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REVIEW PAPER

Cotton fabrics modified for use in oil/water separation: a perspective review

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Abstract This article provides a perspective review on the use of modified cotton fabrics for oil–water separation. The principles of surface hydrophobicity of cotton fabrics are first described, from which the basis for producing superhydrophobic surfaces is presented. Then the preparation methods to convert hydrophilic cotton fabrics to hydrophobic fabrics are reviewed and discussed. Based on literature results the way to design novel preparation methods, the need to summarize testing protocols, and the comprehensive technoeconomic and sustainability analyses, are proposed. A demonstrative cotton fabrics test is used to reveal the significant role of conjugated fluid flows and surface interactions under different application scenarios for determining the separation efficiency of the oil–water mix.

Keywords Cotton fabrics · Hydrophobicity · Oil water separation · Testing

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Introduction

Oil spilling from life, industry and accident can adversely impact environment quality. The oil skimmer, centrifugation and air flotation processes were proposed to mitigate oil spilling pollution (Wang et al. 2020; Lu et al. 2020), which have drawbacks including low separation efficiency, high energy demand and time-consuming procedures (Shang et al. 2018; Dashairya et al. 2018). Oil-in-water wastewaters are generated in petrochemical, mineral and smelting, and food processing industries (Tao et al. 2014). Sufficient de-emulsification to convert stabilized micro droplets to large oil droplets can enhance oil–water separation efficiency in practice (Zhang et al. 2020a).

The porous sorbents, if applicable, can be effective to remediate oil pollution sites at low operational cost (Dashairya et al. 2019). A surface is considered hydrophobic with contact angle of water drop exceeding 90°. A surface is regarded superhydrophobic if the water contact angle is higher than 150° and the contact angle hysteresis is lower than 10° (Wang et al. 2016). A few superhydrophobic surfaces in nature can possess lotus leaf effect (Wei et al. 2010), rose petal effect (Shao et al. 2020) or butterfly wing effect (Bixler and Bhushan 2014). The porous sorbent with superhydrophobic surface should be able to adsorb oil from a water/oil mix (Hadji et al. 2020). For instance, a melamine sponge coated with hydrophobic TiO_2 nanoparticles is shown to preferably separate oils and organic solvents from water (Cho et al. 2016). Cai et al. (2020) fabricated fluorine-free hydrophobic HDTMS-TiO_2 coated mesh with high oil separation efficiency and cost effectiveness. Other modified matrixes including stainless mesh, polymer and melamine foams were applied for separating oil from water (Wang et al., 2020). However, these adsorbents are either produced in an environmentally unfriendly process or their final disposal can lead to secondary pollution (Lyon et al. 2020; Gong et al. 2019). The eco-friendly matrix with superhydrophobic surfaces is desired to tackle the oil spill pollution problems.

Cellulose is the most abundant biopolymer that is used as biodegradable and cost-effective oil adsorbent (He et al. 2020). Owing to the numerous hydroxyl groups presented on the pristine cellulose, hydrophobic modification of its surface should benefit the oil adsorption tendency from the spill (Mai et al. 2018). Cotton fabric is one of the cellulose-rich primary agricultural products, whose modification and use as oil spill adsorbent should have its promise. To apply hydrophilic cotton fabric for oil spill remediation acquires modifications to hydrophobilize the surfaces (Deng et al. 2020). This article summarized the commonly adopted modification methods including dip coating, in situ crosslinking, UV-triggered click reaction, self-affine modification and spray coating for fabricating superhydrophobic cotton fabric surface. A demonstration test with a modified cotton fabric is presented to reveal the needs to revise the testing method and to link to practical use of the fabricated cotton fabrics.

Water wetting and hydrophobicity

The hydrophobicity of a surface is attributed to the surface energy of substrate and determined by differences of the surface energies for the liquid and the solid surface (Krasowska et al. 2009). If a smooth surface, naturally or synthetically made, is subjected to a liquid drop in air; if the surface energy of the solid surface is higher than that of the liquid, the surface will be wet since the solid tends to pull the liquid molecules to it for minimizing their total energy level. Conversely if the surface energy of solid is lower than that of liquid, the solid would try to repel the liquid out to prevent increasing of its energy level.

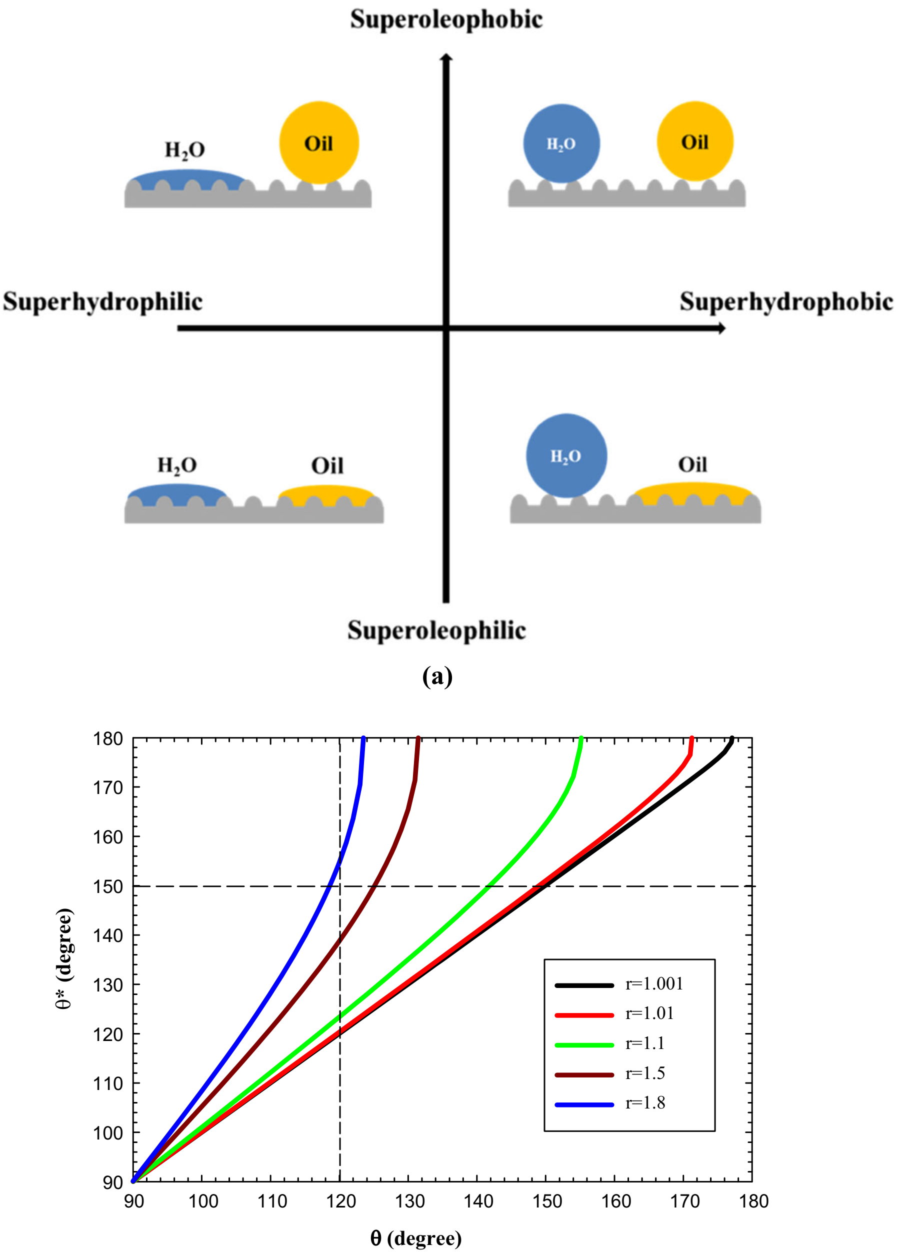
The way to estimate the surface energy is to measure the contact angle between the solid and liquid. When a droplet contacts with a solid surface, three interfacial forces will act on the contact point: interfacial force between liquid and vapor (γ_L), interfacial force between solid and vapor (also mean the surface energy of solid) (γ_S) and interfacial force between solid and liquid (γ_{SL}). The contact angle (θ) is measured by the angle between solid surface and a liquid droplet on the surface (Kwok and Neumann, 1999). The interfacial force balance at the three-phase point for a smooth surface can be presented as the Young's equation as follows:

$$\gamma_L \cos\theta = \gamma_S - \gamma_{SL}, \tag{1}$$

Based on Eq. 1, high θ ($> 90°$) acquires low γ_S, low γ_L and/or high γ_{SL}. Low γ_S indicates a surface with both low polar and dispersive interactions with the surrounding molecules [> 250 mJ/m^2 for metal and glass; 22–35 mJ/m^2 for C/H polymer films (Fenouillot et al. 2009); 18 mJ/m^2 for PFTE with only C and F atoms (Heitz and Dickinson 1999)]; low γ_L suggests a liquid with low inter-molecular interactions (18.4 mJ/m^2 for n-hexane vs 72.8 mJ/m^2 for water @ room temperature); high γ_{SL} corresponding to null work of adhesion, or there is no interaction between solid and liquid. The superhydrophobic surface can have high contact angle (θ) $> 150°$. At $\theta 180^o$, $\gamma_{SL} = \gamma_S + \gamma_L$. with null work of adhesion; restated, the solid and liquid should extremely repel each other. The schematics of superhydrophobicity and superoleophobicity of a solid surface is shown in Fig. 1a.

The theory by Girifalco and Good (1957) estimated minimum $\gamma_S = 6$ mJ/m^2, corresponding to maximum $\theta = 115.2°$. By plotting the plots of literature data with γ_{LV}–γ_S versus cos θ, the limit contact angle is estimated to be 156° for water and 109° for hexadecane on a surface at $\gamma_S = 0$ limit (Shafrin and Zisman 1964). However, since no $\gamma_S = 0$ surface exists, the maximum apparent contact angle on a real-world smooth surface involving a sharp edge is estimated 120° (Oliver et al. 1977). Therefore, by a completely wet, smooth surface it is impractical to yield superhydrophobic surface using pure chemical modification pathways.

If a rough surface, naturally or synthetically made, is placed by a liquid drop in air, very large apparent contact angle (θ^*) can be yielded by the contact angle hysteresis or by the air-trapped cavity to lift the drop from the solid surface. The Wenzel model predicts the link between θ^* and θ as $\cos\theta^* = r\cos\theta$, where r is the



(a)

(b)

Fig. 1 **a** Schematics of superhydrophobicity and superoleophobicity of a surface. Typical superhydrophilic and superoleophilic surfaces are high energy metal surfaces; typical superhydrophobic and superoleophilic surfaces are lotus leaves (Tuteja et al. 2007), typical superhydrophilic and superoleophobic surfaces are ammonia treated heptadecafluorononanoic acid modified TiO_2 coated SiO_2 nanoparticles coated sponge (Xu et al. 2015); typical superhydrophobic and superoleophobic surfaces are 1H,1H,2H,2H-perfluorooctyltrichlorosilane coated porous silicon (Chu and Seeger 2014). **b** θ^* versus θ^* based on Wenzel model. r is the roughness ratio

roughness ratio. Their correspondence is shown in Fig. 1b. For smooth surface (nearly unity r), $\theta^* = \theta$; for very rough surface with $r = 1.8$, hydrophobic surfaces with θ of ca. 120° can yield superhydrophobic surface with $\theta^* = 154°$. Restated, the rough surfaces with hydrophobic properties can generate superhydrophobic effects in need.

Numerous surface modifications were adopted to affect the hydrophobicity of surface, including the doping/grafting nanoparticles (Dashairya et al. 2019b; Rostami et al. 2019), polymers (Qin et al. 2019; Jannatun et al. 2020), fluoroalkylsilanes (Cai et al. 2020) and alkyl silanes (Nanda et al. 2019; Guo et al. 2018a), were used for increasing hydrophobicity of the modified cotton fabrics. For instance, the nanoparticles, particularly zinc oxide nanoparticles with satisfactory biocompatibility and biodegradability, were doped onto targeted surface for increasing local roughness (Bai et al. 2019; Mendoza et al. 2019). The use of polymer, such as polyurethane monolith can be applied to produce interconnected pore structures on a solid surface via phase separation pathway so generating superhydrophobic effects on elastic substrate (Ye et al. 2020). Silane agents can react with the hydroxyl groups of substrates then anchor on the surface, so a hydrophilic surface can be converted into hydrophobicity with the hydrophobic poly silane groups with increased alkyl chain length (Qi et al. 2019). The surface hydrophobicity can be synergistically enhanced by the hierarchical structure and roughness which decreases the apparent surface energy (Wang et al. 2009). The surface grafted by long chain poly(fluoro-methacrylate), if the grafted layer thickness is thick enough to prevent water penetration, would yield > 140° contact angle (Guo et al. 2020). (Apparently the surface of the grafted layer should not be smooth otherwise the contact angle cannot exceed 120°). Wang et al. (2017a) utilized hexadecyltrimethoxysilane (HDTMS) constructing hierarchical structure to fabricate superhydrophobic steel mesh. Huang et al. (2020) fabricated fluoroalkylsilane-modified balsa wood sponge to clean up viscous crude oil.

A superamphiphobic or superomniphobic surface is a superhydrophobic and superoleophobic surface (Chu and Seeger 2014), which was first developed by Tuteja et al. (2007) by employment of re-entrant surface curvature and chemical composition modification. The superhydrophobic and superoleophobic surfaces are ready to foul in the oil–water separation; and since water is denser than oil, the hydrophobic adsorbent would hinder water to flow though (Wang et al. 2015). The superhydrophilic and superoleophobic surface is preferred for oil–water separation so water can flow over but oil can be prevented (Su et al. 2017). However, superoleophobic surface dislikes water, so it is hard to make it hydrophilic. The mesh with coating can make it a hydrophilic and oleophobic matrix for adsorbing oil from flowing through water stream.

Fig. 2 Schematics of modification methods for cotton with superhydrophobic surfaces. **a** Dip-deposition of nanoparticles onto a substrate surface; **b** in situ crosslinking method; **c** thiol-ene click reaction; **d** crosslink with amino-containing silane via Michael addition/Schiff base reactions; **e** spray coating method with modification of HNTs then spray coat HNTs to establish superhydrophobic surface ▶

As stated, the rough surface that can provide air trapped space beneath the drop is essential to generate superhydrophobic and/or superoleophobic effects (Tuteja et al. 2007). The nanoscale or microscale surface roughness can decrease the θ^* if $\theta < 65°$ or increase the θ^* if $\theta > 65°$ (Tian et al. 2013). Numerous methods were applied for generating the surface roughness, random or regular, including nanoparticle deposition, chemical etching or lithography, sol–gel reactions, and galvanic deposition (Chu and Seeger 2014). The natural fabrics such as cotton fabrics have surface roughness of all scales, providing sufficient air reentrant cavities all over the cotton fabric surfaces. The remaining part is to convert the hydrophilic nature of cotton fabrics to hydrophobic. The hydrophobic surface can be synthesized by grating with fluorinate polymers or fluorine monomers, polyhedral oligomeric silsesquioxane (POSS), deposited fluorinated nanoparticles, or other hydrophobic compounds. Xu et al. (2015) prepared a superhydrophobic and superoleophobic surface by dipping PS fabric or polyurethane sponge to SiO_2 nanoparticles and heptadecafluorononanoic acid ($CF_3(CF_2)_7COOH$) modified TiO_2 sol. This coating can become superhydrophilic and superoleophobic after exposure to ammonia, which can adsorb water from bulk oil.

Preparation techniques to oil–water separation by modified cotton fabrics

Cotton fabrics were selected for oil spill separation based on its availability, degradability, and low cost (Singh and Singh 2019). Cotton fabrics are naturally grown fabrics with rough and hydrophilic surface, whose surface needs modification to make them fit the needs for oil–water separation (Zhou et al. 2013). As

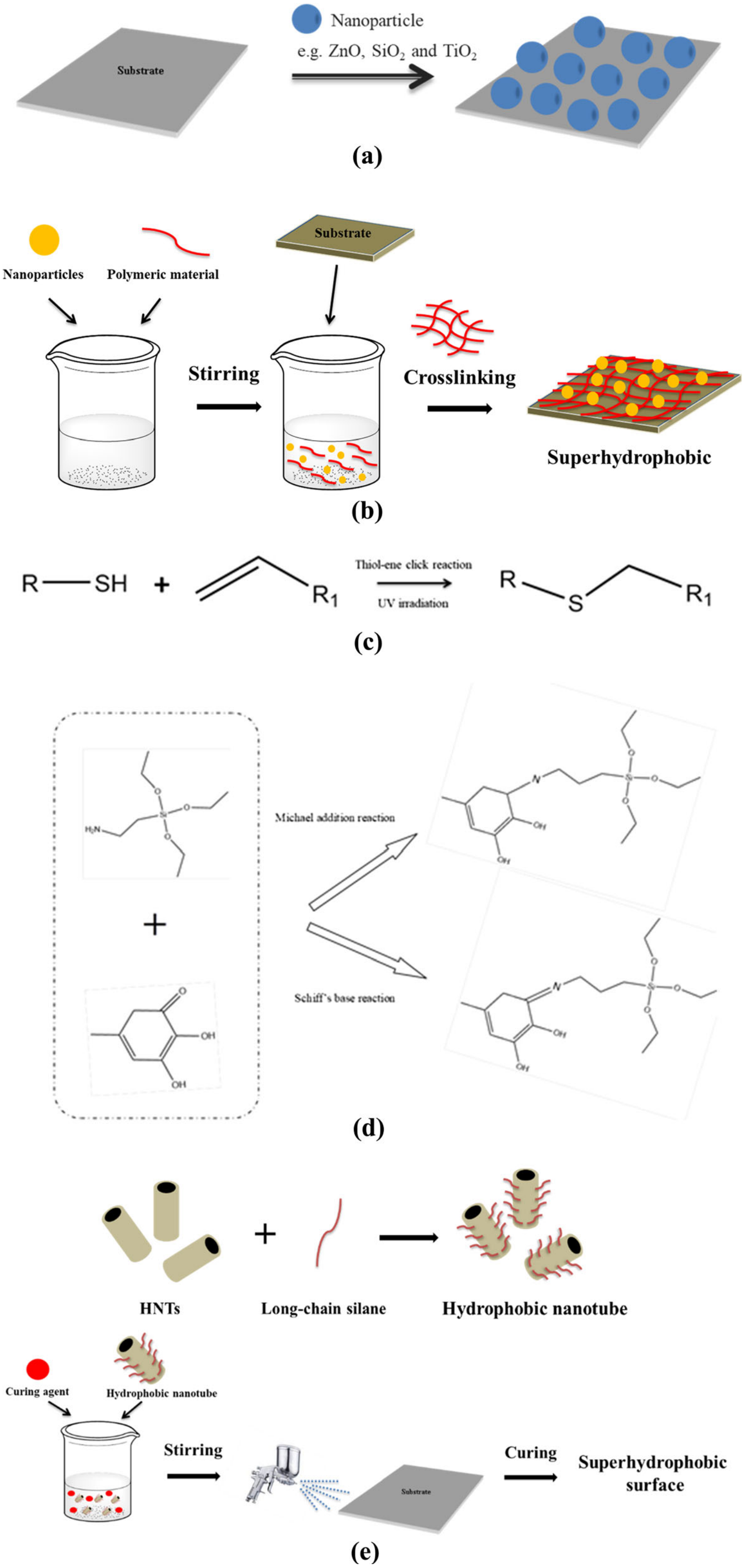

Substrate
Nanoparticle
e.g. ZnO, SiO2 and TiO2
(a)
Nanoparticles
Polymeric material
Substrate
Stirring
Crosslinking
Superhydrophobic
(b)
R—SH + R1
Thiol-ene click reaction
UV irradiation
R S R1
(c)
H2N
O
Si
OH
+
Michael addition reaction
Schiff's base reaction
N
(d)
HNTs
Long-chain silane
Hydrophobic nanotube
Curing agent
Hydrophobic nanotube
Stirring
Substrate
Curing
Superhydrophobic surface
(e)

Fig. 1 lists, when using the modified cotton fabrics as oil adsorbent from oil–water mixture, a superhydrophobic and superoleophilic surfaces such as lotus leaves is desired. If the cotton fabric is used to remove trace water from an oil mix, superhydrophilic and superoleophobic surfaces such as the hot modified alkali treatment which introduce excess hydroxyl groups on the cotton fabric surface as proposed by Rana et al. (2016) is desired. This section categorizes the contemporary works that modified cotton fabrics surfaces for use in oil–water separation.

Dip coating

Nanoparticles ZnO (Boticas et al. 2019; He et al. 2019), SiO_2 (Subramanian et al. 2020), TiO_2 (Rostami et al. 2019) and Ag (Ahmed et al. 2020) were deposit to hydrophilic surfaces for reducing the surface energy of solids, so turning the surface to superhydrophobic (Fig. 2a). Zhang et al. (2013) adopted dip-coating method for fabricating superhydrophobic cotton fabric textiles using modified Zn nanoparticles and polystyrene as the water expeller (Zhang and Seeger 2011). Liu et al. (2013) fabricated superhydrophobic cotton fabrics with SiO_2 nanoparticle deposition and octadecyltrichlorosilane modification. The key of dip coating is the affinity of nanoparticles and the surface of substrate to lead to strong particle attachment. Dip-coating can also be applied for fabricating functional membrane (Wenten et al. 2020) via sol–gel process (Matin et al. 2019) in large scale (Shafiee et al. 2019). This method is commonly simple to implement (Zhang et al. 2013), however, the disadvantages include easy detachment of particles as noted in some applications (Deng et al. 2020), which may yield health concerns and environmental concerns (Lee and Lee 2019). Prolong durability is essential for the success of the nanoparticle deposition method for fabricating superhydrophobic surfaces. To improve the durability, polymer compounds were used as binder between the substrate and nanoparticles (Anjum et al. 2020; Jeyasubramanian et al. 2016).

In situ crosslinking

Jannatun et al. (2020) fabricated SiO_2/PVA/PDMS modified cotton fabric which used boric acid as a crosslinker. Guo et al. (2019) constructed hydrophobic cotton fabric by crosslinking of FAS15 and PDMS. Zhang et al. (2020b) synthesized superhydrophobic surface by immobilizing the ZnO nanoparticles and octyltriethoxysilane on the cotton fabric. In these cases, nanoparticles and polymer monomers can be mixed and crosslinking on the surfaces to form hydrophobic/superhydrophobic layer (Fig. 2b). The CuSA2-modified superhydrophobic cotton fabric that can separate n-heptane/water mixture up to 96% efficiency was fabricated (Pan et al. 2019). Cheng et al. (2019) established superhydrophobic cotton fabric via solvothermal polymerization. Song et al. (2020) synthesized ODTMS-HNTs modified cotton fabric which could maintain high separation efficiency after 40 cycles of oil/water separation. Table 1 lists the typical studies with in situ crosslinking modified cotton fabrics that can be applied for efficient oil–water separation. The modified substrate fabricated via in situ crosslinking possesses excellent durability against abrasion and chemical corrosion. Nevertheless, the fabrication procedure is energy-consuming by the high reaction temperature required.

The UV-triggered reactions were developed with high reactivity under mild and well-controlled reaction conditions (Jiang et al. 2019; Ning et al. 2019). Shen et al. (2019a) adopted ODA to crosslink with polydimethylsiloxane via UV stimulation to synthesize superhydrophobic coating. Shen et al. (2019b) synthesized superhydrophobic cotton fabric via click reaction under UV irradiation. Meng et al. (2020) constructed superhydrophobic surface via thiol-ene click reactions. The POSS (Deng et al. 2020; Yang et al. 2020), fluorine-containing molecules (Jiang et al. 2019; Yang et al. 2019), and long-chain molecules (Chen et al. 2019) were grafted to make superhydrophobic cotton fabrics via thiol-ene reactions under UV irradiation (Fig. 2c). Restated the hydroxyl groups on surface were first converted to thiol groups by thiol-containing silane, and then the so-produced thiol groups are reacted with POSS, fluorine-containing molecular, or long chain molecular to form double bonds under UV irradiation via thiol-ene click reactions. The final groups on surface can efficiently expel water molecules. Deng et al. (2020) fabricated POSS-grafted hydrophobic cotton fabrics via thiol-ene click reaction for oil/water separation. Hou et al. (2018) fabricated robust POSS-based superhydrophobic fabrics to absorb organic solvents from water. Table 2

Table 1 Substrate modified using in situ crosslinking methods for oil/water separation

Material*	Solvent	Drying condition	Contact angle	Shedding angle	Separation cycle	Separation efficiency	Feature	Reference
SiO_2/PVA/PDMS	THF	65 °C; 1 h	156°	1.8	45	90%	1. Superhydrophobic 2. Anti-abrasion 3. Oil–water separation	Jannatun et al. (2020)
PDMS/FAS15/PVP	Dichloromethane	50 °C; 6 h	140°	–	30	96%	1. Hydrophobic 2. Anti-corrosion 3. Anti-abrasion 4. Oil/water separation	Guo et al. (2019)
ZnO/OTES/PGMA	ethanol	120 °C; 6 h	165°	–	–	–	1. Superhydrophobic 2. Anti-abrasion 3. Anti-corrosion 4. Oil/water separation	Zhang et al. (2018)
PDMS/$CuSA_2$	n-hexane	80 °C; 3 h	158°	–	50	96%	1. Superhydrophobic 2. Anti-fouling 3. Anti-corrosion 4. Anti-abrasion 5. UV-resistance 6. Oil/water separation	Pan et al. (2019)
PDVB	ethyl acetate	80 °C; 1 h	158°	–	10	98%	1. Superhydrophobic 2. Anti- abrasion 3. Anti-corrosion 4. Oil–water separation	Cheng et al. (2019)
ODTMS-HNTs/PDMS	Water/HCl	120 °C; 2 h	164°	–	40	99%	1. Superhydrophobic 2. Oil/water separation	Song et al. (2020)

*PVA: polyvinyl alcohol; PDMS: polydimethylsiloxane; FAS15: triethoxy(perfluorooctyl)silane; PVP: polyvinylpyrrolidone; OTES: triethoxy(octyl)silane; PGMA: glycidyl methacrylate; $CuSA_2$: copper stearate; PDVB: polydivinylbenzene; ODTMS: octadecyl trimethoxysilane; HNT: halloysite nanotube

lists the typical UV triggered reactions for superhydrophobilization of surfaces of cotton fabrics.

Self-affine modification

Nature-inspired methods received research interests owing to its sustainability, flexibility, and versatility to applications (Huang et al. 2016). Dopamine, 3,4-dihydroxyphenethylamine, is a hormone that plays essential regulating functions in human body. Inspired by the compositions of adhesive proteins in mussels, dopamine was applied in multifunctional polymer coating and in preparation of heparin immobilizing membrane (Jiang et al. 2010). Polydopamine (pDA)

Table 2 Typical studies using UV-triggered modification on cotton fabrics for separation of oil/water mix

Modification agent*	Solvent#	Reaction	Time	Reaction condition	contact angle	shedding angle	Separating cycle	Separation efficiency	Features	Reference
TiO_2/PFOMA	DMF	Thiol-ene click	1 h	Room temperature; UV (365 nm, 20mW cm^{-2})	157.7°	4°	20	99%	1. Superhydrophobic 2. Self-cleaning 3.Photocatalytic degradation 4. Oil/Water separation	Jiang et al. (2019)
PDMS/ODA	THF	UV crosslinking	60 s	Room temperature; UV (365 nm, 19.8 cm^{-2})	161°	5°	–	–	1. Superhydrophobic 2. Self-cleaning 3. Anti-corrosion 4. Anti-abrasion	Shen et al. (2019a)
PDMS-OH/IPDI/PETA	acetone	Click	90 s	Room temperature; UV (365 nm, 1 kW)	159.5°	7.5°	30	98%	1. Superhydrophobic 2. Anti-abrasion 3. Anti-corrosion 4. Oil/water separation	Shen et al. (2019b)
VPOSS/MPTMS	THF	Thiol-ene click	15 min	Room temperature; UV (365 nm)	142°	-	50	> 96%	1. Hydrophobic 2. UV-resisrance 3. Anti-corrosion 4. Anti-abrasion 5. Oil/water separation 6. High permeation	Deng et al. (2020
TiO_2/MPTES/POSS	Dichloro-methane	Thiol-ene click	1 h	Room temperature; UV (365 nm, 30 mW cm^{-2})	157.6°	–	9	99.0%	1. Superhydrophobic 2. Self-cleaning 3. Anti-corrosion 4. Anti-abrasion	Yang et al. (2020)

Table 2 continued

Modification agent*	Solvent#	Reaction	Time	Reaction condition	contact angle	shedding angle	Separating cycle	Separation efficiency	Features	Reference
ZnO/MTS/ DFMA	DMF	Thiol-ene click	1 h	Room temperature; UV (365 nm, 10 mW cm^{-2})	156.3°	6°	20	98.6%	1. Superhydrophobic 2. Self-cleaning 3. Anti-corrosion 4. UV-resistance 5. Oil–water separation	Yang et al. (2019)
SiO$_2$/ DMAEMA	THF	Thiol-ene click	30 min	Room temperature; UV (360 nm, 40 W)	162°	–	50	99%	1. Superhydrophobic 2. UV-resistance 3. Anti-corrosion 4. Anti-abrasion	Chen et al. (2019)
MPTES/ MPS-SiO$_2$- Fe$_3$O$_4$	Dichloro-methane	Thiol-ene click	2 h	Room temperature; UV (365 nm, 40 W)	156°	4°	-	-	1. Superhydrophobic 2. Anti-corrosion 3. Anti-abrasion 4. Oil/water separation	Fang et al. (2020)
PDMS-OH/ IPDI/PETA	acetone	Click	90 s	Room temperature; UV (365 nm, 1 kW)	159.5°	7.5°	30	98%	1. Superhydrophobic 2. Anti-abrasion 3. Anti-corrosion 4. Oil/water separation	Shen et al. (2019c)

*PDMS: polydimethylsiloxane; ODA: octadecylamine; PDMS: polydimethylsiloxane; IPDI: isophorone diisocyanate; PETA: pentaerythritol triacrylate; PFOMA: perfluorooctyl methacrylate; MTS: 3-mercaptopyltrimethoxysilane; DFMA: dodecafluoroheptyl methacrylate; DMAEMA: dimethylaminoethyl methacrylate; VPOSS: octavinyl-POSS; MPTMS: 3-mercaptopropyltrimethoxysilane; MPTES: methacryloxypropyltrimethoxysilane; MPS: 3-methacryloxypropyltrimethoxysilane; POSS: polyhedral oligomeric silsesquioxanes; THF: tetrahydrofuran; DMF: dimethylformamide can be coated on a variety of inorganic and organic material surfaces, and then the coating can be further modified by numerous functional substances via grafting of macromolecules, deposition of long-chain molecules, and reduction of metal ions (Lee et al. 2007; Ahmed et al. 2020). Dopamine can be easily polymerized into pDA in mild conditions without stimulation (Wang et al. 2017b; Hu et al. 2006; Chen et al. 2019c), so leading to a method with potential for large-scale production (Xi et al. 2009). Xu et al. (2015) utilized silver nanoparticles which is reduced by dopamine to fabricate superhydrophobic melamine foam. Yan et al. (2020) synthesized dopamine-modified superhydrophobic cotton fabric via grafting octadecylamine (ODA). The major disadvantage of applying dopamine as modification agent is its high cost (Chen et al. 2018; Wang et al. 2019a) and excess time for reactions 12 h for TA/APTES, 9 h for TA/Ti (Hu et al. 2020) pDA/GO/PEI for 72 h (Li et al. 2019), and 80 h for pDA/Cu/eumelanin (Ball et al. 2013). In particular, the time to form pDA coating may take days (Zhang et al. 2016). Compared to pDA, tannic acid (TA), an affordable compound, also shows supreme adhesion tendency to wide ranges of substrate surfaces (Wang et al. 2018c). TA is a natural polyphenol which possesses abundant catechol functional groups, which can crosslink with amino-containing silane via Michael addition/Schiff base reactions (Fig. 2d) (Feng et al. 2019). However, the reaction time of tannic acid is still long compared to the UV-triggered reaction. Reducing the reaction time is essential for production efficiency. Other commonly adopted modification agents were also shown in the figure. POSS are organic–inorganic hybrid nanoparticles which have a cage with the general formula $(RSiO_{1.5})_n$ (Sun et al. 2016). Lui el at. (2019) utilized POSS to enhance the thermal stability and water resistance of polymeric film. Qiang el at. (2017) constructed POSS-graft superhydrophobic cotton fabric to improve the durability. The POSS-modified substrates possess UV resistance (Song et al. 2013; Deng et al. 2019), thermal stability (Liu et al. 2007; Huang et al. 2003), anti-corrosion ability (Yang et al. 2020), anti-abrasion (Zeng et al. 2019), low surface energy and good mechanical properties (McMullin et al. 2016).

Spray coating

Spray coating has considered as a promising method owing to its high efficiency (Zhang et al. 2019), simplicity (Lin et al. 2020), feasibility to be performed in large scale (Wang et al. 2019b). Guo et al. (2018a) fabricated superhydrophobic stainless steel mesh with spray coating of hexadecyltrimethoxysilane. For instance, the hydrophilic halloysite clay nanotubes (HNTs) were modified by long-chain silane to form hydrophobic nanotubes (Fig. 2e), and then the modified nanotubes were mix with curing agent and sprayed onto the targeted substrate surface. Superhydrophobic surface could be obtain by curing (Fig. 2e). Using this method, Yuan et al. (2019) synthesized $FSiO_2$/HNTs for spray coating of aluminum plate to form superhydrophobic surface. Foorginezhad et al. (2019) established OV-POSS/PDMS superhydrophobic cotton fabrics that possessed chemical resistance and abrasion resistance. Guo et al. (2018b) constructed hierarchical nano-microstructure via free radical solution polymerization to create superhydrophobic surface. In Table 3, the polymeric materials such as resin (Zhou et al. 2019) or PDMS (Foorginezhad and Zerafat, 2019) were used as binder. Composites with polymeric materials and low surface energy nanoparticles (Gong et al. 2020; Esmeryan et al. 2019), wax (Celik et al. 2020; Torun et al. 2019) were applied to fabricate the superhydrophobic surface. However, the material including nondegradable polymer and detachment of nanoparticle are not friendly to environment (Lee and Lee 2019; Iwata 2015).

Other hydrophobicity modification method

Plasma treatment is efficient (Fu et al. 2018) and reducing waste generation (Kamlangkla et al. 2010). Li et al. (2015) synthesized hydrophobic cotton fabric via plasma-induced graft polymerization. Kamlangkla et al. (2010) utilized SF_6 plasma treatment to fabricate hydrophobic cotton fabric. Kim et al. (2006) deposited hydrocarbon layer on cotton fabric to fabricate hydrophobic surface via plasma treatment. The equipment of plasma treatment is expensive and the plasma treatment lacks scale-up niche. Reducing the cost and making the process continuous instead of batchwise are essential for large-scale implementation of the plasma treatment production (Sarmadi 2013).

Table 3 Typical studies using spray coating method for preparing superhydrophobic surfaces

Substrate	Material*	Solvent	Spraying condition	Drying condition	Highest contact angle	Shedding angle	Separating cycle	Separation efficiency	Features	References
Stainless steel mesh	epoxy/ HDTMS-HNTs	anhydrous ethanol / acetone	under a spraying pressure of 0.3 MPa	120 °C; 2 h	154°	1.5°	25	98%	1. Superhydrophobic 2. Anti-abrasion 3.Oil/water separation	Guo et al. (2018a)
Aluminum plate	F-SiO_2/HNTs	Butyl acetate	under a spraying pressure of 5 bar with distance 20 cm	Room temperature; 1 h	158.1°	1.9°	–	–	1. Superhydrophobic 2. Self-cleaning 3. Anti-fouling 4. Anti-corrosion 5. Anti-abrasion	Yuan et al. (2019b)
Cotton fabric	TiO_2/OV-POSS/PDMS	n-heptane	Spray with distance 20–30 cm	60 °C	168°	< 10°	–	–	1. Superhydrophobic 2. Self-cleaning 3. Anti-adhesion 4. Anti-corrosion 5. Anti-abrasion	Foorginezhad and Zerafat (2019)
Glass	PFA/ ethyl-a-cyanoacrylate	Ethyl acetate/ 1,1,2-trichlorotri-fluoroethane	under pressure of 0.5 MPa with distance 10–20 cm	Room Temperature; 2 h	158°	1.9°	–	–	1. Superhydrophobic 2. High transparency 3. Anti-corrosion 4. Anti-abrasion	Guo et al. (2018b)

*HDTMS: hexadecyltrimethoxysilane; HNT: halloysite nanotube; POSS: polyhedral oligomeric silsesquioxanes; OV-POSS: octavinyl-POSS; PDMS: polydimethylsiloxane; PFA: POSS-based fluorinated acrylate copolymer

Discussions

Design of preparation method

Fabrication of superhydrophobic cotton fabric surfaces is readily achievable with simple method in mild conditions (Sect. 3). Individual method has its own advantage and disadvantage in preparation. Dip coating via sol–gel process to deposit nanoparticles on the substrates is simple and can product in large scale. But the interaction between nanoparticles and substrates is poor so the nanoparticles can be easily detached from the surface. In-situ crosslinking reaction endows the modified substrate with good mechanism resistance and good chemical resistance. However, this process is energy-consuming because of the high reaction temperature. Although the superhydrophobic matrix synthesized via UV-triggered click reaction are well-controlled and efficient, this reaction requires expensive UV irradiation equipment and is energy-consuming and costly, so can only be applied in small scale applications. Self-affine modification is sustainable to environment and is versatile to applications, but the reaction is time consuming compared to other methods. Spray coating is a method which can be applied in large scale with high efficiency; however, the material used in modification such as polymeric substance and fluorine-containing substances may cause adverse impacts on the receiving environment (Table 4).

Design of new preparation method for cotton fabrics can be based on the basic surface characteristics shown in Fig. 1a and the specific incentive to achieve. The pristine cotton fabric surface is superhydrophilic and superoleophobic, at the second quadrant of Fig. 1a. This cotton fabric is unfavorable to extract oil from the oil spilled sites. The modification methods reported in Sect. 3 is to fabricate superhydrophobic and superoleophilic surfaces, located at the forth quadrant of Fig. 1a. The so-generated surface would attract oil but repel water in the feed. If the modification method is to prepare a superhydrophobic and superoleophobic surface (the first quadrant) or a superhydrophilic and superolephilic surface (the third quadrant), separation is not achievable by surface affinity to oil and water phases.

Based on Fig. 1b, if a surface with contact angle exceeding 150° is needed, a rough surface is required. When a smooth surface is to be modified, deposition of nanoparticles to increase the surface roughness is often applied. When using nature cotton fabric surface as the substrate, the inherent surface roughness can be sufficient to serve as the air gap to support water drop with high contact angle.

New superhydrophobic cotton fabric preparation method can be proposed by combining those reported in Sect. 3. For instance, to combine self-affine modification and UV-triggered click reaction into one step can yield a simple cotton fabric preparation process using cost-effective and renewable feedstocks. For instance, the POSS-modified cotton fabrics (POSS-CT) can be fabricated under UV irradiation in one step instead of two steps adopted in literature. By mixing tannic acid, (3-mercaptopropyl)trimethoxysilane (MPTES), POSS and cotton fabrics together tannic acid can react with the thiol groups of MPTES via Michael addition reaction to form the TA hybrid coating that have hierarchical structures and high adhesion to cotton fabrics, and in the same medium POSS can react with the thiol-containing silane via thiol-ene click reaction to form the TA-MPTES-POSS modified cotton fabrics. Since the polymerization of TA by UV irradiation stimulation is a rapid reaction

Table 4 Other hydrophobicity modification method

Material*	Method	Reaction time/ speed	Plasma gas	Working pressure	Power	References
SMA	plasma-induced graft polymerization	5 min	Helium	40 Pa	100 W	Li et al. (2015)
–	SF_6 plasma treatment	5 min	SF_6	0.005–0.5 Torr	50 W	Kamlangkla et al. (2010)
–	CH_4 plasma treatment	5–10 cm/min	Helium/ CH_4	atmospheric pressure	250–400 W	Kim et al. (2006)

*SMA: Stearyl methacrylate

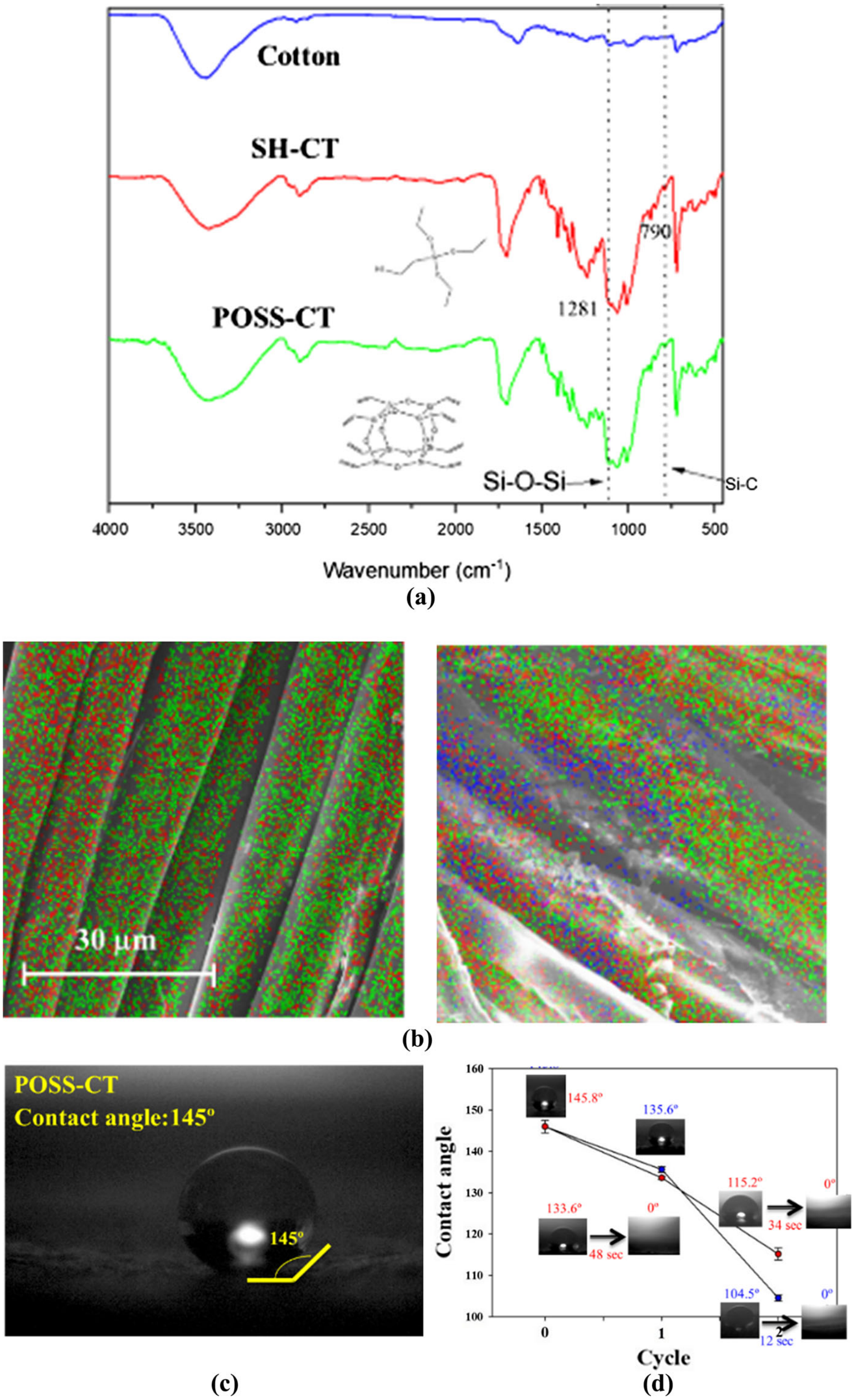


Fig. 3 The characteristics of the pristine and the modified cottons. **a** FTIR spectra for pristine and modified cottons; **b** SEM–EDS (left) pristine cotton; (right) POSS-CT. Red: O, green: C, blue: Si. **c** static contact angle; **d** reusability of POSS-CT cotton after bulk-separation test

(Sadabad et al. 2017), the processing time of the newly prepared cotton fabric surfaces can be reduced. The price for TA is much cheaper than POSS, the replacement of TA to POSS can also sufficiently reduce the fabrication cost and fulfill the merit to apply renewable resources for applications. Shen et al. (2021) also synthesized TA-POSS composite membrane by crosslinking the tannic acid with POSS via Michael addition reactions. Additionally, the superior environmental compactibility of TA to that of POSS is another concern if the end disposal of the used cotton fabrics will be put into consideration.

Proper testing protocol is needed

The oil–water separation efficiencies of the modified cotton fabrics adopted in literature were characterized using various protocols. We synthesized the hydrophobic cotton fabrics by a preparation method as follows: The 3-mercaptopropyltriethoxysilane (MPTS) and EtOH were mixed at weight ratio of 1:20 with pristine cotton fabrics being dipped at room temperature for 2 h. The dipped cotton fabrics was then washed with EtOH and was dried at 80 °C for 2 h to obtain samples SH-CT. The SH-CT samples were mixed in 1.6% w/w POSS solution with 0.16% w/w DMPA and 98.2%% w/w CH_2Cl_2 for 30 min. The dipped samples were then exposed to 365 nm UV light (@ 10 mW/cm^2) to trigger thiol-ene click reaction for 60 min. Then the cotton fabrics were collected and washed with ethanol and further drying at 80 °C for 60 min. The obtained samples were the POSS-CT samples. The FTIR spectra of the cotton fabrics, SH-CT, and POSS-CT were shown in Fig. 3a, showing the attachment of the desired functional groups on the cotton fabric surfaces. The SEM–EDS images for the samples were revealed in Figs. 3b, clearly confirming the presence of C and O on cotton fabric surface, and that of Si on the POSS-CT surface. The produced POSS-CT surface has contact angles with water drops to be 145°., suggesting the successful fabrication of the hydrophobic cotton fabric surface (Fig. 3c). The yielded POSS-CT were used to packed into layer of thickness of 5 mm.

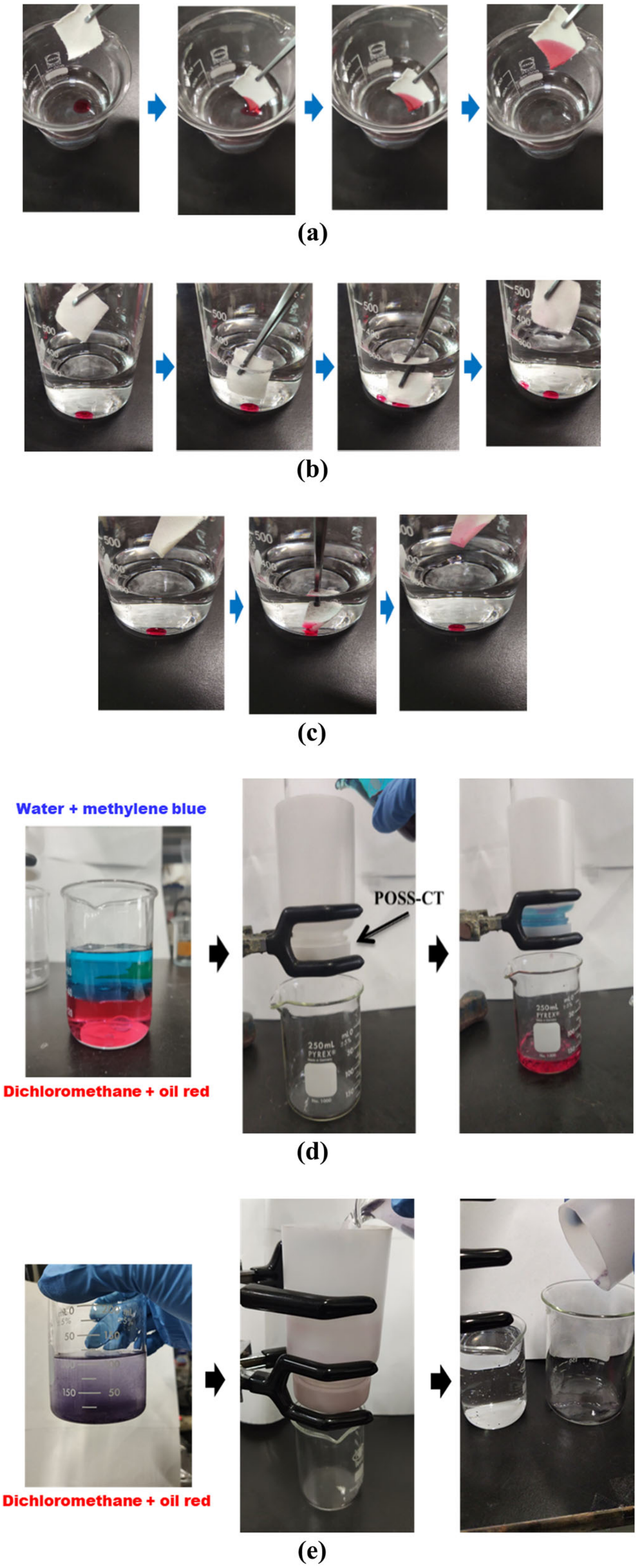
The testing methods for water–oil separation using modified cotton fabrics include the absorption test by applying the cotton fabrics to absorb oil from a clot of oil submerged in a water pool (Hou et al. 2018), and the pour test by pulling a bottle of segregated oil–water layers (a water layer with one or two immersible oil layer) to pass through the cotton fabric matrix for separation (Yang et al. 2020). The hypothesis is that the hydrophobic cotton fabrics would tend to absorb the oil from water (the absorption test) and to reject water with passage of the oil phase (the pour test). The POSS-CT was immersed to contact the DCM (red) at the bottom of the water pool and was noted to efficiently adsorb the DCM by the fabrics (Fig. 4a). This observation suggests that the POSS-CT has hydrophobic surfaces so can be used for oil–water separation. When the pristine cotton fabric was made contacting the DCM (red) under the water layer, the cotton fabric absorbed no DCM, correlating with the superhydrophilic nature of the fabric surface (Fig. 4b). However, when the hydrophilic cotton fabrics were made contacting the hexane (red) floating on the water pool, the cotton fabrics absorbed the hexane (Fig. 4c), contracting to the assumption that the hydrophilic cotton fabrics would not absorb oil compounds. This contradictory result was yielded by the bulk flow induced by capillarity of the macro pores of cotton fabrics with both oil and water phases inflow with minimal roles of surface hydrophobicity. As per the results in Fig. 4b, when the pristine cotton fabrics were immersed in water pool to absorb the DCM underneath the water layer, the macro pores would be filled with water first, so the bulk flow would be minimized, leading to negligible absorption of DCM into the pristine cotton fabrics.

Fig. 4 Tests for oil–water separation for the tested samples. ▶ **a** Absorption test: POSS-CT for DCM (red) under water pool; **b** absorption test: pristine cotton for DCM (red) under water pool; **c** absorption test: pristine cotton for hexane (red) floating on water pool; **d** pour test: POSS-CT for DCM (red, bottom layer) and water (blue, top layer) separation; **e** pour test: POSS-CT for DCM (red) with SDS surfactant in the form of emulsion (looks purple) and water separation

The pour test using DCM (red, bottom layer) and water (blue, top layer) poured on the POSS-CT fabrics. The DCM passed through the fabrics and the water was kept on the POSS-CT, revealing separation (Fig. 4d). The separation efficiency of water from a oil/water mixture was characterized using the following equation:

$$\text{Separation efficiency} = M_2/M_1 \qquad (2)$$

where M_1 and M_2 are the quantities of initial sample


(a)
(b)
(c)
Water + methylene blue
Dichloromethane + oil red
POSS-CT
(d)
Dichloromethane + oil red
(e)

and of collected sample after separation, respectively (Jiang et al. 2019). For instance, the separation efficiently for Fig. 4e was decreased about 30% after two cycles of separation. The durability and cyclability of the cotton fabrics were commonly described by observing the decline in separation efficiency over testing cycles. The mechanical properties of cotton fabrics such as flexural strength, flexural modulus, impact strength and fracture toughness were seldom described in related modification literature for cotton fabrics.

Conversely, when the DCM (red) was mixed with water and 20 mg/L surfactant sodium dodecyl sulfate (SDS) to generate an emulsion, the pour test with the generated emulsion onto the POSS-CT led to no separation, revealing complete passage of emulsion through the fabrics (Fig. 4e). Restated, although also with DCM and water (while the emulsion was with trace quantity of SDS) onto the POSS-CT, the pour tests showed distinct results from that in Fig. 4d owing to the different distribution of oil in the water pool. The pour tests with segregated water and oil layers, since the DCM is heavier than water and flows faster than water to contact the cotton fabrics, the macro pores were filled by DCM which rejected subsequent passage of water through the fabrics. When the oil was dispersed as oil-in-water emulsion, the collision of fine oil droplets with hydrophobic walls of macro pores dominate, leading to full rejection of DCM from the oil–water mix as noted in Fig. 4e. Illustrations in Fig. 4d and e showed that how the oil and water dispersed in the mix can lead to contradictory outcomes for separation.

The separation tests in Fig. 4 reveal the need to set up "standard" testing protocol for the modified cotton fabrics (and other matrix) for oil–water separation. Additionally, the "scenario" of the oil–water separation should also come into play. For instance, for recovery of spilled crude from a tanker on open sea, the as much as possible absorption of oil into the cotton fabrics is the target with the incorporated water and other impurities being non-essential factors and the operation should be considered batchwise or consider the used fabrics non-recyclable. To separate oil droplets in an oil-in-water wastewater from crude refinery, the efficient collisions of oil drops onto the hydrophobic pore surfaces for collection with water to pass though as fast as possible is the design target; however, the accumulation of oil inside macro pores with hydrophobic walls would lead to reduced filtration rate, while the continuous or batchwise deoiling steps from the cotton fabrics should be operated together with the oil–water separation processes. When handing water-in-oil emulsion, Lei et al. (2017) showed that their modified cotton fabrics can allow passage of oil and repel water droplets to accumulate on the top surface of the fabrics. It is clear that the conjugated fluid flow through macro pores and the involvement of surface interactions with the multiphase fluid controls the separation efficiency. Further studies are needed.

Economy and sustainability analyses is needed

Comprehensive technoeconomic and sustainability analyses should be performed to confirm the use of modified cotton fabrics for oil–water separation. The use of cotton fabrics for oil–water separation should be justified on their economy and sustainability based on the fact that cotton fabrics are an economy plant used for textile since pre-historical era, and the modification chemicals used are not all environmentally friendly. If the cotton fabrics are to be used for contamination remediation, the quality of the recovered oil and the efficiency of the separation are not critical. However, if the separation is to be applied in a fine chemical production chain, the purity of the applied cotton fabrics and very high separation efficiency should be the determining factor for the process economy. When handling high-value products, whether the used cotton fabrics can be reused is of no concern. If the oil spill is to be confined, other waste besides cotton fabrics with excess hydroxyl groups may be also candidates for the feasible matrix for oil–water separation. The release of the modified chemicals into the environment after the modified cotton fabrics is disposed of after use.

Conclusions

This article presented a mini-review on the perspectives of applying modified cotton fabrics for oil–water separation. The principles of hydrophobicity of a surface is first stated, with the ways of producing superhydrophobic surfaces being described. The modification methods reported in literature are then summarized. The perspectives of using cotton fabrics for oil–water separation are highlighted. The needs to design new fabrication method, to develop standard testing protocols, and to perform technoeconomic and sustainability analyses for oil–water separation with the use of modified cotton fabrics are addressed.

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Declarations

Conflict of interest No conflicts of interests.

Availability of data and material Can be obtained from corresponding author by reasonable requests.

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