properties due to the existence of hydroxyl groups on the material surface, which can affect its effectiveness and oil absorption in water environment. Substituting hydroxyl groups through chemical reactions can make the water affinity surface of cellulose more hydrophobic. This study presented different methods to enhance the hydrophobic and buoyancy properties of materials, including (1) Physical transformation methods such as mechanical crushing or pressing, heat treatment, and the plasma technique; (2) Chemical modification methods such as mercerization, acetylation, grafting, acidification, aerogel modification, cationic surfactant; and (3) Bioremediation (immobilization of microorganisms). Each method has its own advantages and disadvantages, and selecting the appropriate modifying agent requires considering the characteristics of pollutants and the modification process conditions. Physical and chemical methods are often considered as pre–treatment methods to enhance the potential of functional groups and increase the cellulose content. However, direct chemical modification methods using acetylation or esterification have some drawbacks, including residual odor, material durability loss, cellulose degradation, and environmental harm. Meanwhile, cellulose–based aerogels have high oil absorption capacity that can be up to 170.0 g/g, superhydrophobicity, and potential for reuse more than 80 times. Using cellulase or immobilized microorganisms can also reduce the use of harmful chemicals and replace non–biodegradable oil absorbents.

Some naturally derived adsorbent materials also have high adsorption capacity, but they are not readily available in Vietnam, such as *Calotropis procera* fiber, Kapok fiber, and Silkworm cocoon waste.

4.2. Recommendation for future work

Experimental research in the laboratory will focus on modifying materials from agricultural waste that have not undergone high–level transformation using aerogel and bioremediation methods. The final adsorbent product will be in the form of compressed pellets or freeze–dried aerogel to be suitable for practical conditions.

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