



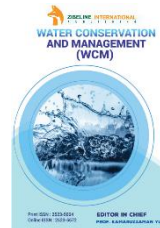
ZIBELINE INTERNATIONAL™

ISSN: 2523-5664 (Print)

ISSN: 2523-5672 (Online)

CODEN: WCMABD

Water Conservation & Management (WCM)

DOI: <http://doi.org/10.26480/wcm.01.2022.06.14>

RESEARCH ARTICLE

HIGHLIGHTS OF OIL TREATMENT TECHNOLOGIES AND RISE OF OIL-ABSORBING MATERIALS IN OCEAN CLEANING STRATEGY

Xuan Phuong Nguyen^{a,*}, Dinh Tuyen Nguyen^a, Van Viet Pham^a, Dinh Tung Vo^{b,*}^a*PATET Research Group, Ho Chi Minh City University of Transport, Ho Chi Minh city, Vietnam*^b*Institute of Engineering, HUTECH University, Ho Chi Minh city, Vietnam***Corresponding author email: phuong@ut.edu.vn; vd.tung@hutech.edu.vn*

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ARTICLE DETAILS

Article History:

Received 05 November 2021

Accepted 24 December 2021

Available online 29 December 2021

ABSTRACT

Nowadays, the problem of oil pollution is becoming a big challenge for the environment. Oil pollution threatens the survival of life on land as well as aquatic life. Aware of the seriousness of oil pollution, there have been many scientific studies on solutions to oil spill treatment. The methods can be mentioned as mechanical treatment, microbiological treatment, chemical treatment, absorbent material treatment. Each method has its advantages and disadvantages. In this short review, recent research activities related to the selection of oil-absorbing absorbents and their application in oil absorption are presented. Then, an extensive list of different oil-absorbing materials from the literature, including polymeric materials, porous inorganic materials, and biomass materials, was provided along with their characteristics. Furthermore, the oil adsorption capacity of such materials for different oils and organic solvents has also been discussed to highlight different factors involved in the selection of adsorbent adsorbents. Oil has been tested to separate oil in oil-water mixtures. Finally, some future trends and prospects for oil-absorbing materials are outlined.

KEYWORDS

oil pollution, cleaning methods, absorbent

1. INTRODUCTION

The world is facing many challenges from population growth and energy consumption demand (Hoang, 2022; Nguyen and Hoang, 2020). The increase in energy demand has led to a dizzying increase in industrial wastewater and domestic wastewater contaminated with oil (Wu et al., 2018; Yong et al., 2017). Oily wastewater is believed to carry organic substances with complex composition, high biotoxicity, and poor biodegradability. Indeed, petroleum is a mixture of hydrocarbon compounds, belonging to the alkanes group, with a very diverse composition (Zhang, 2019; Hoang and Pham, 2018). They may include paraffin compounds, saturated compounds or naphthenes, aromatic hydrocarbons, or aromatics. The number of carbon atoms of the hydrocarbons in the oil is usually between C5 and C60 (Sliwinski, 2017; Yue et al., 2018). Furthermore, the main pollutants in oily wastewater include fats formed from bio-fats and petroleum. These substances can have a very negative impact on both aquatic and terrestrial environments, threatening species in the ecosystem. Another cause of hazardous environments for marine species is the danger of oil spills (Moondra et al., 2021; Toz, 2017). Leaks in the transportation and processing process, incidents caused by oil tanker shipwrecks, have caused terrible pollution to the environment. This pollution has had a great impact on ecosystems, especially marine ecosystems. This pollution can have long-term effects and lead to the extinction of some marine species.

The increase in the density of the shipping fleet along with the increase in the volume of goods transported globally has given rise to many pollution problems from ships (Nguyen, 2020; Hoang and Pham, 2019). In

particular, the pollution of the marine environment by oil leaks and oil spill accidents is of great concern (Toz, 2017). Oil pollution caused by oil tanker accidents or oil pipeline leaks at sea has been threatening the marine ecosystem and environment, especially for the fleet of oil tankers with the huge amount of oil they carry (Chau and Truong, 2021; Pham and Do, 2020). The world's largest oil spill happened in the Gulf of Mexico in 2010 and has had a very bad impact on the marine environment for decades. Studies also show that people living around the area of the oil spill are at risk of many health problems. For example, psychological, biochemical indicators of the body are affected, many symptoms of temporary irritation, cancer risk, and genotoxicity (Figure 1). Gill and Picou monitored the impact of the Exxon Valdez oil spill on people (Gill and Picou, 1998). The data obtained show that a large proportion of people fall into social unrest.

The Prestige oil spill occurred, a group researchers assessed that symptom of lower respiratory tract infection increased with increasing days of exposure (Zock, 2007). Some researchers conducted a study on individuals exposed to oil spills following the Erika oil spill (Amat-Bronnert et al., 2007). The results obtained show that the most common way to absorb fuel gas into the body is through inhalation. Others researchers conducted a study to determine the risk of genotoxicity for food consumers in waters contaminated with polycyclic aromatic hydrocarbons (PAHs) during oil spills (Lemiere et al., 2005). The use of oil-contaminated food contains many dangers to consumers' health. The above studies showed bioaccumulation of oil compounds and their movement into the food chain in oil-contaminated foods, and the results also showed DNA damage. Bro-Rasmussen points out that toxic chemicals

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Access this article online

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DOI:

10.26480/wcm.01.2022.06.14

at low concentrations are not immediately lethal (Bro-Rasmussen, 1996). However, depending on bioavailability and concentrations once they are in the food chain, persistent chemicals can pose hazards to humans in chronic cases. Therefore, it is necessary to study the optimal direction to decontaminate organisms that have come into contact with the oil to ensure safety for consumers.

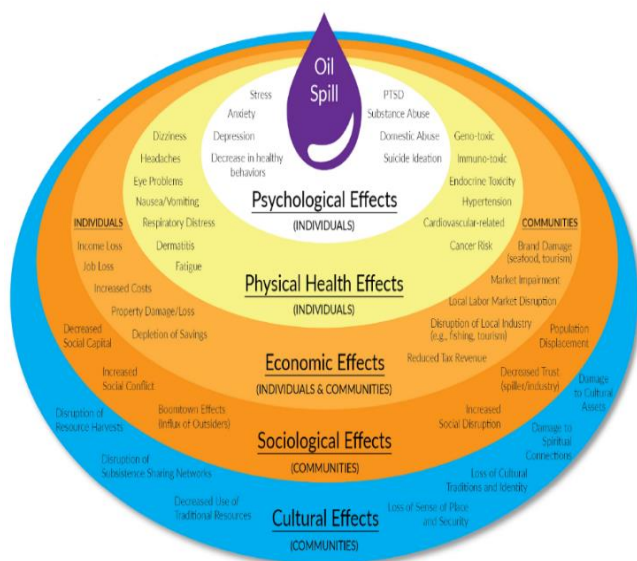


Figure 1: Negative effects of oil spills on human health (Sandifer, 2021)

The size of the oil droplet and the stability of the oil-water mixture were considered to be the determining factors of the separation difficulty of the wastewater. Theories and experiments have classified oil drop sizes into soluble oil drops with diameter less than 0.1 μm , emulsified oil droplet with diameter 0.1–10 μm , dispersed oil droplet with diameter 10–100 μm and floating oil droplet larger than 100 μm in diameter (Tummons et al., 2020). Oil in wastewater and from oil spills is often large and in the form of floating oil. Therefore, oil droplets often tend to stick together and form a film on the surface of the water. Increasing the contact time between oil droplets with water and air will increase the density of the floating oil layers (Dickhout et al., 2018). This makes it easier for the oil to separate from the wastewater. Oil dispersions are easily affected by external conditions such as flow strength. The unstable state in the water of dispersed oils changed their particle size resulting in poor thermal stability. As a result, the dispersed oil can be converted to a floating oil by condensation or converted to an emulsion oil (Hoang and Pham, 2018). The increased density of the emulsion oil means that the surface tension between the oil and the water is decreasing sharply, and at the same time forms a stable film on the water surface where oil and water dispersion is difficult (Long et al., 2017). Therefore, it is difficult for traditional treatment methods to separate oils from water if they are in the form of an oil emulsion. Some of the above-mentioned types of oil-water mixtures can serve as a basis for the search for environmentally and economically efficient oil-contaminated water treatment technologies and materials (Zhao et al., 2021).

Many methods of separation of the oil-water mixture have been proposed such as (1) dispersion and solidification method, (2) in situ combustion method, (3) biological treatment method, (4) mechanical recovery method (Hohl et al., 2017; Lin et al., 2005; Li, 2020; Visco et al., 2021). The main component of the oil is saturated hydrocarbon, so it is difficult to participate in chemical reactions. The use of chemical methods is often an indirect solution through the addition of an oil coagulant. Pollution problems caused by oil leaks can also be solved indirectly by in-situ burning. However, both of the above techniques can cause secondary pollution and waste of resources (Nizetic et al., 2021; Pogorzelski et al., 2020). A safer and cleaner technology is the bioremediation solution. But this technology has low treatment efficiency for floating oil because the conditions for maintaining the growth of organisms are very strict. The mechanical method includes the use of oil skimmers, barriers, and wipers to collect and clean the oil. However, the efficiency of recovery and thorough treatment of contaminated oil is not high (Ghasemi et al., 2020).

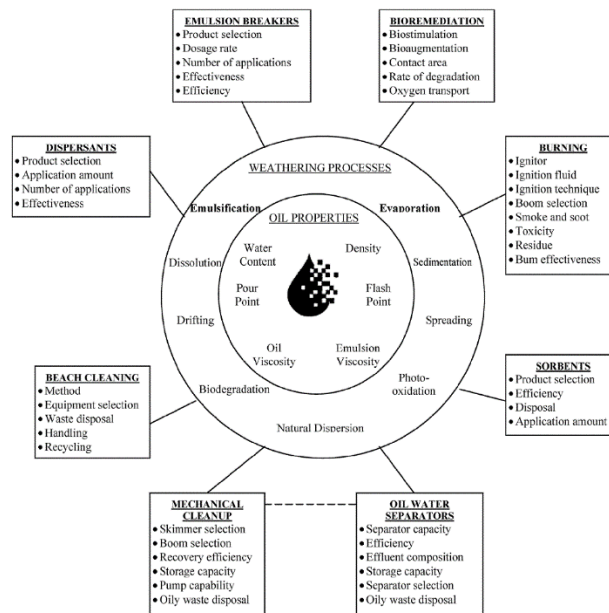


Figure 2: Impact of weather conditions and oil properties on the performance of treatment technologies (Ivshina, 2015)

Currently, the use of oil absorbents is attracting more attention from researchers than most. This technology is considered the most efficient and environmentally friendly, with high economic value (Nguyen et al., 2021). There are many types of traditional materials commonly used to absorb oil in water, including oil-absorbing polymer materials, inorganic materials, biomass materials, etc. Oil-absorbing polymer materials are diverse. They can be natural oil-absorbing materials such as activated carbon, wool fibers, zeolites, rice straw, etc. Besides, they can be synthetic materials such as ester resins, polyurethane, and magnetic materials, and fluoropolymer sponge properties. Recently, oil absorbent materials prepared from lignocellulosic biomass sources have demonstrated high separation efficiency, good recyclability, and outstanding sustainability (Zhang et al., 2020). Materials made from lignocellulosic biomass with fibrous porous structure, large specific surface area, and high surface energy are beneficial for the efficient separation of oil/water mixtures and emulsions (Venkatesh et al., 2021). Furthermore, cellulose-based 3D materials are very flexible and exhibit good tensile properties, good resilience after compression, and corrosion resistance.

Therefore, they can be machined in various forms to meet different industrial production requirements. Each technology to treat contaminated oil in water always has both advantages and disadvantages. It is necessary to consider the characteristics of each technology and tie them to environmental and economic conditions (Kozak, 2008; Nguyen et al., 2021). Current oil spill treatment technologies are strongly influenced by weather conditions. Therefore, when considering the selection of suitable oil spill treatment technology, much attention should be paid to the parameters of weather conditions as shown in Figure 2. In parallel with the prevention of oil spill accidents, anti-leakage of rigs, it is necessary to take measures to handle oil spills on the water surface. Scientists have been trying to find ways to clean up oil-contaminated water. Many solutions have been researched and proposed by scientists. This work presents a brief review of oil spill Clean-up measures and specifically presents the method of oil spill Clean-up with absorbent materials.

2. OIL SPILL CLEAN-UP METHODS

2.1 Mechanical method

As a method of gathering, putting oil in a certain position to avoid oil spreading on a large scale, then combined with other measures to fix the problem (Figure 3). Using a float to keep the oil floating on the water, when the oil is fixed by the float, the next step is to remove the oil from the water surface by combining it with some other methods such as absorption, dispersion, etc. (Wong and Barin, 2003; Wong and Stewart, 2003). Use an oil extractor like a vacuum cleaner, absorbing oil on the surface of the water with attractive affinity or breaking the oil-water physical bond and keeping the oil in a reservoir. This method can only be used for narrow oil slick areas and calm water flows (Fast and Colliander, 1994). Use the burning method with oil spills no more than 3mm thick. This method has been successfully tested in Canada. But this method must be carried out

very carefully. Using a gelling agent to coagulate the oil on the sea surface in the form of a thick film or a mesh, enabling the oil suction machines to recover the oil. Overcoming oil incidents by mechanical methods is considered a prerequisite for responding to oil spills in rivers and seaports to prevent, control, and quickly collect spilled oil on the scene. The two basic means used for oil recovery are the oil sump and the oil recovery device. However, the mechanical method to recover contaminated oil is not thorough (Hoang and Chau, 2018).

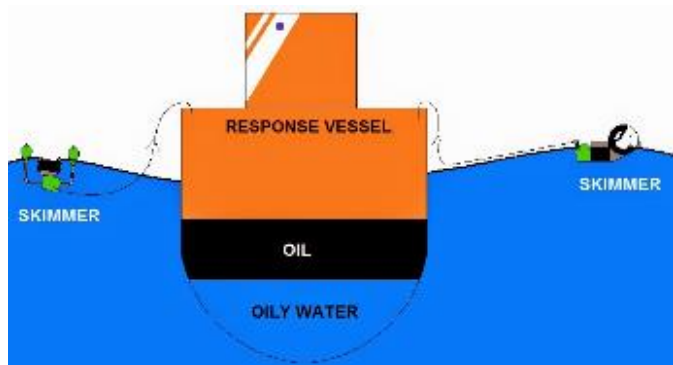


Figure 3: Mechanical oil recovery system by skimmer method (Sedoss Engineering, 2015)

2.2 Biological method

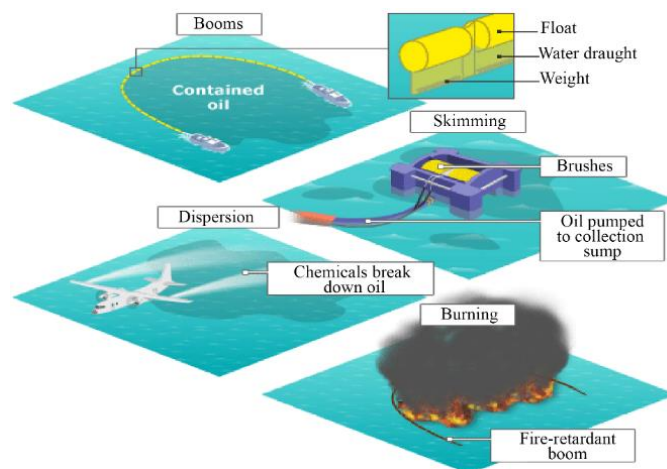
Oil spill clean-up by biotechnology is also a method that is being widely applied and has high efficiency (Feng and Xiao, 2006; Fast and Colliander, 1994). The biological method is a method that uses natural agents or microorganisms (fungi, bacteria, etc.) to promote the decomposition of petroleum hydrocarbons. It is a natural process by microorganisms that break down oil into other substances. The products that can be generated are CO₂, water, or molecules that do not have an adverse effect on the environment. Petroleum is a very special fuel, in their composition mainly straight-chain hydrocarbons (30-35 percent), cyclic hydrocarbons (25-75 percent), and aromatic hydrocarbons (10-15 percent). The chemical components present in petroleum are often difficult to break down. Therefore, the application of biological processes to treat petroleum pollution has very special characteristics. Decomposition, separation of oil spills by natural agents: bacteria or wind and rain washed away, submerged. But because the decomposition process occurs very slowly, in large quantities, the cleaning time is very long and causes many harms before the oil is completely cleaned (Wang et al., 2012). Using microbial agents grown in coal, in oil-absorbing materials, these microorganisms will be the decomposers of oil spills (Hoang, 2022). With a food source of hydrocarbons, at the right humidity, these organisms will grow and biodegrade the oil into harmless substances (Hadiyanto, 2022). However, this method only makes sense for small oil loss incidents at refineries and oil storage facilities. Oil spills on the water in large quantities, this method is not meaningful (Wong and Stewart, 2003).

2.3 Chemical method

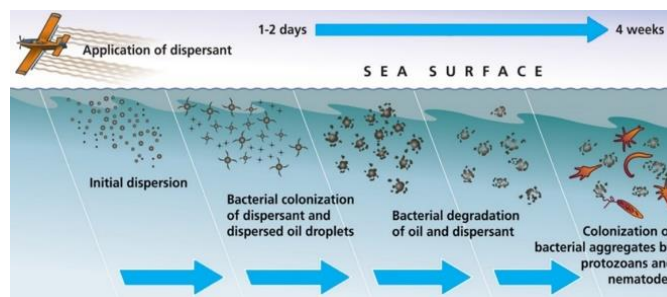
As a method of dealing with oil spills using dispersants, coagulants, oil-absorbing agents, and oil-water emulsifiers. This method is used to clean the oil when it has been or has not been, used for mechanical cleaning, and when the oil has been spilled for a long time. In general, chemicals used are divided into two groups, including dispersants and absorbents (Figure 4) (Aryee, 2012; Katerinap, 2015). Dispersants have the effect of dispersing oil to accelerate the separation of oil from the water surface. They are surfactants, the composition of which includes water and oil acids, and are commonly used in commercial products such as IFO-180, IFO-380, COREXIT-950, etc (Bui and Pham, 2018). The dispersing agent acts as a detergent, which reduces the surface tension between oil and water, creating small oil droplets. This helps speed up the oil's dilution and biodegradation processes and can reduce damage caused by floating oil to certain resources, seabirds, etc. (Chapman et al., 2007). This method has advantages such as: Can be used in extreme weather conditions (such as rough seas, strong winds, and strong waves); It can be used for large areas contaminated with oil, especially where aircraft can be used; in remote areas, this is also an advantage over other methods; dispersants promote oil breakdown by increasing the available surface area of bacteria.

The resulting oil is broken down into a harmless end product; dispersants reduce the ability of the oil to adhere to sediments, shorelines, ships, etc. However, the following limitations still exist: At the time of use, chemical dispersants can cause adverse effects on organisms in contact with dispersants: corals, marine animals, etc. Because of the adverse effects of dispersants on organisms living in the oil Clean-up area and the adverse

effects of dispersants remaining in the water after clean-up, the use of dispersants is limited. The survey results show that the use of dispersants in oil pollution clean-up in EU member countries is lower than in other countries in the world (Chapman et al., 2007). Use chemical decomposition methods to treat oil when the amount of oil spilled is not large, or in some small leaks at the petroleum storage tanks. This method mainly uses catalysts to convert oil under the effect of sunlight. It is possible to use absorbents to absorb oil on the surface of the substance and then use substances with high catalytic ability to oxidize and convert oil (Banerjee et al., 2006).



(a)



(b)

Figure 4: (a) Surface methods used to treat oil spills; (b) Mechanism of dispersion and separation of the oil phase from water by dispersants (Aryee, 2012; Katerinap, 2015).

Chemisorbents are substances that have the ability to absorb oils and separate them from the surface of the water. Oil absorbents allow oil to penetrate gaps in the material or attract oil to the surface of the material. An effective oil adsorbent should have both characteristics: lipophilic and hydrophobic. Oil absorbents have the ability to concentrate and convert liquid phase oils to a semi-solid or solid phase, which can then be conveniently removed from the water and disposed of without significant oil release. This absorbent absorbs spills and spills in any form: whole, partially emulsified or dispersed in water. In particular, the absorbent only absorbs oil, not water. This is one of the most economical and effective methods of oil spill clean-up (Choi and Cloud, 1992; Huang and Lim, 2006; Karana et al., 2011). After conducting oil absorption, it becomes very easy to remove the oil by removing the oil-absorbing structures (Karana et al., 2011). Once the oil absorbents have recovered the oil, they are removed from the water and brought to the surface for Clean-up or cleaning and then reused. Oils extracted from absorbent materials are also properly disposed of or recycled.

Oil sorbents are easy to store, inexpensive, and have no high personnel requirements. They remove the oil by adsorbing oil onto hydrophobic surfaces, thereby preventing the spread, dispersion, or deposition of spilled oil (Gertler et al., 2009; ITOPF, 2011). In most oil adsorbents, there exists a continuum where oil absorption occurs by hydrophobic interactions in the capillary region of the adsorbent, during which oil absorption occurs by capillaries (Hori et al., 2000). The outstanding advantages of the adsorbent are hydrophobicity and lipophilicity, high absorption capacity, and oil is easily recovered by pressing the material. Oil-absorbing materials can be divided into three main groups: natural inorganic substances that absorb oil such as clay, sand, volcanic ash; natural organic matter such as mud, crab hummus, feathers, and some

natural materials containing carbon; synthetic organic material such as polyethylene, porous polyester or polystyrene. The advantage of this type is that it is much more stable than natural organic substances, can be reused, and has high hydrophobicity (Wang et al., 2013). In the chemical method, in addition to using dispersants and absorbents, solidifying agents are also used. Solidifiers are dry materials that react with oil to form a mass that will float to the surface of the water and be recovered with the oil (Hoang, 2018).

3. OIL-ABSORBING MATERIAL

Recently, technology to modify and synthesize new materials to increase hydrophobicity and lipophilicity is being studied. Scientists have fabricated many new materials, and evaluated and compared their oil absorption capacity as well as economic efficiency (Phan et al., 2018). Oil-absorbing materials need to meet a number of technical requirements in order to become commercial products, including High oil absorption capacity, fast absorption rate, and low water absorption; High oil retention during transportation; Good oil recovery and by simple methods; It has good mechanical properties, low density, and high buoyancy in water; It is resistant to common solvents and chemicals, and it does not degrade photochemically; Availability and low cost; ability to be reused numerous times; Meet the standard of oil absorption capacity/1kg of oil-absorbing material (Wang et al., 2021). According to the World Catalog of Oil Spill Response Products, if 1 kg of oil absorbent material absorbs less than 5 kg of oil, the oil absorbent material is classified as poor, uneconomical, and not commercially viable (Lim and Huang, 2007).

If 1 kg of oil absorbent material absorbs about 5-10 kg of oil, the oil absorbent material is classified as good, commercially viable. If 1 kg of oil absorbent material is about 10 kg of oil, the absorbent is classified as good. According to Japanese standards, an oil absorbent material to be accepted as a commercial item must be capable of absorbing at least 6 kg of oil per kg of material (Saito et al., 2003). A group researchers investigated the influence of different properties of oils and absorbent materials on their ability to absorb and retain oil (Husseien et al., 2009). The study summarizes the influence of oil type, material mass, oil film thickness, temperature, and material reusability. Factors that affect the oil absorption capacity of materials include the surface area of the adsorbent, capillary size, shape, the density of fibers constituting the adsorbent, and type of oil (Karana et al., 2011). Capillary size is an important parameter for oil-absorbing materials because it affects the rate at which oil enters and exits the capillary.

The high viscosity of heavy oils significantly affects the capillary penetration of the oil into the pores of the oil-absorbing material (Ansari et al., 2003; Hussien et al., 2009). As the oil becomes more viscous, the pores are obstructed and thus the absorption capacity is reduced. The opposite can be seen in absorbents made from cotton and cotton fibers because thin oil films develop between the fibers, which are more favorable for the absorption of viscous oils (Tanobe et al., 2009; Chau and Le, 2020). Therefore, sorbent structure plays an important role in the oil retention capacity of a sorbent. Absorbent materials with higher porosity have higher initial oil absorption, but low oil retention (Wei et al., 2003). Oil absorption capacity increases with increasing temperatures. This increase may be due to a decrease in oil viscosity at higher temperatures to allow proper penetration into the pores (Husseien et al., 2009). Oils with higher viscosity take longer to penetrate through the pores than less viscous oils (Zaveri, 2004).

The rate of absorption varies with the viscosity of the oil. Light oils are absorbed faster than heavy oils. As the oil film thickness increases, the absorption capacity also increases (Chapman et al., 2007). The weight of the recovered oil can loosen and deform the structure of the absorbent material. When pulled out of the water, the absorbent material can release the oil trapped in its pores. During the recovery of oil absorbents, lighter and less viscous oils can be released from the pores more easily than heavier and more viscous oils. Oil absorbent materials can be classified into three main categories: inorganic materials, natural and modified cellulose materials, and synthetic polymeric materials. In particular, each type of material has certain preeminent properties in oil spill clean-up.

3.1 Material for absorbing inorganic oil

Inorganic materials that absorb oil include clay minerals (vermiculite, diatomite, perlite, quartz sand, crystalline quartz, silica, sodium bicarbonate), amberlite, organic clay minerals, zeolites, fiberglass, graphite, activated carbon, fly ash, etc. A studied the possibility of using modified calcium carbonate powder to clean up oil spills (Arbatan et al., 2011). This powder is treated with fatty acids to give it superhydrophobic, oil-loving properties. The product is suitable for separating oil from

oil/water mixtures and can achieve high separation efficiency. A group researchers used vertically arranged carbon nanotubes to evaluate the oil absorption and recovery by the squeezing method (Fan, 2010). Compared with layered graphite, vertically aligned carbon nanotubes have higher absorption capacity and better recovery efficiency. Research also shows that carbon nanotube porous sheets have quite a high absorption capacity for many oils and organics, up to over 100 g/g (with viscosity in the range of 3–200 shares) (Wei and Mather, 2003).

They still maintain an absorption capacity in the range of 20-40 g/g after 10 cycles of absorption. About 98percent of the oil is recovered by pressing or converted to heat by burning the oil directly inside the foam sheet. Layered graphite can also absorb large amounts of heavy oils in a short period of time. The recovery of heavy oil and graphite was performed by filtration with gentle suction at room temperature (Inagaki et al., 2000). A studied the use of fly ash to clean up oil spills (Karakasi and Moutsatsou, 2010). Two samples of Ca and Si-rich fly ash were modified with sodium oleate to improve buoyancy. In addition, many other materials have also been modified to increase oil absorption, such as peat, silica aigel (Cojocararu et al., 2011; Wang et al., 2012). Inorganic materials generally have the advantage of being readily available and cheap. However, the disadvantage that is difficult to overcome is that it has a high density, cannot be reused, absorbs water, and has poor oil lipophilicity. Therefore, the material has low oil absorption capacity, is difficult to transport and use, and the oil absorption efficiency is much worse than other types of oil-absorbing materials.

3.2 Oil-absorbing materials based on natural cellulose and denaturation

Agricultural products and wastes such as cotton fibers (cotton, cotton, etc.), cotton grasses, peat moss, rice straw, corn cobs, bagasse, sawdust, fiber wood, some types of bark, and many other modified cellulose-based materials that are porous, lightweight, and capable of absorbing oil. Some types, such as milkweed and rice cotton, absorb about 8-20 times more oil than their weight. A group researchers used cotton grass fibers, a by-product of the peat extraction process, like an oil spill absorber (Suni et al., 2004). Cotton grass fibers are pressed into the form of the mat (a type of carpet). Cotton grass mats have 2-3 times higher oil absorption capacity and a 2-3 times faster absorption rate than synthetic absorbents. The separation efficiency of diesel oil on the surface of water reaches more than 99 percent with the ability to absorb about 20 times its weight.

A studied using different plant fibers such as leaf residue mixture (from native plants), sawdust, pineapple fiber, coir, loofah, and silk as steaming materials, crude oil collection (Annunciado et al., 2005). The absorption capacity of these fibers decreases in the following order: silk, pineapple fiber, and sawdust, coir, loofah, leaf residue. Silk has a fast oil absorption rate and a very high oil absorption capacity, up to 85 g/g, has a high degree of hydrophobicity, and has low water absorption. Absorption capacity is 8.5-12 times higher than commercial products from peat. Cotton fiber is also an agricultural product with high oil absorption properties. A group researchers systematically studied the oil absorption capacity and the hydrophilic-hydrophobic properties of rice cotton fibers (Lim and Huang, 2007). The hydrophobic/oil-loving property of rice cotton fibers is due to their waxy surface, while the large cavities of the fiber contribute to increased oil absorption and retention.

The oil absorption capacity of natural fibers mainly depends on the surface void ratio and surface composition. Therefore, the modification of plant fibers is often studied to increase the hydrophobicity and oil absorption capacity of the fibers. A group researchers treated rice cotton fibers with different solvents such as water, HCl, NaOH, NaClO₂, and chloroform (Wang et al., 2012). The results showed that in addition to chloroform, rice cotton fibers treated with solvents increased oil absorption. In addition, the fibers after Clean-up have better reusability, demonstrating a very high potential for oil recovery. Superhydrophobic and lipophilic properties are also achieved by carrying nano-sized silica particles onto rice cotton fibers and then hydrophobically modified by hydrolyzed dodecyltrimethoxysilane. This modified fiber can rapidly absorb diesel and soybean oil with capacities up to 46.9 and 58.8 g/g (Li, 2012). A studied the modification of the sawdust surface by grafting fatty acids (oleic acid, stearic acid, and decanoic acid) and vegetable oils (beaver oil, and rapeseed oil) (Banerjee et al., 2006). Oil absorption is enhanced by surface modification. The results show that oleic acid grafted sawdust has the largest crude oil absorption capacity.

Modification of cellulose fibers by esterification is a fairly common method to increase the hydrophobicity and oil absorption of fibers. Among them, acetylation by acetic anhydride is the most used. The reaction can be carried out under homogenous or heterogeneous conditions with the

participation of different catalysts. A studied the acetylation of cotton fibers under solvent-free conditions using a pyridine catalyst and in dimethyl sulfoxide (DMF) solvent using an N-bromosuccinamide (NBS) catalyst (Wang et al., 2013). Modified fibers have a higher ability to absorb diesel and soybean oils than raw fibers. A group researcher compared the oil absorption capacities of natural and synthetic fibers, including cotton, milkweed, and PP fibers (Run et al., 2004). The fiber-filled columns are evaluated for their ability to absorb high-density oil and diesel oil. Stuffing materials include 100 percent cotton yarn, milkweed yarn, PP yarn, and blends between cotton and PP yarn (70/30), and blends between milkweed yarn and PP yarn (70/30). The porosity of the fiber-filled columns determines the absorption capacity. With a porosity < 0.98, PP fiber gives the largest absorption capacity. When the porosity is high > 0.98, PP fiber gives poor absorption capacity because the pores inside the fiber are large.

In general, the above-mentioned natural oil-absorbing organic materials have the advantages of low cost, environmental friendliness, and biodegradability. Most of the organic materials that absorb the oil of natural origin have a fibrous structure that can be easily processed into fibers and thereby form various products such as buoys, pillows, blankets, towels, etc. Convenient for oil spill response work. However, the disadvantage of this material is its poor buoyancy because of its high density, high hydrophilicity, low lipophilicity, and low recyclability (Karana et al., 2011).

3.3 Oil-absorbing materials based on synthetic polymers

At present, the group of synthetic organic adsorbents is more commonly used due to the relatively fast absorption rate, large absorption volume, high oil absorption selectivity, and the ability to be reused many times, in which Polyolefin-based materials are commonly used. Oil-absorbing synthetic polymers must be hydrophobic polymers. Hydrophobicity and lipophilicity are common features of these polymers. Oil-absorbing materials usually exist in the form of gels, which are soluble but not soluble in oil or non-polar organic solvents. When the material is usually gelled, it is also called "organic gel", that is, the gel is neutralized in organic solvents (Peppas and Harland, 2012). When exposed to a non-polar solvent, the liquid particles diffuse into the structure within the lattice and are retained in it. Depending on the gel strength, the gel has a rubbery, gelatinous, or brick-like appearance if the density is low (Harris, 2002; Restrepo et al., 2006).

Oil-absorbing polymers are composed of polymer molecules that are latticed by reticulating agents that are unsaturated bifunctional monomers to form polymers with a three-dimensional network structure. The characteristics of this material are that it has a porous structure, a flexible three-dimensional network that can shrink before absorbing oil, so its initial volume is usually small, convenient for transportation and storage. In this state, the polymer can be dispersed well on the surface of the water, and when in contact with the oil floating on the water, the oil will diffuse into the spatial network inside the material and be retained in it.

Normally, absorption takes place in three steps. First, the oil molecules diffuse to the outer surface of the material. The oil molecules then migrate from relatively small areas of the outer surface to the pores inside the material. Most of the oil absorption usually occurs in the pores because here the available surface area is very large. Finally, the oil molecules adhere to the inner surface of the pores and are trapped inside the material. The ability of the material to absorb oil is attributed to the effect of Van Der Waals forces between the hydrophilic groups and the floating oil. During stretching, two phenomena occur simultaneously: the penetration of oil into the internal spaces of the polymer structure and the expansion of the polymer chain. The vasodilation depends on the interacting forces within the polymer structure, which are in fact repulsive forces. This repulsive force is the result of different forces such as polar forces, van der Waals forces, molecular forces... The gel properties are determined by the balance between two types of forces: the gravitational force caused by the osmotic pressure of the solvent and other forces present in the gel (Harris, 2002; Restrepo et al., 2006).

Many polymers are made from alkyl acrylate, alkyl methacrylate, and cyclic hydrocarbon derivatives, which are groups with a high affinity for oils. Acrylates are commonly used to absorb organic solvents, oils, chlorinated solvents, or aromatic solvents. The oil absorption capacity of these grafted copolymers depends on many factors: temperature of polymerization, copolymerization, initiator concentration, the molar ratio of monomers, and meshing density in the copolymer, an affinity for oils of groups present in the structure of polymers, etc. Some copolymers of this type, such as lauryl acrylate and butyl methacrylate copolymers, with the

netting agent of ethylene glycol dimethacrylate; copolymers stearyl methacrylate and divinyl benzene have been published in scientific journals, with applications for absorbing organic matter discharged from oil refineries (Shan et al., 2003; Zhou and Cho, 2002; Yang et al., 2005).

The research results showed that the oil absorption capacity is mainly influenced by the lattice density and the lipophilicity of the units in the copolymer (Xu and Xiao, 2011). Copolymers with longer alkyl acrylate chains will have better oil absorption, but the absorption limit of this material is still less than 15g oil/g material. The research on synthesis and application of alkyl acrylate-based copolymers was also conducted (Ji et al., 2011). In this work, the authors studied the influence of reaction parameters, factors such as comonomer, initiator, reticulate, emulsifier, and dispersing agent on the copolymerization process. The obtained copolymer product showed a high oil absorption capacity. The absorption study result for toluene of the copolymer was 17.6 g/g.

Some researchers investigated the oil absorption capacity of copolymers produced by copolymerization of styrene with some long-chain alkyl acrylates (Jang and Kim, 2000). The authors believe that the oil-absorbing role is mainly due to the alkyl group having a great affinity for oil, the oil-absorbing ability of some alkyl groups follows a certain order: stearyl acrylate, lauryl acrylate, 2-ethylhexyl acrylate, butyl acrylates. They have a strong affinity for oil and linear hydrocarbons. Acrylate and methacrylate compounds containing these substituents also have high polymerization and copolymerization ability. The oil absorption capacity of the alkyl methacrylate group in the copolymer is greater than that of the alkyl acrylate (Shan et al., 2003; Zhou and Cho, 2002; Yang et al., 2005). A group researchers synthesized a fibrous butyl methacrylate-lauryl methacrylate copolymer with an oil-absorbing function by heat netting technology after fiber-forming with hydroxyethyl methacrylate reticulate (Feng and Xiao, 2006). The ratio of monomers has a direct effect on the oil absorption capacity of the material, especially the ability to absorb fatty oils. The maximum absorbance of this material for oils is 15 g/g for toluene, 8 g/g for kerosene, 34.75 g/g for chloroform.

A group researchers synthesized a resin with high oil absorption function on the basis of styrene copolymerization with the monomers dodecanol methacrylate and octanol methacrylate in combination with the process of creating chemical grids (with ethylglycol dimethacrylate) and the process of creating physical grids (with polybutadiene meshing agent) to create a type of network recovery that increases oil absorption (Shan et al., 2003). A studied how to synthesize oil-absorbing composites based on the copolymerization of butyl methacrylate and lauryl acrylate in the presence of divinylbenzene reticulum by suspension polymerization (Li et al., 2004). Ferromagnetic particles of Fe₃O₄ surface modified by oleic acid are added to the copolymer composition to improve oil recovery and separation by magnetic separation technique (magnet) without using conventional mechanical separation techniques. The absorption capacity of this polymer composite material is about 20 g/g for chloroform and 10.5 g/g in toluene when the ferromagnetic content is 5 percent. Suda and Panaosak [92] have created absorbent materials for cyclic compounds based on copolymerization of styrene (St) with divinylbenzene (Kiatkamjornwong et al., 2001). When tested for absorption in toluene solvent, the maximum absorption was 7.3 g/g. Some oil-absorbing polymers are synthesized from primary monomers like styrene, alkyl acrylate, and alkyl methacrylate in the presence of a meshing agent (Shan et al., 2003; Jang and Kim, 2000).

However, the study of materials capable of absorbing oil is still a large area of research that needs attention in order to create new materials that can meet the increasing needs of society, and environmental issues are considered very urgent. The absorption capacity of synthetic oil-absorbing materials is much higher than that of other materials, and at the same time, it is easy to transport, easy to use and can be recycled many times. The main drawbacks of these materials are either non-biodegradable or very slow-degrading compared to natural materials.

4. CHALLENGES AND PROSPECTS

Searching for oil-water treatment solutions in wastewater and water contaminated by oil has always been an open issue for researchers and environmentalists. Various techniques for oil in wastewater treatment and oil separation in oil spill waters have been discussed in this paper. However, each treatment technology carries with it both advantages and disadvantages when placed in the context of a technical-environmental-economic synergy. As a short-term solution, mechanical or chemical oil recovery technologies can be effective in separating oil from oil-water mixtures. But they also require huge capital investment and depend a lot on both science and technology and finance. Moreover, the issue of environmental sustainability has always been a bottleneck for both

current mechanical, chemical, and biological methods (Saravasan, 2020). In terms of medium and long-term solutions, changing the energy use strategy is considered the key to the problem. Governments are working hard in the transition to the integration and use of renewable and clean energy sources (Xuan and Viet, 2021). Especially in the current context, when disasters from climate change are challenging all of humanity. The Covid-19 pandemic has forced humanity to change the way we behave and use energy on Earth (Nguyen et al., 2021; Chen, 2021).

A promising trend comes from the recycling, reuse, and recirculation of biomass raw materials and organic solid waste in the circular economy and biorefinery (Hoang, 2021; Moondra et al., 2021; Liu, 2021). Hydrogen fuel produced from refineries is being viewed as ambitious innovations that could help the shipping industry meet its commitments to reduce greenhouse gas emissions and pollutant emissions (Murugesan, 2021; Subramaniam, 2021). Moreover, when maritime transport ships all move to use new renewable fuels such as hydrogen, fuel cells, the concerns about oil spill hazards and oil leakage from ships will disappear (Bui et al., 2021; Filippov and Yaroslavtsev, 2021). Finally, with the current huge biomass reserves, Vietnam is considered one of the countries with advantages in converting sustainable forms of energy from biomass (Nguyen et al., 2021; Jiang et al., 2021). Furthermore, oil and heavy metal absorbent materials produced from biomass will also be value-added products contributing to the sustainable energy development strategy (Yue et al., 2022). The

development of new generations of absorbent materials from lignocellulosic biomass can create breakthroughs in water purification in the near future (Wang et al., 2021).

5. CONCLUSIONS

Oil spills often occur during activities of searching, exploring, exploiting, transporting, processing, distributing, storing oil and gas and petroleum products adversely affecting the ecosystem and causing great damage to economic activities. Recently, the technology of modifying and synthesizing new materials to increase hydrophobicity and oil absorption has been focused on research. Many new materials have been fabricated, evaluated, and compared in terms of oil absorption capacity as well as economic efficiency. Oil-absorbing materials based on natural, environmentally friendly, and easily degradable polymers are being studied by scientists. Natural-origin organic oil-absorbing materials have the advantages of low cost, reusability, environmental friendliness, and biodegradability. Most natural organic oil-absorbing materials have a fibrous structure, so they can be easily processed into fibers and thereby into various products such as buoys, pillows, blankets, towels, etc., which are convenient for oil spill response work. Besides, they have some disadvantages, such as poor buoyancy because of their high density, high hydrophilicity, and low lipophilicity, so they need to be modified to improve oil absorption.

Table 1: Biomass-based sorbent used for the degradation and removal of oils

Sorbent	Method	Oils	Sorption Capacity/Removal	Ref.
Rice Husks	White Ash (Pyrolysis)	Diesel	5.02 g/g	(Doshi et al., 2018; Vlaev et al., 2011)
		Crude Oil	6.22 g/g	
	Black Ash (Pyrolysis under N ₂ or inert atmosphere)	Diesel	2.78 g/g	
		Crude Oil	2.98 g/g	
Rice Husks	Pyrolysis (Carbonized) at 480 °C	Gasoline	3.7 kg/kg	(Doshi et al., 2018; Angelova et al., 2011)
		Diesel	5.5kg/kg	
		Light crude oil	6.0 kg/kg	
		Motor oil	7.5kg/kg	
		Heavy Crude oil	9.2 kg/kg	
Rice Husks	Alkaline treatment	Marine diesel	19 g/g	(Doshi et al., 2018; Bazargan et al., 2014)
Silkworm cocoon waste	Drying (110 °C), Cutting, and Milling	Motor oil	42-52g/g (51%)	(Doshi et al., 2018; Moriwaki, 2009)
		Vegetable oil	37-60g/g (54%)	
Banana skins	Drying (sunlight and 70 °C), crushing and Sieving	Crude oil	5-7 g/g	(Doshi et al., 2018; El-Din et al., 2018)
Potato peel	Drying (70 °C) and Crushing	Waste lubricating oil	2.15 g/g	(Doshi et al., 2018; Tontiwachwuth et al., 2016)
Luffa (an agricultural waste)	Cutting, Sieving, washing, and drying (105°)	Diesel Oil	>85%	(Doshi et al., 2018; Abdelwahab, 2016)
		Heavy Crude Oil	>50%	
M.Rouxii (32.7% Chitosan)	Shaking flask method, Autoclave and drying	Mineral oil	77.2 mg/g (77%)	(Doshi et al., 2018; Srinivasan and Viraraghan, 2010)
		Vegetable oil	95.5 mg/g (93%)	
		Cutting Oil	84 mg/g (84%)	
A.Coerulea(10.4% Chitosan)		Mineral oil	72%	
		Vegetable oil	91%	
		Cutting Oil	80%	
Hybrid peel waste (Banana skins + orange peel)	Hybridization of peels with NaOH and drying at 70 °C	Lubricant oil	38%	(Doshi et al., 2018; Abdullah et al., 2016)
		Petrol oil	32%	
Walnut shells	Media	Mineral Oil	0.56 g/g	(Doshi et al., 2018; Srinivasan and Viraraghavan, 2008)
		Vegetable oil	0.58 g/g	
		DoALL Bright-Edge Oil	0.74 g/g	

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