Synthesis and application of modified commercial sponges for oil-water separation

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Abstract

Oil adsorption sponges have attracted great attention in the field of oil-water mixture treatment, due to their low cost, high porosity, flexible design, and three dimensional skeleton structures. However, the hydrophilic-oleophilic of commercial sponge limits its application for oil-water treatment. To improve the adsorption performance and simplify the oil recovery process, various modification methods have been studied recently. Herein, we discuss the fabrication of hydrophobic/oleophilic sponges and summarize the recently reported modify materials onto sponges. Furthermore, some additional material properties are also presented, which make sponges superior capacity for addressing oil spills under extreme conditions. In addition, some oil collecting devices based on sponges, which are used to facilitate the oil collecting processes, are also shown. Meanwhile, the outlooks and challenges are also offered for future applications of sponges in the remediation of oil spill.

Keywords: modified melamine sponge; polyurethane sponge; oil-water separation; oil spill; modification; application

1. Introduction

The frequent occurrence of oil spill and chemical leakage has caused severe environmental pollution and ecological problems, which make oil-water separation a increasing serious challenge [1, 2]. Currently, several methods have been studied to lower the adverse influences of oil spill and chemical leakage in environment, including in situ burning [3], skimming [4], bioremediation [5-9], chemical dispersion [10], and adsorption [11-13]. Among the methods mentioned above, adsorption has aroused considerable interests because of its facile operation, low cost and flexible design [14-17]. An ideal adsorbent material for oil-water separation should possess excellent water repellency, superior adsorption performance, outstanding reusability and low material cost [18]. However, many conventional adsorbent materials, such as vegetable fibers [19], wool [20], modified organophilic clay [21], exfoliated graphite [22], and cellulose-based materials [23], exhibited various defects including low oil adsorption efficiency, poor recyclability and high materials cost [24]. These drawbacks limit their practical application in oils removal. Therefore, it is necessary to explore adsorbents with excellent adsorption performance, low cost, and good recyclability.

Recently, commercial sponge adsorbents, including polyurethane (PU) sponge and melamine sponge (MS), have attracted great attention for oil-water separation due to their low cost, good flexibility, three dimensional structure, and facile design [25]. Given the outstanding properties, sponge adsorbents have been developed as alternative adsorbing materials. However, both PU sponge and MS are naturally amphipathic, which restricts the removal of oils from oil-water mixtures [26]. Therefore, recent studies have focused on the surface modification of PU sponge and MS, which altered the hydrophilic surface to hydrophobic surface. Such surface would make the sponge high oil adsorption capacity and good water repellency in the separation process of oil-water. To this end, various materials have been selected to decorate the commercial sponges, including silanes [25], hydrophobic polymers [27, 28], hydrophobic nanoparticles [29, 30], carbon materials [31-33] and so on. These modification materials could successfully switch the hydrophilic sponge surface to hydrophobic, making the sponge materials promising candidates for oil spill cleanup. More recently, sponge adsorbents with additional properties, such as magnetic property, stimuli-responsiveness, and excellent durability under harsh conditions, have also been explored to address the oil spill cleanup in some specific situations.

To date, a good deal of work has been done to explore the modification of sponge surface for enhancing its adsorption performance. However, these reports are scattered which is not conducive to a comprehensive understanding of the related information on the synthesis and application of modified commercial sponges. Recent related reviews mainly focus on various advanced oil sorbents. For example, Ge et al. [34] discussed the design considerations for the fabrication of oil sorbents and summarized recently developed oil sorbents. However, only a fraction of his work relate to commercial sponges, which can not satisfy the further understanding of design and fabrication of hydrophobic commercial sponges. Moreover, we note that an all-round overview summarized the modification methods and materials of commercial sponge is still absent. Based on this consideration, this review presents the recent advances related to the materials used for modification, followed by a discussion of methods of modification. In addition, we address other important properties of sponge adsorbents to treat with oil spill under harsh conditions, and list some oil collection devices. Finally, the outlooks and challenges for fabrication and application of sponge adsorbents are also discussed to expect the better future applications of sponge adsorbents in the oil-water separation. Herein, we deem that this review will provide a good perspective for the design and application of ideal sponge adsorbents for oil spill treatments.

2. Methods for sponge modification

In order to satisfy the requirement of application in oils and solvents removal, it is imperative to develop facile and efficient method to tune the hydrophilic sponge surface to hydrophobic. In this part, we will summarize the general synthetic methods reported in the literatures, including dip coating, chemical vapor deposition, in situ chemical reaction, carbonization, and other methods.

2.1 Dip coating

Dip coating is the most widely used method to fabricate desired sponges. It is not required sophisticated technique or equipment and simple to operate. Moreover, the process is time-saving and only takes a few hours to obtain the hydrophobic sponge. In the dip coating processes, sponges were immersed in solution which containing modified materials for a few times, followed by a dry process, and then the hydrophobic sponges were obtained (Fig. 1a). Organosilicons modified sponges were usually prepared by dip-coating method. Pham [25] silanized melamine sponge with octadecyltrichlorosilane via dip coating method. The prepared sponge exhibited outstanding superhydrophobicity, high adsorption capacity up to 163 g/g for chloroform, excellent recyclability and good adsorption capacity retention. Similarly, polydimethylsiloxane (PDMS)-functionalized sponge was fabricated by dip coating [35].

Besides, dip coating was also used in the preparation of nanomaterials functionalized sponge. Qiang et al. [36] fabricated a functionalized graphene nanoribbon/PU sponge via a facile dip coating method, the obtained sponge exhibited good water repellency, excellent electrical resistance, and high adsorption performance. In the process of dip coating method, to facilitate the nanomaterials coated on the skeletons of sponges, binding agents were used. For example, cellulose nanowhiskers (CNWs) were used to strengthen the binding force between graphene sheets and sponge surface [37]. The as-prepared sponge exhibited superhydrophobicity with water contact angle (WCA) of 152°, which endowed the modified sponge good oil adsorption efficiency for oil-water separation. The thin PDMS films were also used to cement the graphene nanosheets onto the sponge skeletons [38]. Moreover, Ge et al. [39] coated hydrophobic SiO_2 NPs and polyfluorowax (PFW) on the PU sponge skeletons to form superhydrophobic surface via dip coating process. In his work, PFW was used to improve the adhesion between SiO_2 NPs and sponge. In addition, dip coating was also used to prepare magnetic sponges. For example, Fe_3O_4 NPs and lignin incorporated with melamine sponge via dip coating not only exhibited good water repellency, high adsorption capacity, and superior self-cleaning performance, but also could be magnetically driven to remove the oils from water surface [29].

2.2 Chemical vapor deposition

Chemical vapor deposition (CVD), a competent technique used to produce high purity and excellent performance materials, has been widely used to fabricate hydrophobic sponges [40, 41]. Wu et al. [42] prepared a robust SiO_2/Fe_3O_4/PU sponge via CVD. The modified sponge exhibited magnetic property and superhydrophobicity/superoleophilicity. Pyrrole modified sponges always were fabricated by CVD method. For example, a thin layer of PTES-polypyrrole (PPy) was used to modify the sponge via a simple CVD process (Fig.1b) [27]. The obtained sponge exhibited high porosity, good flexibility, excellent adsorption performance, and especially selective absorption for oil removal. Similarly, Chen et al. [43] synthesized a superhydrophobic melamine sponge, and pyrrole was used to modify the MS sponge by a simple vapor-phase deposition. Compared with dip coating method, an advantage of CVD is that it can control the turbulent modification process, resulting in a density and orderliness coating on the sponge surface [44, 45].

2.3 In situ chemical reaction

In the in situ chemical reaction process, the commercial sponge with three dimensional structure can act as a template, and the functional groups on sponge surface suggest that it can serve as a reactant to react with modification materials to form hydrophobic sponge surface. As such the in situ chemical reaction method can significantly improve the binding force between the modification materials and sponge skeleton. Herein, we discussed four categories, including wet chemical reaction, polymerization, thermal treatment, and electroless deposition, which are commonly used to fabricate hydrophobic sponges.

2.3.1 *Wet chemical reaction*

In the wet chemical reaction, the sponge can serve as an carrier or a reactant to participate in the chemical modification process [46]. Wang et al. [47] grafted PU sponge with polymer molecular brushes to fabricate a superhydrophobic PU sponge via wet chemical reaction. The obtained sponge exhibited high oil adsorption rate, excellent oil retention, and outstanding recyclability. Similarly, oleophilic monomer Lauryl methacrylate (LMA) was also used to modify the sponge surface via in situ chemical reaction [48], due to its long-chain alkyl group which has a good affinity to oils [49-51]. Moreover, urea cross-link modified sponge was fabricated through in situ chemical reaction. The obtained sponge showed superhydrophobicity with WCA of 153.4°, high adsorption capacity (54-100 g/g), and outstanding selectivity [52].

2.3.2 Polymerization

Polymerization is also a common method to fabricate a superhydrophobic sponge in recent years. In this reaction, polymer is crosslinked to form polymer chains, which can serve as crosslink agent to connect hydrophobic materials with sponge surface to produce hydrophobic sponge surface. Dopamine is the most commonly used crosslink agent, which could self-polymerize and produce stable covalent or noncovalent bonding force with materials [53]. For example, inspired by the remarkable adhesive ability of dopamine, Wang et al. [54] prepared a superhydrophobic and superoleophilic carbon nanotubes (CNTs) reinforced PU sponge, which involved the oxidative self-polymerization of dopamine. The obtained sponge exhibited adsorption capacity for oils up to 34.5 g/g, and it remained high adsorption performance after 150 times reused for the removal of oils from oil-water mixtures.

Similarly, Lei et al. [55] immersed the MS into dopamine solution for 12 h to form PDA via polymerization, and the zeolitic imidazolate framework-8 (ZIF-8)/MS was fabricated via Knoevenagel reaction. Besides, furfuryl alcohol (FA) can serve as a modifying agent to decorate raw MS because it not only possess good miscibility of water and many organic solvents, but also can polymerize to form hydrophobic poly(furfuryl alcohol) (PFA) at different harsh conditions [56]. FA modified commercial MS via a polymerization process without using additional toxic and expensive modifying agents or solutions has been reported [57]. Such an FA modified MS exhibited excellent water repellency due to the polymerization of FA onto the MS surface. In the modification process, the abundant hydroxyl and carbonyl groups of FA could react with melamine through hydrogen bonding, and the PFA could be polymerized onto the sponge surface skeletons.

2.3.3 Thermal treatment

Thermal treatment is a relative green and cost-effective method because no additional hazardous reducing reagent is required. This method is commonly used to fabricate graphene modified sponge. Zhu et al. [58] immersed melamine foam in GO suspension and GO was reduced by the thermal reduction treatment at 180°C for 6 h, (Fig. 1c). In the thermal reduction process, GO was reduce to form graphene sheets, which could self-assembly on the sponge surface and tightly bonding with sponge surface skeletons under the van der Waals force. Besides, low temperature thermal treatment is also facile to fabricate superhydrophobic sponge. The as-prepared sponge exhibited superhydrophobicty with WCA of 153°[59]. Similarly, Zhou et al. [60] modified PU sponge with (3-Mercaptopropyl)trimethoxysilane and GO via a thermal treatment, the obtained sponge possessed a superior water repellency with WCA exceeding 160°, and the oil separation efficiency was greater than 99.5%. A superhydrophobic reduced graphene oxide coated melamine sponge was also prepared via thermal treatment [61]. The modified sponge showed WCA of 162°, and exhibited adsorption capacity for oils to 120 g/g.

2.3.4 *Electroless deposition*

Zhu et al. [62] utilized the electroless deposition method to modify sponge with superhydrophobic coating. In his work, a film of copper was selected to modify the pristine sponge via electroless deposition. In this process, copper crystalline was produced to coat on the pristine sponge surface, which could act as a cross-linking agent between the superhydrophobic coatings and sponge skeletons. The reported sponge showed high water repellency even after floating on corrosive solution for 19 h, indicating it possess great potential to apply in the extreme conditions.

The in situ chemical reaction has more advantages than other conventional methods, such as CVD, electrostatic deposition, because it does not need specific equipment and condition. Moreover, compared with dip coating, it can form stronger bonding force between the modified materials and sponge surface. In general, the in situ chemical reaction is a convenient, economical, flexible, and environmental friendly method to fabricate an ideal hydrophobic sponge [27, 63, 64].

2.4 Carbonization

Carbonization is a simple and efficient method to obtain modified sponges, which is suitable for the modification of melamine sponge to possess flame retardancy property. Appropriate carbonizing treatments are conducive to improve the oil adsorption capacity, chemical stability and temperature tolerance than dip-coated sponges [65]. Recently, several researchers reported a carbonization treatment to pristine MS. In the carbonization process, the raw MS was treated with high temperature under inert atmosphere to form hydrophobic sponge surface. The strategy includes pyrolysis temperatures over 800°C [66, 67], pyrolysis pre-processing with coating additives [68, 69], and pyrolysis post-processing with additives [70]. Yao et al. [71] treated melamine sponge with deoiled asphalt (DOA) before sponge was pyrolyzed at 600°C for 2 h under argon atmosphere (Fig. 1d). Similarly, an ultralight melamine foam was fabricated by pyrolyzing sponge at 400°C for 1 h under nitrogen atmosphere and post hydrophobic modified with chlorotrimethysilane, the as-prepared sponge exhibited excellent hydrophobicity, ultralight weight, and outstanding fire-resistance [70]. Moreover, in order to explore the effect of pyrolysis temperature on the sponge properties, melamine sponge was heated in a given temperature from 300°C to 800°C for 1 h under nitrogen atmosphere [72]. The results showed that a carbonization treatment limited to 500-600°C and without the introduction of additives or further treatment is enough to get hydrophobic sponges for oil-water separation.

2.5 Other methods

Besides the methods mentioned above, other methods, including rapid microwave method [73], atom transfer radical polymerization (ATRP) [74, 75], and radical copolymerization [76] have also been reported to prepare superhydrophobic sponges in recent years. For instance, Tran and Lee [73] laden zinc oxide (ZnO) flakes on the PU sponge surface via a rapid microwave method, this approach towards engineering a suitable surface roughness. After a facile ZnO coating step performed with a commercial microwave, the hydrophilic PU sponge surface was tuned to hydrophobic surface, making the obtained sponge high adsorption capacity for oils removal. Lei et al. [77] fabricated poly (4-vinylpyridine) on the skeleton surface of MS through ATRP for oil adsorption and desorption. In addition, poly(N-isopropylacrylamide) (PNIPAAm) could also be grafted onto the surface of MS skeletons via ATRP [75]. Besides, spiropyran methacrylate monomers were integrated with the MS via a radical copolymerization process, and the obtained sponge showed superhydrophobicity with WCA of 155.5°, and exhibited high adsorption capacity of 70-154 g/g [76].

3. Research on sponge modification for oil/water separation

Hydrophobicity and oleophilicity play a pivotal role in the application of sponge adsorbents for oil-water separation. Recently, hydrophobic materials have been widely used to modify the hydrophilic sponge surface, and the as-prepared sponges possess a significantly improvement of the adsorption performance, which attracted more and more attention [27, 62, 78, 79]. In the following, we summarized materials that have been used to modify the surface of sponges, including organosilicons, organic monomers, polymers, carbon materials, and hydrophobic nanoparticles.

3.1 Organosilicons modified sponges

Organosilicons are used as the coating material because of its hydrophobic nature.

The present organosilicons available for design of hydrophobic sponges mainly include organosilanes and polysiloxanes. Organosilanes, such as vinyltrimethoxysilane (VTMS) [80], octadecyltrichlorosilane [25, 81], methyltrichlorosilane [82], heptadecafluoro-1,1,2,2-tetrahydrodecyltrimethoxysilane (FAS-17) [83], hexadecyltrimethoxysilane [84], fluoroalkylsilane [85], and dodecafluoro heptyl-propyl-trimethoxylsilane (Actyflon-G502) [1], are commonly selected to decrease the surface energy of sponge surface. Ke et al. [81] fabricated an octadecyltrichlorosilane silanized melamine sponge for oils removal in oil-water system. The reported sponge displayed excellent superhydrophobicity with a WCA of 153°. Also, methyltrichlorosilane salinized sponge exhibited a WCA of 143° and the adsorption capability of diesel is 65 g/g [82].

Polysiloxanes are good materials to modify the sponge surface due to its low energy surface and hydrophobic functional groups. For example, polydimethylsiloxane (PDMS) was utilized to modify the melamine sponge in a simple process by Chen et al. [35], who presented a superhydrophobic PDMS-functionalized sponge for oil-water separation. After modification, the obtained sponge exhibited superhydrophobic (WCA>150°) properties, high adsorption capacity (45-75 g/g), and good adsorption recyclability. In addition, Zhang et al. [26] prepared polymethylsilsesquioxane modified sponge, the wettability of the sponge with WCA reached 156° and showed good performance (58-127 g/g) in removal of various oils. As shown in Fig. 2a, the polymethylsilsesquioxane became covalently bound to the MS surface via a polycondensation reaction. After silanization, the modified sponge possessed superhydrophobic property and retained its inherent three-dimensional and porous structure.

3.2 Organic monomers modified sponges

Organic monomers with hydrophobic properties could be used as modified material for the surface of sponges, due to the organic monomers could give the sponge surface a long aliphatic chain, which endow the sponge with hydrophobicity [86]. Li et al. [48] fabricated hydrophobic PU sponges by a grafting of oleophilic monomer Lauryl methacrylate (LMA). As shown in Fig. 2b, LMA is an alkyl acrylate monomer with a long alkyl group chain. After the modification of LMA, the oil adsorption capacity of modified PU cubes was enhanced significantly, meeting the demand of oil spill cleanup. Except for LMA, other organic monomers, such as spiropyran methacrylate [76], furfuryl alcohol [57], octadecylamine (ODA) [54], dodecanethiol [87], and 1H, 1H, 2H, 2H-perfluoradecanethiol [88] are also competent to change the wettability of sponge surface to hydrophobicity.

3.3 Polymers modified sponges

Polymers with hydrophobic nature can be selected as modification materials to form hydrophobic sponge surface. Wang et al. [47] improved the hydrophobicity of a PU sponge by grafting a layer of polymer molecular brush. The WCA of the grafted PU sponge reached 152°. In addition, poly 4-vinyl pyridine (PVP) is also efficient for increasing the hydrophobicity of sponge surface (Fig.2c) [77]. Fluoropolymers is a good hydrophobic material to decrease the hydrophilicity of sponge surface due to its low surface energy of the -CF_3 group. Jiang et al. [89] coated sponge with a porous coordination polymer to fabricate a hydrophobic sponge. Because the abundant CF_3-groups of the polymer could significantly decrease the surface energy of sponge surface, which was conducive to improve the hydrophobicity of the modified sponge.

3.4 Hydrophobic materials modified sponges

3.4.1 Carbon materials

Given the intrinsic hydrophobicity, carbon materials have been widely used in the field of oil spill treatment. Based on this consideration, coating carbon materials onto sponge surface can easily turn hydrophilic surface to hydrophobic sponge surface. For example, CNTs are a commonly used modification materials due to its inherent water repellency [90-93]. Wang et al. [79] anchored the hydrophobic CNT/ poly-(dimethylsiloxane) PDMS onto sponge skeletons to alter its hydrophilic surface to superhydrophobic surface. The WCA of the as-prepared CNT/PDMS-coated PU sponge was 162°, making the sponge a promising candidate for oils adsorption. In addition, nanodiamonds (NDs) are one of the most promising new carbon-based materials for a wide range of applications. NDs modified PU sponge possessed superhydrophobic property (WCA=160°), good oil selective adsorption and excellent adsorption capacity (15-60 g/g) [94].

Besides CNTs and NDs, graphene, a kind of atomic-thick carbon nanosheets, has attracted tremendous attentions due to its hydrophobic nature and outstanding properties, and has also been used as a hydrophobic material to hydrophobilize sponge surface [95, 96]. More recently, superhydrophobic graphene modified sponges with superior oil adsorption performance have also been reported [37, 97-99]. Tjandra et al. [100] reported two types reduced graphene oxide/PU sponge. In his synthesis process, c-methacryloxypropyl trimethoxy silane (KH-570) was used to decorate PU sponge surface, the obtained sponge showed superhydrophobic property [101]. In addition, (3-Mercaptopropyl) trimethoxysilane (MPS) was applied to reduce GO to obtain MPS functionalized graphene/PU sponge, the modified sponge possessed superhydrophobicity with WCA larger than 160° [60]. Graphene coated on sponge surface can not only switch sponge surface from hydrophilic to hydrophobic, but also enhance the roughness of the surface and improve the adsorption performance.

3.4.2 Hydrophobic particle materials

Hydrophobic particle materials attached on the smooth surface of sponge can form rough and hydrophobic surface. The particles used in this field including TiO_2 [30], SiO_2 [84], Ag [102, 103], ZnO [73], Fe_3O_4 [104], attapulgite particles [105], kaolinite [106], and so on. Before particles used to modified sponge, they always need pretreat process to obtain the hydrophobic particle. Cho et al. [30] pretreated TiO_2 NPs with oleic acid and trifluoroacetic acid (TFAA) to obtain hydrophobic TiO_2 NPs, then coated the as-prepared TiO_2 NPs onto melamine sponge. The modified sponge exhibited superhydrophobic property with WCA of 161° and excellent adsorption capacity up to 88.1 g/g. Superhydrophobic Fe_3O_4/PU sponge was also fabricated. In a typical process, Fe_3O_4 nanoparticles were pre-treated with silanes and then coated on the PU sponge to obtain Fe_3O_4/PU sponge [104]. The prepared sponge exhibited good water repellency with WCA of 154.7° and adsorption capacity up to 43 g/g. Similarly, superhydrophobic attapulgite (APT) modified PU sponge was synthesized as shown in Fig. 2d. The coated PU sponge exhibited robust water repellency with WCA of 160° under various extreme conditions, indicating it has widely applications in the separation of oil-water mixtures under different environmental conditions [105].

Hydrophobic materials not only tuned the hydrophilic sponge surface to hydrophobic surface, but also increased the adsorption capacity of modified sponge because the hydrophobic materials can adsorb various oils and organic solvents [106].

4. Desirable properties of modified sponges

The properties of modified sponges play a vital role in the application of as-prepared sponges. Hydrophobic property is essential for the modification of sponge. Moreover, high adsorption capacity is regarded as a prerequisite for an ideal adsorbent. Meanwhile, sponges with additional properties are beneficial to apply in different conditions. In this part, we will not only summarize the adsorption capacity of modified sponges, but also introduce the additional properties, including adsorption capacity, magnetic property, adaptive capacity, stimuli-responsive property, emulsion separation capacity, and durable capacity. All these properties enabled the modified sponges could apply to a variety of extreme environment.

4.1 Adsorption capacity

The adsorption capacity is an important factor for the design of hydrophobic sponges. The commercial sponges possess high adsorption capacity due to its high porosity. In general, the porosity of PU sponge and MS is over 97% and 99%, respectively [25, 27]. After modification, the porosity of as-prepared sponge was slightly decreased compared to pure sponge, because of the surface loading of modified materials. For example, lignin shells were aggregated on the sponge surface to improve hydrophobicity, whereas the porosity of as-prepared sponge was 96.5%, which was slightly decreased compared to the

99% porosity of pure sponge. The adsorption capacity of obtained sponge was 18-51 g/g for various oils and organic solvents [107]. In comparison, a silanized sponge with the porosity of 99.48%, which was nearly identical compared to the porosity of pure sponge (99.49%), exhibited high adsorption capacity for various oils and organic solvents from 82 to 163 times its own weight [25]. The modified sponge retained its high porosity is highly beneficial to improve adsorption capacity.

In addition, it is an effective approach to increase the adsorption performance of sponges by combining an oil adsorbing material with sponge. Recently, large number of reports were focused on the graphene modified sponges, graphene can not only turn amphiphilic sponge to hydrophobic/oleophilic sponge, but also significantly increase the adsorption capacity of modified sponge because graphene is a good adsorbent to absorb wide range of oils and organic liquids [60, 108]. For example, Cho [97] prepared a few layer graphene based sponge, which showed high adsorption capacity up to 153 g/g for the chloroform. A graphene melamine sponge showed adsorption capacity of 130 g/g for phenixin [109]. Similarly, the TiO_2 NPs, attapulgite, and kaolinite are good adsorbents. After combination with sponges, the modified sponges showed improved adsorption capacity for oils. For example, kaolinite modified sponge showed adsorption ability for DMF and diesel oil of 89 g/g and 76 g/g, respectively [106]. Recently reported modification materials, modification methods, adsorption capacity, and the WCAs of the prepared hydrophobic sponges are summarized in Table 1.

4.2 Magnetic property

Sponges with magnetic property can be controlled by a magnet to remove and collect oils, which have been designed for oil-water separation [99, 110]. Liu et al. [111] immersed the PU sponge with the mixture solution of graphene oxide and $FeSO_4·7H_2O$ in Teflon vessel to fabricate magnetic graphene PU sponge. The as-prepared sponge possessed superhydrophobicity with WCA of 158 ± 1° (Fig. 3a). Similarly, Liu et al. [1] fabricated a magnetic sponge by dipping the pristine sponge in a solution containing Fe_3O_4 magnetic nanoparticles and low-surface-energy compound Actyflon-G502 (dodecafluoroheptyl-propyl-trimethoxylsilane) to construct the superhydrophobic surfaces for PU sponge [104]. These magnetic modified sponges not only possessed excellent adsorption performance, but also could be easily controlled by a magnet to facile separate oils from oil-water mixtures.

4.3 Adaptive capacity

4.3.1 Anticorrosive property

Recently, studies concerning coating anticorrosion materials on the sponge materials for specific application in oil recovery have attracted great attention. It is well known that corrosive solutions have an effect on the structure or chemical composition of adsorbent materials, resulting in an adverse impact on the hydrophobicity and adsorption performance of sponges. Liu et al. [83] introduced the hydrophobic molecules heptadecafluoro-1,1,2,2-tetrahydrodecyltrimethoxysilane (FAS-17) on the Fe_3O_4/PU sponge. The obtained PU sponge not only displayed superior water repellency, magnetic responsiveness, and more favorably possessed outstanding stability against corrosive solutions. Moreover, it showed excellent separation performance in the complex environment under magnetic actuation (Fig. 3b), and still remained the hydrophobicity after immersed in corrosive liquids (pH values from 1 to 14) for 24 h. A superhydrophobic sponge by modifying PDA-coated MS with dodecanethiol (DDT) was reported [87]. And the obtained sponge was tested in various solutions with different pH for a day to investigate its anticorrosive property, the results showed that the sponges remained hydrophobicity with the WCAs about 150°, indicating the as-prepared sponges have good resistance toward corrosive solutions.

4.3.2 Temperature tolerant property

It is uncertain that the oil spill will occur on high or low temperature. Based on the consideration, sponges with temperature tolerant are in demand. Compared with PU sponge, melamine sponge is an ideal temperature tolerant and fire-resistant material due to its high nitrogen content [112]. The modified melamine sponge also showed a similar property, which could apply to adsorb oils from oil-water mixtures in a high temperature condition. Wan et al [113] presented a MoS_2@RTV sponge, which exhibited excellent hydrophobicity (WCA > 145°) even exposed to 200°C and -20°C. Ruan et al. [88] fabricated a 1H,1H,2H,2H-perfluorodecanethiol modified PDA-coated sponge, which inherited the fire resistance of the pristine MS. It showed highly stable and robust against various harsh conditions and could work in a temperature between -196°C and 200°C (Fig. 3c). Moreover, a multiple walled carbon nanotubes modified melamine sponge was fabricated, which not only showed superhydrophobicity with WCA of 159.8°, but also featured excellent flame resistance [93]. This property makes the as-prepared sponge a promising adsorbent which can apply to harsh conditions for oil-water separation.

4.4 Stimuli-responsive property

Recently, stimuli-responsive materials, such as pH-responsive, thermoresponsive, and light-responsive, have attracted great attention and been reported by many researchers [114, 115]. The stimuli-responsive materials decorated on sponges could endow the modified sponges with a switching property for changing its wettability under specific conditions. For example, Lei et al. [77] grafted poly (4-vinylpyridine) on the MS skeleton surface to fabricate the pH-responsive sponge. This responsive sponge showed superhydrophilicity underwater at pH values of 1.0, and highly hydrophobicity at pH values of 7.0. This pH responsiveness endowed the modified sponges could adsorb the oil pH = 7.0 and then released the adsorbed oil underwater at pH = 1.0 to remove the oils from oil-water solutions (Fig. 4a). In addition, pH-responsive poly(2-vinylpyridine) (P2VP) and hydrophobic PDMS were used to modify PU sponge [116].

A temperature responsive melamine sponge was synthesized by Lei et al. [75]. Poly (N-isopropylacrylamide) (PNIPAAm) is a thermoresponsive material, thus the PNIPAAm/MS possessed a smart surface with controllable wettability between superhydrophilicity and superhydrophobicity under defined temperature (Fig. 4b). Simultaneously, the obtained sponge could adsorb oils under water at 37°C, and then released the adsorbed oil under water at 20°C by taking advantage of this property, indicating the modified sponge could finish the adsorb-desorb process by changing the temperature without additional procedures.

Recently, light-responsive materials have aroused great interests because light is easily acquired. The light-responsive material modified sponge would make the obtained sponge a good conversion switch between hydrophobic and hydrophilic by giving its visible light or UV irradiation. For example, Spiropyran, a commonly used light-responsive compound, exhibited hydrophobicity under visible light, while it could switch to hydrophilicity under UV irradiation (λ = 365 nm) [117]. Zhu et al. [76] fabricated an intelligent light-responsive spiropyran methacrylate derivative (SPMA) modified melamine sponge (Fig. 4c). The amphiphilic MS was modified with vinyl, and then the pretreated sponge was copolymerized with SPMA to fabricate the SP-MS. The modified sponge showed a controllable wettability between superhydrophobicity and superhydrophilicty under light-initiation and UV light, respectively. It provided a novel way to collect oils by simple UV light irradiation, as shown in Fig. 4d.

4.5 Emulsion separation capacity

The fabricated sponge with oil/water emulsion separation capacity is important for water pollution treatment. In general, the selective oil wettability of modified sponge makes sponge absorb tiny oil droplets from oil/water emulsions. Wang et al. [118] tested the separation efficiency of as-prepared sponge for the n-Hexane-in-water and toluene-in–water emulsion. After adsorbed by sponge, the emulsion became transparent while the initial emulsion was milky liquid. The results showed that the transparency of n-hexane-in-water and toluene-in–water emulsion increased to 85.4% from 6.2% and 80.7% from 14.7% respectively. Moreover, the separation capacity of a superhydrophobic sponge for five emulsions has been investigated [87]. The results displayed that the transparency of n-hexane, toluene, and chloroform rise up to 93.8%, 92.7%, and 91.5% respectively, whereas the transparency of emulsions formed by gasoline and diesel was 76.6% and 84.2%, which lower than that of solvent oil-in-water emulsion due to the complex chemical constituents of the two oils. The emulsions mentioned above are surfactant-free emulsions. Besides, the separation capacity of modified sponge for surfactant-stabilized emulsion has also been studied. Tween 80-stabilized oil-in-water emulsion was selected as object of study. Li et al. [105] fabricated a superhydrophobic sponge, which exhibited a separation efficiency for five Tween 80-stabilized oil-in-water emulsions, including kerosene-, diesel-, petroleum-, toluene-, and n-hexane-in-water emulsions, was up to 99.87%. In addition, three types of Tween 80-stabilized hexane-, hexadecane-, and soybean-in-water emulsions were tested by Kong et al [119]. As shown in Fig.5a, the original milky emulsions have been demulsified by modified sponge, and the separation efficiency for the three types emulsion were greater than 99.90%, even up to 99.96% for hexadecane-in-water emulsion. The high separation efficiencies of modified sponges for various oil-in-water emulsions make it a good candidate material for oil/water emulsion treatment.

4.6 Durable capacity

4.6.1 Mechanical property

An ideal sponge adsorbent should exhibit excellent mechanical property, which is important to increase the lifetime of adsorbent. The raw sponge shows good elasticity and flexibility, which makes sponge an excellent template to fabricate sponge adsorbent for oil/water separation. Ji et al. [109] repeated 1000 times of cyclic compression test to evaluate the mechanical property of the as-prepared sponge, The cyclic stress–strain curves with a maximum stress of 15 kPa and strain of 60%. As shown in Fig. 5b, even after 1000 cyclic compression cycles, the sponge could completely return to its initial shape, indicating no plastic deformation and outstanding elasticity and flexibility. Even, the modification of sponge could improve the compressive strength of the modified sponge, which has been reported at Zhou's work [60]. In his work, the modified sponge loaded nearly twice compressive stress than that of pure sponge, whereas exhibited slower decline rate than the initial sponge, suggesting the flexibility of modified sponge was not decrease compared with pure sponge. Moreover, the modified sponge also showed good elasticity under high compressive stress. The obtained sponge displayed 80% of strain when the stress was 61 kPa, and the sponge almost returned to its initial shape even after 500 compression cycles [119]. The excellent mechanical property of the modified sponge makes it a reusable adsorbent material for practical application.

4.6.2 Recycling property

The recycling property of sponge adsorbent is a very vital consideration for the oil/water separation in practical application. The excellent deformability of sponge means that the sponge could multiple reuse. In general, the recyclability of the as-prepared sponge was evaluated by repeated adsorption-squeezing processes, the sponge adsorbed oil to saturate then manually squeezing oil and the sponge was used for next cycle. Pan et al. [1] reported that the adsorption capacity of as-prepared sponge decreased within 6 cycles, after 6 cycles, even reach up to 50 cycles, the adsorption capacity has no obvious decrease. Moreover, sponge after modification maintained its elasticity and flexibility endow sponge superior recyclability with adsorption capacity retention up to 93% after 100 cycles, and more than 90% after 1000 cycles for various oils and organic solvents (Fig. 5c) [25]. Except for squeezing, combustion and distillation also can be utilized to test the recycling property of sponge adsorbents. Huang et al. [120] illustrated the recyclability of the as-prepared melamine sponge by three recycling process. Cyclohexane saturated sponge was removed by combustion for 100 cycles, n-hexadecane saturated sponge was removed by squeezing for 100 cycles, and heptane saturated sponge was removed by distillation for 50 cycles. The results showed that the modified melamine sponge possessed high reusable capacity recovered by the three approaches, which makes the modified sponge great potential in treating different kinds of oils and organic pollutants.

5. Oil collection devices

The initial way of collecting oils from oil-water mixtures by sponges was achieved by a simple mechanical squeezing process (Fig. 6a). Obviously, this process is toilsome and time-consuming work, and it is easy to destroy the three dimensional structure of sponge, resulting in the decrease of oils adsorption capacity. Recently, although great improvement has been achieved to develop sponges with high oils adsorption performance, the repeated adsorption-squeezing process are extremely laborious. In this section, we discussed two types oil collection device which were commonly used to addressing the above problems.

5.1 Filter type oil collection device

The modified sponges possess hydrophobicity and lipophilicity to adsorb oils from oil-water mixtures. For this property, sponge materials can serve as filter to separate oil from water. As shown in Fig. 6b, in the separation of oil-water mixtures, the sponge surface would adsorb oils and repel water so that the oils could be collected while the water is retained in the sponge surface [87]. For instance, Zhu et al. [58] fabricated graphene modified sponge, which fixed with a customized glass funnel to test its oil/water separation efficiency (Fig. 6c). When an oil/water mixture was poured into the glass funnel, the red organic rapidly adsorbed and then collected in the bottle, whereas the blue water was repelled. This device is facile to operate and only several simple equipment are required. Moreover, the device can save the process of collection and desorption, and reduce the adsorbents consumption.

5.2 Pump type oil collection device

The separation capacity of filter type oil collection device is restricted. For the purpose of continuous separation of oils from oil-water mixtures, there is a need to explore another oil collection devices. Recently, Yu et al. [121] have verified that pumping through porous oil adsorbent could realize the continuous separation of oils in situ from oil-water mixtures, which significantly improve the separation efficiency, reduce the sponges consumption and simplify the oil recovery process. Therefore, it is a good choice by introducing a pumping force to the modified sponges to selectively and continuously collect oil from water surface in a practical oil spill treatment. According to this assumption, Jiang et al. [89] have simulated a lab-scale apparatus (Fig. 7a). Once the pump is turned on, the oils surrounding the sponge continually adsorbed and is pumped into the recovery vessel. When the oils are almost completely absorbed, the water is repel to penetrate the sponge, thus the oils are successfully removed from oil-water mixtures. (Fig. 7b). To evaluate the applicability of the apparatus to marine oil spill, they tested 10 h successive recovery of hexane and 8 h recovery of oil from water, the results showed that the modified sponge retained well separation efficiency during the experiment. In addition, they tested the stability and practicability of the apparatus under the harsh conditions, such as strong winds, powerful waves and extreme weather, which were given in the sea, as presented in Fig. 7c. The results showed that the oil separation efficiency was slightly affected, indicating that utilizing pumping force to achieve continuously separation of oils from water surface is a promising technique to treat with oil spill cleanup in practice. Similarly, Li et al. [122] utilized the vacuum pump to continuous remove the oils, the results showed that a small piece (0.32 g, 1.5 × 1.5 × 3.0 cm^3) of sponge could successively collect more than 13.0 L oils, which amount to 32000 times its own weight. Moreover, the device is also appropriate for removing oils underwater. The successive collection of CCl_4/water mixture has been tested [119]. The flow rate was 8.9 × 10^4 $L \cdot m^{-2} \cdot h^{-1}$, and the separation efficiency was reach up to 99.99%. All the results suggest that the modified sponge is suitable for the removal of large amounts of oils and organic solvents in oil spillage.

6. Conclusions and outlooks

In summary, commercial sponge materials have been explored for the design and application as adsorbents for the removal oils. Herein, we have discussed the modify considerations for sponges to tune the hydrophilic surface to hydrophobic surface. In order to remove oils from water, the modified sponge must possess hydrophobic and oleophilic properties. Based on this consideration, numerous materials and methods have been developed to fabricate desired sponges. Recently, various hydrophobic materials and deposition methods have been applied to modify sponges, which has not only simplified the process of surface modification, but also greatly improved the adsorption performance of sponges and decreased the cost. In addition, we have also discussed sponges with additional properties that are conducive to be used for specific application, which greatly expand the application of modified sponges in different environments. Moreover, we have also presented two types oil collecting devices which were commonly used. These devices have not only significantly decreased the consumption of sponges, but also simplified the oil collecting processes, and saved time and labor. Although numerous sponge adsorbents and oil collecting devices have been successfully developed, a great deal of issues need further exploration before these sponges and devices being applied in remediation of oil spills. Herein, some important issues are highlighted for further studies.

(1) Research the practical application of hydrophobic sponges in the real treatment of oil spill: These sponges and devices mentioned above are lab-scale, whereas oil spill is large scale. For the convenience of experiments, sponges were cut into small cubes. However, it is uneasy for small sponges to adsorb the massive oil spill. The small sponges float at random on the sea and will be floating freely on the sea, especially under harsh sea conditions.

(2) Fabricate sponges with superior oils adsorption performance of high viscosity oils and organic solvents: Most reported sponges exhibited excellent adsorption capability of low viscosity oils [54]. However, oils with high viscosity are difficult to pass through the sponges. Therefore, it is a meaningful research to explore a hydrophobic sponge adsorbent for the removal of high viscosity oils and organic liquids.

(3) Consider the effects of changeable ocean weather on the adsorption performance

of modified sponges: For example, Jiang et al.[89] highlighted the oil spill is often take place in the sea with complicated conditions, such as powerful waves, strong winds and extreme weather, thus sponge adsorbents should have the ability to overcome these conditions. Therefore, it is necessary for sponges have good mechanical strength and environment adaptability, and the oil collecting devices have long term stability and recyclability to withstand those environments.

(4) Explore a facile approach to disposal the oil-soaked sponges: Although various methods have been proposed to fabricate sponges, which exhibit excellent adsorption performance, these methods are still not efficient and the redundant solutions are not environmental enough since the reagents are toxic and harmful [34]. In addition, the oil-water separation efficiency of sponges would decrease after being used for a period of time, and sponges cannot be reused indefinitely. The waste sponges need to be solved for preventing second pollution. In a word, many challenges still exist for the fabrication of ideal sponge adsorbents, and considerable efforts should be devoted to better apply sponges to oil spill cleanup in practice.

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Table 1. Summary of modification methods, adsorption capacity, and WCAs for reported sponge adsorbents.

Sponge sorbents	Modification method	Adsorption capacity (g/g)	WCA	Ref.
Silanized melamine sponge	Dip coating	82-163	151°	[25]
Acylated melamine sponge	Dip coating	66.6-168.2	140°	[46]
PDMS-melamine sponge	Dip coating	45-75	>150°	[35]
Polymethylsilsesquioxane modified sponge	Dip coating	58.2-127.6	156°	[26]
Octadecyltrichlorosilane modified sponge	Dip coating/ polymerization	42-68	153°	[81]
MoS$_2$@RTV sponge	Dip coating/curing	56-104	152 ± 2°	[113]
SiO$_2$/GO-PU sponge	Dip coating	80-180	145°	[14]
RGO@PU sponge	Dip coating/thermal treatment	24.2-37.6	153°	[123]
SiO$_2$@VTMS sponge	Dip coating	60-109	-	[80]
Functionalized GO-PU sponge	Dip coating	16-35	152 ± 1°	[124]
SiO$_2$-PU sponge	Dip coating	33.9-55.8	>150°	[122]
SiO$_2$-PFW-PU sponge	Dip coating	22-75	156°	[39]
SiO$_2$-PDA-PU sponge	Polymerization/ dip coating	18.3-46.8	154°	[125]
TiO$_2$-melamine sponge	Dip coating	37.2-88.1	161.1°	[30]
Fe$_3$O$_4$-PU sponge	Dip coating	39.3-46.3	154.7°	[104]
Attapulgite coated PU sponge	Dip coating	17-45	160°	[105]
lignin/Fe$_3$O$_4$-melamine sponge	Dip coating	-	147°	[29]
Actyflon-G502 / Fe$_3$O$_4$-PU sponge	Dip coating	25-87	153°	[1]
GO/PU sponge	Dip coating	25-95	153°	[126]
Thiolated graphene sponge	Dip coating	29.5-90	160.5°	[98]
Few-laywer graphene sponge	Dip coating	57-153	143.5°	[97]

CNWs/GN/PU sponge	Dip coating	31-46	152°	[37]
MWCNTs-PDMS sponge	Dip coating	38-127	159.8°	[93]
CNT/PDMS-coated PU sponge	Dip coating	15-25	162 ± 2°	[79]
NDs-PU sponge	Dip coating	15-60	160°	[94]
FAS-17/ Fe_3O_4-PU sponge	Dip coating/ polymerization	10-35	153.7 ± 2.7°	[83]
PDA-ZIF-8 melamine sponge	Polymerization/ dip coating	10-38	140°	[55]
Furfuryl alcohol modified sponge	Dip coating/ polymerization	75-160	138-145°	[57]
Fluoroalkylsilane modified PU sponge	In situ chemical reaction	-	155°	[85]
Polymer molecular brush/PU sponge	In situ chemical reaction	17-40	152°	[47]
Fe_3O_4-graphene PU sponge	In situ chemical reaction	9-27	158 ± 1°	[111]
Graphene melamine sponge	In situ chemical reaction	99.0	-	[127]
USTC-6@GO@sponge	In situ chemical reaction	12-43	132°	[89]
Fe(III)-TA melamine sponge	In situ chemical reaction	69-176	157.0 ± 0.3°	[120]
Urea modified sponge	In situ chemical reaction	54-100	153.4°	[52]
NMP /graphene PU sponge	Polymerization	40-80	151.8 ± 0.5°	[119]
PU–CNT–PDA–ODA sponge	Polymerization	22-34.9	158°	[54]
LMA-PU sponge	Polymerization	50-69	-	[48]
DDT-PDA-melamine sponge	Polymerization	45.2-98.6	158°	[87]
1H,1H,2H,2H-perfluorodecanethiol-PDA sponge	Polymerization/dip coating	79-195	163.4°	[88]
Cu/$C_{11}H_{23}COOAg$/PU sponge	Electroless deposition	13	171°	[62]
Graphene melamine foam	Thermal reduction	60-140	151.4°	[58]
Silaned graphene PU sponge	Thermal treatment	25-44	>160°	[60]
rGO@MF sponge	Thermal treatment	90-120	162°	[61]
Carbonzied melamine sponge	Carbonization	-	146°	[69]
Carbonized asphalt-melamine sponges	Dip coating /carbonization	73-140	134.5°	[71]
Melamine-derived carbon sponge	Carbonization	90-200	140°	[72]
Lignin-melamine sponge	Dip coating /carbonization	98-217	142.5°	[68]
PPy-PTES-sponge	CVD	21.8-31	153.7°	[27]
PPy-PA PU sponge	CVD	22-62	140°	[41]
MF/PPy/Ag/F sponge	CVD	40-100	156.1°	[43]
PU@Fe_3O_4@SiO_2@FP Sponges	CVD	13.26-44.50	157°	[42]
SP-MF sponge	Radical copolymerization	74-154	155.5°	[76]
MF-OTS/PNIPAAm sponge	ATRP	35-70	-	[75]
PVP-melamine sponge	ATRP	49	135°	[77]

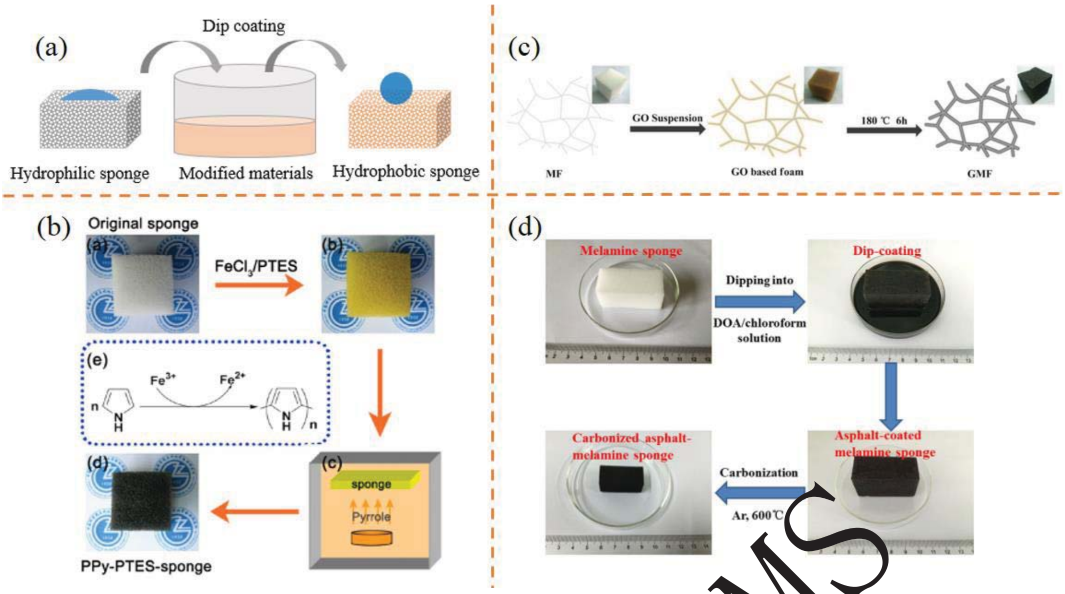

Fig. 1. (a) Dip coating methods to fabricate hydrophobic sponge. (b) Preparation of PPy-PTES-sponge via the chemical vapor deposition. (b) Reproduced with permission [27]. Copyright © 2013, American Chemical Society. (c) The in situ chemical reaction method to modify sponge. (c) Reproduced with permission [58]. Copyright © 2015, Small. Published by Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Carbonization of melamine sponge. (d) Reproduced with permission.[71] Copyright © 2016, Society of Chemical Industry.

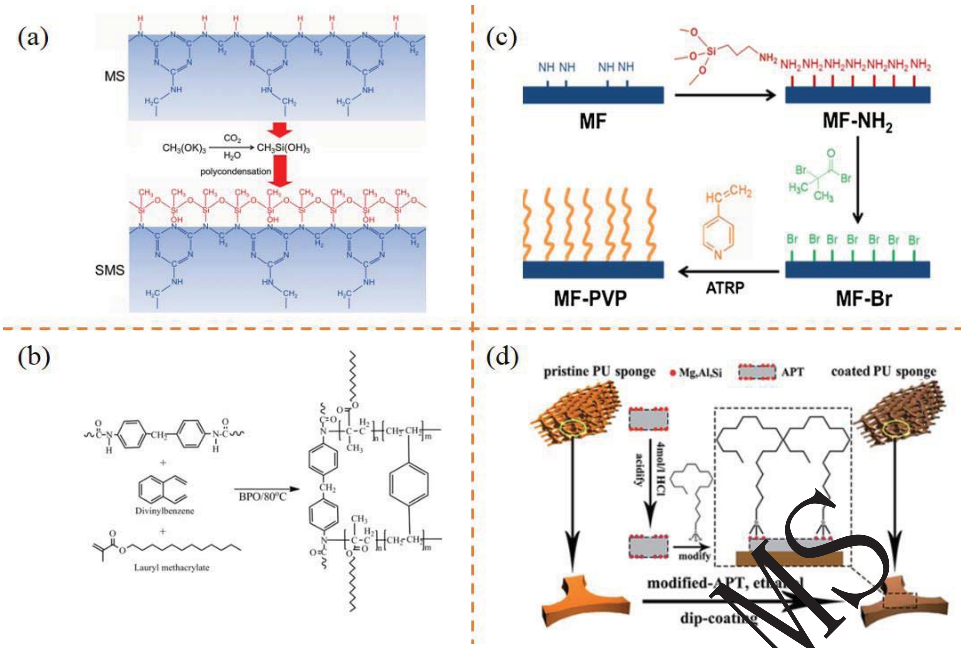

Fig. 2. (a) Schematic of the formation of a hydrophobic melamine sponge. (a) Reproduced with permission [26]. Copyright © 2017, published by Journal of Materials Science. (b) Reaction scheme for polymerization of LMA onto PU sponge surface. (b) Reproduced with permission [48]. Copyright © 2012, Marine Pollution Bulletin. Published by Elsevier B.V. (c) Preparation of MF-PVP sponges. (c) Reproduced with permission [77]. Copyright © 2017, Applied Surface Science. Published by Elsevier B.V. (d) Preparation of the hydrophobic PU sponge by coating APT onto the raw sponge. (d) Reproduced with permission [105]. Copyright © 2016, Journal of Materials Chemistry A. Published by the Royal Society of Chemistry.

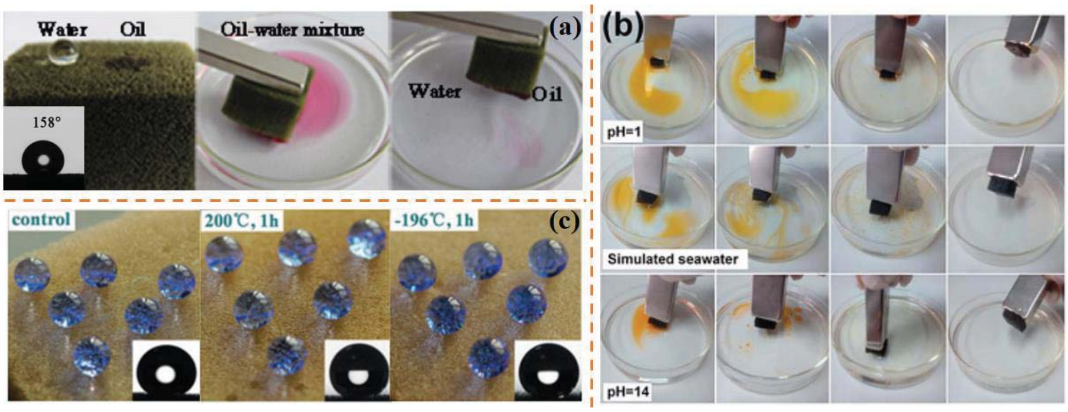

Fig. 3. (a) Superhydrophobic property of as-prepared sponge and the removal of oil from water under magnetic field. (a) Reproduced with permission [111]. Copyright © 2014, published by Elsevier B.V. (b) Photographs of dyed water droplets (10 mL) on the surface of the superhydrophobic MF sponges before and after low-/high-temperature exposure for 1 h. Inset: the corresponding water contact angle. (b) Reproduced with permission [88]. Copyright © 2014, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) The oil/water separation experiment under magnetic actuation in three different media such as acidic solution (pH = 1), simulated seawater, and basic solution (pH = 14). (c) Reproduced with permission [83]. Copyright © 2015, published by The Royal Society of Chemistry.

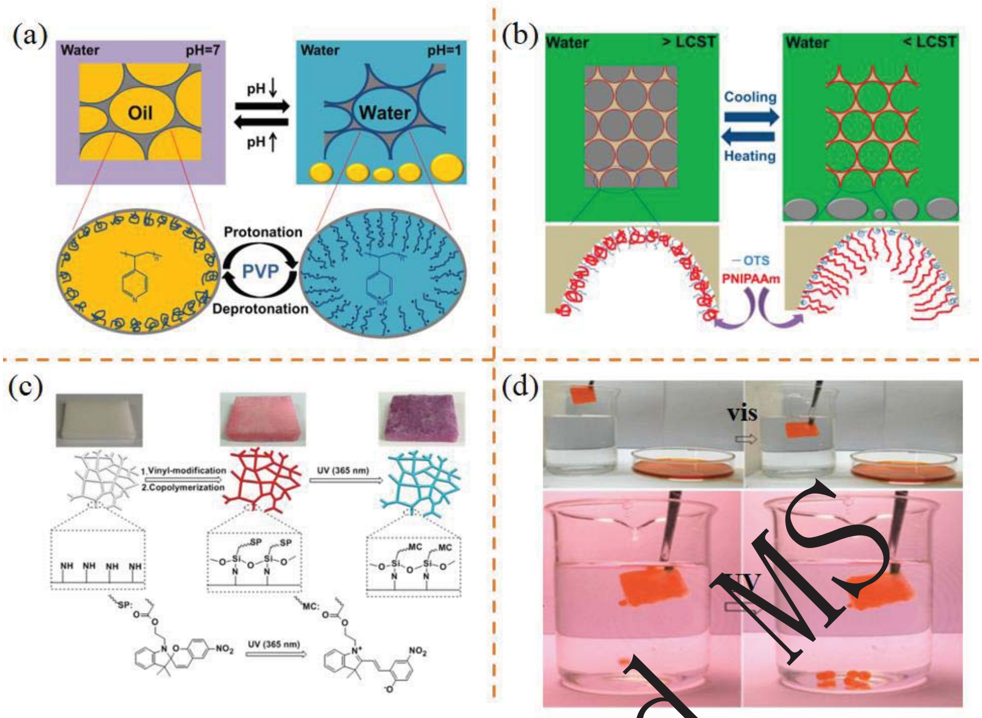

Fig. 4. (a) Schematic diagram of oil adsorption and desorption by MF-PVP sponges underwater at different pH values. (a) Reproduced with permission [77]. Copyright © 2017, Applied Surface Science. Published by Elsevier B.V. (b) Schematic diagram of the absorption performance of MFOTS/PNIPAAm sponges at different temperatures. (b) Reproduced with permission [75]. Copyright © 2017, American Chemical Society. (c) The synthetic route of a light-responsive superhydrophobic SP-MF sponge. (d) Oil desorption under vis and UV irradiation. (c-d) Reproduced with permission [76]. Copyright © 2015, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

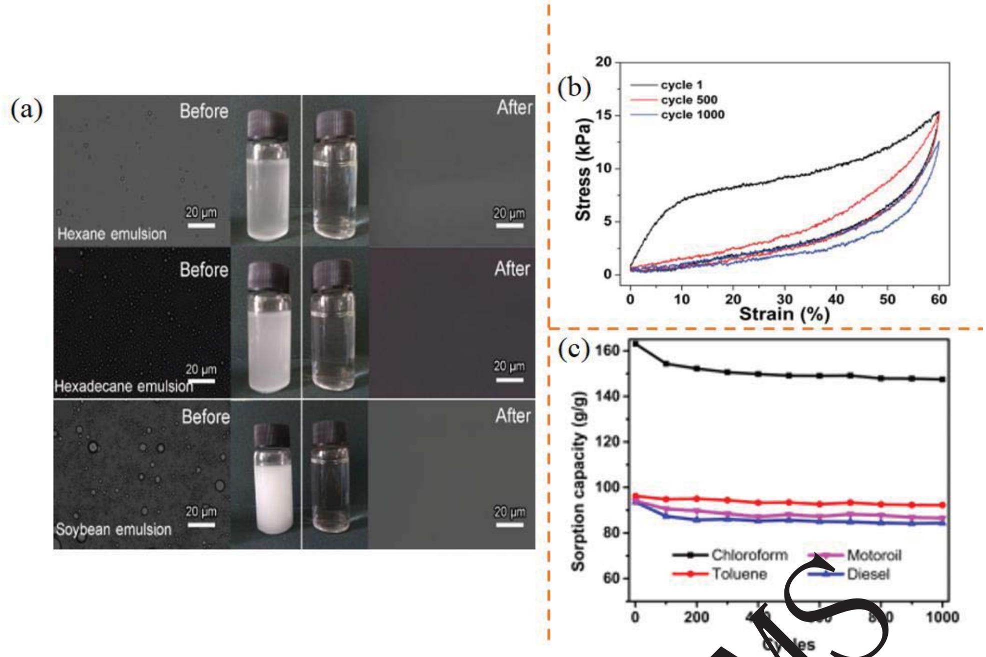

Fig. 5. (a) Demulsification of oil-in-water emulsions by modified sponge. (a) Reproduced with permission [119]. Copyright © 2017, Nano Research. Published by Springer. (b) Compressive stress–strain curves of the as-prepared sponge. (b) Reproduced with permission [109]. Copyright © 2017, Journal of Materials Chemistry A. Published by Royal Society of Chemistry. (c) Sorption recyclability of sMS-10 for various organic solvents and oils. (c) Reproduced with permission [25]. Copyright © 2014, Applied Materials & Interfaces. Published by American Chemical Society.

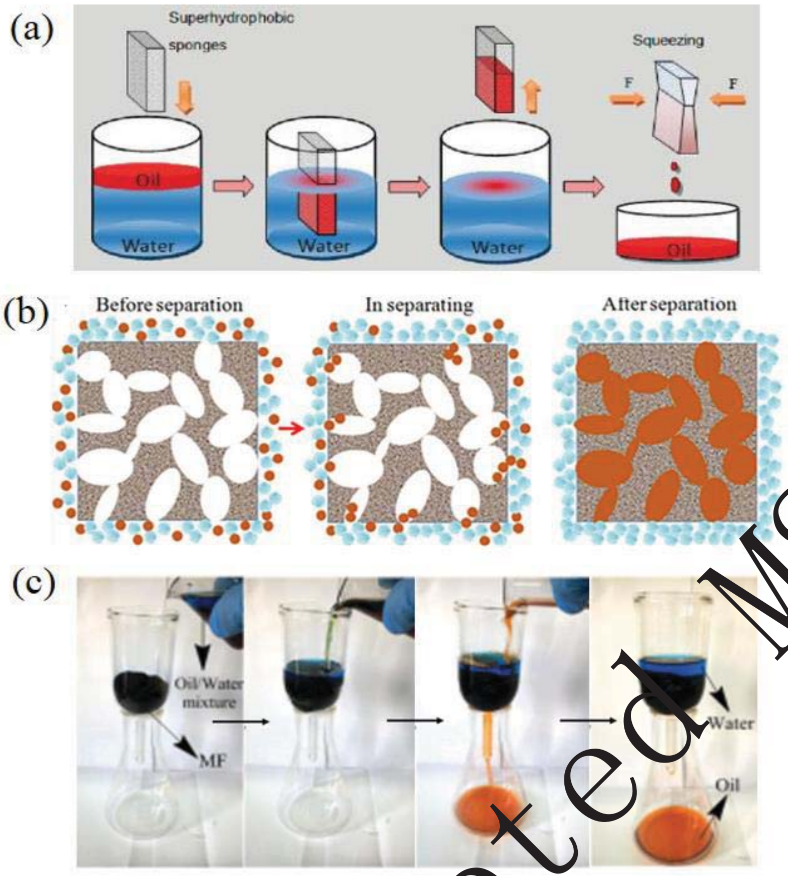

Fig. 6. (a) Illustration for the removal and collection of oils from the water surface by superhydrophobic sponges. (a) Reproduced with permission [62]. Copyright © 2011, American Chemical Society. (b) Schematic of the separation process of the oil-water mixture. (b) Reproduced with permission [87]. Copyright © 2017, Marine Pollution Bulletin. Published by Elsevier Ltd. (c) The oil/water separation process using the GMF (oil and water were stain with Sudan red and methyl blue, respectively). (c) Reproduced with permission [58]. Copyright © 2015, Small. Published by Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

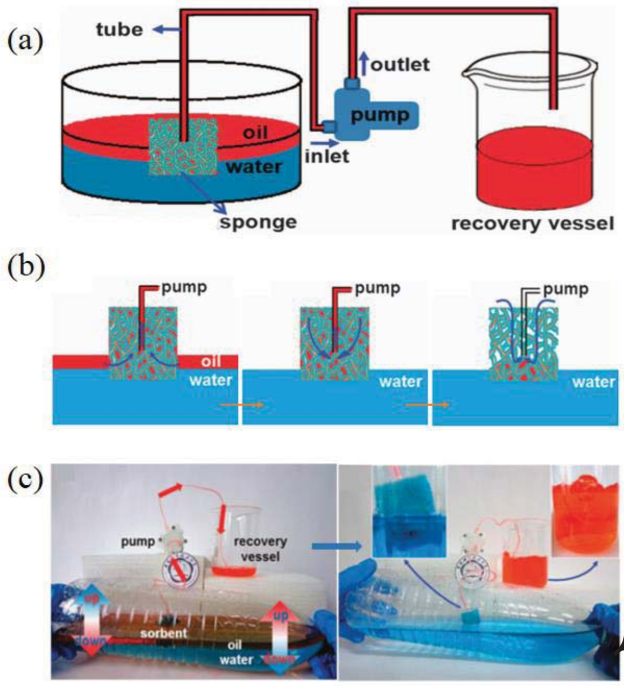

Fig. 7. (a) Illustration of the experimental apparatus for the in situ consecutive collection of oil on the surface of water into the recovery vessel. The oil and the water are colored blue and red, respectively. (b) An illustration of the oil recovery process with the three main stages accompanied by a pressure change in the oil–air and oil–water interfaces. (c) The apparatus continuously collecting the floating oil (dyed red) on the surface of water. (a-c) Reproduced with permission [89]. Copyright © 2016, NPG Asia Materials.