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Recent Progress in Fabrication of Candle Soot Modified Superhydrophobic/Superoleophilic Surfaces for Oil-Water Separation

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Abstract
Continually occurred the oceanic oil spill accidents and discharging huge amount of industrial oily wastewater cause a serious threat to the environment. Oil and organic pollutants in water has a severe problem for aquatic life and human being. There is a need to develop technology for oil-water separation. Recently, superhydrophobic/superoleophilic sponges, metal meshes, membranes and porous materials plays crucial role to separate oil from oil-water mixture. The micro and nanopores of substrate facilitate to enter liquid into it and superhydrophobic/superoleophilic property of substrate surface resist water and allows oil to enter into the porous substrate. Carbon soot nanoparticles are hydrophobic (water repellent) in nature and has the advantages of cost-effectiveness and production scalability over other carbons like graphene, carbon nanotubes (CNTs), carbon nanodots (CNDs), etc., in their synthesis. Carbon soot based superhydrophobic/superoleophilic surfaces have outstanding water repulsion and oil absorption capacity, highly selectivity, chemical inertness and excellent recyclability. In this paper, we discussed recent progress of carbon soot based superhydrophobic/superoleophilic sponges and meshes for oil-water separation.

Keywords: Carbon soot, oil-water separation, sponge, stainless steel mesh and superhydrophobic.

Introduction

The oil spillage and industrial pollutants has emerged as critical issue to an ecosystem, human health, economic growth, etc. To address this challenge, several efforts have been done to separate oil and organic solvents from oil contaminated water [1-2]. The various methods such as spray coating, dip-coating, hydrothermal method, chemical vapour deposition, surface etching, solvothermal method, layer-by-layer assembly and electrochemical treatment have been used to prepare superhydrophobic/superoleophilic coating on sponge and mesh surface [3]. The ideal superhydrophobic/superoleophilic meshes and sponges must have following characteristics, such as fine selectivity toward various oils, high separation efficiency, reusability and along with high mechanical, chemical, and thermal stability [4].

The superhydrophobic/superoleophilic surfaces have shown water contact angle (WCA) over 150°, sliding angle < 10° and oil contact angle <5° [5]. The traditional technologies, including in situ burning, bioremediation, chemical dispersant methods, skimming, and sorbents are used for oil-water separation. However, many of these technologies involve energy intensive and slow processes, have low oil-water separation capacities or create secondary pollution during the oil-water separation process, restricting their widespread practical applications [6-8]. Different strategies used for formation of hierarchical structures such as sol-gel coating, chemical vapor deposition, plasma etching, template processing, lithographic patterning, etc. have been adopted [9-10].

Carbon soot (CS) generated from incomplete combustion of paraffin wax has demonstrated the advantages of cost effectiveness and production scalability over carbon nanotubes (CNTs), graphene and activated carbons in their synthesis. Hence, the carbon soot coated superhydrophobic/superoleophilic surfaces shows excellent oil-water separation ability rather than carbon nanotubes, carbon nanodots and graphene [11].

Song *et al.* [12] have fabricated carbon soot (CS) coated mesh using dip-coating method. The cleaned stainless-steel mesh (SSM) was subsequently dipped in the glue solution for 10 min and CS-dispersion, finally dried at 80°C to obtain the CS-glue coated mesh. The CS coating is close packed because of using superglue (EVO-STIK Serious Glue) as a binder. The CS-glue coated mesh revealed the separation efficiency higher than 99.95%. Even after 20 cycle separation tests, it was shown excellent reusability and durability. The different chemical methods are used for the fabrication of the superhydrophobic/superoleophilic meshes/sponges for efficient oil-water separation. As the carbon nanotubes are hydrophobic in nature and it shows strong affinity toward oil, Lee *et al.* [13] adopted chemical vapor deposition (CVD) technique to deposit vertically aligned CNTs on stainless steel (SS) mesh. The as-prepared SS-CNT mesh effectively separates oil from water-in-oil emulsions with efficiency higher than 80%. Gu *et al.* [14] prepared polystyrene (PS) and carbon nanotubes coated superhydrophobic/superoleophilic membrane. The as-prepared superhydrophobic/ superoleophilic membrane shows mechanically robust PS-CNT surface shows excellent oil separation from oil-water mixture with good repeatability and separation efficiency of more than 99%. Electrospinning technique can be effectively used to form superhydrophobic/superoleophilic surfaces. Wang *et al.* [15] prepared the polyurethane (PU) sponge from hydrophilic to superhydrophobic by dip coating it from the nanocomposite of CNT/poly- (dimethyl siloxane) (PDMS). The as-prepared superhydrophobic/superoleophilic sponge shows the continuous removal of various oils such as Soybean oil, motor oil, diesel, n-hexadecane, gasoline, and n-hexane from the surface of the water with high separation efficiency.

In this article, we will discuss on the simple, low-cost, rapid and innovative methods for the fabrication of superhydrophobic/superoleophilic carbon soot nanoparticle coated sponges and meshes for efficient oil-water separation application.

Recent Developments for Effective Oil-Water Separation Superhydrophobic-Superoleophilic Sponge

Hydrophobic candle soot (CS) particles can be collected easily and cost effectively from the candle flame. Li *et al.* [16] obtained the hydrophobic CS particles by incomplete combustion of hydrocarbons from the mid-candle flame. The polyurethane sponge immersed in the mixture of CS, hydrophobic silica nanoparticles, and PU resin to achieve stable superhydrophobicity. The CS-SiO_2-PU sponge showed excellent oil separation capacity from hot water and acidic, alkaline, and salt solutions. The CS-SiO_2-PU sponges could absorb a various oil efficiently and selectively, and show high absorption capacity that is up to 65 times of its own weight. Furthermore, the CS-SiO_2-PU sponge possesses stable superhydrophobicity and excellent ability of selective absorption to oil even at various harsh conditions, including acid (1M HCl) and alkali (1M NaOH), and salt (1M NaCl) aqueous solutions at mechanical agitation condition, hot water and ice/water mixtures. The CS-SiO_2-PU sponge combination with a vacuum system could continuously absorb and remove oil from water surface. As shown in (Fig. 1a), the contact angle of coloured water droplet on the PU sponge is nearly 100° and the coloured kerosene droplet contact angle is about 0°. However, the CS-SiO_2-PU sponge shows superhydrophobicity and superoleophobicity simultaneously (Fig. 1b). The kerosene droplet was absorbed by the CS-SiO_2-PU sponge immediately when it was dropped on the surface of sponge, while the water droplet stayed on the surface of as-prepared sponge and kept its spherical shape. As shown in (Fig. 1c and d), the water contact angle on the CS-SiO_2-PU sponge is up to 155° and the SA is as low as 7°. Therefore, the water was repelled by the as-prepared sponge, whereas the oil absorbed through the sponge quickly.

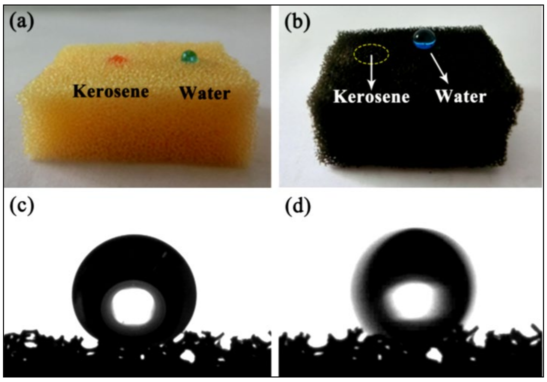

Fig 1: (a and b) Schematic illustration of the fabrication the wettability of the pristine PU sponge and the CS-SiO_2-PU sponge to water and oil droplets, respectively. (c and d) The CA and SA of water on the CS-SiO_2-PU sponge, respectively [16], with permission from J. Sol-Gel Sci Technol., Copyright 2017.

Beshkar *et al.* [17] prepared durable magnetic superhydrophobic PU sponge by immersing it in the colloidal solution of straw candle soot (carbon nanoparticles) and further modification was done by Fe_2O_4 nanoparticles and PDMS. The waste oil was easily collected from the surface of water through guiding the magnetic superhydrophobic polyurethane (PU) sponge by a bar magnet. The wettability of sponge remained unchanged even after 30 absorption/desorption cycles. The oil-water separation experiments were performed using the as-prepared optimized superhydrophobic sponge to absorbing of waste oil. The schematic of experimental protocol for preparation and operation superhydrophobic/superoleophilic magnetic straw soot sponge is shown in Fig. 2. It was indicated that the superhydrophobic sponge is taken by a magnet bar and absorbs waste oil completely from the oil-water mixture within 10 seconds.

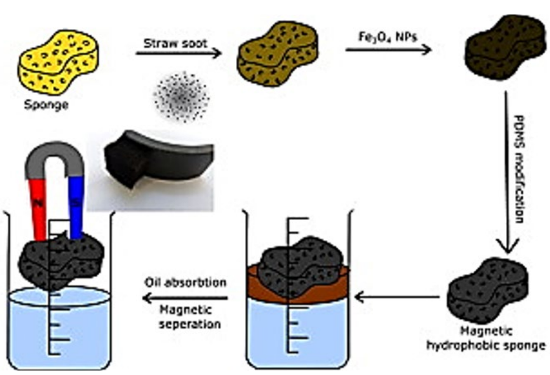

Fig 2: Schematic illustration of preparation of modified polyurethane sponge and oil separation process. Images reprinted from [17], with permission from Elsevier, Copyright 2017.

Gao *et al.* [18] obtained the candle soot (CS) particles from combustion flame and dispersed it in 1,2-dichloroethane (DCE). The superhydrophobic/superoleophilic melamine sponge prepared by a uniform coating of as-grown CS-DCE solution using dip-coating method. The CS-modified sponge exhibited strong water repellency and oil absorbency without further chemical modification (Fig. 3. a and b). An easy and fast recovery of engine oil floating on the water surface by CS-modified sponge confirms its high oil-water separation efficiency (Fig. 3. c–e). The CS-modified sponge could absorb numerous oils and organic solvents with an absorption capacity of 25-80 times of its own weight. Oil can be recycled for more than 10 times with absolutely no change in an oil absorption capacity and wettability of the CS-modified sponge.

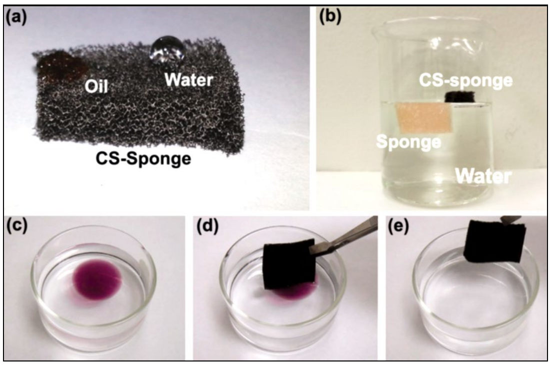

Fig 3: Schematic showing (a) Photograph of water and oil droplets on the surface of a CS-sponge. (b) Photograph of pure and CS-sponges placed on water. (c-e) Photographs demonstrating the removal of an oil droplet from water using a CS-sponge [18], with permission from American Chemical Society, Copyright 2014.

Yue *et al.* [19] the superhydrophobic surface was fabricated by using PVDF and candle soot via sugar template method. It was shown the water contact angle of 158° and roll-on angle of <6°. The oil quickly absorbed by superhydrophobic sponge shows the superoleophilic property. The sponge shows excellent oil-water separation property even after 25 cycles. The strong elasticity, high stretching resistance confirms that the modified superhydrophobic surface is highly mechanical durable. The modified sponge maintains the 89% of recovery rate even after 10 cycles. The absorption capability recovered up to 96% without obvious change of morphology of the sponge surface. This method was used to prepare a photothermal, porous PVDF/CS sponge with structural, chemical and mechanical property [19]. Fig. 4 shows that the preparation process of the porous PVDF/candle soot sponge. In short, sugar particles were placed into a mixture of PVDF/DMF/CS solution, followed by removing sugar templates via the water dissolving. It is important to note that a simple sugar-templating process was used for formation of superhydrophobic sponge, it requires only a simple as well as eco-friendly preparation process.

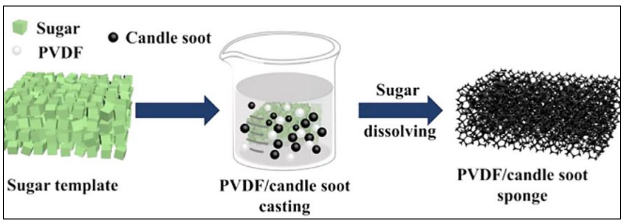

Fig 4: Schematic illustration of the fabrication of porous PVDF/candle soot sponge using sugar template. Images reprinted from [19], with permission from Elsevier, Copyright 2021.

Superhydrophobic/Superoleophilic Mesh

Li *et al.* [20] deposited hydrophobic CS particles on a SS mesh by holding it in the mid-flame of paraffin candle and then spray coated the hydrophobic silica nanoparticles (50 nm) on it. The as-prepared carbon soot/silica nanoparticles (CS/SiO_2) hybrid mesh revealed strong repellence toward pure water and droplets of pH ranging from 1 to 14, whereas the oil drops quickly spread on the surface. The hybrid mesh revealed strong ability to separate various oils and organic liquids mixed in pure water, hot water (92°C), and strong corrosive solutions (1M HCl, NaOH, and NaCl) with more than 98% of separation efficiency. The oil/water separation was performed as shown in Fig. 5. Because of the excellent mechanical flexibility of the stainless-steel mesh, the mesh was folded into a three-dimensional structure, then coated with CS and hydrophobic silica for oil/water separation. The coated mesh was placed on beaker and mixture of kerosene and water was poured onto the three-dimensional superhydrophobic mesh, as shown in Fig. 5a-c. Kerosene was dropped into the beaker, while water was repelled and retained above the mesh. Fig. 5d exhibited that there are no oily droplets were present in the water. After the separation, nearly equal amount of water and kerosene were collected, which was suggested that extremely high separation efficiency of the coated mesh as shown in Fig. 5e.

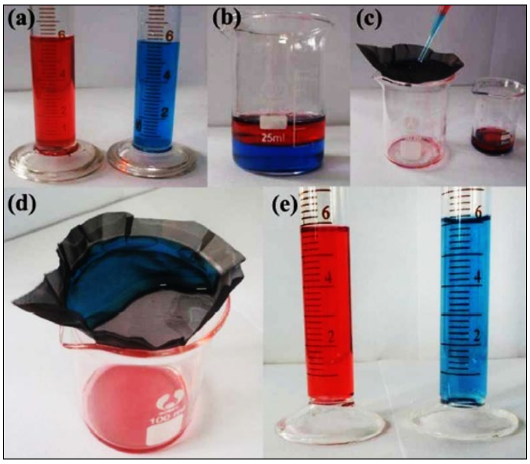

Fig 5: Schematic illustration of the separation process of oil (kerosene)/water mixture based on the CS and silica overlap coated mesh. (a-b) water is dyed with methylene blue and oil is dyed with Oil Red O and then they are mixed before separation. (c-d) the separation process of the oil/water mixture using the overlap coated mesh. (e) The water and oil volume keep nearly the same after separation [20], with permission from RSC Nanoscale, Copyright 2016.

Cao *et al.* [21] used the electrodeposition technique for the formation of superhydrophobic/superoleophilic copper meshes and then held it above candle flame to deposit carbon soot (CS) nanoparticles. The agglomerated CS nanoparticles formed a chain like rough hierarchical structure on which water drop exhibited contact angle higher than 153°. The CS deposited copper mesh quickly separated various oils such as silicone oil, cyclohexane, n-hexane, n-heptane, petroleum ether, and liquid paraffin from oil-water mixtures with oil separation efficiency greater than 90% even after 30 separation cycles. The rough mesh was placed on 1.5 cm height of flame of burning candle for deposition for 5 minutes, the whole preparation process is shown in the fig. 6, the superhydrophobic mesh surface was formed by using candle soot.

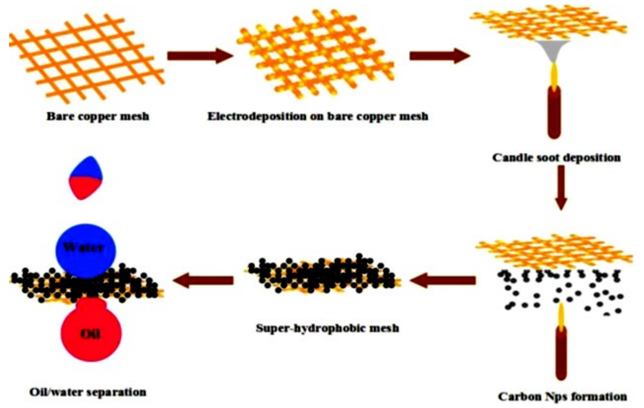

Fig 6: Schematic illustration of the preparation process of super-hydrophobic and super-oleophilic copper mesh, and the application in oil-water separation [21], with permission from Colloids and Surfaces A: Physicochem. Eng. Aspects, Copyright 2017.

Zulfiqar *et al.* [22] deposited cheaply available sawdust on polychloroprene adhesive-coated SS mesh with subsequent deposition of silicone polymer by dip coating. Thereafter, a thin layer of CS particles was applied on the as-prepared mesh by holding above a candle flame. The carbon soot (CS) nanoparticles uniformly deposited on silicone covered SD exhibited highly rough and porous morphology required for superhydrophobicity and superoleophobicity. As depicted in Fig. 7, a superhydrophobic SS mesh easily and rapidly separated oil-water mixture. No trace of water was observed in separated oil confirming its potential in oil-water separation capability. Apart from excellent oil-water separation efficiency (>95%), the superhydrophobic SS mesh exhibited good recyclability, reusability, and mechanical stability. The potential of these coatings for oil/water separation is shown in Fig. 7. The figure typically shows a mesh coated with superhydrophobic material holding water without any leakage. The superhydrophobic mesh was used for the separation of oil from oil-water mixtures as shown in Fig. 7. The mesh readily separated the oil while blocking the flow of water through its pores. The water content was not found in the separated oil, which confirms the excellent superhydrophobic/superoleophilic property. The steel mesh with excellent mechanical stability modified with durable superhydrophobic coating can be used as a medium for oil/water separation.

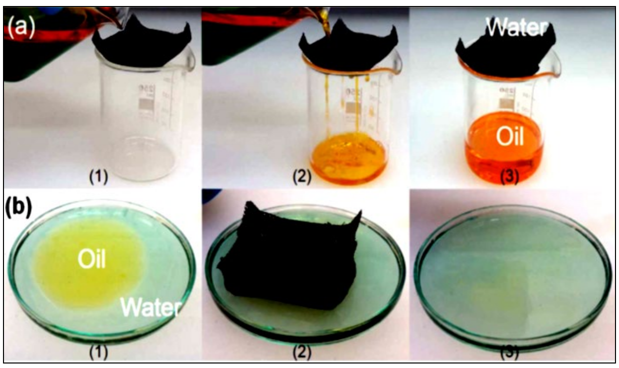

Fig 7: Schematic illustration of a superhydrophobic mesh demonstrating (a) the separation of oil/water mixture and (b) collection of oil from oil/water mixture [22], with permission from Colloids and Surfaces A: Physicochem. Eng. Aspects, Copyright 2017.

Chen *et al.* [23] synthesized a superhydrophobic surfaces by simply placing the stainless steel (SS) on the middle of candle flame for deposition of carbon soot (CS) on the SS mesh. The candle soot coated substrate together with $SiCl_4$ was placed in a drier for chemical vapour deposition. Then, though calcination at 600°C for half in air, CS composed NPs thermally degraded and diffused through the silica shell gradually. It was shows excellent oil-water separation efficiency even after 30 times recycling use of modified superhydrophobic surfaces. The superhydrophobic coating revealed excellent separation efficiency even after 6 times reuses of same superhydrophobic surface. It could be potentially used in optical and visual application scenarios were in harsh, oily environments, like goggles, building façade, visual oil-water separation device, touch screen, etc. As shown in Fig. 8, the cheap and accessible candle soot were used for formation of a rough surface template to directly deposition on the substrate. The candle soot was formed carbon nanoparticles forming by incompletely burned paraffin, which loosely bounded on the substrate and formed a rough surface. Finally, a hard and porous silica shell was deposited on the candle soot template through chemical vapor deposition of $SiCl_4$ like Stober reaction, which completely formed the roughness of the template and a robust rough surface.

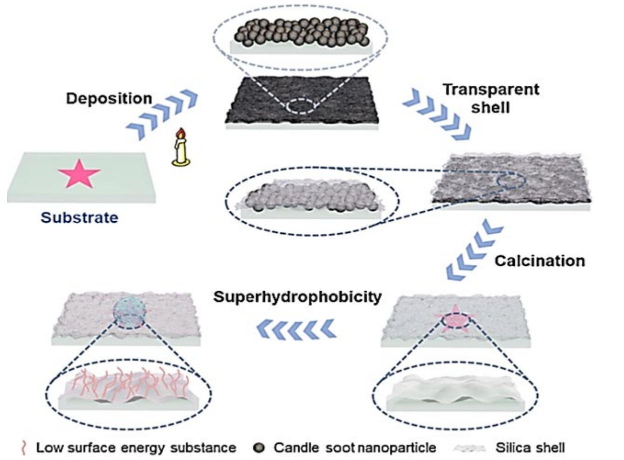

Fig 8: Schematic of preparation and properties of the transparent and robust superhydrophobic coating and oil–water separation. Images reprinted from [23], with permission from Nature, Copyright 2022.

Conclusion

As this review highlights, carbon soot nanoparticles are unique in that, their fabrication requires little control of external parameters. It is beneficial economically, facile and straightforward to synthesize. The carbon soot nanoparticles coated sponges and mesh has been developed by using carbon soot nanoparticles and different polymers. The absorption/separation investigation demonstrates that, the carbon soot surface is highly efficient and stable in absorbing a wide range of oil and organic solvents. It can be believed that, the carbon soot coated superhydrophobic materials are very useful for oil-water separation. The carbon soot synthesis, carbon soot coated sponge/mesh preparation, procedures are simple, cost-effective and scalable. The absorption/separation investigation demonstrates that, the carbon soot sponge/mesh is highly efficient and stable in absorbing a wide range of oil and organic solvents. It can be believed that, the carbon soot coated superhydrophobic surfaces are very useful for oil-water separation. It shows various tremendous results with carbon soot-polymer composite in various mechanical conditions. A carbon soot nanoparticle shows significant surface area to volume ratio, high electronic and ionic conductivity. Carbon soot is produced by simply burning of candles and hence, it is ecofriendly, economical and useful. Carbon soot coated sponge and mesh can show stability, durability, reusability and reproducibility. Hence, this review is helpful to upcoming researchers to develop highly scalable superhydrophobic surfaces for efficient oil-water separation applications.

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