Review Article

Innovations in Oil Spill Clean-up Techniques

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Abstract

As the world population continues to grow, so do the production, refining, and distribution of crude oil or petroleum. During the course of processing and transporting oil and oil based products across different regions of a country, continent, or the world, a phenomenon known as oil spill arises. Oil spill pollutes the environment-mostly land and aquatic habitats with contaminants capable of harming man, crops, and animals. In order to clean up oil spills, a number of conventional methods are employed. These methods are often considered inefficient. In this paper, we reviewed a number of innovative approaches applicable for use in effectively and efficiently cleaning up oil spills. These techniques are either currently part of ongoing researches or are suitable for small scale applications.

Keywords: Bioremediation, Crude Oil, Nanotechnology, Oil Spill, Sorbent, Techniques

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Introduction

Crude oil is a dark brown to black, naturally occurring viscous rock fluid obtained from the thermal and biological decay of plant and animal remains, in a process spanning millions of years [1]. Since crude oil was first discovered in commercial quantities, its production, processing, and worldwide distribution have been ever increasing. Crude oil is processed into a wide range of consumer and industrial products via a technique regarded as fractional distillation. This technique gives universally significant products such as gasoline, kerosene, petro-diesel and lubricating oil, and feed stocks or raw materials for petrochemical industries, such as ethylene and butylenes.

The production, refining, and distribution of crude oil and products based on it involves a series of complex processes and procedures. A phenomenon referred to as "oil spill or spillage" usually arises in the course of undertaking these processes. Oil spill is a form of environmental pollution defined as the release of oil and oil based products into the environment in such amounts which have the capability of harming man, animals, crops, and marine and terrestrial habitats. Oil spill occurrence is often as a result of natural or man-made disasters, human negligence, and acts of terror by certain individuals. It can also be caused by accidents when oil is being loaded or discharged from tankers at seaports, oil tanker accidents at sea, oil discharge in areas near oil wells as well as at oil storage facilities, equipment failure, land runoff, vessels and pipeline accidents, wars, sabotage, oil theft, and natural disasters during the production, transportation, storage and use of oil [1, 5, 10, 7, 29, 2, 18].

Transportation of crude oil, as well as the refined products from the production point to the consumer bears great degree of complexity. There is hence continuous spill of oil which results in devastating environmental consequences. Serious concerns have thus been raised widely on the possible environmental impact of extreme oil spills [2, 18, 33, 25, 12]. Oil spills can as well affect negatively the vast marine ecology which includes sea mammals, algae, sea grass and coral. They also results to serious health hazards to human beings, mostly those living in nearby coastal zones. Furthermore, enormous economic losses are incurred by oil companies and origin states as a result of oil spill disasters.

Several solutions have been adopted in dealing with the problems of oil spill. These remediation technologies for marine oil spills are basically classified into three to four categories which include physical/mechanical, chemical, thermal, and biological remediation technologies. There involves a number of techniques such as the use of mechanical means like skimmers, pumps, booms, and mechanical separators; using microorganisms to break down the oil; adopting the use of chemical dispersants like detergents to break large oil slicks into tiny droplets; in-situ burning of spilled oil; and application of sorbents to remove oil from water through absorption and/or adsorption (**Figure 1**) [35, 25, 34, 10]. Each technique comes with its advantages and disadvantages, and the choice of a particular technique depends on several factors such as type of oil spilled, temperature, wind speed and direction, and amongst others topographic location of the oil spill [10].

However, due to the massive oil spills, conventional techniques are not adequate to solve this problem. There has been more than ever before increased attention towards developing and implementing innovative technologies for the removal of oil contaminants in the environment [25, 33]. In recent years, there has been emergence of potential sources of innovative solutions in oil spill cleanup. Most of such techniques are considered to be green ways to clean up oil spills, although a large part of them are still in the process of research, have only been proposed, or are only in use in small scale. This paper is aimed at reviewing the various innovative technologies available for cleaning up oil spills.

The paper would provide concise but detailed explanation about the mechanism of the various innovative techniques, while also in certain cases would report research findings (on innovation in oil spill cleanup) of scientists who have had their works published in journals.

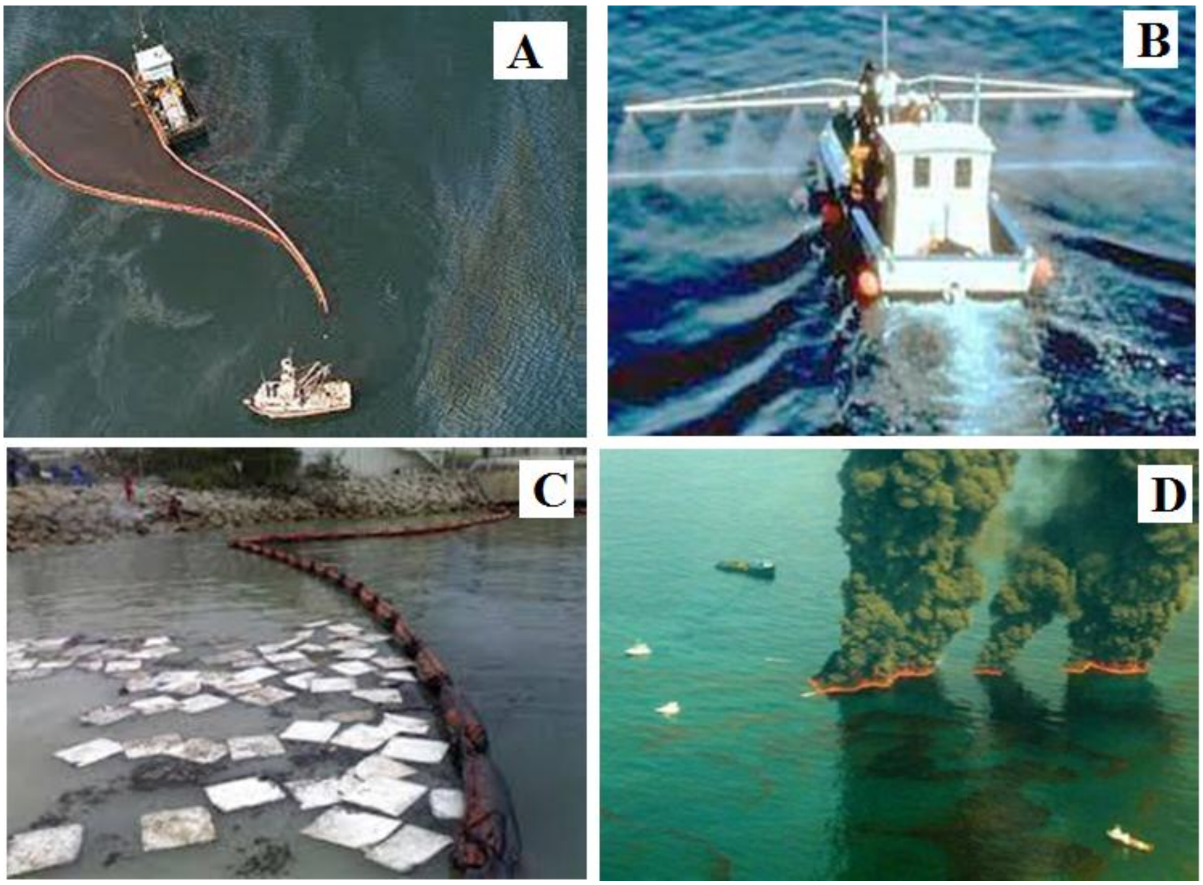

Figure 1 Conventional methods of oil spill cleanup: (A) oil containment with booms; (B) a vessel spraying chemical dispersants; (C) using adsorbents; (D) in-situ burning [10, 25, 34, 35]

Innovative Materials
Sorbent Materials

Karan *et al* defined a sorbent as "an insoluble material or mixture of materials used to recover liquids through the mechanisms of absorption or adsorption, or both" [18]. Sorption is a popular technique applied in order to treat spillage of oil. It is a very effective technique which contributes to the total clean-up of spilled oil in ambient environments. Common sorbents include straw, chalk powder, micas, ekoperl, sawdust, foams of polyether or polyurethane, nylon fibers, and polyethylene [16].

Investigation is being carried out to develop hydrophobic, biodegradable, and cellulosic sorbent materials for cleaning up oil spills. Large amounts of natural sorbents and a wide variety of natural organic products like wood, cotton, wool fibers, rice straw, peat moss, and amongst others corn cob have for a while now attracted attention for applications in oil spill cleanup [11].

It has been demonstrated by Adebajo and Frost that the 4-dimethylaminopyridine (DMAP) catalyzed acetylation of raw cotton samples with acetic anhydride without solvent, prepares a sorbent with enormous hydrophobic characteristic and high oil sorption capacity [11]. They have predicted a clear future through the potential usage of raw cotton fibers acetylated into hydrophobic sorbent materials for cleaning up oil spills.

There are millions of tons of paper industry waste and these could end up as reusable green solution to clean or mop up oil. With annual paper mill sludges produced in the European Union (EU) estimated at 18 million tons (most of which is disposed by burning), the EU organization had a few years back funded a research project whose objective was to convert the waste sludge into a highly absorbent material with capability of cleaning up oil spills [32]. This new absorbent is capable of absorbing any oil or fluid spilled on hard or water surfaces. So many successful tests have already been carried out using this innovative sorbent, and several countries have begun their own production of the paper mill sludge absorbent. The effectiveness of paper mill sludge in absorbing large amounts of oil from water was also successfully assessed by Likon and Saarela, in which the absorbent material was perceived as having a very efficient lifecycle [23]. This is because after carrying out a controlled incineration, the used paper mill sludge sorbent was converted into an inorganic, inert meta-kaolin product which could be further used as hydrophilic sorbent material.

Hussein et al. have studied the oil sorption in aqueous media by low quality grade cotton, focusing on environmental acceptability and ease of recovery of the material. The sorption capacity (g oil / g fiber) and water pickup of low grade cotton fibers were compared with a pad containing low grade cotton fibers. It was found that the low grade cotton fibers oil sorption capacity depended on a number of factors such as temperature conditions. Results showed that loose fibers have "a higher sorption capacity compared to the pad containing the low grade cotton fibers due to the higher surface area of the loose fibers [16]." They have concluded based on various other results obtained that the pad and loose cotton fibers possess excellent potential as commercial sorbents for oil spillage cleanup.

The claim of cotton being an abundant, low cost, sustainable, biodegradable, and effective oil spill sorbent has been furthered by the research carried out by Ramkumar et al. (2013) [31]. The oil sorption properties of low micronaire cotton were studied by them. Low micronaire cotton refers to a form of unprocessed cotton with relatively low commercial value. An estimated 30 pounds of crude oil was reportedly sopped up and held by each pound of the material. Both absorption and adsorption were ways employed by the cotton fibers in taking up oil. It was concluded that raw cotton possesses high crude oil sorption capacity and is a green or environmentally friendly sorbent for cleaning up oil spills.

Innovative clay based technologies are being developed to remove and treat oil spills and leaks as reported by Frost *et al* [14]. The clay is being designed to treat common and normally small oil spills of various types, and to clean up larger catastrophic spills in land and aquatic environments. These technologies are based upon sorption of oil into clay material which has been modified to be oleophilic (oil attracting). In order to obtain an efficient oil sorption material, hydrophobicity is normally induced in natural clays like bentonites (which contain metal cations that impart hydrophilic character) [30]. This modification is achieved by treatment with quaternary amines [19].

Frost *et al*. synthesized organo-clays via the ion exchange of sodium in Wyoming Na-Montmorillonite (SWy-2-MMT) with three given surfactants: didecyldimethylammonium bromide [DDDMA ($C_{22}H_{48}BrN$)], octadecyltrimethyl ammonium bromide [ODTMA ($C_{21}H_{46}NBr$)] and di (hydrogenated tallow) dimethylammonium chloride (tallow). The ability of these organo-clays to adsorb hydrocarbon was then tested [13]. The effectiveness of these sorbent materials was evaluated for a selected range of petroleum products with the likeliest possibility of getting involved in land-based oil spills. High petroleum sorption capacity was obtained using the organo-clays, with greater adsorption reported as the hydrocarbon chains in the surfactant increased. Certain analysis carried out also revealed that organo-clays ranked above any of the competing sorbents, including commercial benchmark materials. The research team has found out that the use of organo-clays in oil spill cleanup is highly feasible, due to the high hydrocarbon sorption and retention capacities, and hydrophobicity of the material. It was concluded that although organo-clays have so many desirable properties, the negative connotations of using them in cleaning up oil spill include their cost and problem of recyclability.

To combat oil spills, a technology centered on a cross-linked polyolefin terpolymer was developed by Yuan and Chung (2012) [38]. The polymer contained styrene, 1-octene, and divinylbenzene units. The technology had superabsorbent characteristics due to the presence of aliphatic and aromatic side chains, giving the polymer similar solubility parameters with the hydrocarbon components present in crude oil. Oil was rapidly absorbed by the terpolymer which swelled to reach a capacity of about 45 times its original weight. The oil-swelled gel was eventually recovered, containing approximately 98% of oil and 2% of the polymer. It has been deduced from this study that this technique is suitable for oil spill cleanup processes due to its economic value, leaving behind no waste and no pollutant.

Lastly, oil could be sop up from the environment using complete, parts, or products of plants and animals. As reported in the *Inhabitat*, first a mixture of mushroom and hair is used to fabricate mats which offer a totally organic and effective way to sop up oil in water [17]. This uncommon but quite effective technique was employed in the cleanup operations of the Cosco Busan oil spill of 2007 [17]. Second spilled oil is soaked up with hyper absorbent peat moss [17]. The method in which this works is that the super absorbent natural peat moss is scattered on spills to absorb oil, after which it is scooped right out of the water along with the oil. **Figure 2** shows these animal/plant based sorbents.

Figure 2 (a) Mat made from a mixture of mushroom & hair, and (b) hyper absorbent peat moss [17]

Bioremediation Technologies

Biological method of oil spill cleanup involves using living organisms or by-products of living organisms to assist in cleaning up the mess left over after oil has been spilled, either in the marine or land environment. The most common biological method in oil spill cleanup is bioremediation, a technique which could simply be described as the use of organisms to cause the breakdown and further detoxification of dangerous chemicals or contaminants present in the environment [25].

Bioremediation takes place in different forms [1, 25]. There is bioaugmentation which involves the addition of bacterial culture to the contaminated soil or water [1]. In other words, it is the addition of microorganisms which have the capability of bio-transforming or biodegrading given contaminants [25]. Biostimulation on the other hand involves the addition to a contaminated environment of microbial nutrients which are needed for increase in microbial activities of indigenous flora and fauna [11]. The nutrients added are mostly major nutrients such as carbon, oxygen, nitrogen, and phosphorus which boost the number and/or activity of naturally occurring microorganisms readily available for bioremediation [3, 25].

There is great amount of attractiveness in biological alternatives or biodegradation of oil substances given the enormous negative consequences of some physicochemical methods of oil degradation, as compared by Bello [5]. Microorganisms aid in breaking chemical bonds and transferring electrons away from organic compounds in an oxidation-reduction reaction process [3, 25]. Although bioremediation is a very slow process which requires weeks and months for effective cleanup, it is a universal method of oil spill cleanup due to its cost effectiveness and less requirement of high skilled technology and technical know-how. In addition, minimal environmental impact is expected from bioremediation because it is a natural process.

In 1972, a number of the first studies carried out on bioremediation were the employment of microbes to successfully impact oil degradation in sea water, as seen in Lu et al report [25]. There have since been more extensive studies and applications of bioremediation. Several researches are therefore also ongoing on developing better, and more efficient and effective bioremediation techniques to cleanup spills [33].

According to Dennis and Osatohamhen, Poultry droppings biostimulant was used in remediating a crude oil contaminated soil in a study carried out by Dennis and Osatohamhen, in order to investigate the impact of the age of the poultry manure particulate on the contaminated soil [1]. The results obtained by the duo revealed that poultry manure particulates are very effective in the amendment of crude oil polluted soil, suggesting that lower aged particulates produced the best outcome. In conclusion, the study has shown the significant effect of poultry manure particulates in reducing to tolerable limits the amount of hydrocarbons in the soil. Part of the findings made by Dennis and Osatohamhen are shown on **Tables 1** and **2**, which represent total amounts of hydrocarbons remaining in soil after the bioremediation process and percentage reduction in hydrocarbon content respectively.

A comparative mesocosm trial of bioremediation of contaminated soil of a petroleum refinery was undertaken by Couto *et al*. The experiment (spanning a period of nine months) was carried out inside the refinery area so as to achieve reproducible results. Oil contaminated soil samples were packed in different containers and treated using a number of techniques. Results showed that petroleum hydrocarbons' degradation was most efficient using bioaugmentation in the presence of nutrients and surfactant amendments. Total degradation of petroleum hydrocarbons was higher at the soil surface due to the combination of sufficient dioxygen which favors aerobic degradation, with solar radiation which is required in order to produce strong photochemical oxidants like ozone. The study was concluded with the statement that factors such as revolving of the soil (like tillage to expose different parts of contaminated soil to oxidative atmosphere); bioaugmentation, and nutrients and surfactant amendments may be combined in order to achieve a cost effective and improved petroleum hydrocarbon degradation rate [9].

Table 1 Total Hydrocarbon Content of crude oil contaminated soil bioremediated with poultry manure of different ages [1]

Sample	Age of Sample (days)	Time (Weeks)								
		0	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
A	3	84.10	63.90	48.90	34.80	25.30	17.40	10.20	4.30	0.70
B	28	82.90	65.80	49.20	36.80	26.70	18.90	11.10	5.40	1.60
C	42	84.10	63.50	48.20	33.90	25.70	18.30	11.40	5.20	1.10
D	126	83.40	61.80	47.30	34.20	26.40	19.90	12.10	6.60	1.90

Table 2 Percentage reduction in Total Hydrocarbon Content (THC) of soil bioremediated with poultry manure particulates of different ages

Sample	THC reduction (%)
A	99.17
B	98.07
C	98.69
D	97.72

It has been demonstrated in a study by Genovese et al. that in order to efficiently clean petroleum-contaminated anaerobic sediments, an *in situ* oxygenation or aeration (exposing the sediment to aerobic environment) can be carried out to stimulate their self-cleaning potential due to the reawakening of autochthonous aerobic obligate marine hydrocarbonoclastic bacteria (OMHCB) (2014). A 98% success (in the removal of petroleum hydrocarbon contaminants) was obtained in their study at the end of the aeration period.

In addition to already identified methods, Bello indicated that the use of biosurfactants could as well serve as an effective bioremediation technique [5]. Biosurfactants simply put are surface-active-agents produced by micro-organisms. These agents aid the emulsification of hydrocarbon in growth medium, reducing surface and interfacial tensions. There are wide reports about the ability of biosurfactants to emulsify hydrocarbon-water mixtures.

Nanomaterials/Nanotechnology

Bhawana and Fulekar defined nanotechnology as "the design, characterization, production, and application of structures, devices and systems by controlling the shape and size at the nanometer scale (10^{-9})" [6]. A nanoparticle follows suit as a microscopic particle whose size is measured in nanometers (nm). It is defined as a particle with at least one dimension less than 100nm [6]. The usage of nanostructures, materials and devices of their like for the remediation of oil contaminated soil and water has gained so much popularity in recent years due to the great advantages which nanoparticles bear over conventional methods [6, 20, 26, 28]. This is as a result of their much larger surface area on a mass basis. A characteristic structure and certain electronic properties also make nanoparticles excellent adsorbents of most common land and aquatic pollutants.

Highly dense order cuprous oxide (Cu_2O) nanorods on phosphor-copper mesh were prepared from simple aqueous solution-immersion process, and the surface of the Cu_2O nanostructures was modified with 1-dodecanethiol which impacted superhydrophobicity and superoleophilicity, as reported by Kong [22]. This structure was used to make a miniboat which floated freely on water surface and *in situ* collected oil from the water surface. The demonstration has shown its promise for application in oil-water separation and offshore oil spill cleanup.

A tea-bag shaped nanocomposite system was developed by Avila et al. for oil spill cleanup using the nanotechnology approach [4]. The nanocomposite system combined superhydrophobic nanomembranes and oleophilic graphite nanoplatelets. Results obtained have shown an average oil sorption capacity peak of 32 g/g. The tea-bag shape design ensures that the system possesses high efficiency and easy recyclability.

Oil spillage on seawater also requires the development of oil sorbents with the following characteristics: high sorption capacity, high selectivity, low cost, and scalable fabrication. Lin et al. reported the use of nanoporous polystyrene (PS) fibers which were prepared through a one-step electrospinning process as oil sorbents for oil spill cleanup [24]. The nano-sorbent had an oleophilic-hydrophobic character. It had highly porous structures and a motor oil sorption capacity of 113.87 g/g was reported. Comparative analysis showed that these sorbents had 3-4 times more sorption capacity than other common sorbents.

A low density, highly porous, mechanically stable, highly hydrophobic and superoleophilic carbon nanofiber (CNF) aerogel was reported by Wu as an efficient sorbent for cleaning up oil spillage [36]. The aerogel has high sorption capacity and is easily recycled. Its ability to operate over a wide range of temperature makes the CNF aerogel highly suitable for use in cleanups under harsh conditions.

It has also been discovered, as reported by Knapik and Stopa, that silica nanopowders have great suitability for use in permanently removing oil spills from the surface of water [21]. These nanopowders which are based on nano-silica prepared in a laboratory experiment exhibited 95 wt % separation efficiency. Suggestion that further investigations should be carried out on these nanopowders paves way for optimizing the nanosorbent properties of the powders and the development of technologies for their large scale usage.

Nanowires made of potassium manganese oxide have not long ago been developed to clean up oil in water without restricting oil recovery, as reported by Yuan et al [37]. The presence as well of certain chemical contaminants in water after clean-up operations can be addressed through the usage of carbon nanotubes (CNT) [6]. These nanomaterials serve as powerful adsorbents for a vast amount of organic compounds left ove in aquatic environments, such as polynuclear aromatic hydrocarbons (PAHs), chlorobenzenes and other methane and ethane-based compounds. Reports also have it that nanostructure ZnO films have the capability of simultaneously detecting and degrading organic compounds in water [6]. The many different techniques which have been developed using nanoparticles for a variety of approaches are summarized in **Figure 3**.

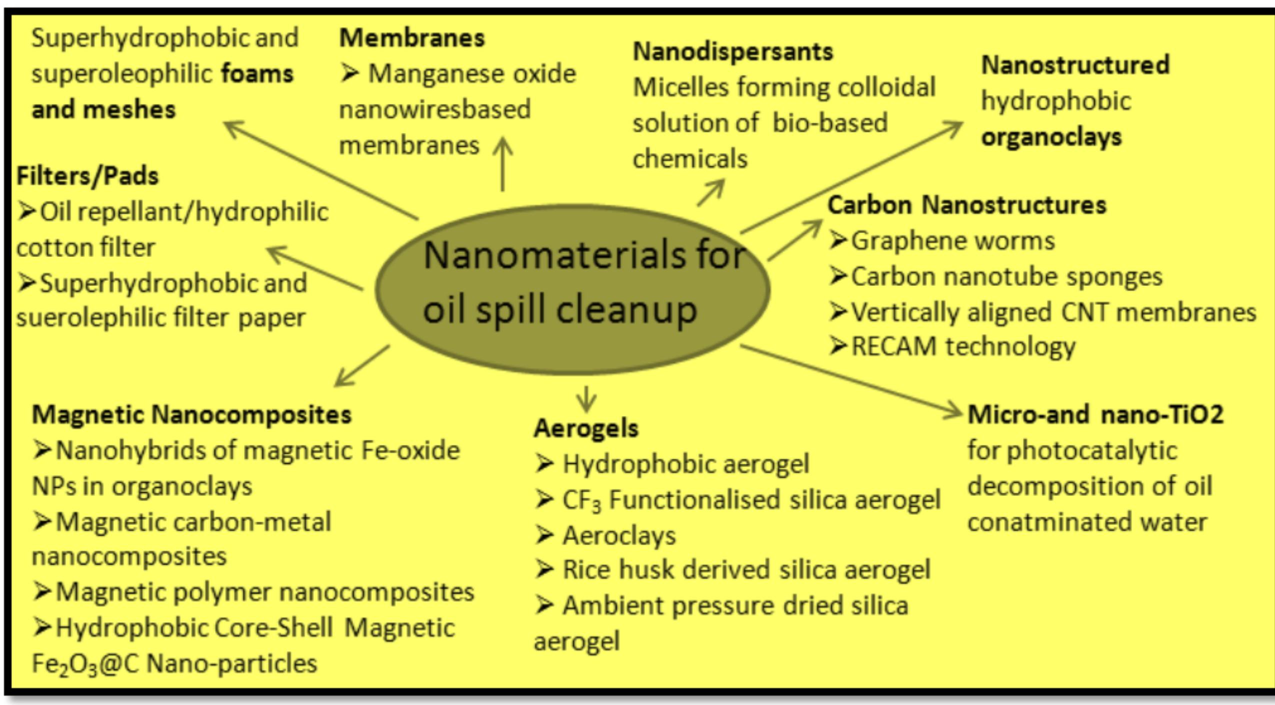

Figure 3 Nanotechnology-based solutions for oil spills [6]

Oil Spill Detection and Cleanup via Other Technologies

It is possible to fabricate a multifunctional device which has the capability of combining the functions performed by the previously listed conventional techniques of oil spill cleanup. By combining electroless metal deposition with self-assembled monolayers, Cheng et al came up with a device which “integrated the functions of oil containment booms, oil-sorption materials, oil skimmers, and water-oil separating devices [8].” The device is less dense than water to enable it float like a conventional oil boom. By taking up oil over 3 times its weight, the device possesses very good oil-sorption potential. The final characterization of the device is its ability to yield a 92% water-oil separation.

In a report which appeared on the Cable News Network (CNN) in 2010, a start-up in San Francisco by the name Liquid Robotics created a robot which is capable of sensing oil leakage around drilling platforms [27]. The robot according to this report is powered by the sun and waves, and it is very costly. There is also a report which shows scientists have developed a magnetic soap to address the problems caused by the use of dispersants [27]. The environmentally friendly soap is said to be iron-rich and salty, enabling it whenever magnetic force is applied to rise to the water surface with large amounts of oil trapped in it.

Lastly, Zahugi *et al*. presented a paper on a multi-robot system which works on water surface to facilitate oil spills clean-up [39]. The main aim of the system is to contain the oil spill in certain a position for quick, effective and easy clean-up, as well as to prevent further oil spreading. In their research, the team designed the robot to operate on water surface. The robot was equipped with a half oval shape skimmer to push a floating oil spill to some position for better skimming and easier cleaning up. **Figure 4** illustrates how a swarm of sea robots can be used in collecting an oil spill in the sea.

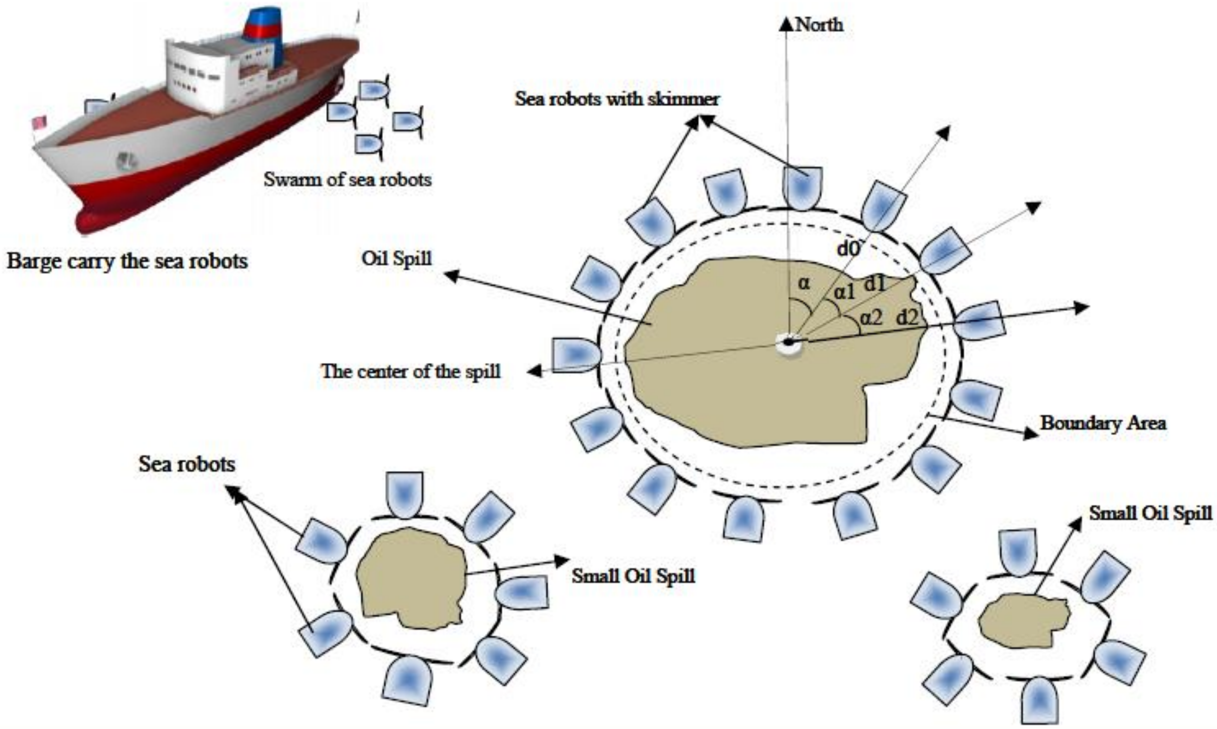

Figure 4 Dispatch swarm of sea robots around oil spills [39]

Conclusion

This review has highlighted a number of innovative techniques which have enormous potential of cleaning up oil and other contaminants in areas of oil spillage. These techniques offer better quality clean up than conventional methods such as containment booms, skimmers, and use of dispersants amongst others. Their superiority is attributed to their unique properties such as cost-effectiveness, biodegradability, recyclability, and higher yield of oil-water mixture separation. It could be deduced from this review that the use of innovative sorbent materials remains the most widely used technique and the most effective in oil recovery from areas of oil spillage. This is echoed by the comparative analysis carried out by Dave and Ghaly in which they pointed out that oil recovery with mechanical methods (of which use of sorbent materials is part of) is the most effective response for marine oil spill, followed by the application of dispersants and bioremediation [10]. Most of the techniques reviewed in this paper are still being researched on or can only be used for small scale applications. Further extensive researches and adequate financing would ensure that these techniques are fully replicated to large scale operations, opening up ground for efficient and effective oil spill cleanups the likes of which have never been seen before.

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