

Vol. 22

Review Research Paper

doi https://doi.org/10.46488/NEPT.2023.v22i01.004

Open Access Journal

Advanced Synthetic and Bio-Based Sorbents for Oil Spill Clean-up: A Review of Novel Trends

M. G. Gote*[†], H. H. Dhila* and S. R. Muley**

*School of Chemistry, Dr. Vishwanath Karad MIT World Peace University, Pune, Maharashtra, India **Nuvation Bio Inc., 1500 Broadway, New York, NY, USA †Corresponding author: M. G. Gote; meghna.gote@mitwpu.edu.in

Nat. Env. & Poll. Tech. Website: www.neptjournal.com

Received: 18-08-2022 Revised: 06-10-2022 Accepted: 19-10-2022

Key Words: Oil/water mixtures Sorbents Surface modification Polymers

ABSTRACT

Due to immense population growth and economic development, the use of crude oil for various energy applications has escalated in the past few decades. This has led to the largescale exploitation of oil reserves which has further resulted in the accidental release of large amounts of oil into our oceans. In recent years, significant emphasis has been placed on processes involving oil sorption by various natural and synthetic sorbents. Several sorbent materials based on synthetic polymers such as polypropylene, polyurethane, polystyrene, etc., possessing three-dimensional porous structure, large surface area, high mechanical strength, and exhibiting good oil recoverability and reusability, have been employed for oil-water separation processes. Conversely, many of these materials in their native or pristine form are amphipathic, which prevents their large-scale use in oil spill clean-up. This has led to researchers exploring surface modifications of commercially available sorbent polymeric materials to enhance their oleophilicity and hydrophobicity. This review article summarizes and discusses recent advances in the strategies for the fabrication of newer surface-modified synthetic polymeric materials and natural bio-based sorbents, and further highlights their effectiveness in dealing with the oil/water separation challenges.

INTRODUCTION

Water is an essential component of all living beings on this planet. Earth's surface has 97% saline water, and only 3% fresh water is available for us to use. Therefore, water needs to be recycled and re-utilized in the best possible manner across the globe. In the last few decades, the rise in water pollution is seriously threatening the environment and onethird of all water pollution is due to oil spillage and untreated industrial discharge (Zhang et al. 2019). Crude oil is an important source of energy and is currently an indispensable component of industrial development and modernization. Several complex processes are involved in the extraction, refining, and transportation of oil to consumers (Speight 2006). However, accidental spillage of crude oil and oil distillate products occurs quite frequently in seas and oceans as a result of large offshore drilling and oil transportation (Rogowska & Namieśnik 2010). There are numerous reasons for the occurrence of these oil spills which include

ORCID details of the authors:

M. G. Gote https://orcid.org/0000-0002-7862-9621 oil discharge near oil wells, pipelines and other equipment failures, land runoff, natural disasters, etc. Most of the accidents are due to human error and some may be due to natural disasters such as earthquakes and hurricanes which cause oil tanker ships to be damaged. The various oils that get mixed with water are mostly lipids, hydrocarbons, and fractions of petroleum products such as diesel oil, gasoline, kerosene, etc. In oil-containing wastewater, oils may be mainly present in three forms: free oil (oil droplet size > 150 μ m), dispersed oil (20 μ m < oil droplet size < 150 μ m), and emulsified oil (oil droplet size $< 20 \ \mu$ m). The amount of oil that is spilled during such accidents may range from a few hundred tons to several hundred thousand tons. A recent report from ITOPF 2020 indicated that the number of incidences with spills larger than 700 tonnes has decreased significantly over the last few decades. The annual average number of spills reported in the 2010s was 1.8 spills, which in comparison to the 1970s shows a 90% reduction in the number of average spills, with a loss of 164,000 tonnes of oil from tanker spills \geq 7 tonnes in 2010s. These oil spills have a deteriorating effect on soil, water and air. The BP Deepwater Horizon disaster which occurred in 2010, is considered to be the worst and the largest oil spill ever (Middlebrook et al. 2012) recorded. Another recent mishap near the California

coastline (2021) released 25,000 gallons of crude oil into the waters. Although there is a trend of decreasing the number and quantity of oil spilled due to better drilling technology and more safeguards against spills in recent years, there remains a significantly high risk of large oil spills causing environmental havoc.

Oil spills are a catastrophe for marine wildlife as it suffocates fish and other aquatic fauna. Oil-coated birds and marine mammals may die from hypothermia. Photosynthesis by marine plants is also hindered due to oil spills blocking sunlight from penetrating the water's surface. In addition, textile and chemical industries release a large amount of oily wastewater that contaminates aquatic bodies causing serious damage to the ecosystem. The cost of oil spills is very high in terms of both the economy and ecology. Oil spills are a serious environmental crisis that needs to be dealt with relentlessly to save our ecosystem. The remediation of oily wastewater is extremely critical and of paramount importance. Oil clean-up and recovery of oil from the spill is however, a complicated task that is influenced by numerous factors such as quantity and type of oil spilled, water temperature, air currents, waves, type of shorelines, etc., which may take a long time to clean up (Usman & Okoro 2017).

CONVENTIONAL METHODS OF OIL-SPILL CLEAN UP

Many conventional techniques are in use to tackle the problem of oil-water separation. These methods include mechanical, chemical, thermal, and biological treatments

(Fig. 1) (Padaki et al. 2015). Various devices such as skimmers, pumps, booms, etc. are being used as mechanical methods (Jamaly et al. 2015, Ramanathan et al. 2021). Additionally, chemicals such as detergents are employed to emulsify the spilled oils to enable their biodegradation. In-situ, localized burning of oil has also been carried out, but needless to say, this method can have devastating effects on the environment (Potter & Buist 2008). Biological remediation, wherein certain oil-eating bacteria are employed, causes the breakdown and further detoxification of dangerous chemicals or contaminants present in the polluted water (Atlas & Hazen 2011). Other technologies which have been developed include electrochemical methods specifically electrocoagulation and electroflotation, wherein by passing an electric current through an emulsion, oil-water separation can be achieved. Oily waste water treatment has also been conducted through membrane filtration which involves applying pressure to separate the liquids via a membrane usually made of ceramic and polymeric materials (Fingas 2012). These oil-water separation techniques are not very efficient due to ineffective oil/water separation, use of energy-intensive processes, high operations costs, timeconsuming processes, the release of secondary pollutants, and various other challenges (Pavlatou 2020).

Sorbents for oil-spill clean-up, owing to their structure are materials capable of capturing a liquid primarily in their cavities, fibers, and surfaces. The word "sorbent" comes from the Latin word "sorbere" which means material that soaks with liquid. Liquid sorption occurs till the system



Fig. 1: Common methods used in oil spill clean up.

reaches the equilibrium or saturation point. In comparison with other conventional techniques, the application of sorbent materials has proven to be a more effective way of dealing with environmental concerns and recovering oil in preference to water (Wu et al. 2014, Zhang & Seeger 2011). The mode of action of these sorbents is either absorption or adsorption. Absorbents work like sponges and absorb oil by capillary action or suction and are useful for less viscous oils. On the other hand, adsorbents have a large surface area, high porosity, and high chemical affinity for spilled oil and work best for heavy, sticky oils. Some sorbents may use both modes of action to sequester oil. A wide array of techniques based on sorbent materials has recently been developed as promising solutions to the oil spill problem (Shang et al. 2016). These absorbing or adsorbing materials are generally easily available, inexpensive, and environmentally friendly (Singh et al. 2013, Yang et al. 2015). In addition, they also have high regeneration ability and the oil can be subsequently recovered and reused, making the sorbents crucial in the removal of oil from water.

Oil sorbents fall into three basic categories: organic, inorganic synthetic materials, and biomaterials available in nature. The carbon-based biomaterials include rice husk, sugarcane bagasse, sawdust, straw, cellulose, etc. (Yue et al. 2019). The natural inorganic sorbents include clay, perlite, glass wool, sand, zeolites, etc. (Ruan et al. 2014, Wang & Geng 2015, Zhang et al. 2019). However, these sorbents suffer from certain drawbacks such as being amphipathic i.e., absorbing both oil and water, poor oil recoverability, poor buoyancy, and poor recyclability.

SYNTHETIC SORBENTS

Several novel synthetic sorbents have been developed in the form of foams, sponges, fibers, meshes, nanoparticles, etc. to achieve high absorption capacity, oil/water selectivity, and reusability for efficient separation and recovery of oil from water surfaces (Gui et al. 2013, Zhang et al. 2014). Recently, synthetic polymers such as polypropylene (PP), polystyrene (PS), and polyurethane (PU), have received a lot of attention as viable sorbent materials among others since they have reasonably good oil sorption capacity (Sarbatly et al. 2016, Zhang et al. 2018). They are substantiated by properties such as lower density, high porosity, larger surface area, and excellent mechanical strength. Polymers have several advantages in the manner in which they can be easily processed, molded, and functionalized in comparison with other materials (Guo et al. 2017, Zhou et al. 2013). However, certain drawbacks, such as poor recyclability, low oil retention, and poor oil recoverability still limit their performance in oil spill clean-up. Hence, it has become imperative to modify these polymeric materials to enhance their hydrophobicity and oleophilicity to improve

their performance in oil/water separation (Khosravi & Azizian 2015, Li et al. 2018).

One of the research strategies has been to modify the surface of synthetic, polymeric sorbents by using certain low surface energy materials to enhance their surface roughness to convert them into more hydrophobic and oleophilic materials to achieve higher oil absorption capacity (Lü et al. 2016). Generally, if the water contact angle (WCA) is lower than 90°, the solid surface is considered to be hydrophilic, and if the WCA is greater than 90°, it is considered hydrophobic. Highly hydrophobic surfaces made of low surface energy materials may achieve WCA as high as $\approx 120^{\circ}$. Some of the materials with highly rough surfaces may achieve a static WCA greater than 150° and a dynamic WCA less than 10° due to the existence of air pockets under the liquid drops and have been termed superhydrophobic surfaces. Several reports have been published which illustrate the superior performance of superhydrophobic materials in oil-water separation applications due to their super-antiwetting and self-cleaning properties. Extensive efforts have been dedicated in the last couple of decades to researching the preparation and synthesis of durable superhydrophobic materials for efficient separation of oil and water during the clean-up of global oil-spills. One of the disadvantages of the use of hydrophobic, highly porous polymeric materials in oil-spill clean-up is the tendency of highly viscous oil to be trapped in the pores of the material due to their low surface tension, resulting in decreased regeneration and reusability of the material to continue adsorbing the oil. Therefore, modification of polymeric materials to enhance their adsorption capacity, surface self-cleaning, and regeneration ability have to be evaluated in depth. In addition, challenges related to the decreased thermal stability of polymeric materials at high temperatures and low energy conversion efficiency need to be overcome. Future research should also aim at making the commercial manufacture of functional polymeric foams more efficient, economical, widely available, and easy to deploy and use.

Given the plethora of research in the field of environmental science in curbing the effects of aquatic pollution specifically due to oil spills, this review evaluates the latest advances in the strategies for modification of polymeric materials to make them more efficient in oil-spill clean-up and assesses their effectiveness and commercial applicability.

STRATEGIES EMPLOYED IN THE FABRICATION OF MODIFIED SYNTHETIC SORBENTS

Polyurethane

Polyurethane (PU) sorbents are some of the popular sorbents due to a combination of their characteristics including low cost, high porosity, good elasticity, large internal surface area, smaller cross-linked structure, and excellent microstructure. PU polymers are usually synthesized by reacting a di- or tri-isocyanate with a polyol and are classified as alternating copolymers since PU monomers polymerize one after the other in a sequence. The possibilities for the preparation and modification of PUs are numerous, which combined with an extensive choice of additives and processing conditions available, make the use of PU very widespread. However, due to the presence of groups like carboxyl and amino groups in these polymers, PU is amphiphilic. Increasing the oil absorption capacity of PU sponge is therefore an essential requirement for its applicability in oil spill clean-up. The efficiency of PU sponges as sorbents is greatly enhanced when their wettability is improved upon modification of its surface with low surface energy hydrophobic materials. Surface-modified polyurethane has been shown to exhibit promising results in terms of oil/water separation owing to its good stability, high sorption capacity, and recyclability. Specifically, methods that generally involve the coating of various materials onto the sorbents should aim at transforming the surface functions and not alter their bulk properties. The following sections discuss various strategies for the modification of PU and other polymers to help improve their sorbent efficiency. A summary of modified polyurethane sorbents is given in Table 1.

In recent years, polymerization has been employed as a facile technique to construct superhydrophobic sponge surfaces. The polymerization reactions involve covering the surface of the sorbent with crosslinked polymer chains to produce oil-sorbing surfaces. This technique results in polymeric chains formed on the surface to facilitate the adhering of other hydrophobic materials. Graft copolymerization has been conducted with an oleophilic monomer as lauryl methacrylate (LMA) to modify PU using divinylbenzene as the cross-linker (Li et al. 2012) to get a highly oleophilic polyurethane. The sorption efficiency of LMA microspheres coated onto PU cubes was also evaluated. The surface roughness of the modified PU sorbents was observed to increase upon grafting or coating

Table 1: Modified Polyurethane Sorbents.

Sorbent	Treatment Material	Method	Sorption Capaci- ty (g/g)	Sorption Experimental Conditions	Reference
PU foam	Lauryl methacrylate (using DVB as a linker)	Graft copolymerization	50–69	Sorption conducted at $23 \pm 4^{\circ}$ C by immersing sorbent into oil	Li et al. 2012
PU	Perfluorinated polydopa- mine-coated nanodiamonds	Controlled dripping/dry- ing process	Diesel: 31.2 Gasoline: 20.5	NDs-fPDA loading at 60%, highest sorption capacities observed at pH 1 and 13	Cao et al. 2017
PU	Polydopamine- hexamethyl disila- zane (HMDS)	PU sponge coated with PDA nano-aggregates grafted with HMDS	Acetone: 36 Chloroform: 53	Sorption conducted at room temperature (RT). No specif- ic test conditions described	Liu et al. 2019
PU	Acidified carbon nanotubes-Poly- dopamine	Electrospun PU nanofib- ers immersed in ACNTs followed by self-polym- erization of dopamine	Separation Effi- ciency: 99.9%	Water Permeation Flux: 4195 – 7240 L m ⁻² h ⁻¹ at driving pressure 0.5 bar for heptane, toluene & cyclohex- ane; SDS used as emulsifier @ 0.001% w/w	Huang et al. 2020
PU Foam Waste	Lauryl methacrylate & hexade- cene, using crosslinkers like divi- nyl benzene (DVB) & 1,1,1-tri- methylolpropane trimethacrylate (TPT)	Free radical polymeri- zation	Diesel:132.4 Crude oil: 125.1	Sorption with immersion for ~60 min at RT at 2% w/w nanoparticles loading	Keshawy et al. 2020
PU	Poly(dimethylsiloxane) (PDMS)- TiO ₂	Sol-gel method followed by in-situ polymerization	Diesel & Pump Oil > 16.7	Sponge size $1 \times 1 \times 2$ cm ³ , sorption determined at RT with TiO ₂ sol-gel loading of 13.6g/mL	Shuai et al. 2015
PU sponge	1,3-oxazolidine, octadecyl trichlo- rosilane (OTS) self-assemblies	Solution immersion	Bean Oil: 25 Lubrication Oil: 23	Oil absorption occurred within few seconds at RT	Liang et al. 2019
PU sponge	PU@ZnO@Fe3O4@SA	Microwave and Dip-coating	Diesel: 81 Vacuum Oil: 110	Evaluated on a round sponge, 7 cm diameter & 2 cm thick at RT	Tran & Lee 2017

Sorbent	Treatment Material	Method	Sorption Capaci- ty (g/g)	Sorption Experimental Conditions	Reference
PU sponge	TiO ₂ nanoparticles	Immersion	Motor Oil: 105 Diesel: 90	Sponge added to oil film 2-6 mm thick at $20 \pm 2^{\circ}C$ & stirred at 500 rpm for 1h.	Wu et al. 2014
PU	TiO ₂ nanoparticles coated PU followed by GO amidated with tetradecyl amine (TDA)	Immersion	Crude Oil: 26 Chloroform: 62	Carbon content 69%; Maxi- mum sorption occurred in <5 min at RT	Wei et al. 2018
PU sponge	Asphaltene-capped hydrophobic silica nanoparticles	Emulsion technique	Crude Oil: 50	PU: HSNP-2 at 1:1 ratio using sponge 2 cm thick	Atta et al. 2019
PU sponge	Carbon nanofibers	Grafting	Hexane: 27 Toluene: 50	Dynamic separation of oil by Pumping oil/water mixture through a pipe fitted with sponge	Baig et al. 2019
PU sponge	Carbon nanofibers & methylene diphenyl diisocyanate oligomers	Grafting	Pure System: 5.25 Mixture: 2.56	1 cm ³ foam in contact with 50 mL water, diesel, or wa- ter/diesel mixture for 30 min	Visco et al. 2021
PU sponge	Crosslinked reduced graphene oxide	Immersion	Not Available	Oil/water emulsion poured over sponge strip at RT	Zhu et al. 2016
Graphene/ PU sponge	Graphene/N-methyl pyrrolidone	<i>In-situ</i> polymerization of polyols & diisocy- anate in the presence of graphene/N-methyl pyrrolidone	Sorption Capaci- ty: NA, Continu- ous collection of various oils	Engine oil and crude oil filtered through PU sponge at 8.9×10^4 and 6.4×10^4 L*m ⁻² *h ⁻¹ respectively, at 30 kPa applied pressure	Kong et al. 2017
PU	Octadecylamine coating using polydopamine-reduced graphene oxide for adhesion	Solution immersion	Crude Oil: 25 Silicone Oil: 30	Sorbent foam in contact with oil dispersed on the water surface at RT	Oribayo et al. 2017
PU sponge	Polydimethylsiloxane (PDMS) layer and carbon dots	Solar-mediated grafting	Castor Oil: 34 Kerosene: 52	3 mm thick sponges placed in oil/water mixtures and irradiated at 1 kW/m ² (wave- length 350-1800 nm) at 25°C	Singh & Jelinek, 2020
PU foam	Crosslinked siloxane	Immersion followed by pyrolysis	Pump Oil: 500 – 2500% wt. gain	Felt piece size $3 \times 1 \times 0.5$ cm ³ , sorption determined @ 25°C	Biesuz et al. 2020
PU sponge	SiO ₂ /GO nanohybrid	Dip-coating	180	Cubical sorbent specimen of $5 \times 5 \times 2 \text{ mm}^3$ dipped in 20 ml oil for 5 min.	Lü et al. 2016
PU foam	Zeolitic imidazolate (ZIF-8) framework (MOF)	In situ self-assembly method	Silicone Oil: 28 Methylbenzene: 33	No specific test conditions described; sorption conduct- ed at RT	Zhao et al. 2019

the PU surface with LMA. The grafting copolymerization with LMA was observed to be less efficient or incomplete compared to the coating of PU with LMA microspheres resulting in greater sorption of oil by PU-LMA microspheres in comparison to PU-g-LMA and blank PU cubes. A novel and promising PU sponge coated with superhydrophobic poly(dimethylsiloxane) (PDMS)-TiO₂ was reported (Shuai et al. 2015) for the treatment of oil spills. An easy fabrication was carried out on the surface of the PU sponge via solgel growth of TiO₂, followed by immersing the PU-TiO₂ construct in PDMS solution for in situ polymerization. The surface was found to be superhydrophobic as proven by the WCA of 154°. The sorption capacity was observed to increase to 16.7 g.g⁻¹. The rate of sorption and sorption selectivity was found to be greatest for diesel oil out of the various oils evaluated. The absorbed oil was recovered from the sponge by a mechanical extrusion process and the recovered sponge displayed excellent reusability of more than 60 times.

There has been a surge in the exploration of multifunctional polydopamine (PDA) nanocoatings on a variety of substrates due to their versatility. In addition, they are a practical and effective approach to transforming the surface properties of various materials (Zhang et al. 2019). Out of the several cross-linking agents, dopamine has been especially useful as it can self-polymerize and result in the fabrication of stable covalent and non-covalent bonds between the materials (Peng et al. 2019). Taking advantage of the adhesion property of dopamine, a practical approach was developed by Cao et al. (2017) to fabricate stable superhydrophobic nanodiamond particles (NDs) coated with polydopamine which were subsequently perfluorinated. Nanodiamonds are a new class of carbonaceous material, which exhibit high surface area, non-reactivity, and extensive surface chemistry, and hence show great promise for a variety of applications. Self-polymerization of dopamine was first carried out on the surface of NDs. The catechol and amino groups present on the polydopamine film assisted in a facile Michael addition reaction of the -SH groups of 1H,1H,2H,2H-perfluoro-1-decanethiol (PFDT) to prepare superhydrophobic perfluorinated (NDs-fPDA) particles. The performance of the modified sponge was improved due to its superhydrophobicity, and high oil/water separation properties. The sponge shows an absorption capacity 15 to 60 times greater than its weight for a variety of solvents and oils. It also exhibited good recyclability and good oil recoverability. The low cost, easy fabrication method, and excellent performance of this superhydrophobic sponge make it a good candidate for oil spill clean-up.

A unique flame-retardant polydopamine (PDA) coated PU sponge was fabricated (Liu et al. 2019) via a one-step polymerization with further surface modification carried out using hexamethyl disilazane (HMDS). The PU-PDA-HMDS sponge demonstrated superhydrophobicity with a contact angle of 153°. Pristine PU sponges were dipped into dopamine solution which self-polymerized. The nano aggregates of PDA rendered the surface quite rough. However, due to the presence of hydroxyl groups on the PDA film, the sorbent was observed to be quite hydrophilic. Further grafting of HMDS on the PU-PDA surface was conducted which led to the attachment of methyl groups across the surface transforming the hydrophilic surface into an oleophilic surface. The surface modification led to an enhanced oil absorption capacity in the range of 21 to 53 g.g⁻¹ for various oils and organic solvents evaluated. Stable absorption performance was demonstrated by the surfacemodified PU sponge with repeated absorption-desorption cycles. Measurements showed that the PU-PDA-HMDS sponges exhibited excellent heat stability and flameretardancy in comparison with the original PU or PDAcoated PU. The flame-retardant property was attributed to the formation of a silica-hybrid char residue layer during the combustion process. The compact nature of the char residue layer can prevent flames from reaching the interior bulk of the sponge, thereby preserving the latter's structural integrity and exhibiting good flame-retardant capabilities during a real fire.

Membrane technology processes are rapidly emerging as highly viable solutions for treating oily wastewater due

to the ease with which they can be used, as well as their high separation effectiveness. Other advantages of using membrane separation processes include reproducible product quality, lower operating costs, and a smaller carbon footprint, i.e., lower energy consumption over long durations required for oil-water separation processes (Karami et al. 2020). However, the hydrophobic properties of the membrane which make it an efficient oil-adsorbing agent can also shorten the life of the membrane due to increased membrane fouling. As a result, modification of membranes, specifically the enhancement of their hydrophilicity is also essential to improve the membrane performance. Superhydrophilic/ underwater superoleophilic PU nanofibrous membranes decorated with acidified carbon nanotubes (ACNTs), and coated with polydopamine with a core/shell structure have been fabricated by (Huang et al. 2020). Excellent stretching ability and surface stability were achieved due to extensive interfacial hydrogen bonding between the various components of the nanofibres. Better anti-fouling properties and excellent oil-water separation with outstanding recyclability were exhibited by the fabricated nanofibres. Core/shell composites of PDA/ACNTs and polymer nanofiber exhibited great potential in oil spill clean-up.

Another pressing environmental pollution problem we are facing is the daunting task of dealing with an enormous quantity of plastic waste generated around the world. The usage of plastic waste as a feed for the fabrication of oil sorbents could therefore solve the dual problems of minimizing and management of plastic waste, as well as containment of oil spills. To further investigate this approach, Keshawy et al. (2020) evaluated the functionalization of the surface of polyurethane foam waste via a facile copolymerization of hydrophobic monomers, namely lauryl methacrylate & hexadecene, using crosslinkers as divinyl benzene (DVB) and 1,1,1-trimethylolpropane trimethacrylate (TPT). Furthermore, nano magnetite was incorporated into the coated PU foam waste which enhanced its oleophilicity. Increasing the nanoparticle concentrations also increased the sorption capacity for diesel and Egyptian crude oil to 132.4 g/g and 125.1 g/g respectively. Higher oil sorption was observed for DVB crosslinked coated samples compared to TPT crosslinked samples. It was inferred that a denser network could be achieved with TPT in comparison to DVB as the crosslinker.

Liang et al. (2019) have prepared via a facile, mild, and inexpensive process, 1,3-oxazolidine-modified polyurethane sponges were immersed in a solution of octadecyl trichlorosilane (OTS) to generate self-assemblies. Though the OTS functionalization could not alter the original structure of the PU sponge, it still made its surface quite rough, thus rendering it more hydrophobic. The sponge showed good absorption ability for various oils and organic solvents, such as lubricating oil, bean oil, acetone, peanut oil, and n-hexane. High selectivity and absorption capacity of up to 25 times the original weight were observed for the modified sorbent in addition to oil retention of up to 92% with good recyclability.

Surface Modification Using Nanomaterials

In the pursuit of finding efficient sorbents to assuage the environmental pollution caused by the petroleum industry, newer technologies utilizing super oleophilic and superhydrophobic nanomaterials have been developed for modification of PU and other polymeric materials to improve the sorbents' oil sorption capacities. Sorbents have been functionalized by incorporating various nanoparticles along with low surface energy molecules containing long hydrophobic groups to impart them with superhydrophobic properties. These nanostructures have been shown to perform much better than conventional materials in resolving the issue of oil spillage. Tran & Lee (2017) investigated a novel, facile, and environmentally friendly method for the fabrication of a durable, magnetic, and superhydrophobic PU@ZnO@ Fe₃O₄@stearic acid sponge. This work highlighted that the functionalized sponge had high surface roughness, low surface energy, and magnetic responsiveness necessary for high oil absorption capacity. A large static WCA of 161°, and a very low dynamic WCA of 7°, were exhibited by the fabricated sponge. Oils of varying densities and viscosities were absorbed to different extents by the fabricated and modified sponge. The efficiency of the fabricated sponge in oil-water separation exceeded 99%, and easy recoverability was observed upon simple squeezing of the sponge. The superhydrophobicity and oil absorbency of the fabricated sponge were unaltered after multiple rounds of stretching and compression.

The fabrication of PU sponges modified with TiO₂ sol has been reported (Wu et al. 2014). TiO₂ nanoparticles attached to the PU sponge were able to reduce the hydrophilicity of the sponge to a great extent. An oil absorption capacity of 95–110 g.g⁻¹ was achieved by TiO₂ -PU sponges with negligible uptake of water under both static and dynamic conditions. Good reusability of up to 12 cycles with a holding capacity of 70% was observed for the modified sponge compared to the uptake capacity of the original PU sponge. Similarly, the preparation of PU foam coated with TiO₂ nanoparticles (TPU), and its further treatment with graphene oxide (GO) amidated with tetradecyl amine (TDA) to obtain TPU-GO-TDA foam has been described (Wei et al. 2018) as depicted in Fig. 2. Due to an extensively interconnected three-dimensional pore structure in foams which are also supported by pore mesh to lend them good structural stability, the pores in these foams were able to store large amounts of absorbed oil. The authors hypothesized that the enhancement in the oil uptake capacity was probably due to the long hydrophobic chains of tetradecyl amine. The surface roughness was enabled by coating with TiO₂ nanoparticles and GO nanosheets. It was observed that the hydrophobicity



Fig. 2: Schematic illustration for the fabrication of TPU-GO-TDA foam sorbent. (Reprinted with permission from Wei et al. (2018). Copyright © 2018, American Chemical Society).

was achieved only after the treatment with GO-TDA since oil sorption increased in comparison to unmodified TPU foam. This was also evident from the high-WCA of around 140°.

Improved hydrophobicity of the PU sponge has also been achieved by coating asphaltene-capped hydrophobic silica nanoparticles onto the surface of the PU sponge. This modified PU sponge has been used for heavy Arabian crude oil spill clean-up (Atta et al. 2019). The PU sponge modification was conducted by emulsification technique, and it was hypothesized that the hydrophobicity of the PU sponge was due to the interaction between the PU sponge and the reactive groups of superhydrophobic silica. Excellent and rapid collection of oil by the sponge was enabled by the hydrophobic PU sponge, which was also reusable and exhibited an excellent recovery.

Biesuz et al. (2020) developed efficient oleophilic nanofelt by impregnating PU foams with a liquid preceramic siloxane followed by controlled pyrolysis, wherein the siloxane converted into an amorphous SiOC foam at a temperature below 800°C, and at temperature ~ 1500°C it decomposed to give Si₃N₄ fibers. Oil separation from the water was easily achieved by the nanofelt. The results showed a 500% to 2500% increase in weight after oil absorption. Simple thermal treatment was able to clean the material. The fibers developed from small pore foams were found to be extremely stable even after repeated oil-absorption cycles.

Carbon Materials

Owing to their intrinsic hydrophobicity, carbon-based materials such as graphene-reduced GO, carbon nanotubes and carbon nanofibres have been explored for oil spill treatment. These carbon materials coated onto the sponge surface can render the original hydrophilic surface into a hydrophobic surface. Carbon nanofibres (CNFs) display exceptional hydrophobicity but have not been evaluated in detail for their application as PU modifiers for oil spill clean-up. The hydrophobic characteristics of CNFs and the adsorption properties of PU can be combined to improve the efficiency of the latter for oil sorption from oily wastewater. To leverage the hydrophobic nature of CNF, Baig et al. (2019) have investigated the grafting of CNF onto a PU surface using a dip-coating method. The formation of additional pores by the CNF on the surface of PU resulted in a decrease in the average pore size of the PU sponge, resulting in a significant increase in the surface area and a greater uptake of oil. The surface area of PU increased by about 31 times from 9 m².g⁻¹ to 276 m².g⁻¹ owing to the grafting process, which resulted in an increase in the oil absorption capacity by approximately 50 times its weight. The study demonstrated the CNF-PU sponge's repeated

ability for oil absorption and its recoverability due to the improved flexibility and mechanical stability of the sorbent.

In a recent study, Visco et al. (2021) fabricated polyurethane-based nanocomposite foams by incorporating varying amounts of ball-milled carbon nanofibres and employed them for the selective recovery of oil from water. The fabrication of the grafted PU foams was carried out by mixing methylene diphenyl diisocyanate oligomers with carbonaceous filler followed by the addition of polyether polyols (PEP) to obtain the crosslinked polymer. CNFs were loaded on to PU foams up to 15% by weight. A contact angle greater than 90° was observed for nanocomposite sponges indicating low surface energy between liquid and solid. Water/diesel mixtures were used to study the efficiency of the sorbents. The absorption values for the sponges determined by Visco et al. (2021) contrasted significantly with those obtained by Baig et al. (2019) probably because the former evaluated a sponge structure possessing a larger pore size distribution (range of 130-190 µm) with oils which varied significantly in their chemical composition and viscosity. Maximum absorption of diesel from the oil/water mixture was demonstrated for a foam with 1% w/w loading of the carbonaceous filler as 1: 5.25, oil-water ratio.

Graphene aerogels and CNTs can significantly improve the hydrophobic-oleophilic wettability of a PU surface due to their porous interconnected structures. However, they suffer from a few disadvantages which limit their use as good PU surface modifiers, which include their high cost which makes large-scale manufacture of CNT and graphene-grafted PU more expensive, and poor stability of the grafted sorbent due to weak CNT/graphene-PU interactions which decrease their oil absorption capacity. Therefore, carbon derivatives that can bond more strongly with PU are required. One such derivative is the reduced graphene oxide which itself can be further modified appropriately to obtain a sorbent with greater oil absorption capacity. The presence of groups such as carboxyl, epoxy, and hydroxyl in the two-dimensional structure of graphene oxide (GO) makes it a good swelling material. Its applicability as a feasible sorbent for oil spill clean-up has been explored also because of its large specific surface area and excellent sorption capability (Junaidi et al. 2021). However, the hydrophilic nature of GO has to be chemically modified to render it more hydrophobic. Zhu et al. (2016) have fabricated a porous PU sponge cross-linked with reduced graphene oxide (rGO). This was achieved by the addition of calcium carbonate nanoparticles and graphene oxide nanoparticles during PU foam synthesis, which upon etching with a mild acid resulted in the formation of a PU sponge with a nanosized porous framework which is not present in a normal PU sponge. A thin layer of rGO was then coated onto the porous PU sponge via a crosslinking procedure. The results indicated that PU@ rGO sponge has excellent potential use in oil-water separation. The rGO surface coating in the porous PU@rGO structure was found to have good electrical conductivity which prompted the authors to evaluate the construct to disrupt the oil in water emulsion.

A microporous PU sponge coated with SiO₂/GO nanohybrid material displayed a flexible structure with an excellent oil sorption capacity of about 180 times its own weight and good recyclability (Lü et al. 2016). A highperformance graphene/PU sponge was prepared by in-situ polymerization of polyols and diisocyanate in the presence of graphene/N-methyl pyrrolidone (NMP) suspension (Kong et al. 2017). The strong dipole interactions between graphene sheets and NMP facilitated the introduction of graphene sheets into the framework of the PU sponge, which led to better physical and chemical strength of the graphene/PU sponge. Continuous oil removal from the water was achieved by using a vacuum pump. The graphene/PU sponge was found to have good filtration capabilities for continuous removal of oil due to its superior mechanical strength and oleophilicity as evidenced by its magnificent adsorption capacities up to 40 to 80 times its own weight.

A PU foam derived from lignin (LPU) a plant-based material, using green technology was subjected to surface modification by treating it with octadecyl amine (ODA) using adhesive polydopamine-reduced graphene oxide (rGO) (Oribayo et al. 2017). The synthetic methodology led to grafting long hydrophobic methylene chains and alkyl groups from ODA molecules onto the surface of LPU foam through an adhesive coating of polydopamine-rGO, resulting in a 3-D porous structure. GO contains distended structural hydrophilic groups such as hydroxyl, epoxy, and carboxyl groups which are responsible for its significant swelling, intercalating and ion-exchange capabilities. An outstanding oil sorption capacity of 26-68 times its own weight could be achieved with the LPU foam which was prepared by covalently attaching hydrophilic GO to LPU, followed by reducing it to hydrophobic rGO via oxidative self-polymerization of dopamine to polydopamine. This oil sorption capacity was found to be much higher than that of a non-woven commercial polypropylene sorbent. ODA along with the low surface energy and hydrophobic rGO rendered the foam with varying degrees of wettability for oil and water. rGO was responsible for introducing surface roughness which affected the interaction of the foam with a wetting or non-wetting liquid. LPU-rGO-ODA foam sorbent also showed good oil selectivity and reusability.

A new class of carbon nanoparticles called the carbon dots (C-dots) have been recently identified as having

great potential in oil-spill containment due to their simple synthesis, unique optical properties, and environmentfriendly characteristics. An innovative strategy was demonstrated involving the application of a composite material consisting of a PU sponge embedded with a polydimethylsiloxane (PDMS) layer and C-dots prepared from p-phenylenediamine for solar-mediated oil uptake from oil spills (Singh & Jelinek 2020). The combination of the high porosity of the PU sponge structure, hydrophobicity of PDMS, and the conversion of sunlight into heat by carbon dots, facilitates greater absorption of oil from the spills. C-dots absorb sunlight in the visible region. An excess of electrons on the C-dot surface results in a reduction of the HOMO-LUMO gap which further results in increased light absorption. The C-dots were immobilized in the PU-PDMS matrix such that the energy absorbed by them upon exposure to sunlight increased the temperature of the matrix which facilitated increased oil absorption. The carbon dot-PDMSpolyurethane composite sponge exhibited good chemical stability and recyclability.

Surface Modification Using Metal-Organic Frameworks

Metal-organic frameworks (MOFs) are porous materials composed of a metal ion or a cluster of metal ions and an organic molecule as a linker. They have large pores and open metal sites, thereby making them attractive for applications such as the adsorption of pollutants from water (Fig. 3). However, MOFs have certain drawbacks which include low mechanical strength and poor processability, which have been overcome by integrating them with more sturdy and inert materials to produce sorbents for efficient oil-water separation. Zeolitic imidazolate (ZIF-8) framework, a type of MOF grown on flexible PU foam (MFPU) was synthesized by Zhao et al. (2020) through an in situ self-assembly method. The increased surface roughness of MFPU foams resulted in a higher contact angle, which improved the hydrophobicity and lipophilicity of the parent MOF. Various types of oils and organic solvents could be absorbed into these foams quite efficiently with an extraordinary absorption capacity reaching 33 times its own weight. The ZIF-8 coating was also observed to significantly enhance the flame retardancy of MFPU foams, which was determined by cone calorimeter test, vertical burning test, and limited oxygen index test.

Polystyrene

Yu et al. (2016) have reported a durable and modified magnetic polystyrene foam (DMMPF), synthesized via an inexpensive two-step immersion method by using oleic acid coated Fe_3O_4 particles and PS microspheres which exhibited excellent superhydrophobicity, superoleophilicity, and

fast magnetic response. The modified magnetic PS foam exhibited an oil absorption capacity of nearly 40 times its original weight. Simple mechanical extrusion allowed recovery of absorbed oils and solvents from the DMMPF foam which was shown to retain its high oil absorption capacity even after 60 use cycles. Porous PS fibers work as effective oil sorbents by either adsorption or capillary action and promising prospective in wastewater remediation. Oils and solvents like diesel, lubricating oil, vegetable oil, DCM, cyclohexane, benzene, etc. were found to be absorbed by the as-fabricated foam. Authors claim that the absorption capacity of DMMPF is higher than magnetic graphene foam and cryogels based on graphene. Damavandi & Soares (2022) grafted polystyrene (PS) chains on the surface of silica-coated super magnetic iron oxide nanoparticles using a blending technique using SI-ATRP. The use of SI-ATRP conferred high hydrophobicity and oleophilicity to the PS graft. Oil was absorbed rapidly and to an extent of 5x in 5 minutes by a nanocomposite in which 90% of the PS-SiO₂-IONP was replaced with polystyrene. The fast oil absorption is explained by van der Waals forces, hydrophobic effects, and stacking interactions between polystyrene molecules grafted on PS-SiO₂-IONP and the oil molecules. Nanofibrous syndiotactic polystyrene (sPS) monoliths were fabricated and were demonstrated to possess superhydrophobicity by Gui et al. (2019). These fibers were prepared by solidifying non-aqueous high internal phase emulsions (HIPEs) through crystallization-induced gelation. The HIPEs were formed by dispersing glycerol in 1,2,4-trichlorobenzene (TCB), stabilizing with sulfonated sPS at a relatively high temperature of 120 °C. Cooling of HIPEs resulted in their solidification due to the crystallization of sPS within the continuous phase to produce porous sPS monoliths. The morphological study of the prepared sPS monoliths showed hierarchically porous structures (Fig. 4). They were found to exhibit a high specific surface area of approximately 420

 m^2 .g⁻¹ and also good mechanical strength with Young's moduli ranging from 157.7 to 2638.0 kPa. The sorbent material exhibited excellent oil absorbing capacities of up to 81.3, 44.4, and 41.9 g.g⁻¹ for chloroform, olive oil, and diesel, respectively.

A superhydrophobic sorbent was fabricated from PS foam waste via a high internal phase pickering emulsion (HIPPE) technique in a one-step process (Yu et al. 2019). Silica and Span 80 were used as surfactants for stabilizing the emulsion in which the PS packaging waste was introduced as the crosslinker due to its low surface energy, thereby making the process more facile. The as-fabricated SiO₂@PS sorbent had a high porosity which enabled good removal of oily contaminants from the water. The sorbent displayed a high adsorption capacity of 20.4–58.1 g.g⁻¹ and retained its recyclability for 10 clean-up cycles. Polymeric nanofibres owing to their properties such as layered structure have great potential to collect oil from oily wastewater. Such nanofibres are fabricated through various techniques such as electrospinning, melt-blowing, bicomponent spinning, and force spinning.

Out of the various techniques widely used for the construction of nanofibres, electrospinning (Sarbatly et al. 2016) has found its niche in the manufacture of sorbents for efficient oil-water separation. The process involves subjecting a jet of polymer solution to electrostatic forces to obtain the polymeric fibers with diameters in the range of 5 to 500 nm. Polystyrene-carbon nanotubes (PS-CNTs) prepared using the electrospinning technique were found to possess high oleophilicity (Wu et al. 2017). Studies indicated that the oil absorption occurred due to intra-particle diffusion. The super-hydrophobic-oleophilic PS-CNT sorbents showed reusable potential. These sorbents proved to be efficient sorption materials for the clean-up of oil spills. Various oils such as sunflower oil, peanut oil, and motor oils were



Fig. 3: Polyurethane foam coated with a metal-organic framework to trap oil.



Fig. 4: (a) Schematic Illustration of Fabricating Emulsion-Templated sPS Monoliths through Crystallization Induced Gelation, (b) Porous sPS Monoliths with Superhydrophobicity/Oleophilicity, Extremely Low Density, and Nanofibrous Structures, (c) Absorption of Oils on Water Surface, Underwater Oils and Emulsified Oils Using the sPS Monolith (Reprinted with permission from Gui et al. 2019. Copyright © 2019, American Chemical Society).

used to study the oil sorption capacity which was found to be 116, 123, and 112 g.g⁻¹, respectively. An increase of 65% in oil uptake capacity was observed for the composite in comparison to PS sorbent without CNTs. Alnaqbi et al. (2020) demonstrated that polymer blends have greatly enhanced properties and distinct advantages over original polymers as components of oil-sorbent materials. Their study highlighted the application of hydrophobic polymers prepared through polymer blending, wherein blends were prepared by using PS as the matrix polymer, to which poly (vinyl chloride) (PVC) and polyethylene (PE) were added individually at 5-20 % w/w. Electrospinning of the blends was carried out to obtain microfibers. Good removal of crude oil, diesel, and motor oil was achieved. In comparison to other commercially available polypropylene (PP) fibrous materials for oil absorption, the blended electrospun fibers were far more efficient for a wide range of oils.

The removal of various oils from aqueous solutions by PS nanofiber membranes synthesized by two methods namely electrospinning and impregnation filled up with different types of sawdust was demonstrated by Ghiasvand et al. (2021). The particle size of sawdust in the range of 40-60 mesh showed the highest sorption capacity. Separator pads were fabricated comprising of an outer covering made of PS nanofiber membranes sandwiching sawdust and used for the removal of oil spills from the surface of the water. An increase in temperature led to a decrease in the sorption capacity of the PS nanofiber membrane. The sorption capacity also decreased with an increase in pH up to neutral pH while increasing slightly with a further increase in pH above 7. These membranes prepared by the electrospinning technique offer better sorption capability than the ones prepared via impregnation. This variation in the performance of the membranes may be explained in terms of the process of impregnation that does not leave many pores open for the oil to be taken up since they are covered by polystyrene. A magnetic PS/Fe₃O₄ fibrous membrane fabricated by the electrospinning technique showed high selectivity and excellent oil adsorption. The magnetic response from Fe₃O₄ and superiority in hydrophobicity and oleophilicity of polystyrene combined to enhance the separation efficiency as well as adsorption capacity of magnetic fibrous sorbent in oil/water mixtures (Song et al. 2017). The highly porous structured PS fibrous sorbent shows a motor oil sorption capacity of 113.87 g.g⁻¹, approximately 3-4 times that of natural sorbents and nonwoven PP fibrous

membranes. Additionally, the nanoporous fiber sorbents also exhibited high sorption capacity for edible oils, such as bean oil (111.80 g.g⁻¹) and sunflower seed oil (96.89 g.g⁻¹) ¹) (Lin et al. 2012). Yadav et al. (2019) demonstrated the recyclability of thermoplastic PS using orange peel extract. The method involves extracting liquid from the outer rind of the peel (primary extract) which was further processed into three layers. PS was dissolved in the layers to get viscous solutions. These PS solutions were then processed into fibers that exhibited high contact angle. The oil sorption capacity was found to be higher for fibers obtained using the top layer of extract (TLF) compared to the ones obtained from using the primary extract (PEF), primarily due mainly to a greater surface area of TLF. Expanded polystyrene (EPS) waste was utilized as a sorbent to clean up a crude oil spill from an aquatic environment (Nasir & Hameed 2014). The sorption capacity of these sorbents increases with the increase in surface area and sorption time. It was also observed that the rough surface of EPS adsorbed more oil than the smooth surface. The punctured hole diameter in EPS sorbent (1.65 mm) also enhanced the oil sorption capacity.

Zhu et al. (2011) investigated the viability of fibers made from blended PVC-PS as a unique oil sorbent prepared by an electrospinning process. Up to a 9 times increase in the oil, uptake was noted for these sorbent fibers as compared to commercial PP sorbent due to the voids present in PS-PVC fibers. A one-step electrospinning process has been demonstrated for the fabrication of nanoporous polystyrene fibers by Lin et al. (2012). Due to the high porosity of PS fibers, a maximum oil sorption capacity of 113.87 g.g⁻¹ was obtained for motor oil. The uptake efficiency of certain edible oils was also found to be good with an oil absorption capacity of about 111.80 g.g⁻¹ achieved for bean oil.

Melamine

Melamine sponges consist of formaldehyde-melaminesodium bisulfite copolymer. Owing to the high porosity (>99%), extensive pore structure, large surface area, low density, and ease of chemical modification, they are finding application in the treatment of oily wastewater as sorbents (Wang et al. 2015). A highly oleophilic composite comprising melamine, MOF (HPU-13) crystals, and carboxymethyl cellulose (CMC) was prepared by Xu et al. (2019). CMC sodium-treated melamine sponge (MS) sponge was able to facilitate the attachment of HPU-13 onto the MS leading to the formation of an excellent hydrophobic material proven by its oil uptake capacity of up to 13000%. A hydrophilic cellulose polymer, carboxymethylcellulose sodium (CMC) was used to impart adequate coordination of functional groups on the surface of MS. CMC was envisioned to wrap

the branches of the sponge with hydroxyl and carboxylic groups enabling greater interaction with metal ions to strongly link HPU-13 to MS. Feng et al. (2017) modified a commercially available melamine sponge with eco-friendly material, furfuryl alcohol. The MS was soaked in furfuryl alcohol solution followed by acidic polymerization to achieve surface modification. This method resulted in a reaction of furfuryl alcohol with MS along with polymerization of furfuryl alcohol onto the surface of MS, thereby making the sponge hydrophobic. The modified MS displayed excellent sorption capacities for various oils and organic solvents (75-160 g.g⁻¹), resulting in a good hydrophobic, environmentfriendly modified melamine sponge. Xue et al. (2021) investigated a unique superhydrophobic and superoleophilic tetradecyl amine-MXene functionalized melamine sponge for effective oil/water separation through selective oil adsorption. The results showed an absorption capacity of 60-112 times the weight of the sorbent. The functionalized melamine sponge exhibited high mechanical strength, and chemical stability, as well as good reproducibility and recyclability for oil-water separation. Arunagiri et al. (2021) fabricated a chemically modified 3D porous MS with polycaprolactone and poly-D, L-Lactic acid via inexpensive freeze-drying. The sponge displayed high hydrophobicity with a water contact angle of 162°. The composite material showed an absorption capacity in the range of 3.3-8.7 g.g⁻¹ for several oils and organic solvents such as crude oil, soyabean oil, n-hexane, n-octane, etc.

POLYMERIC SORBENTS AS SOLIDIFIERS

Conversion of spilled oil into a solid coherent mass using solidifiers could be a viable option for efficient oil-water separation. Though quite an attractive possibility, solidifiers have remained an underutilized approach to alleviate the effects of oil spills. Solidifiers can potentially help in oilwater separation by containing the oil spill, preventing it from spreading, and promoting its removal from the environment, as well as aiding the recovery of oil. Shojaei et al. (2021) demonstrated that oil spills can be solidified using polypropylene and polyethylene fibers and granules. The performance of these polymeric fibers and granules was analyzed by gravimetric and spectrophotometric methods. The results showed a high sorption capacity of 12.5 g.g⁻¹ for polypropylene fibers. The Freundlich isotherm model is consistent with the sorption characteristics of PP and PE granules owing to the heterogeneous surface of these two solidifiers. Oil adsorption tendency was attributed to the heterogeneous surface of polymeric granules with varying adsorption energies which resulted in enhanced oil uptake. Motta et al. (2019) demonstrated an inexpensive and simple method for the fabrication of a bio-based subclass of solidifiers constructed from sorbent amylopectin-graftpoly (methyl acrylate) (AP-g-PMA) polymer. The results indicated that the rapid solidification of diluted bitumen in terms of contact time and solidifier-to-oil ratio could be achieved within a few minutes by AP-f-PMA. The combination of the biodegradability of amylopectin and the hydrophobicity of the synthetic polymer holds a lot of potential. Another recent study by (Lv et al. 2020) showed the application of polymer-grafted wheat bran for oil gelling capability. Vinyl tri ethoxy silane was used to introduce vinyl functionality onto the wheat bran, which was further polymerized with divinyl benzene monomer, using azo bis isobutyronitrile as the initiator. The methodology rendered the wheat bran with a large BET surface area of 176 m².g⁻ ¹ and a maximum absorption capacity of 12.0 g.g⁻¹ for various oily liquids. The sorbent material was found to exhibit excellent hydrophobicity and oleophilicity due to the presence of a multi-scale porous structure.

SORBENT GELS FOR OIL SPILL CLEAN-UP

Unique and efficient oleophilic sorbent gels based on glycerol propoxylate, synthesized through a bulk polymerization technique without the use of an activator, an initiator, or a catalyst, have been reported by (Kizil & Sonmez 2017). Various organic solvents and oils were selected to study the absorption tendency of the synthesized gels. Varying concentrations of the crosslinkers were examined for their effect on the above-mentioned parameters wherein star-type propoxylate monomers were reacted with different concentrations of tris [3-(trimethoxysilyl) propyl] isocyanurate (ICS) crosslinker. For the PDMS sorbents, the capability to swell was found to depend on the amount of cross-linking in the polymers. Fully swollen

material obtained after 24 h, was analyzed for the oil/solvent retention ability by determining the polymer weight loss avoiding the use of any external forces such as squeezing or exposing the sorbents to high temperatures. Janqamsari et al. (2021) developed ecofriendly sorbent aerogels based on recycled polyethylene terephthalate (PET) fibers chemically modified with the polymer polyvinyl alcohol (PVA) and further with carbon nanotubes (CNTs). The incorporation of CNTs in the structure of nanocomposite sorbent increases the oleophilicity and thereby increases the oil sorption capacity. The surface area, entanglements among the fibers and the hydrophobic interaction between the sorbents and oil droplets significantly affect the sorption capacity. The nanocomposite sorbent shows efficient oil sorption capacity and reusability even after four cycles. They investigated the efficiency of the sorbent aerogels for their density, porosity, and oil sorption capacity which were found to be 0.043-0.097 g.cm⁻³, 92.93-96.87%, and ~ 12 g.g⁻¹ for kerosene oil respectively. In another study, Shen et al. (2021) demonstrated the applicability of superelastic and strong polyimide (PI) nanofiber-based aerogels (NFAs) aided by a three-dimensional (3D) structure and porosity that can also be altered. The aerogels were prepared by freeze-drying followed by solvent-vapor treatment as illustrated in Fig. 5. Silicone nanofilaments (SiNFs) were then formed on the surface of PI fibers upon treatment with trichloromethyl silane. SiNFs-coated NFAs exhibited very low density, high porosity, and very high oil sorption capacity of 159x its own weight. Separation of stable water-in-oil emulsions was also achieved indicating that the method was very energy efficient. Ali (2019) has synthesized and demonstrated the use of an oleophilic gel based on butyl acrylate polymer for the remediation of oily wastewater. The polymer, poly (n-butyl acrylate) was synthesized by solution polymerization and various parameters such as reaction time,



Fig. 5: Schematic illustration of the preparation of SiNFs/PI-NFAs. (Reprinted with permission from Shen et al. 2021. Copyright © 2021, American Chemical Society).

the concentration of the monomer, initiator, crosslinker, and the temperature were optimized for improved yield of the polymer sorbent. The sorption capacity of the sorbent under different conditions like dry system, static system, and the dynamic system was found to be quite promising for various oils such as gasoline, kerosene, diesel, xylene, octanol, and Mobil oil.

MISCELLANEOUS SYNTHETIC SORBENTS

Fabrication of poly(dimethylsiloxane) based hydrophobic sorbents through the condensation reaction of poly(dimethylsiloxane) and tris[3-(trimethoxysilyl)propyl] isocyanurate cross-linker via bulk polymerization was demonstrated by Askin et al. (2021). High oil absorption capacity and a high rate of absorption-desorption were achieved. The sorbents could be easily reused many times without any major loss in the oil-uptake capacity. The hydrophobic polymeric sorbents have an additional advantage in that they selectively absorb oil from the water surface which increases their potential use in water clean-up applications. Additionally, Askin et al. (2021) also developed a unique hybrid sorbent via a bulk polymerization reaction between poly (dimethylsiloxane) and fluorinated alkyl silane. The highly hydrophobic structure of the material resulted in good oil uptake from both the surface and the bulk of water. High oil sorption capacity in the range of 500-1800% and fast uptake and release of oil were also observed indicating promising clean-up of oil spills. Krishnan et al. (2022) have recently demonstrated through in-situ one-step bulk copolymerization, the fabrication of a composite absorbent with a three-dimensional cross-linked network comprised of polystyrene-polymethyl methacrylate/divinyl benzene (PS-PMMA/DVB) copolymer and 2D-nanosheets of BN₃ (Boron Nitride)/graphene. The as-prepared composite material was found to efficiently remove various organic solvents and oils from the spills and showed good flexibility. The absorption capability was found to be dependent on the amount of DVB used. Interestingly, the PS-PMMA/DVB composites with BN₃ nanosheets exhibited higher absorption capacities in comparison to the ones with CG composites. The polymer composites with lower nanosheet content, i.e., 0.1 wt% were observed to have higher absorption capacities compared to the ones with higher nanosheet content (2.0 wt%).

The conversion of nonwoven polyester fibers into hydrophobic oil-absorbing material by treating the fibers with poly (methylhydrodimethyl) siloxane copolymers has been

Table 2: Other Modified Polymeric Sorbents.

Sorbent	Treatment Material	Method	Sorption Capacity (g/g)	Sorption Experimental Conditions	Reference
PS foam	Oleic acid-coated Fe ₃ O ₄	Two-step immersion	40	Maximum absorption capacity reached in 60 s	Yu et al. 2016
PS fibers	Silica-coated iron oxide nanoparticles	Grafting	Diesel: 3 Diluted Bitu- men: 5	20 mg of the sorbent for 5 minutes at 23°C.	Damavandi & Soares 2022
Syndiotactic PS (sPS) mon- oliths	Trichlorobenzene (TCB) & Glycerol	High internal phase emulsion crystalliza- tion-induced gelation	41.9-81.3	Cubic monoliths $(\square 1.0 \times 1.0 \times 1.0 \times 1.0 \text{ cm}^3)$ with known mass immersed in 20 mL of o/w emulsion for 12 hours.	Gui et al. 2019
PS foam waste	Silica and Span 80	High internal phase pickering emulsion (HIPPE)	20.4-58.1	0.1 g cylindrical-shaped foam sample was immersed in oil at RT for 5 min.	Yu et al. 2019
PS	Carbon nanotubes	Electrospinning	111.45-122.82	Sorbent immersed in pure oil for 90 min at RT.	Wu et al. 2017
PS fibers	Blended with PVC and PE individually	Electrospinning	112-119	0.1 g of sorbent was placed onto the oil surface for 15 minutes.	Alnaqbi et al. 2020
PS -PVC blended fibers	Not Applicable	Electrospinning	81-146	1 g of sorbent was placed on top of 300 mL of oil at 23 ±4 °C for 60 min. and drained for 2 min before weighing.	Zhu et al. 2011
PS nanoporous fibers	Not Applicable	Electrospinning	Motor oil: 113.87 Bean oil: 111.80 Sunflower seed oil: 96.89	0.1 g of sorbent was dropped onto the oil film and drained for 10 s.	Lin et al. 2012

Table cont....

Sorbent	Treatment Material	Method	Sorption Capacity (g/g)	Sorption Experimental Conditions	Reference
PS nanofibres	Sawdust	Electrospinning	Motor oil: 1.41	Reaction temperature = 25°C,	Ghiasvand
			Diesel oil: 1.4	pH = 4, contact time = 1 min.	et al. 2021
			Furnace oil: 1.5		
			Crude oil: 1.1		
		Impregnation	Motor oil: 1.00		
			Diesel oil: 0.63		
			Furnace oil: 0.97		
			Crude oil: 0.59		
PS/Fe ₃ O ₄ nanofibres	Fe ₃ O ₄ nanoparticles	Electrospinning	Edible oil: 87 Saxoline: 65 Dimethicone: 94	Sorbent drained for 2 minutes before weighing.	Song et al. 2017
rPS fabric	Orange peel extract	Electrospinning	40.5±3.6	0.2 g sorbent in 100 mL of oil for 60 min	Yadav et al. 2019
Expanded PS waste	Mechanical modification with holes puncture	Mechanical Punc- turing	Crude oil: 3.26 Lubricating oil: 4.9 Distilled water: 0.32	0.9 g of sorbent was placed on the surface of the oil/water mixture at RT	Nasir & Hameed 2014
PS/PVC fabric	PVC	Electrospinning	Motor oil: 146 Peanut oil: 119 Diesel: 38 Ethylene Glycol: 81	1 g of sorbent was placed on the top layer of 300 ml oil at 23±4°C for 60 min and drained for 2 min before final weighing.	Zhu et al. 2011
Melamine sponge	MOF crystals	Not Applicable	13000%	Not Specified	Xu et al. 2019
Melamine sponge	Furfuryl alcohol	Immersion followed by polymerization	75-160	Not Specified	Feng et al. 2017
Melamine sponge	Tetradecylamine-MXene	Immersion	60-112	Sorbent immersed into oil/ water mixture at RT. Thermal and chemical stability tested at pH range 2-11 for 24 hours.	Xue et al. 2021
Melamine sponge	Polycaprolactone and poly-D, L-Lactic acid	Freeze drying	3.3-8.7	Not Specified	Arunagiri et al. 2021
PP and PE fib- ers & granules	Not Applicable	Not Applicable	12.5	Crude oil (0.5 mL, 0.49 g) in 80 mL water. Volumes of crude oil and saline water were kept constant, while the solid- ifier mass varied depending on the SOR ratio.	Shojaei et al. 2021
Poly(methyl acrylate)	Amylopectin	Not Applicable	Not Available	The sorbent was tested at RT on a 0.2 mm thick oil layer on top of fresh water by using a contact time of 8 min for crude oil while 32 min for diluted bitumen.	Motta et al. 2019
Divinyl ben- zene-vinyltri- ethoxysilane	Wheat bran	Polymerization	12	Oil gelling experiment of the bran sample conducted for 10 min at RT	Lv et al. 2020
Glycerol propoxylate	Tris[3-(trimethoxysilyl) propyl]isocyanurate (ICS) as crosslinker	Bulk Polymerization	Not Available	Wire mesh basket sorbent soaked in an organic solvent for sorption studies at RT.	Kizil & Sonmez 2017

Table cont....

Sorbent	Treatment Material	Method	Sorption Capacity (g/g)	Sorption Experimental Conditions	Reference
Recycled polyethylene terephthalate fibers	Polyvinyl alcohol followed by carbon nanotubes	Not Applicable	12	Performance of rPET-based oil sorbents studied at RT	Janqamsari et al. 2021
Polyimide aerogels	Silicone nanofilaments (SiNFs) formed on the surface of PI fibers	Freeze drying fol- lowed by solvent-va- por treatment	159	Surfactant (Span 80, 10 mg) dissolved in 100 mL of organic liquid and homogenized at 10,000 rpm for 5 min.	Shen et al. 2021
Poly(n-butyl acrylate)	Not Applicable	Solution Polymeri- zation	Not Available	Not Specified	Ali 2019
Poly(dimethyl- siloxane)	Tris[3-(trimethoxysilyl) propyl] isocyanurate cross-linker	Bulk Polymerization	Not Available	Maximum absorbancy of 85- 90% achieved in 40 minutes.	Askin et al. 2021
Polydimethyl- siloxane	Fluorinated alkyl silane.	Bulk Polymerization	500-1800%	Highest sorption capacity of 1800% observed for dichloro- methane due to its high polarity	Askin et al. 2021
PS-polymethyl methacrylate/ divinyl benzene	2D-nanosheets of BN ₃ / graphene	Bulk Polymerization	Not Available	Sorption efficiency depends upon the polarity of adsorbate and the study was conducted in the temperature range of 30-60 °C.	Krishnan et al. 2022
Polyester fibers	Poly(methylhydrodimethyl) siloxane copolymers	Heterogeneous polym- erization	Dodecane: 5.52	Square-shaped sorbent immersed in oil and reaches saturation in 15 min and drained for 30 s before weighing and tested at 23 ± 4 °C.	Cojocaru et al. 2017
			Motor oil: 10.03		
Polysiloxane	Viscose-maghemite/ goethite magnetic nano- particles	Not Applicable	Liquid HC: 15-17	Sorbent sample (0.15-0.2 g) dipped in oil for 15 min till saturation and drained for 30 s before weighing.	Rotaru et al. 2019
			Motor oils: 35-37		
PP/polyester micro/ nano- fibres	Not Applicable	Three-step process including blending, drawing and etching	21.9	CCl ₄ /water mixture was passed through the sorbent as a filter set between two glass tubes. CCl ₄ passed through the filter in 60 s and water retained in the upper glass tube.	Zhang et al. 2020
Polyether sul- fone (PES)	Zeolitic imidazolate (ZIF-8) framework (MOF)	One-step phase inver- sion method	Paraffin Oil: 37.6% (v/v)	Sorption at RT for 30 min.	Abbasi et al. 2017
			Olive Oil: 35.6% (v/v)		

demonstrated by Cojocaru et al. (2017). Novel polysiloxane copolymers were synthesized which were then used to treat polyester fibers to make them more hydrophobic. The hydrophobic polyester fibers were evaluated for their oil-absorbing capacities, which were found to be 5.52 g.g⁻¹ and 10.03 g.g⁻¹ for dodecane and motor oil, respectively. Optical microscopy identified the presence of inter-fiber voids which were responsible for oil absorption. Good recyclability of the sorbent was achieved as demonstrated by centrifugation tests.

The technique of engraving or etching has been used to modify the surface morphology of micro- and nanofibres to improve their oil sorption capacity. Another hydrophobic modification of polysiloxane with viscose-maghemite/ goethite magnetic nanoparticles has been reported by Rotaru et al. (2019). The material was found to exhibit a sorption capacity of 15-17 g.g⁻¹ for liquid hydrocarbons and 35-37 g.g⁻¹ for motor oils uptake. A multi-step procedure was employed to obtain the polymeric composite. Groovelike rough morphology was engraved on nonwoven polypropylene/polyester microfibrous and nanofibrous material by Zhang et al. (2020), using a three-step process that involved blending, drawing and etching. The oil adsorption results revealed that the nonwoven nanofibres had oil-saturated adsorption of 21.9 g.g⁻¹ and an oil retention rate of 10.3 g.g⁻¹. An excellent oil/water separation efficiency of 99% was obtained driven solely by gravity. Coating of the zeolitic imidazolate framework (ZIF-8) on polyether sulfone (PES) was successfully carried out by Abbasi et al. (2017) through a simple one-step phase inversion method. Phase inversion refers to the process in which a homogenous solution of a polymer (in solvent) is reverted from a single phase into a two-phase system. The two-phase system consists of a solid polymer phase which forms the membrane structure and a liquid (polymer-poor) phase which forms the pores in the final film. In the composite material, the ZIF-8 particles were held together by the PES, and the ZIF-8 was responsible for effectively absorbing oil. A summary of other modified polymeric sorbents is given in Table 2.

CHEMICALLY MODIFIED NATURAL SORBENTS

Natural adsorbents used for oil spill cleanup have been reported as the most economical, eco-friendly, and costeffective promising technique. Some of the bio-based sorbents used for crude oil removal include chitosan, bagasse from sugar cane and pith, cotton, dried straws, fibers of palm, banana trunk and vegetables, byproducts of cork, rice hulls and fibers, onion and garlic peels, kapok, coconut fibers, and shells of various nuts, etc. Chemical modifications of these bio-based sorbents by acid and alkaline treatments have been reported to increase their sorption capacity for various pollutants (Cao et al. 2017, Gheriany et al. 2020, Zamparas et al. 2020).

Chitosan

Biobased aerogels/sorbents offer great advantages in terms of natural abundance, low cost, environment-friendly, and biodegradability as compared to synthetic polymers. Chitosan is a naturally abundant polymer that has attracted great attention worldwide due to its extraordinary properties as biodegradability and the presence of reactive functional groups (Vidal & Moraes 2019). Hu et al. (2020) successfully fabricated aerogels of graphene oxide-modified chitosan nanosheets, followed by freeze drying of the nanosheets with silicon particles and PDMS, to impart hydrophobicity to the modified aerogels for improved oil absorption. The composite aerogel showed promising adsorption capacity in the range of 18-45 g.g⁻¹ for various oils and solvents. Since the recovery of adsorbed oil is usually achieved through the mechanical extrusion of the oil-loaded sorbents, the aerogels with better compressibility and stability exhibited the highest oil recovery. In another study, Yi et al. (2020) constructed a chitosan aerogel composite in which methyl trimethoxy silane was bonded to the aerogel through chemical vapor deposition. The rapid absorption of various oils and solvents and oil absorption capacities in the range of 31-63 g.g⁻¹ were observed. The chitosan-based aerogel also displayed

excellent reusability and recyclability, with rapid recovery of oil by mechanical squeezing. Li et al. (2018) investigated the applicability of a unique chitosan-cellulose aerogel which was prepared by oxidation, crosslinking, freezedrying, and cold plasma modification for remediation of oily wastewater. A cellulosic aerogel was prepared by freeze drying, followed by oxidation with NaIO₄ and exposure to an acetic acid solution containing 2% w/w of chitosan. The amino groups from chitosan reacted with aldehydic groups of oxidized cellulose resulting in crosslinking between the two molecules, followed by treatment with trimethyl chlorosilane (TMCS) to enhance the aerogel's hydrophobicity. Due to the rapid absorption rate, high absorption capacity (13.77-28.20 g.g⁻¹), high oil/water selectivity, and good reusability demonstrated by the chitosan aerogel, it can be considered a promising sorbent for oil/water separation.

Modified Celluloses

Cellulosic sorbents are natural plant-based materials and have been more widely used as effective sorbents due to their biodegradability compared to other synthetic materials. Cellulose and lignin materials have been hydrophobically modified by physical or chemical methods (Valdebenito et al. 2018). Physical modifications include cold plasma coating, surface crystal growth, grafting, and sol-gel conversion treatments (Galblaub et al. 2016, Koch & Barthlott 2009). Chemical modifications include hydrophobic modifications with nanomaterials and polymers. Cotton fabric was rendered superhydrophobic upon treating the fabric with dopamine in presence of FeCl₃, causing self-polymerization of dopamine (polydopamine (PDA)). The PDA-cotton fabric was then treated with octadecyl amine (ODA) (Yan et al. 2020) to form a nano construct that exhibited a 2x surface roughness with greater strength and robustness compared to untreated cotton fabric. The oil separation efficiency was observed to increase by 96% which could be attributed to the high adhesion capacity of PDA. An additional improvement in oil/water separation ability could be achieved by further wrapping the Fe/PDA/ODA fabric around the PU sponge to fabricate an adsorption bag.

A superhydrophobic magnetic cellulose sponge was fabricated by deposition of Fe_3O_4 nanoparticles on the cellulose sponge which was further modified by immersion in an ethanolic solution of hexadecyltrimethoxysilane (Peng et al. 2016). The oil sorption capacity and oil separation efficiency of natural cellulose were increased by more than 95% due to the acetylation of the modified sponge.

Modified and unmodified date palm fibers (PFs) were used as natural sorbent material for oil spill removal by Abdelwahab et al. (2017). Various oils such as diesel oil, crude oil, and vegetable oil were chosen to study the efficacy of the sorbent fibers. Simple chemical modification was carried out on PFs by soaking them in 2% w/v NaOH and 2% v/v H₂SO₄ separately at room temperature overnight, followed by drying at 80°C. The acid-base treatment made the fiber surface rough which affected the interaction between the inner surface of the fibers and the oil such that high sorption efficiency was achieved. In another study by Neelamegan et al. (2020), chemical modification of cellulose and lignin was carried out in the presence of carbon nanotubes (CNT) under basic (NaOH) conditions using CNT-COC1. The material was further reinforced with iron oxide to enable its purification. The increased hydrophobicity of the sorbent was proven by an increased water contact angle in the range of 1300135°. Sorption capacities for CNT-g-Cellulose/Fe₃O₄ and CNT-g-Lignin/Fe₃O₄ were found to be 15-25 and 10-23 g.g⁻¹ for a variety of oils and organic solvents.

Rice Husk

Wang et al. (2020) demonstrated the biological delignification of cellulose derived from rice husk followed by acetylation to prepare a hydrophobic sorbent. Both fungi and bacteria were employed to conduct the delignification process. The fabricated cellulosic sorbent exhibited good hydrophobicity which was evident in a sorption capacity of 20 g/g. In this study, Langmuir and Freundlich isotherms were used to analyze the sorption capacity, which indicated that the oil sorption capacity of rice husk fit the Langmuir model better than the Freundlich model. Through simple acetylation of oil palm empty fruit bunch (OPEFB) and cocoa pod (CP) fibers under mild conditions, Onwuka et al. (2018) fabricated a sorbent material for the sequestration of oil from water. A comparison of the acetylated and unacetylated sorbents indicated that the differences in their sorption capacities were dependent on time and concentration. Acetylation improved the oil sorption capacity of CP fibers. Kinetic studies demonstrated that the oil sorption was diffusion-controlled for both physisorption and chemisorption.

Thermally enhanced rice husk montmorillonite combination (TRMC) was evaluated for handling crude oil pollution (Akpomie et al. 2018). Studies revealed that the modified sorbent (TRMC) was more efficient in treating oilcontaminated water compared to the unmodified sorbent. It was observed that the sorption efficiency was significantly dependent on pH, temperature, the concentration of crude oil, sorbent-oil contact time, and dosage of sorbent. TRMC exhibited the maximum sorption potential as compared to the other reported sorbents which have been used for remediation of the crude oil spill.

Other Natural Sorbents

Seashells and epoxy resins have been used to modify PU sponges to create eco-friendly, robust, superhydrophobic, and super oleophilic adsorbents for the removal of oil from polluted water (Jamsaz & Goharshadi 2020). The clean PU sponge was immersed in a suspension of treated seashell powder followed by the addition of epoxy resin. The modified PU sponge was then freeze dried resulting in a sorbent material that exhibited good recyclability and high oil adsorption capacity in the range of 28.03-42.17 g.g⁻¹.

Eze et al. (2019) have shown the use of a unique biomass i.e., *Dialium guineense* (velvet tamarind) seed husk (DGSH) chemically treated with ammonium sulfate (AS-DGHS) for the remediation of oily water collected from the Niger Delta region in Nigeria. The surface morphology of both modified and unmodified sorbents exhibited a certain degree of porosity, with AS-DGSH possessing a greater porosity than DGSH indicating that the chemical modification was able to alter the surface of the biomass material. Around 50% of oil sequestration was achieved from polluted water at a pH of 6.1. However, a significant decrease in sorption was observed for both DGSH (41.2 to 20.3%) and AS-DGSH (52.6 to 27.2%) with an increase in pH from 3.0 to 9.0.

The adsorption capacity of raw luffa fibers was evaluated for various oil/water emulsions (Abdelwahab et al. 2014). High selectivity was exhibited by luffa fibers for various oils along with excellent water repellency in its original form. The contact time and dose of adsorbent were observed to have a significant effect on the sorption capacity such that it decreased as the amount of adsorbent increased and vice versa. A maximum sorption capacity was observed at 40°C for diesel oil. Other parameters that affected the efficiency of the fibrous sorbent were surface properties, temperature, the concentration of the crude oil, and the amount of fiber used. Raw luffa fibers were not very reusable since their sorption efficiency was observed to decrease after three sorption trials.

The sorption capacity of raw and acetylated sugarcane bagasse was determined for crude oil (Behnood et al. 2016). It was observed that the sorption capacity of acetylated bagasse was 90% greater than that of raw bagasse mainly due to the oleophilicity of acetylated bagasse. A sorption capacity of 11.3 g.g⁻¹ of crude oil and 9.1 g.g⁻¹ of oil layer in an emulsion was observed. The sugarcane bagasse, therefore, offers a significantly improved, low-cost method for the treatment of hydrocarbon impurities in aqueous systems (Boni et al. 2016). The porous nature and fibrous network structure of natural luffa sponge were observed to assist in the removal of dye and other solvents through filtration and absorption mechanisms (Wang et al. 2017). Surface modification of

luffa with polyhedral oligomeric silsesquioxane (POSS) imparted superhydrophobicity to the luffa sponge and significantly enhanced its oil absorption capacity.

CONCLUSIONS

This review article evaluates the advances made in oil spill clean-up techniques based on the use of modified polymeric and natural sorbents. The methods reported by various researchers show promising prospects in dealing with such a hard pressing problem as the remediation of oil spills and oily wastewater. The treatment of oily wastewater requires a great deal of research in developing cost-efficient, recyclable and effective materials that can be employed in cleaning oil spills. The sorption characteristics of polymeric or natural oil sorbents depend on material morphology, especially for sponge-type sorbents which have a three-dimensional porous structure. Therefore, an effort should be made to understand the structure-property relationships of porous materials with respect to the effect of pore size, shape, and porosity on the uptake capacity and kinetics of various oils and organic solvents. An assessment of the performance of various sorbent materials developed through innovative processes which function in more efficient ways as compared to other typical methods used for oil spill clean-up has been the focus of this article. Surface roughness and high porosity are some of the characteristics gained from the surface modification of polymeric sorbents which have resulted in an enhancement of their oil sorption capacities. In addition, the ability to recover the oil from the sorbent and its reusability have also been discussed. The recyclability of the sorbents in efficient and continued extraction and separation of oil from spills and wastewater which will allow the recovery of potentially lost oil are critical endeavors in oils/organic solvents cleanup applications.

Identification of materials with excellent separation efficiency, elasticity, recyclability, and durability is therefore imperative for the viability of these sorbents in large-scale usage for remediation of global oil spill disasters. The development of super-wetting materials that are highly stable and durable is necessary for continued advancement in spill-cleanup technology. Additionally, sorbents with flame-retardant properties will help minimize fire hazards that usually occur during an active oil spill and minimize the spread of flammable gas and the generation of toxic smoke.

Significant challenges still exist when it comes to practical applications for synthetic sorbents, such as the fabrication cost, environmental impact, the strength of absorbent materials, and the oil treatment after recovery. Efforts are therefore needed to make the sorbent modification processes and techniques for functionalization, inexpensive, and

relatively easy to conduct on a large scale. Furthermore, the environmental and economic impact of the techniques used in the fabrication of various chemically modified sorbents should be thoroughly evaluated. The techniques discussed herein have so far been studied on a lab scale, and further evaluation at an industrial scale needs to be conducted. A combination of a variety of sorbent modification techniques may be considered in identifying a cost-effective solution for mitigating an oil spill, while also considering environmental and economic implications. Further advancements in this field will include the generation of new functional oil-absorbing materials which are suitable for optimized, low-cost, large-scale manufacture of sorbents that are themselves environment-friendly for widespread usage during oil spill containment and oily wastewater treatment. Extensive research and funding will be needed to achieve these goals.

REFERENCES

- Abbasi, Z., Shamsaei, E., Fang, X. Y., Ladewig, B. and Wang, H. 2017. Simple fabrication of zeolitic imidazolate framework ZIF-8/polymer composite beads by phase inversion method for efficient oil sorption. Journal of Colloid and Interface Science, 493: 150-161. https://doi. org/10.1016/j.jcis.2017.01.006
- Abdelwahab, O. 2014. Assessment of raw luffa as a natural hollow oleophilic fibrous sorbent for oil spill cleanup. Alexandria Engineering Journal, 53(1): 213-218. https://doi.org/10.1016/j.aej.2013.11.001
- Abdelwahab, O., Nasr, S. M. and Thabet, W. M. 2017. Palm fibers and modified palm fibers adsorbents for different oils. Alexandria Engineering Journal, 56(4): 749-755. https://doi.org/10.1016/j. aej.2016.11.020
- Akpomie, K. G., Ezeofor, C. C., Olikagu, C. S., Odewole, O. A. and Ezeorah, C. J. 2018. Abstraction and regeneration potential of temperatureenhanced rice husk montmorillonite combo for oil spill. Environmental Science and Pollution Research, 25(34): 34711-34719. https://doi. org/10.1007/s11356-018-3425-9
- Ali, S. 2019. Synthesis and characteristics of oleophilic gel polymer for oil spills cleanup. African Journal of Engineering Research, 7(3): 85-93. https://doi.org/10.30918/AJER.73.19.032
- Alnaqbi, M. A., Al Blooshi, A. G. and Greish, Y. E. 2020. Polyethylene and polyvinyl chloride-blended polystyrene nanofibrous sorbents and their application in the removal of various oil spills. Advances in Polymer Technology, 1-12. https://doi.org/10.1155/2020/4097520
- Arunagiri, V., Prasannan, A., Udomsin, J., Lai, J.Y., Wang, C.-F., Hong, P.-D. and Tsai, H. C. 2021. Facile fabrication of eco-friendly polycaprolactone (PCL)/Poly-D, L-Lactic acid (PDLLA) modified melamine sorbent for oil-spill cleaning and water/oil (W/O) emulsion separation. Separation and Purification Technology, 259: 118081. https://doi.org/10.1016/j.seppur.2020.118081
- Askin, S., Kizil, S. and Bulbul Sonmez, H. 2021. Creating of highly hydrophobic sorbent with fluoroalkyl silane cross-linker for efficient oil-water separation. Reactive and Functional Polymers, 167: 105002. https://doi.org/10.1016/j.reactfunctpolym.2021.105002
- Askin, S., Kizil, S. and Bulbul Sonmez, H. 2021. Hydrophobic crossIlinked poly(dimethylsiloxane)Ibased sorbents for oil spill applications. Macromolecular Materials and Engineering, 306(2): 2000556. https:// doi.org/10.1002/mame.202000556
- Atlas, R. M. and Hazen, T. C. 2011. oil biodegradation and bioremediation: a tale of the two worst spills in U.S. history. Environmental Science and Technology, 45(16): 6709-6715. https://doi.org/10.1021/es2013227

- Atta, A. M., Abdullah, M. M. S., Al-Lohedan, H. A. and Mohamed, N. H. 2019. Novel superhydrophobic sand and polyurethane sponge coated with silica/modified asphaltene nanoparticles for rapid oil spill cleanup. Nanomaterials, 9(2). https://doi.org/10.3390/nano9020187
- Baig, N., Alghunaimi, F. I., Dossary, H. S. and Saleh, T. A. 2019. Superhydrophobic and superoleophilic carbon nanofiber grafted polyurethane for oil-water separation. Process Safety and Environmental Protection, 123: 327-334. https://doi.org/10.1016/j.psep.2019.01.007
- Behnood, R., Anvaripour, B., Jaafarzadeh, N. and Farasati, M. 2016. Oil spill sorption using raw and acetylated sugarcane bagasse. Journal of Central South University, 23(7): 1618–1625. https://doi.org/10.1007/ s11771-016-3216-8
- Biesuz, M., Tomasi, M., Santhosh, B., Sglavo, V. M. and Sorarù, G. D. 2020. Polymer-derived Si3N4 nanofelts as a novel oil spills clean-up architecture. Journal of Environmental Chemical Engineering, 8(5): 104134. https://doi.org/10.1016/j.jece.2020.104134
- Boni, H. T., de Oliveira, D., Ulson de Souza, A. A. and Ulson de Souza, S. M. A. G. 2016. Bioadsorption by sugarcane bagasse for the reduction in oil and grease content in aqueous effluent. International Journal of Environmental Science and Technology, 13(4): 1169-1176. https://doi. org/10.1007/s13762-016-0962-y
- Cao, N., Yang, B., Barras, A., Szunerits, S. and Boukherroub, R. 2017. Polyurethane sponge functionalized with superhydrophobic nanodiamond particles for efficient oil/water separation. Chemical Engineering Journal, 307: 319-325. https://doi.org/10.1016/j. cej.2016.08.105
- Cao, S., Dong, T., Xu, G. and Wang, F. 2017. Oil spill cleanup by hydrophobic natural fibers. Journal of Natural Fibres, 14(5): 727-735. https://doi.org/10.1080/15440478.2016.1277820
- Cojocaru, C., Pricop, L., Samoila, P., Rotaru, R. and Harabagiu, V. 2017. Surface hydrophobization of polyester fibers with poly(methylhydrodimethyl)siloxane copolymers: Experimental design for testing of modified nonwoven materials as oil spill sorbents. Polymer Testing, 59: 377-389. https://doi.org/10.1016/j.polymertesting.2017.02.024
- Damavandi, F. and Soares, J. B. P. 2022. Polystyrene magnetic nanocomposite blend: An effective, facile, and economical alternative in oil spill removal applications. Chemosphere, 286(April 2021). https:// doi.org/10.1016/j.chemosphere.2021.131611
- El Gheriany, I. A., Ahmad El Saqa, F., Abd El Razek Amer, A. and Hussein, M. 2020. Oil spill sorption capacity of raw and thermally modified orange peel waste. Alexandria Engineering Journal, 59(2): 925-932. https://doi.org/10.1016/j.aej.2020.03.024
- Eze, S. I., Akpomie, K. G., Ezeofor, C. C., Mmadubuike, N. V. and Ojo, F. K. 2019. Remediation of oil spill polluted water from Niger Delta Nigeria by sorption onto ammonium sulfate modified Dialium guineense seed husk. Petroleum Science and Technology, 37(15): 1830-1838. https:// doi.org/10.1080/10916466.2019.1608240
- Feng, Y., Wang, Y., Wang, Y. and Yao, J. 2017. Furfuryl alcohol modified melamine sponge for highly efficient oil spill clean-up and recovery. J. Mater. Chem. A, 5(41): 21893-21897. https://doi.org/10.1039/ C7TA06966A
- Fingas, M. 2012. The Basics of Oil Spill Cleanup. CRC Press. https://doi. org/10.1201/b13686
- Galblaub, O. A., Shaykhiev, I. G., Stepanova, S. V. and Timirbaeva, G. R. 2016. Oil spill cleanup of water surface by plant-based sorbents: Russian practices. Process Safety and Environmental Protection, 101: 88-92. https://doi.org/10.1016/j.psep.2015.11.002
- Ghiasvand, S., Rahmani, A., Samadi, M., Asgari, G., Azizian, S. and Poormohammadi, A. 2021. Application of polystyrene nanofibres filled with sawdust as separator pads for separation of oil spills. Process Safety and Environmental Protection, 146: 161-168. https://doi.org/10.1016/j. psep.2020.08.044
- Gui, H., Zhang, T. and Guo, Q. 2019. Nanofibrous, emulsion-templated syndiotactic polystyrenes with superhydrophobicity for oil spill

cleanup. ACS Applied Materials and Interfaces, 11(39): 36063-36072. https://doi.org/10.1021/acsami.9b10467

- Gui, X., Zeng, Z., Lin, Z., Gan, Q., Xiang, R., Zhu, Y., Cao, A. and Tang, Z. 2013. Magnetic and highly recyclable macroporous carbon nanotubes for spilled oil sorption and separation. ACS Applied Materials and Interfaces, 5(12): 5845-5850. https://doi.org/10.1021/am4015007
- Guo, J., Wang, J., Zhang, S., Ma, X., Qiu, Z., Peng, X., Ying, J., Wang, Y. and Wu, G. 2017. One-step modification of PU sponges for selective absorption of oil–water mixtures. New Journal of Chemistry, 41(1): 90-96. https://doi.org/10.1039/C6NJ03239G
- Hu, J., Zhu, J., Ge, S., Jiang, C., Guo, T., Peng, T., Huang, T. and Xie, L. 2020. Biocompatible, hydrophobic and resilience graphene/ chitosan composite aerogel for efficient oil-water separation. Surface and Coatings Technology, 385: 125361. https://doi.org/10.1016/j. surfcoat.2020.125361
- Huang, X., Zhang, S., Xiao, W., Luo, J., Li, B., Wang, L., Xue, H. and Gao, J. 2020. Flexible PDA@ACNTs decorated polymer nanofiber composite with superhydrophilicity and underwater superoleophobicity for efficient separation of oil-in-water emulsion. Journal of Membrane Science, 614: 118500. https://doi.org/10.1016/j.memsci.2020.118500
- Jamaly, S., Giwa, A. and Hasan, S. W. 2015. Recent improvements in oily wastewater treatment: Progress, challenges, and future opportunities. Journal of Environmental Sciences, 37: 15-30. https://doi.org/10.1016/j. jes.2015.04.011
- Jamsaz, A. and Goharshadi, E. K. 2020. An environmentally friendly superhydrophobic modified polyurethane sponge by seashell for the efficient oil/water separation. Process Safety and Environmental Protection, 139: 297-304. https://doi.org/10.1016/j.psep.2020.04.042
- Janqamsari, Y., Ashjari, M. and Niazi, Z. 2021. Carbon nanotube promoted porous nanocomposite based on PVA and recycled PET fibers for efficient oil spills cleanup applications. Chemical Papers, 75(7): 3443-3456. https://doi.org/10.1007/s11696-021-01587-x
- Junaidi, N. F. D., Othman, N. H., Fuzil, N. S., Mat Shayuti, M. S., Alias, N. H., Shahruddin, M. Z., Marpani, F., Lau, W. J., Ismail, A. F. and Aba, N. D. 2021. Recent development of graphene oxide-based membranes for oil-water separation: A review. Separation and Purification Technology, 258: 118000. https://doi.org/10.1016/j. seppur.2020.118000
- Karami, P., Khorshidi, B., McGregor, M., Peichel, J. T., Soares, J. B. P. and Sadrzadeh, M. 2020. Thermally stable thin film composite polymeric membranes for water treatment: A review. Journal of Cleaner Production, 250. https://doi.org/10.1016/j.jclepro.2019.119447
- Keshawy, M., Farag, R. K. and Gaffer, A. 2020. Egyptian crude oil sorbent based on coated polyurethane foam waste. Egyptian Journal of Petroleum, 29(1): 67-73. https://doi.org/10.1016/j.ejpe.2019.11.001
- Khosravi, M. and Azizian, S. 2015. Synthesis of a novel highly oleophilic and highly hydrophobic sponge for rapid oil spill cleanup. ACS Applied Materials and Interfaces, 7(45): 25326-25333. https://doi.org/10.1021/ acsami.5b07504
- Kizil, S. and Sonmez, B. 2017. Oil loving hydrophobic gels made from glycerol propoxylate: Efficient and reusable sorbents for oil spill cleanup. Journal of Environmental Management, 196: 330-339. https://doi. org/10.1016/j.jenvman.2017.02.016
- Koch, K. and Barthlott, W. 2009. Superhydrophobic and superhydrophilic plant surfaces: an inspiration for biomimetic materials. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 367(1893): 1487-1509. https://doi.org/10.1098/ rsta.2009.0022
- Kong, Z., Wang, J., Lu, X., Zhu, Y. and Jiang, L. 2017. In situ fastening graphene sheets into a polyurethane sponge for the highly efficient continuous cleanup of oil spills. Nano Research, 10(5): 1756-1766. https://doi.org/10.1007/s12274-017-1484-8
- Krishnan, M. R., Aldawsari, Y. F. and Alsharaeh, E. H. 2022. 3D-poly (styrene-methyl methacrylate)/divinyl benzene-2D-nanosheet

composite networks for organic solvents and crude oil spill cleanup. Polymer Bulletin, 79(6): 3779-3802. https://doi.org/10.1007/s00289-021-03565-5

- Li, D., Gou, X., Wu, D. and Guo, Z. 2018. A robust and stretchable superhydrophobic PDMS/PVDF@KNFs membrane for oil/water separation and flame retardancy. Nanoscale, 10(14): 6695-6703. https:// doi.org/10.1039/C8NR01274A
- Li, H., Liu, L. and Yang, F. 2012. Hydrophobic modification of polyurethane foam for oil spill cleanup. Marine Pollution Bulletin, 64(8): 1648-1653. https://doi.org/10.1016/j.marpolbul.2012.05.039
- Li, Z., Shao, L., Hu, W., Zheng, T., Lu, L., Cao, Y. and Chen, Y. 2018. Excellent reusable chitosan/cellulose aerogel as an oil and organic solvent absorbent. Carbohydrate Polymers, 191: 183-190. https://doi. org/10.1016/j.carbpol.2018.03.027
- Liang, L., Xue, Y., Wu, Q., Dong, Y. and Meng, X. 2019. Self-assembly modification of polyurethane sponge for application in oil/water separation. RSC Advances, 9(69): 40378-40387. https://doi. org/10.1039/c9ra05855a
- Lin, J., Shang, Y., Ding, B., Yang, J., Yu, J. and Al-Deyab, S. S. 2012. Nanoporous polystyrene fibers for oil spill cleanup. Marine Pollution Bulletin, 64(2): 347-352. https://doi.org/10.1016/j. marpolbul.2011.11.002
- Liu, C., Fang, Y., Miao, X., Pei, Y., Yan, Y., Xiao, W. and Wu, L. 2019. Facile fabrication of superhydrophobic polyurethane sponge towards oil-water separation with exceptional flame-retardant performance. Separation and Purification Technology, 229: 115801. https://doi. org/10.1016/j.seppur.2019.115801
- Lü, X., Cui, Z., Wei, W., Xie, J., Jiang, L., Huang, J. and Liu, J. 2016. Constructing polyurethane sponge modified with silica/graphene oxide nanohybrids as a ternary sorbent. Chemical Engineering Journal, 284: 478-486. https://doi.org/10.1016/j.cej.2015.09.002
- Lv, P., Yasin, A., Hao, B. and Ma, P.-C. 2020. Wheat bran/polymer composites as a solidifier to gel oil on water surface. Composites Communications, 22: 100471. https://doi.org/10.1016/j.coco.2020.100471
- Middlebrook, A. M., Murphy, D. M., Ahmadov, R., Atlas, E. L., Bahreini, R., Blake, D. R., Brioude, J., de Gouw, J. A., Fehsenfeld, F. C., Frost, G. J., Holloway, J. S., Lack, D. A., Langridge, J. M., Lueb, R. A., McKeen, S. A., Meagher, J. F., Meinardi, S., Neuman, J. A., Nowak, J. B. and Ravishankara, A. R. 2012. Air quality implications of the deepwater horizon oil spill. Proceedings of the National Academy of Sciences, 109(50): 20280-20285. https://doi.org/10.1073/pnas.1110052108
- Motta, F. L., Stoyanov, S. R. and Soares, J. B. P. 2019. Development and application of an amylopectin-graft-poly(methyl acrylate) solidifier for rapid and efficient containment and recovery of heavy oil spills in aqueous environments. Chemosphere, 236: 124352. https://doi. org/10.1016/j.chemosphere.2019.124352
- Nasir, M. I. and Hameed, Z. M. 2014. Utilization of expanded polystyrene (EPS) Waste as sorbent for oil spill cleanup. Society of Petroleum Engineers - 30th Abu Dhabi International Petroleum Exhibition and Conference, ADIPEC 2014: Challenges and Opportunities for the Next 30 Years, 5, 3375-3385. https://doi.org/10.2118/172000-ms
- Neelamegan, H., Yang, D.K., Lee, G.-J., Anandan, S. and Wu, J. J. 2020. Synthesis of magnetite nanoparticles anchored cellulose and ligninbased carbon nanotube composites for rapid oil spill cleanup. Materials Today Communications, 22: 100746. https://doi.org/10.1016/j. mtcomm.2019.100746
- Onwuka, J. C., Agbaji, E. B., Ajibola, V. O. and Okibe, F. G. 2018. Treatment of crude oil-contaminated water with chemically modified natural fiber. Applied Water Science, 8(3): 86. https://doi.org/10.1007/ s13201-018-0727-5
- Oribayo, O., Feng, X., Rempel, G. L. and Pan, Q. 2017. Synthesis of ligninbased polyurethane/graphene oxide foam and its application as an absorbent for oil spill clean-ups and recovery. Chemical Engineering Journal, 323: 191-202. https://doi.org/10.1016/j.cej.2017.04.054

- Padaki, M., Surya Murali, R., Abdullah, M. S., Misdan, N., Moslehyani, A., Kassim, M. A., Hilal, N. and Ismail, A. F. 2015. Membrane technology enhancement in oil–water separation. A review. Desalination, 357: 197-207. https://doi.org/10.1016/j.desal.2014.11.023
- Pavlatou, E. A. 2020. Commercial sponges as a novel technology for crude oil removal from seawater and industrial wastewater: a review. Biomedical Journal of Scientific and Technical Research, 25(5). https:// doi.org/10.26717/BJSTR.2020.25.004251
- Peng, H., Wang, H., Wu, J., Meng, G., Wang, Y., Shi, Y., Liu, Z. and Guo, X. 2016. Preparation of superhydrophobic magnetic cellulose sponge for removing oil from water. Industrial and Engineering Chemistry Research, 55(3): 832-838. https://doi.org/10.1021/acs.iecr.5b03862
- Peng, M., Zhu, Y., Li, H., He, K., Zeng, G., Chen, A., Huang, Z., Huang, T., Yuan, L. and Chen, G. 2019. Synthesis and application of modified commercial sponges for oil-water separation. Chemical Engineering Journal, 373: 213-226. https://doi.org/10.1016/j.cej.2019.05.013
- Potter, S. and Buist, I. 2008. In-situ burning for oil spills in arctic waters: State-of-the-Art and Future Research Needs, 23-39. https://doi. org/10.1007/978-1-4020-8565-9_5
- Ramanathan, R., Abdullah, L., Mohamed, S. and Fauadi, M.H.F.M. 2022. A review of user-centred design methods for designing a portable oil spill skimmer. Nat. Env. Poll. Tech., 21(4).
- Rogowska, J. and Namieśnik, J. 2010. Environmental implications of oil spills from shipping accidents. Reviews of Environmental Contamination and Toxicology, 95-114. https://doi.org/10.1007/978-1-4419-6260-7_5
- Rotaru, R., Fortuna, M. E., Cojocaru, C., Samoila, P., Pricop, L. and Harabagiu, V. 2019. Viscose-maghemite/goethite polymeric composite as sorbent for oil spill cleanup. Environmental Engineering and Management Journal, 18(6): 1193-1200. https://doi.org/10.30638/ eemj.2019.114
- Ruan, C., Ai, K., Li, X. and Lu, L. 2014. A superhydrophobic sponge with excellent absorbency and flame retardancy. Angewandte Chemie, 126(22): 5662-5666. https://doi.org/10.1002/ange.201400775
- Sarbatly, R., Krishnaiah, D. and Kamin, Z. 2016. A review of polymer nanofibres by electrospinning and their application in oil-water separation for cleaning up marine oil spills. Marine Pollution Bulletin, 106(1-2): 8-16. https://doi.org/10.1016/j.marpolbul.2016.03.037
- Shang, B., Wang, Y., Peng, B. and Deng, Z. 2016. Bioinspired polydopamine particles-assisted construction of superhydrophobic surfaces for oil/ water separation. Journal of Colloid and Interface Science, 482: 240-251. https://doi.org/10.1016/j.jcis.2016.07.081
- Shen, Y., Li, D., Wang, L., Zhou, Y., Liu, F., Wu, H., Deng, B. and Liu, Q. 2021. Superelastic polyimide nanofiber-based aerogels modified with silicone nanofilaments for ultrafast oil/water separation. ACS Applied Materials and Interfaces, 13(17): 20489-20500. https://doi. org/10.1021/acsami.1c01136
- Shojaei, N., Aminsharei, F. and Abbastabar Ahangar, H. 2021. Application of hydrophobic polymers as solidifiers for oil spill cleanup. International Journal of Environmental Science and Technology, 18(6): 1419-1424. https://doi.org/10.1007/s13762-020-02882-y
- Shuai, Q., Yang, X., Luo, Y., Tang, H., Luo, X., Tan, Y. and Ma, M. 2015. A superhydrophobic poly(dimethylsiloxane)-TiO₂ coated polyurethane sponge for selective absorption of oil from water. Materials Chemistry and Physics, 162: 94-99. https://doi.org/10.1016/j. matchemphys.2015.05.011
- Singh, S. and Jelinek, R. 2020. Solar-mediated oil-spill cleanup by a carbon dot-polyurethane sponge. Carbon, 160: 196-203. https://doi. org/10.1016/j.carbon.2020.01.016
- Singh, V., Kendall, R. J., Hake, K. and Ramkumar, S. 2013. Crude oil sorption by raw cotton. Industrial and Engineering Chemistry Research, 52(18): 6277-6281. https://doi.org/10.1021/ie4005942
- Song, B., Zhu, J. and Fan, H. 2017. Magnetic fibrous sorbent for remote and efficient oil adsorption. Marine Pollution Bulletin, 120(1-2): 159-164.

https://doi.org/10.1016/j.marpolbul.2017.05.011

- Speight, J. G. 2006. The Chemistry and Technology of Petroleum. CRC Press. https://doi.org/10.1201/9781420008388
- Tran, V.-H. T. and Lee, B.K. 2017. Novel fabrication of a robust superhydrophobic PU@ZnO@Fe3O4@SA sponge and its application in oil-water separations. Scientific Reports, 7(1): 17520. https://doi. org/10.1038/s41598-017-17761-9
- Usman, A. D. and Okoro, L. N. 2017. Chemical science review and letters innovations in oil spill clean-up techniques. Chem. Sci. Rev. Lett., 6(23): 1908-1916.
- Valdebenito, F., García, R., Cruces, K., Ciudad, G., Chinga-Carrasco, G. and Habibi, Y. 2018. CO₂ adsorption of surface-modified cellulose nanofibril films derived from agricultural wastes. ACS Sustainable Chemistry and Engineering, 6(10): 12603-12612. https://doi. org/10.1021/acssuschemeng.8b00771
- Vidal, R. R. L. and Moraes, J. S. 2019. Removal of organic pollutants from wastewater using chitosan: a literature review. International Journal of Environmental Science and Technology, 16(3): 1741-1754. https://doi. org/10.1007/s13762-018-2061-8
- Visco, A., Quattrocchi, A., Nocita, D., Montanini, R. and Pistone, A. 2021. Polyurethane foams loaded with carbon nanofibres for oil spill recovery: Mechanical properties under fatigue conditions and selective absorption in oil/water mixtures. Nanomaterials, 11(3): 1-14. https:// doi.org/10.3390/nano11030735
- Wang, C.F., Huang, H.-C. and Chen, L.-T. 2015. Protonated melamine sponge for effective oil/water separation. Scientific Reports, 5(1): 14294. https://doi.org/10.1038/srep14294
- Wang, J. and Geng, G. 2015. Highly recyclable superhydrophobic sponge suitable for the selective sorption of high viscosity oil from water. Marine Pollution Bulletin, 97(1-2): 118-124. https://doi.org/10.1016/j. marpolbul.2015.06.026
- Wang, Z., Ma, H., Chu, B. and Hsiao, B. S. 2017. Super-hydrophobic modification of porous natural polymer "luffa sponge" for oil absorption. Polymer, 126: 470-476. https://doi.org/10.1016/j. polymer.2017.05.068
- Wang, Z., Saleem, J., Barford, J. P. and McKay, G. 2020. Preparation and characterization of modified rice husks by biological delignification and acetylation for oil spill cleanup. Environmental Technology, 41(15): 1980-1991. https://doi.org/10.1080/09593330.2018.1552725
- Wei, Q., Oribayo, O., Feng, X., Rempel, G. L. and Pan, Q. 2018. Synthesis of polyurethane foams loaded with tio2 nanoparticles and their modification for enhanced performance in oil spill cleanup. Industrial and Engineering Chemistry Research, 57(27): 8918-8926. https://doi. org/10.1021/acs.iecr.8b01037
- Wu, D., Wu, W., Yu, Z., Zhang, C. and Zhu, H. 2014. Facile preparation and characterization of modified polyurethane sponge for oil absorption. Industrial and Engineering Chemistry Research, 53(52): 20139-20144. https://doi.org/10.1021/ie5032327
- Wu, J., An, A. K., Guo, J., Lee, E. J., Farid, M. U. and Jeong, S. 2017. CNTs reinforced super-hydrophobic-oleophilic electrospun polystyrene oil sorbent for enhanced sorption capacity and reusability. Chemical Engineering Journal, 314: 526-536. https://doi.org/10.1016/j. cej.2016.12.010
- Wu, L., Zhang, J., Li, B. and Wang, A. 2014. Magnetically driven super durable superhydrophobic polyester materials for oil/water separation. Polymer Chemistry, 5(7): 2382. https://doi.org/10.1039/c3py01478a
- Xu, Z., Wang, J., Li, H. and Wang, Y. 2019a. Coating sponge with multifunctional and porous metal-organic framework for oil spill remediation. Chemical Engineering Journal, 370(April), 1181-1187. https://doi.org/10.1016/j.cej.2019.03.288
- Xu, Z., Wang, J., Li, H. and Wang, Y. 2019b. Coating sponge with multifunctional and porous metal-organic framework for oil spill remediation. Chemical Engineering Journal, 370: 1181-1187. https:// doi.org/10.1016/j.cej.2019.03.288

- Xue, J., Zhu, L., Zhu, X., Li, H., Ma, C., Yu, S., Sun, D., Xia, F. and Xue, Q. 2021. Tetradecylamine-MXene functionalized melamine sponge for effective oil/water separation and selective oil adsorption. Separation and Purification Technology, 259: 118106. https://doi.org/10.1016/j. seppur.2020.118106
- Yadav, S., Mattaparthi, S., Sreenivasulu, K., Khandelwal, M., Majumdar, S. and Sharma, C. S. 2019. Recycling of thermoplastic polystyrene waste using citrus peel extract for oil spill remediation. Journal of Applied Polymer Science, 136(33): 1-8. https://doi.org/10.1002/app.47886
- Yan, X., Zhu, X., Ruan, Y., Xing, T., Chen, G. and Zhou, C. 2020. Biomimetic, dopamine-modified superhydrophobic cotton fabric for oil-water separation. Cellulose, 27(13): 7873-7885. https://doi. org/10.1007/s10570-020-03336-x
- Yang, Y., Yi, H. and Wang, C. 2015. Oil absorbents based on melamine/ lignin by a dip adsorbing method. ACS Sustainable Chemistry and Engineering, 3(12): 3012-3018. https://doi.org/10.1021/ acssuschemeng.5b01187
- Yi, L., Yang, J., Fang, X., Xia, Y., Zhao, L., Wu, H. and Guo, S. 2020. Facile fabrication of wood-inspired aerogel from chitosan for efficient removal of oil from Water. Journal of Hazardous Materials, 385: 121507. https:// doi.org/10.1016/j.jhazmat.2019.121507
- Yu, C., Lin, W., Jiang, J., Jing, Z., Hong, P. and Li, Y. 2019. Preparation of a porous superhydrophobic foam from waste plastic and its application for oil spill cleanup. RSC Advances, 9(65): 37759-37767. https://doi. org/10.1039/c9ra06848a
- Yu, L., Hao, G., Zhou, S. and Jiang, W. 2016. Durable and modified foam for cleanup of oil contamination and separation of oil–water mixtures. RSC Advances, 6(29): 24773-24779. https://doi.org/10.1039/C5RA27370F
- Yue, X., Li, Z., Zhang, T., Yang, D. and Qiu, F. 2019. Design and fabrication of superwetting fiber-based membranes for oil/water separation applications. Chemical Engineering Journal, 364: 292-309. https://doi. org/10.1016/j.cej.2019.01.149
- Zamparas, M., Tzivras, D., Dracopoulos, V. and Ioannides, T. 2020. Application of sorbents for oil spill cleanup focusing on natural-based modified materials: a review. Molecules, 25(19): 4522. https://doi. org/10.3390/molecules25194522
- Zhang, B., Matchinski, E. J., Chen, B., Ye, X., Jing, L. and Lee, K. 2019. Marine oil spills-oil pollution, sources and effects. In World Seas: an Environmental Evaluation (pp. 391-406). Elsevier. https://doi. org/10.1016/B978-0-12-805052-1.00024-3
- Zhang, H., Zhen, Q., Cui, J.-Q., Liu, R.-T., Zhang, Y.-F., Qian, X.-M. and Liu, Y. 2020. Groove-shaped polypropylene/polyester micro/ nanofibrous nonwoven with enhanced oil wetting capability for high oil/water separation. Polymer, 193, 122356. https://doi.org/10.1016/j. polymer.2020.122356
- Zhang, Jiaoyuan, Chen, R., Liu, J., Liu, Q., Yu, J., Zhang, H., Jing, X., Zhang, M. and Wang, J. 2019. Construction of ZnO@Co₃O₄-loaded nickel foam with abrasion resistance and chemical stability for oil/water separation. Surface and Coatings Technology, 357: 244-251. https:// doi.org/10.1016/j.surfcoat.2018.09.042
- Zhang, Junping and Seeger, S. 2011. Polyester materials with superwetting silicone nanofilaments for oil/water separation and selective oil absorption. Advanced Functional Materials, 21(24): 4699-4704. https:// doi.org/10.1002/adfm.201101090
- Zhang, P., Hu, W., Wu, M., Gong, L., Tang, A., Xiang, L., Zhu, B., Zhu, L. and Zeng, H. 2019. Cost-effective strategy for surface modification via complexation of disassembled polydopamine with Fe(III) ions. Langmuir, 35(11): 4101-4109. https://doi.org/10.1021/ acs.langmuir.9b00245
- Zhang, X., Wang, X., Liu, X., Lv, C., Wang, Y., Zheng, G., Liu, H., Liu, C., Guo, Z. and Shen, C. 2018. Porous polyethylene bundles with enhanced hydrophobicity and pumping oil-recovery ability via skin-peeling. ACS Sustainable Chemistry and Engineering, 6(10): 12580-12585. https:// doi.org/10.1021/acssuschemeng.8b03305

- Zhang, Z., Sèbe, G., Rentsch, D., Zimmermann, T. and Tingaut, P. 2014. Ultralightweight and flexible silylated nanocellulose sponges for the selective removal of oil from water. Chemistry of Materials, 26(8): 2659-2668. https://doi.org/10.1021/cm5004164
- Zhao, S., Yin, L., Zhou, Q., Liu, C. and Zhou, K. 2020. In situ selfassembly of zeolitic imidazolate frameworks on the surface of flexible polyurethane foam: Towards for highly efficient oil spill cleanup and fire safety. Applied Surface Science, 506: 144700. https://doi. org/10.1016/j.apsusc.2019.144700
- Zhou, X., Zhang, Z., Xu, X., Men, X. and Zhu, X. 2013. Facile fabrication of superhydrophobic sponge with selective absorption and collection

of oil from water. Industrial and Engineering Chemistry Research, 52(27): 9411-9416. https://doi.org/10.1021/ie400942t

- Zhu, H., Chen, D., Yang, S., Li, N., Xu, Q., Li, H., Wang, L., He, J., Jiang, J. and Lu, J. 2016. A versatile and cost-effective reduced graphene oxide-crosslinked polyurethane sponge for highly effective wastewater treatment. RSC Advances, 6(44): 38350-38355. https://doi.org/10.1039/ C6RA05450A
- Zhu, H., Qiu, S., Jiang, W., Wu, D. and Zhang, C. 2011a. Evaluation of electrospun polyvinyl chloride/polystyrene fibers as sorbent materials for oil spill cleanup. Environmental Science and Technology, 45(10): 4527-4531. https://doi.org/10.1021/es2002343