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During the past two decades, developing superwetting materials has emerged as an advanced technology for the efficient separation of oil/water mixtures. Among various special wettability materials, textiles and fabrics have been proposed as flexible porous membranes for the treatment of oily wastewater and crude oil spills. Superwetting textiles modified with polydopamine (PDA) coatings are regarded as a sustainable class of smart multifunctional materials for the treatment of stratified and emulsified oil/water mixtures. PDA coatings offer advantages such as low cost, versatility, biodegradability, and a simple fabrication that avoids the use of harsh chemicals/conditions. The PDA coating plays a critical role in the fabrication of durable superwetting textiles by enhancing the adhesion and surface roughness and offering diverse post-functionalization with low/high surface energy materials.

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Sustainable and Durable Superwetting Textiles Coated with Polydopamine for Oil/Water Separation

Abdul Kalam Azad[1], Nedal Y. Abu-Thabit[1,*], Mahmoud H. Abu Elella[2]

[1]Chemical Engineering Department, Jubail Industrial College, Jubail Industrial City 31961, Saudi Arabia
[2]Chemistry Department, Faculty of Science, Cairo University, Giza 12613, Egypt

*Corresponding author: abuthabit_nidal@yahoo.com
Tel: +966 13 340 5400

Current Postal address:
Dr. Nedal Abu-Thabit
P.O. BOX: 10099
Jubail Industrial College
Chemical Engineering Department, Office Number (AD-301)
Jubail Industrial City, Jubail
Saudi Arabia 31961

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Abstract:

The industrial revolution and the frequent crude oil spills have caused tremendous contamination of the natural water resources with oily waste. Superwetting textiles have been proposed as an advanced functional material for selective oil separation from oily wastewater. Superwetting textiles have been fabricated by engineering smart coatings that allow for the modified textile's optimal surface roughness and chemical composition. During the past decade, polydopamine (PDA) has been proposed as a sustainable, biodegradable, and biomimetic coating for the fabrication of superwetting textiles with superhydrophobic and/or underwater superoleophobic properties. PDA coatings offer advantages including a simple coating procedure in a wide pH range, use of various organic/inorganic oxidants, high adhesion strength to various substrates, increased surface roughness of the coated substrates, and diverse post-functionalization *via* different ionic/ covalent interactions. Thus, bioinspired sustainable and durable superwetting textiles have been designed and fabricated by controlling the surface roughness and the surface energy of the PDA-coated textiles. This review highlights the recent advances in fabricating superwetting PDA-coated textiles for the separation of various oil/water mixtures. Two classes of superwetting textiles are discussed including superhydrophobic (SHB) and superhydrophilic/underwater superoleophobic (SHL/UWSOB) PDA-coated textiles. In each case, the discussion focuses on the fabrication methods, coating procedure, separation performance, recyclability, and enhanced mechanical and chemical durability due to the synergistic effect of the deposited and/or functionalized PDA coating. The discussion includes the separation of emulsified mixtures, current challenges, proposed solutions, and future perspectives of PDA-coated textiles for practical and large-scale oil/water separations.

Keywords:

Polydopamine, emulsion, superoleophilic, superhydrophobic, superoleophobic, textile, fabric, separation.

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1. Introduction:

The industrial revolution has led to increased consumption of freshwater and produced significant oily wastewater by discharging oily waste into various water streams (1–6). In a similar scenario, frequent crude oil spills have discharged vast quantities of toxic liquid hydrocarbons into the sea or coastal areas, resulting in significant environmental pollution and posing a direct threat to aquatic systems and an indirect risk to public health and communities (7–10). Oil can be separated from oily wastewater using traditional methods including gravity filtration, chemical precipitation, flocculation, coagulation, centrifugation, or thermal incineration (11–15). However, these methods suffer from low separation efficiency, large footprint requirements, high operation cost, and/or generation of secondary pollution (16). Therefore, there is a crucial need to develop new and cost-effective materials for efficient separation of oil/water mixtures.

Special wettability materials have superhydrophobic or superhydrophilic superwetting surfaces for liquids on liquid/air/solid or water/oil/solid interfaces with a water contact angle (WCA) greater than 150º or less than 10º, respectively (17,18). During the past two decades, special wettability materials with superhydrophobic (SHB) or superhydrophilic and underwater superoleophobic (SHL/UWSOB) surfaces have been proposed as an advanced functional material for the separation of emulsified and stratified oil/water mixtures (19–21). Typically, the construction of superwetting surfaces can be achieved by tuning the surface roughness and surface energy (22,23). The common substrates that have been modified as superwetting materials for oil/water separation include fabrics and textiles (24–26), sponges (27–30), metal meshes (31–34), and polymeric membranes (35–38). Compared to other substrates, woven and nonwoven textiles have been

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proposed for efficient separation of oil/water mixtures in harsh environments due to their low cost, high porosity, high permeability, high flexibility, good mechanical durability, and corrosion resistance (39,40). Unlike superwetting sponges which have been mainly employed as absorption materials for oil/water separation (27), superwetting textiles have been utilized as filter membranes to separate oil/water mixtures by filtration strategy which is beneficial for continuous and large-scale separations (41–43).

The surface wettability of various textiles has been modified by using different strategies such as grafting, dip-coating, spray-coating, chemical vapor deposition, solution immersion, and *in situ* polymerization. The fabrication of superhydrophobic textiles has been achieved by using a wide range of materials/chemicals/techniques, which aim to: (1) increase the surface roughness (44), and (2) reduce the surface free energy of the modified fabrics. The presence of strong adhesion between the deposited materials (e.g., nanoparticles) and the surface of the modified textiles plays an important role in making durable superwetting textiles with superior mechanical abrasion resistance, good recyclability, and corrosion resistance (45,46). Good adhesion can be promoted *via* strong inter/intramolecular ionic interactions, covalent bonds, or ionic/covalent crosslinks (47,48).

Polydopamine coating, invented by Lee and Messersmith in 2007, has been employed extensively for fabricating various durable superwetting substrates including fabrics and textiles (49–51). In general, PDA coating is prepared by the chemical oxidative polymerization in the absence (pH = 8.5) or presence (neutral /acidic conditions) of organic/inorganic oxidants (52–54). The main requirement for chemical oxidative polymerization is the presence of an electron donor atom (e.g., O, N, S) with a high

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oxidation tendency to induce the initiation of the chain growth *via* the formation of cation/ cation radical species (55). Chemical oxidative polymerization has been employed for the fabrication of electrically conducting polymers such as polyaniline, polypyrrole and polythiophene (56–60). The oxidative polymerization of polydopamine is depicted in **Fig. 1a**. The versatility of polydopamine coating is due to its ease of application as well as its ability to adhere to various surfaces (metals, plastics, ceramics, etc..) by simple solution immersion and *in situ* deposition process (61–63). Depending on the modification process, materials used and the chemical composition of the textile surface, the adhesion of polydopamine to the fabric surface can be achieved *via* different surface interaction mechanisms including metal coordination, Michael-type additions, and/or Schiff-base formations, hydrogen bonding, and π–π stacking (48,52,64–67), **Fig. 1b**. This review highlights the most recent literature studies related to the fabrication of superwetting textiles modified with polydopamine coating for oil/water separation applications. The first part of the review is related to SHB textiles, the second part is devoted to the SHL/UWSOB textiles, and the final section discusses the separation of emulsions. In addition, the recent related studies have been presented and summarized in **Tables 1 and 2**.

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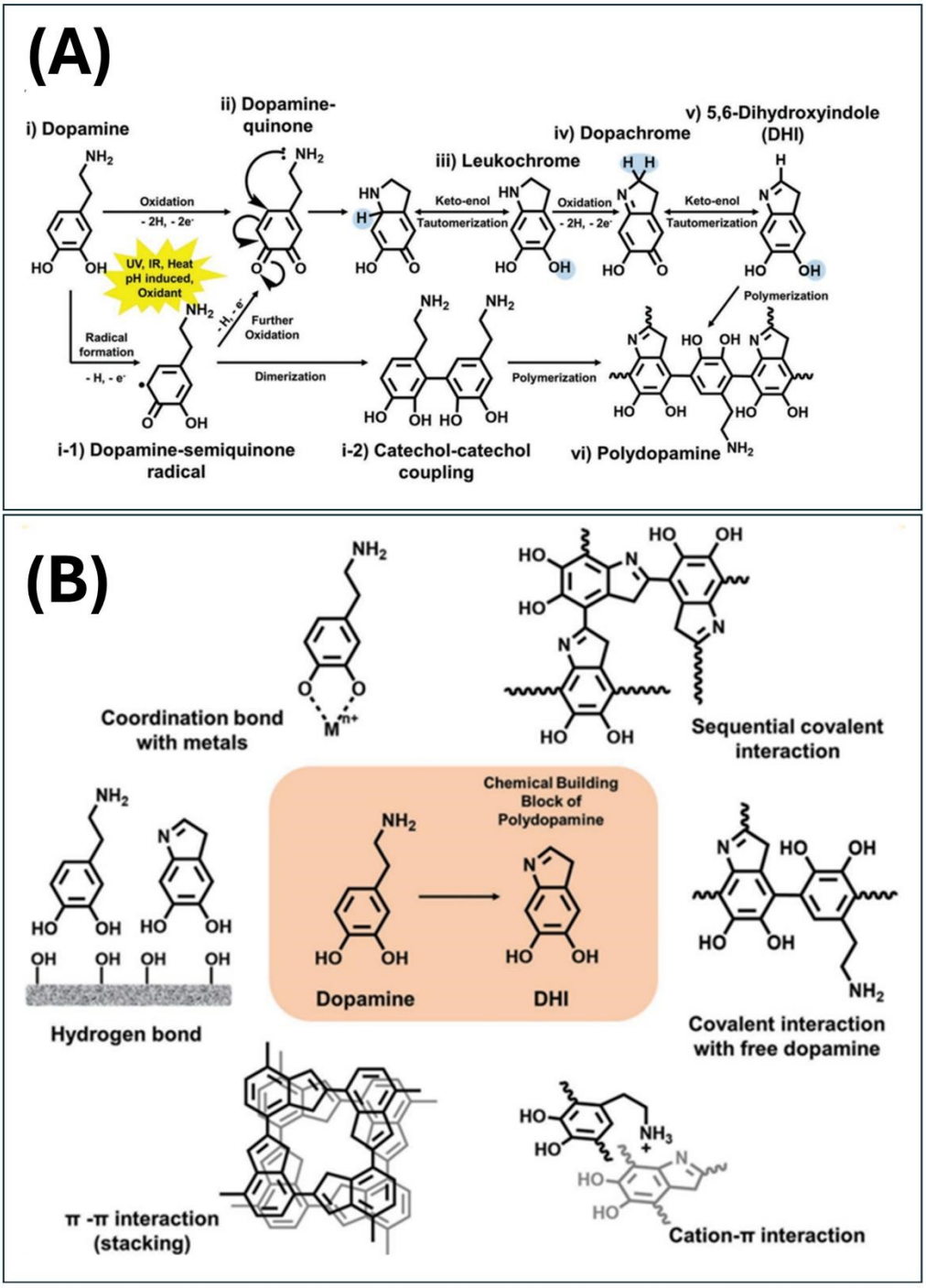

Figure 1 Polymerization and interaction mechanisms of dopamine. A) Polydopamine formation: the copolymer consisting of 5,6-dihydroxyindole (DHI) and dopamine. B) Diverse interactions of dopamines and DHI, (64).

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2. Review for the models of wettability:

The effectiveness of separating oil/water mixtures highly depends on the membrane surface's wettability, which was determined by how easily the liquid could penetrate the solid surface. When the liquid droplet contacts a solid surface in the atmosphere, it creates a three-phase boundary where the solid, liquid, and gas phases intersect (68). Notably, the characteristics of the wetted surface are frequently described via the contact angle (CA) and the sliding angle (SA). The CA indicates the equilibrium of surface tension forces among the solid, vapor, and liquid interfaces, and the solid substrate behaves as a static surface. Conversely, the SA is utilized to gauge the dynamic behavior of a solid surface concerning wetting. Significantly, in the wetting process, the system achieves its lowest energy state when the droplet reaches the equilibrium (69). Based on the value of the CA (θ), the wetting behavior is categorized into four primary types, **Fig. 2**. The surface is classified as hydrophilic if the CA falls in the range of 10° - 90°, **Fig. 2a**. However, the superhydrophilic surfaces have CA lower than 10 degrees. They exhibit a strong affinity for water, resulting in the spontaneous spreading of water droplets and full surface wetting. As shown in **Fig. 2b**, hydrophobic surfaces exhibit contact angles in the 90° - 150° range, allowing the liquid droplets to not spread uniformly within the surface. As a result, they reject water and exhibit a lower affinity for liquids. In comparison, superhydrophobic surfaces represent an extreme form of hydrophobicity. These surfaces have contact angles greater than 150°, causing water droplets to form almost perfect spheres and roll off the surface easily. Superhydrophobic surfaces are characterized by their outstanding water-repellent properties and self-cleaning capabilities (70,71).

The wetting phenomena have been described by Young, Wenzel, and Cassie-Baxter models. Thomas Young introduced the concept of the CA for solid/liquid interface. He discussed the cohesion of superficial particles at both solid and liquid surfaces. Additionally, once the tension at

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the interfaces reaches equilibrium, as illustrated in **Fig. 2c**, the correlation can be represented by Young's equation (26), as indicated in equation (1):

$$\cos\theta_Y = (\gamma_{SG} - \gamma_{SL})/\gamma_{LG} \quad (1)$$

In this context, θ represents the equilibrium contact angle, while; γ_{SG}, γ_{SL}, and γ_{LG} represent the interfacial energies of the solid-gas, solid-liquid, and liquid-gas interfaces, respectively. Young's equation assumes an ideal scenario of a perfectly smooth, rigid, and homogeneous solid surface, which doesn't accurately reflect real-world surfaces that are often rough and heterogeneous. When water encounters a rough solid surface, it can exist in two states: (i) where water fully adheres to the solid surface, or (ii) where water adheres to the combined surface of solid and air, forming droplets known as fakir droplets (72). Wenzel's model proposed that due to varying degrees of rough microstructure on the solid surface, the liquid would completely occupy the surface microstructure upon contact with the solid. He incorporated a roughness coefficient (R) into Young's equation, thereby creating the Wenzel model shown in equation (2):

$$\cos\theta_W = R\cos\theta_Y \quad (2)$$

Where θw is the apparent rough surface's contact angle, θ_Y is the plane's inherent Young contact angle, and R is the roughness factor, which represents the proportion of the actual surface area compared to the surface area of a rough solid substrate. According to the above equation, the surface roughness has an impact on wettability, which can be enhanced or decreased by R. If a liquid droplet prefers to wet surface i.e. $\cos\theta_y > 0$, the roughness factor (r) will exceed one. In this context, the surface roughness amplifies the wetting behavior, resulting in an apparent contact angle (θ_w) that is larger than the equilibrium contact angle (θ_y). This situation is referred to as the "Wenzel wetting regime." Conversely, if the liquid forms droplets on the surface i.e. $\cos\theta_y < 0$, R will be <1. Surface roughness inhibits wetting behavior, leading to a lower apparent CA (θw) than the equilibrium CA (θ_y). This phenomenon is known as the "Wenzel non-wetting regime" (**Fig. 2d**). The Wenzel model's principle focuses on understanding surface roughness's influence on wetting

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behavior. Thereby, it enables the prediction of the apparent CA on rough surfaces by considering the properties of the smooth surface and the roughness factor (70,73).

Increasing surface roughness and reducing feature size can cause a liquid droplet to be suspended above the rough surface structure, trapping few amounts of air beneath the liquid in pits or grooves. This phenomenon was explained by the Cassie-Baxter model according to the following Equation (3) and (**Fig. 2e**):

$$\cos\theta_{CB} = f_{SL}\ \cos\theta_1\ + f_{LG}\ \cos\theta_2 \quad (3)$$

where, θ_{CB}, $\theta_1$and θ_2 are the apparent CA of the droplet on the rough surface, the solid surface, and the air pockets or gaps, respectively. f_{SL} and f_{LG} are the area fraction of the solid-liquid phase and liquid-gas phase, respectively. The model considers the contributions of both the solid surface and the air pockets to the overall contact angle, leading to a higher apparent contact angle compared to the Wenzel model, which assumes the total wetting phenomenon of a rough surface (70,74).

Contact angle hysteresis (CAH) refers to the difference between the advancing contact angle (θ_a) and the receding contact angle (θ_r) of the droplet. The θ_a is the angle generated as the droplet spreads on the surface, whereas the θ_r is created when the droplet retracts from the surface. Contact angle hysteresis exists because contact lines usually do not slide freely. Real surfaces are amorphous, deformable, rough, and show different crystalline orientations which lead to the observed contact angle hysteresis.

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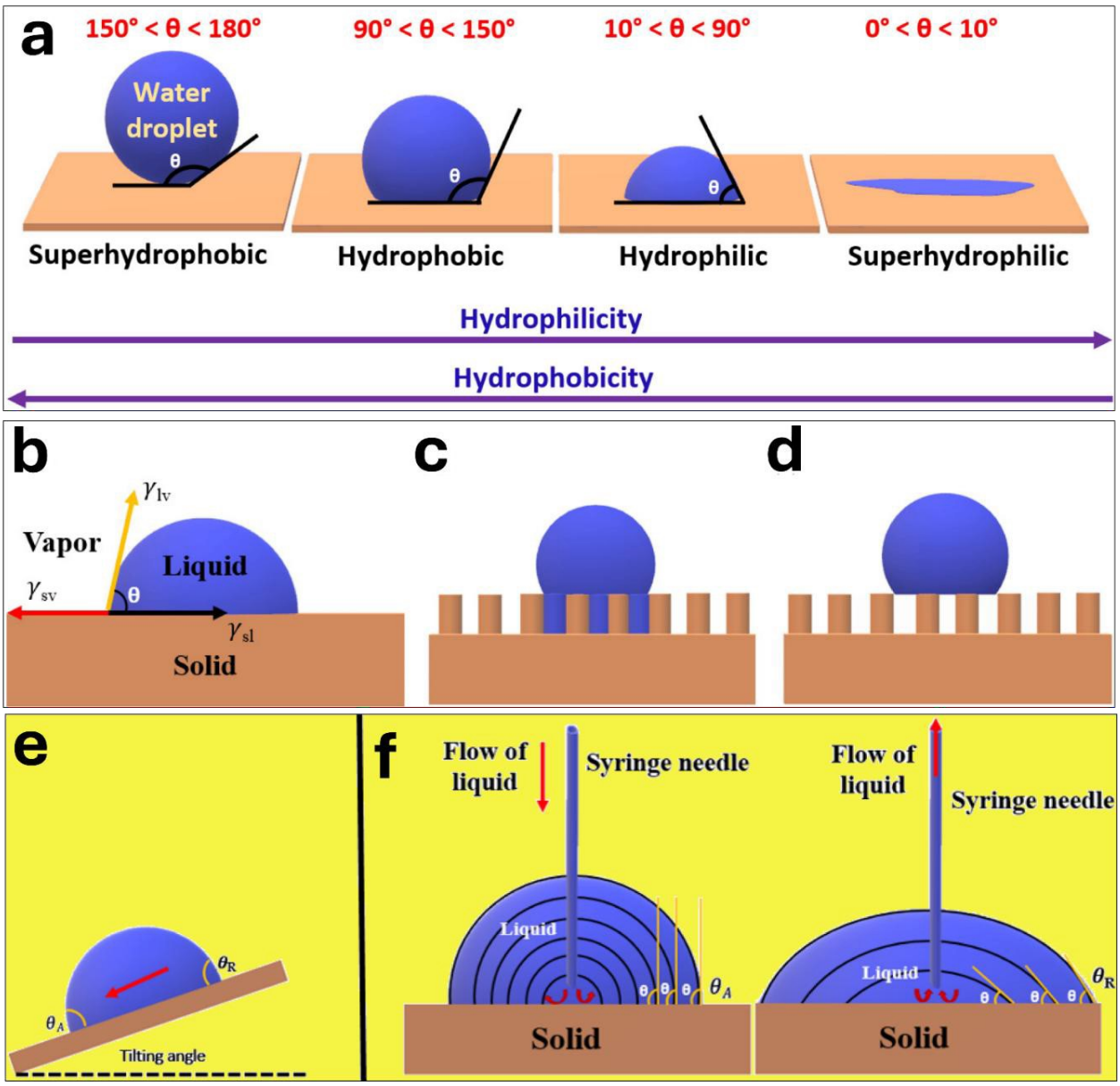

Figure 2 Schematic illustration of various wettability states (a), an illustration of wetting models based on Young's model (b), Wenzel's model (c), and Cassie-Baxter's model (d); Measuring the advancing and receding angles by (e) tilting the substrate; and (f) changing the droplet volume by gradually introducing or extracting liquid from a sessile droplet, Ref. (75).

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3. PDA-coated superhydrophobic (SHB) textiles for oil/water separation

3.1. Cotton textiles

Wang et al. fabricated a PDA-coated superhydrophobic fabric, composed of a polyester (65%) and cotton (35%) blend, by using a two-step modification process (76), **Fig.3 A**. The surface roughness was achieved by using folic acid (FA) as a structure directing agent (SDA) during the deposition of PDA NPs, while octadecylamine (ODA) was employed as a low surface energy material which endowed the modified textiles with a superhydrophobic character as revealed from the high WCA (162º), and the low WSA (7º). The construction of hierarchal rough morphology was achieved by the controlled addition of DA and FA, which was followed by the addition of the tris-HCl buffer solution after a stirring period of 1 hour. As illustrated in **Fig.3 A**, self-driven, energy-saving, and highly efficient crude oil spill cleanup was achieved by using a mini boat made from the modified superhydrophobic textiles and floated on the top of crude oil polluted water. This mini boat absorbed all the crude oil and accumulated inside the cavity for 10 min with a cleanup efficiency of 97.1%.

Liu et al. fabricated self-healing superhydrophobic cotton fabrics, coated with PDA microcapsules, by using a one-step modification process (77). The pristine cotton fabric was dipped into an emulsion containing a low surface energy material (ODA or octadecanethiol ODT), tris-HCl buffer, and DA (0.5 mg/mL) for a total period of 12 hours. After plasma treatment, the modified superhydrophobic fabric lost its hydrophobicity and became a hydrophilic one, **Fig.3 B-C**. However, upon mechanical action on the fabric (compression/stretching), the self-healing property was demonstrated with a full recovery

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of its superhydrophobic nature. This self-healing property was attributed to the release of the low surface energy materials (ODA or ODT) embedded in the coated PDA microcapsules, **Fig.3B-C**.

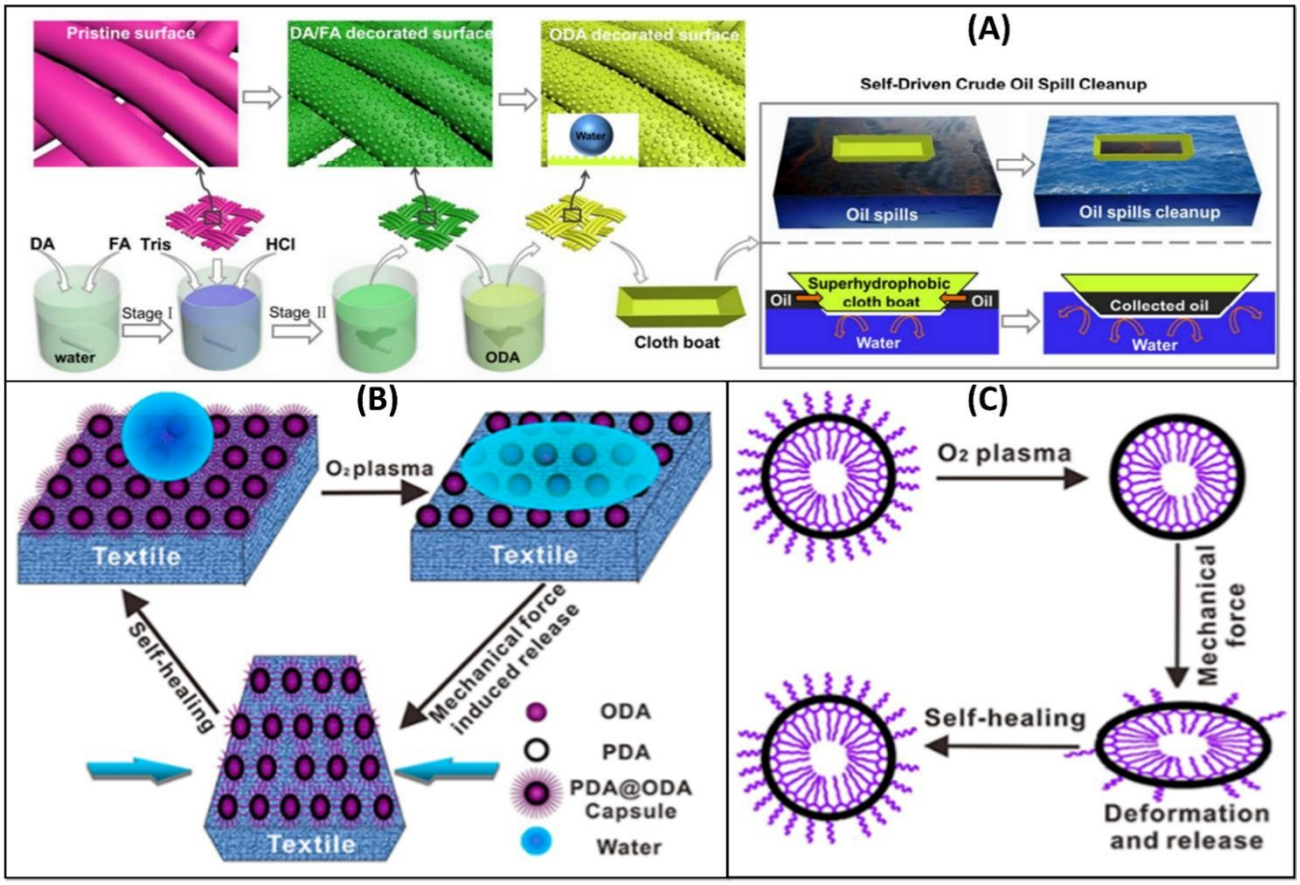

Figure 3 Schematic illustration of the fabrication of superhydrophobic fabric with micro-nanostructure via a novel mussel-inspired approach and superhydrophobic cloth boat made by the fabric for self-driven oil spill cleanup (A), Reproduced with permission from reference (76).Regeneration of Hydrophobicity through Mechanical Stimulation (B) and Mechanism in the Self-Healing Process during Compression (C), Reproduced with permission from Ref. (77).

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Guo *et al.* reported a simple one-pot method to fabricate a robust superhydrophobic cotton fabric by solution immersion method (78). The surface roughness was achieved by the combined effect of PDA and silicon dioxide (SiO_2) through the copolymerization process at room temperature and the lower surface energy was attained by the presence of methyl groups on the PDA/SiO_2-coated superhydrophobic fabric. The modified fabric exhibited a separation efficiency of 99.9% and a permeation flux of ~ 4000 L/m^2.h with a stable performance even after 20 separation cycles. The modified fabric showed excellent mechanical durability for various tests (abrasion, peeling, and ultrasonic treatment), **Fig.4 A-D.** The functionalized fabrics showed high durability under harsh chemical environments, UV radiation, and boiling water **Fig.4 E-H**.

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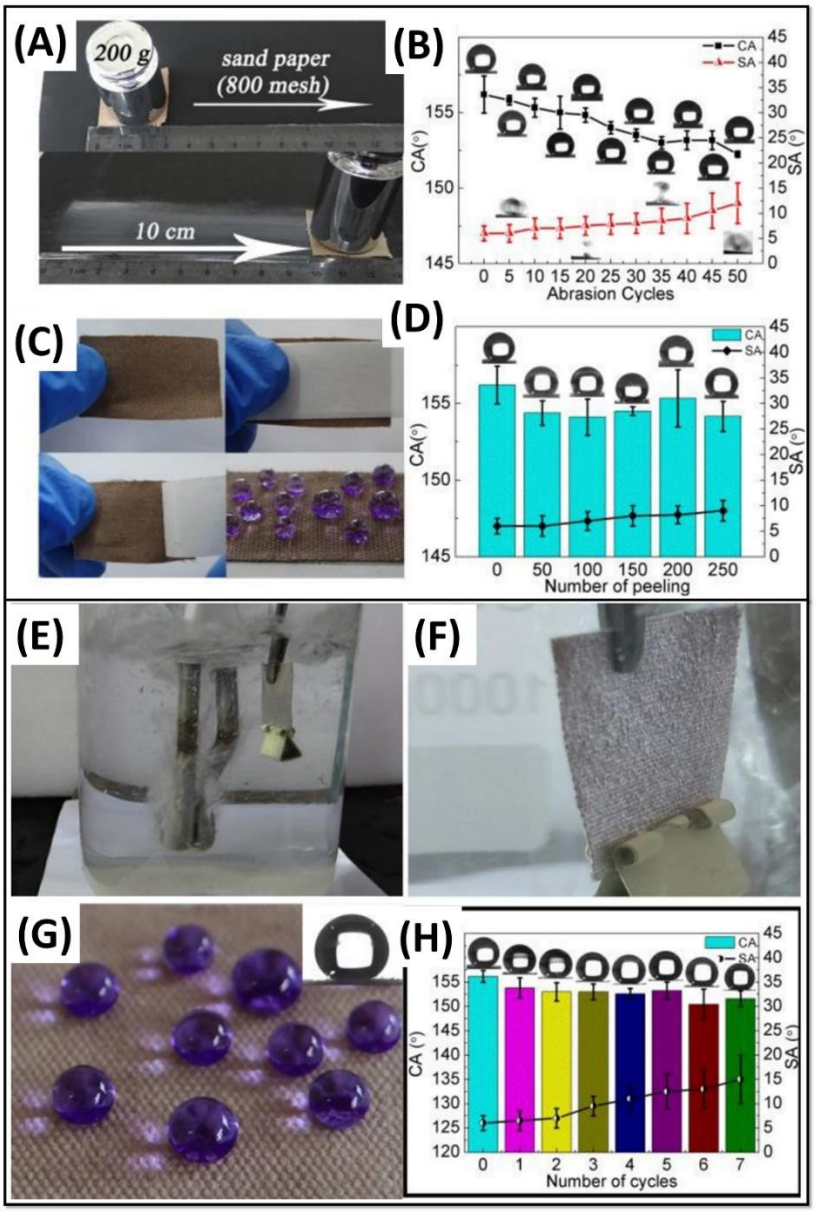

Figure 4 (A) Photographs showing the abrasion process. (B) Contact angle and sliding angle of superhydrophobic fabric after each abrasion cycle. The optical images of the static water droplets (5μL) after each abrasion cycle are shown in the insets. (C) Photographs showing the tear test process and water droplets (dyed with crystal violet) on the fabric surface after the test. (D) Contact angle and sliding angle of superhydrophobic fabric after each tear test cycle. The optical images of the static water droplets (5μL) after each cycle are shown in the insets. (E) Photograph of the superhydrophobic PDA@SiO2 coated cotton fabric immersed in boiling water. (F) A stable silver mirror-like phenomenon of the fabric immersed in boiling water. (G) Photograph of water (dyed with crystal violet) standing on the fabric treated in boiling water for 5 min. Optical image of the static water droplets (5μL) after 5 min boiling-water treatment is shown in the inset. (H) Water contact angle and sliding angle of the tested fabric after each 5 min boiling-water treatment. Optical images of the static water droplets (5μL) after each 5 min boiling-water treatment are shown in the inset, reproduced with permission from Ref. (78).

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Luo et al., reported the fabrication of a flame retardant superhydrophobic polyethylene imine (PEI) grafted cotton fabric coated with the biologically derived materials of tannic acid (TA), borax, and PDA by a dip-coating method (79), **Fig.5 A.** Borax was utilized to attain the flame-retardant property. However, when it contacts with water while washing will dissolve the borax. Hence, TA was used to make a complexation with borax with the additional benefit of oxygen-free radical scavenging and carbonization ability during combustion. The functionalized cotton fabric displayed a WCA of 153.3° ± 1.2°, WSA of 9° ± 0.8°, and oil/water separation efficiency of >98% after 30 cycles. The residual content during thermal studies at 800°C under nitrogen and oxygen atmosphere is about 48.9% and 27.9%, respectively, clearly showing the improved thermal stability of the modified fabric as depicted in **Fig.6 A-D.**

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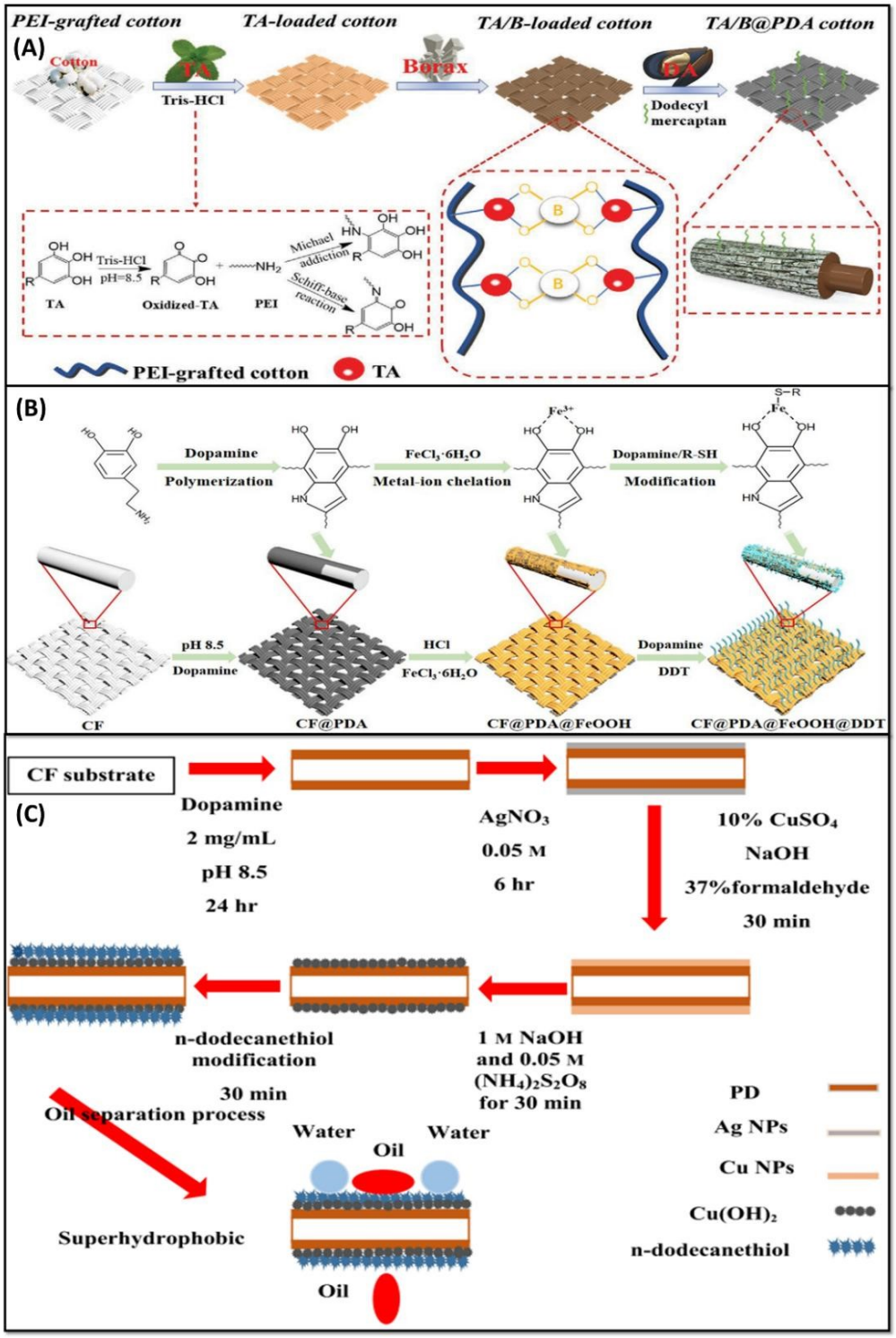

Figure 5 Schematic illustration for the preparation of superhydrophobic cotton fabric (A) TA/Borax coating, Reproduced with permission from reference (79); CF/PDA/FeOOH/DDT (B), Reproduced with permission from reference (80); CF with aligned $Cu(OH)_2$ nanoneedles (C), Reproduced with permission from Ref. (81).

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Cheng *et al.* deposited β-FeOOH nanorods on the PDA-coated cotton fabric by an *in-situ* hydrothermal process to provide roughness on the surface (82), **Fig.5B.** Superhydrophobicity (WCA~151°) and lipophilicity (OCA~0°) were achieved by the addition of 1-DDT on the PDA/β-FeOOH deposited cotton fabric to reduce the surface energy. The modified fabric exhibited higher oil-water separation efficiency for all types of oils, which is greater than 98% after 50 cycles. Enhanced thermal stability and excellent photocatalytic performance after 5 cycles were observed in the modified fabric due to the development of an even layer of β-FeOOH nanorods on the PDA-coated fabric.

Belal *et al.* synthesized copper hydroxide Nano needles on the PDA-coated cotton fabric by solution immersion process (83), **Fig.5C.** Insitu polymerization of DA was carried out on the cotton fabric, followed by the addition of Ag nanoparticles (reduction) and Cu nanoparticles (oxidation) endowed the surface roughness. Thereafter, the low surface energy material of n-dodecanethiol (n-DDT) was coated on the functionalized fabric to achieve superhydrophobicity. The functionalized fabric exhibited a higher WCA of 168° together with a higher oil-water separation efficiency of above 99% after 100 cycles. The attained values of WCA >166° after the solvent immersion (32 days) and acidic/basic condition (48 hours) reveal water repellency and stability of the functionalized fabric.

Yan *et al.* reported a two-step procedure for fabricating PDA-coated superhydrophobic cotton fabric grafted with ODA through Schiff base reaction in the presence of $FeCl_3$ by solution immersion method (84). The modified fabric showed excellent stability under harsh environments of varied pH, laundering, and organic solvent with a WCA >150° and WSA <14°. However, it shows poor acid resistance due to the hydrolysis of amino groups in ODA and poor resistance to carbon tetrachloride (CCl_4) as the ODA will be leached out

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in CCl_4. The functionalized fabric showed an oil/water separation efficiency of above 99% after 20 cycles. As illustrated in **Fig.6 E-F,** the functionalized fabric showed self-cleaning properties with good mechanical durability.

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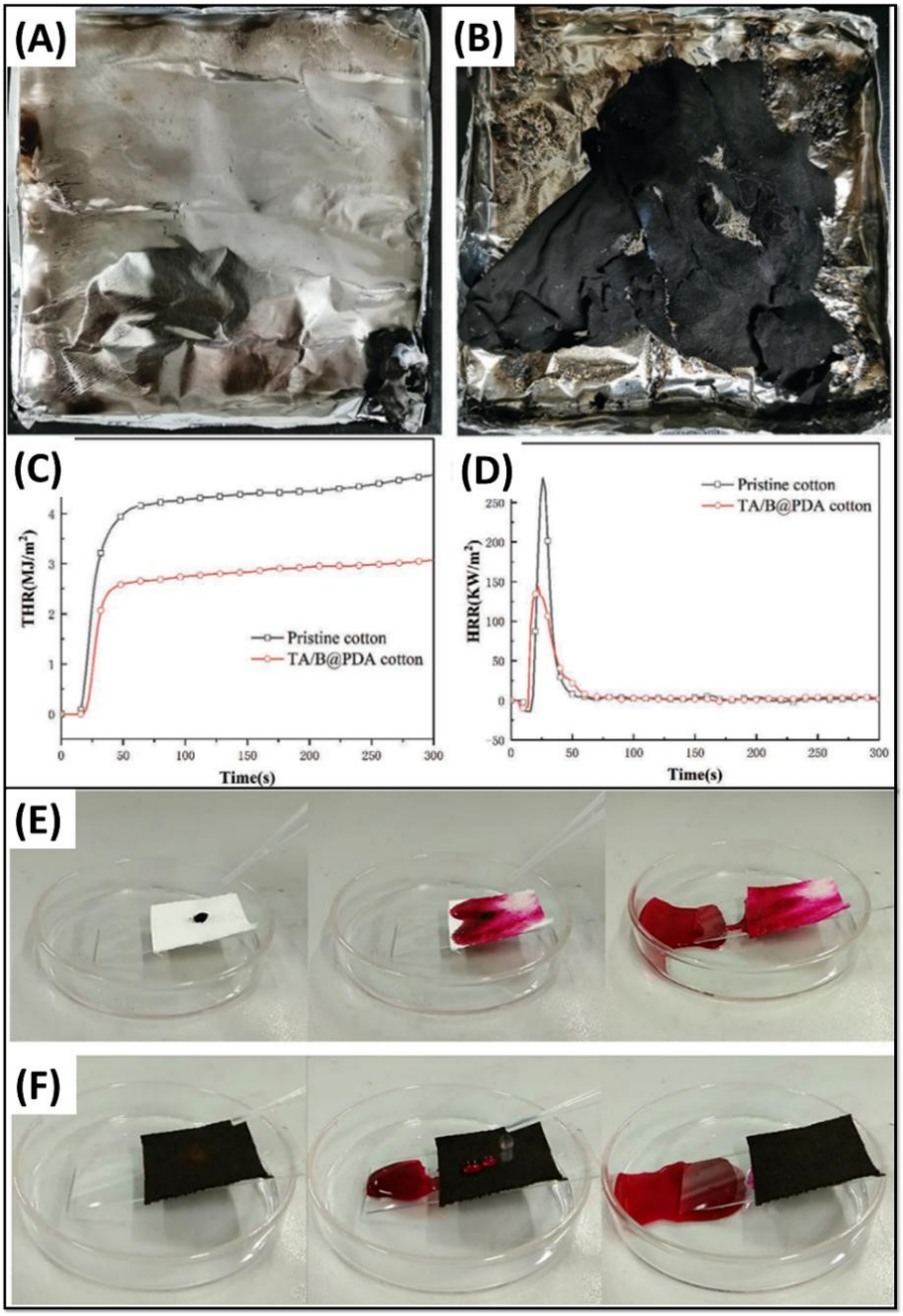

Figure 6 Digital photos of char residues of (A) pristine cotton and (B) TA/B@PDA cotton fabric after CCT. (C) THR and (D) HRR of cotton samples, reproduced with permission from Ref. (79); Illustrations of fabric with reactive red dye powder under the rinsing of water (E) original cotton fabric, (F) Fe/PDA/ ODA cotton fabric, reproduced with permission from Ref. (5).

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Chen and Guo fabricated the PDA-coated superhydrophobic fabric composed of polyester (65%) and cotton (35%) blend, by solution immersion method (81). They employed sodium periodate as an oxidant, with a molar ratio of the half, for obtaining a thick and rough PDA coating on the modified fabric. In the second step, superhydrophobicity was achieved by the incorporation of low surface energy ODT molecules. The functionalized fabric showed excellent superhydrophobic and self-cleaning properties with a WCA of 154° and WSA of 9.5°. The modified fabric showed a separation efficiency of greater than 97% and a permeation flux of 4500 $L.m^{-2}.h^{-1}$ even after 20 absorption/ squeezing cycles.

Wang *et al.* employed a solution immersion method and thiol-ene click reaction to fabricate a superhydrophobic fabric composed of a blend of 50% cotton fiber and 50% nylon-56 fiber for oil-water separation with a self-cleaning ability (80). The modified fabric was prepared by immersing the PDA-coated fabric into the mercaptopropylmethyl dimethoxy silane (MPMDS) under prescribed conditions. Thereafter, superhydrophobicity was achieved by treating the mercapto-modified fabric with a mixture of hexadecyl mercaptan, octavinyl-POSS, and 2,2-Dimethoxy-2-phenylacetophenone (DMPA) to provide low surface energy along with the micro-nano rough surface. The modified fabric showed WCA and WSA of 162° and 8°, respectively, with an excellent oil-water separation capacity. The durability of the modified fabric was evaluated by exposing the sample to mechanical abrasion, UV irradiation for 16 hours, immersion in an organic solvent for 24 hours, immersion in varying pH solution (1 to 13) for 48 hours, and laundering test for 4.5 hours. The results from the formed tests showed that the modified

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fabric did not lose its hydrophobicity with the WCA > 150° and WSA < 15°, which revealed the durability of the superhydrophobic fabric.

Xu *et al.* fabricated different PDA-coated superhydrophobic fabrics by using oxygen-induced polymerization of DA and solution immersion method (85). Deposition of PDA and silver nanoparticles provided the surface roughness and further superhydrophobicity was achieved by the incorporation of 1H,1H,2H,2H-perfluorodecanethiol to attain the rough hierarchical structure and low surface energy. The modified fabrics showed WCA >150° and WSA<10° with excellent oil-water separation and self-cleaning ability.

Dong *et al.* fabricated the eco-friendly, solar-induced self-healing superhydrophobic cotton fabric by solution immersion and dip-coating method (86). An activator ($CuSO_4/H_2O_2$) was used to deposit PDA onto the cotton fabric to attain a stable and uniform rough surface. Stearic acid (SA) with a concentration of 0.1 wt% was employed to reduce the surface energy due to its long alkyl chain. The modified fabric exhibited a WCA of 162° and WSA of 7.8° with an oil-water separation efficiency of ~100% after 5 cycles. Durability and stability of the modified fabric were evaluated by abrasion test for 1000cycles, laundering test for 5 cycles, and immersion in acidic/basic/alkaline solution for 24 hours shows that the modified fabric did not lose its superhydrophobicity with the WCA >150°. Furthermore, UV transmittance is <1% for the modified fabric and recovers its superhydrophobicity after 6 cycles of plasma treatment and light irradiation. The above results reveal the UV protection and self-healing property of the modified fabric.

Zhu et al., prepared the antibacterial composite cotton fabric by dip-coating method (87). Surface roughness was created by self-polymerized PDA nps and the antimicrobial effect was attained by the in-situ growth of ZIF-8 nanomaterials which acts as a photo thermal

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agent followed by incorporating PDMS to reduce the surface energy through dip-coating process on cotton fabrics. The prepared composite fabric shows the WCA of 153° at 1wt% PDMS which shows the successful attainment super wettable characteristics. Durability was assessed through 5 washing cycles and a friction distance of 100cm which exhibits the WCA >140°. Additionally, ZIF-8 nanomaterials on the prepared fabric release positively charged Zn^{2+} under near-infrared light irradiation to rupture the bacterial cells which enhances the antibacterial effectiveness.

Long et al., (88) fabricated the multifunctional super hydrophobic membrane with significant photocatalytic effect on the cotton fabric through impregnation and water bath heating method. They employed β-FeOOH nps to achieve the photocatalytic characteristics and rough structure on the PDA coated cotton material. Further, surface energy was reduced by the incorporation of hexadecyltrimethoxysilane at 60°C for 5 hours to attain the Hexadecyltrimethoxysilane/FeOOH/polydopamine/cotton fabric (HFPC) membrane. The as-prepared HFPC membrane guards the permeation of various liquids of muddy water, tea, urine, coca-cola and milk with the WCA of 153 - 158° and the SA of 6 - 7° and rejects the various organic solvents which clearly reveals the successful attainment of superhydrophobicity and Antifouling property. As shown in **Fig. 7,** the outstanding chemical durability and mechanical robustness were assessed through various methods like tape peeling (1000 times), finger wiping (60 kPa/1000 times), knife scratches (1000 times), abrasion test (1000 wears), hammering test (500 g/1000 times), water impact test (2.5 m/sec for 5 min), kneading, hand twisting, stretching, chemical immersion (24 hours), UV aging (12 hours) and outdoor weathering test (5 months). Moreover, it has an efficient catalytic activity under visible light irradiation and catalytic

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degradation ability of 99.5% under UV light irradiation through photo fenton reaction. Additionally, it can separate oil-water mixtures continuously with an efficiency of up to 99.5%.

Zhang and Guo (89), developed the self-healing durable cotton fabric for oil-water separation by two step process. They incorporated ZIF-8 on the cotton fabric along with PDA as the combination of ZIF-8/dopamine (DA) can achieve quick assembly within 30 minutes on the different material surfaces in the neutral pH which can enhance the self-healing properties. Beside this, multiwalled carbon nanotubes were added along with ZIF-8/DA through solution immersion process on the cotton to attain mechanical stability.

The as-prepared fabric shows the separation efficiency of 99% with the WCA of 158.9±1.4° and SA of 4±0.5°. In addition, it exhibits excellent resistance towards acidic (12 hrs), alkali solution (6 hrs), ultrasonication (14 hrs) and abrasion (35 cycles) as the WCA is greater than 145° after the above-mentioned test. The as-prepared fabric surface maintains its superhydrophobicity until 6 healing cycles as revealed by oxygen-plasma test due to the deposition of PDMS on the structural pores which migrate rapidly to the surface driven by heat. However, self-healing of microstructure on the modified fabric was not possible as it will be demolished after abrasion as detected by XPS.

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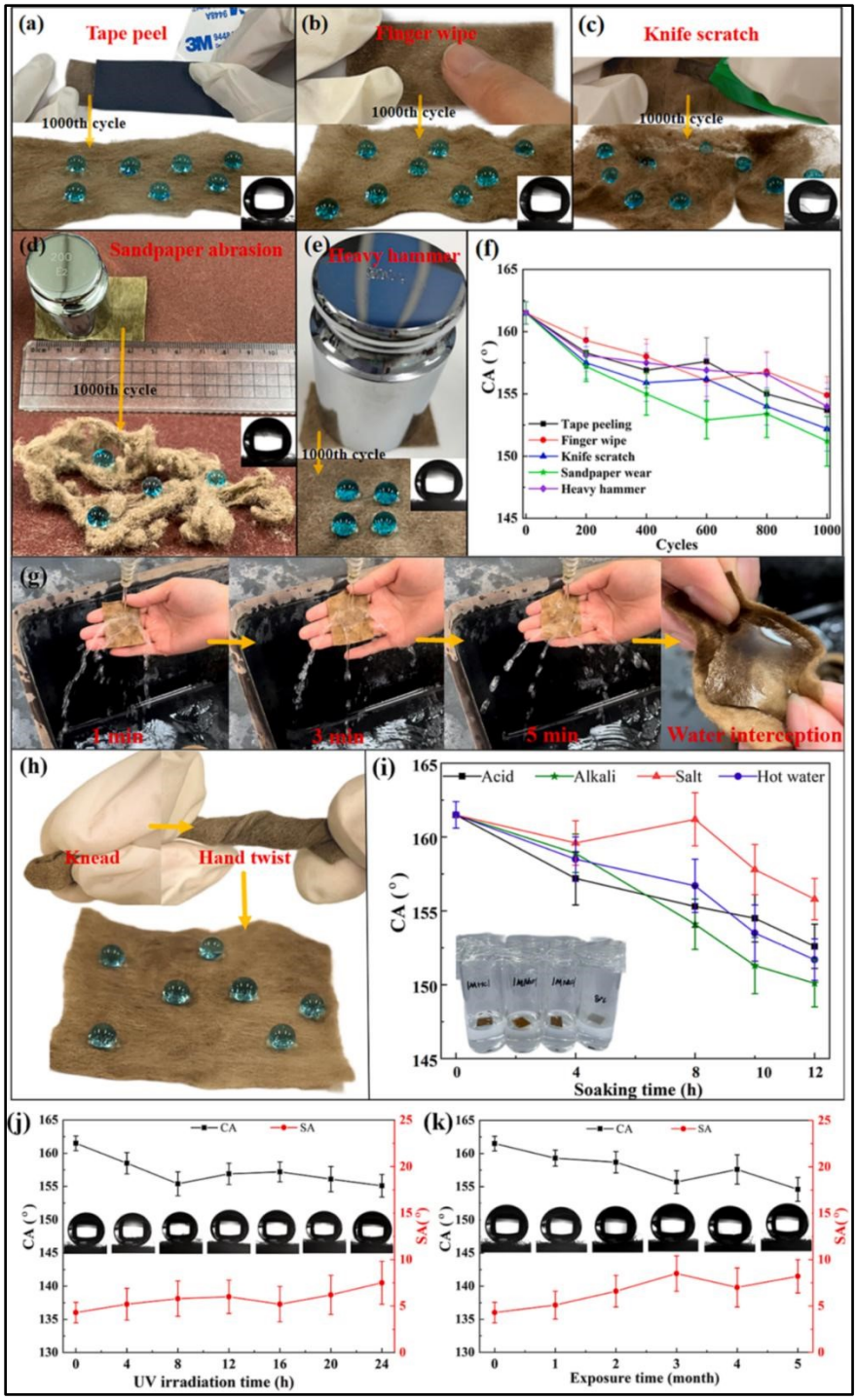

Figure 7 Robustness of HFPC membranes. (a) Tape peel, (b) finger wipe, (c) knife scratch, (d) sandpaper abrasion, (e) heavy hammering, and superhydrophobic photographs of the corresponding 1000 cycles. (f) The corresponding CAs changes after 1000 cycles. (g) Effective water interception even after 5 min of running water impact. (h) Elasticity test. (i) The superhydrophobic behavior after soaking in organic solvents (1 M H_2SO_4, 1 M NaOH, 1 M NaCl) and 80°C hot water for 12 h. Changes in CAs and SAs after (j) UV irradiation for 24 h and (k) outdoor weathering for 5 months. Reproduced with permission from Ref.(88)

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3.2. Other SHB textiles

During the last few decades, researchers have focused on simple, eco-friendly, and sustainable methods to fabricate superhydrophobic material. Eventually, mussels have drawn attraction due to their uniqueness of discharging an adhesive protein to bind with all types of substrates (90). PDA has been employed among researchers to mimic the mussel adhesion effect as it can be easily deposited on various substrates to achieve surface roughness. Additionally, it has the property of biocompatibility, oxidation resistance, and stability in near-infrared UV, and visible light, as well as a high conversion efficiency of photothermal activity (91).

Xue et al. fabricated a superhydrophobic PET fabric by using a two-step fabrication process (92), **Fig.8**. The surface roughness was attained *via* COP of DA in Tris buffer which resulted in the deposition of PDA in the form of nanostructured papillae, while the superhydrophobicity was achieved in the subsequent step by immersing the modified textiles in 1% ethanolic solution of perfluorodecyl trichlorosilane (PFDTS). In an approach to mimic the lotus leaf nanostructured morphology, PET textiles were coated initially with DA in a phosphate buffer (pH = 8.5), followed by a second coating of DA in a Tris buffer. The former approach led to an increase in the WCA from 150º to 160º due to the hierarchal nanostructured morphology of the coated textiles caused by the production of larger PDA nanoparticles during the COP in phosphate buffer solution. The modified PET textiles exhibited properties including UV-shielding, good mechanical durability, and chemical stability in acidic and alkaline environments.

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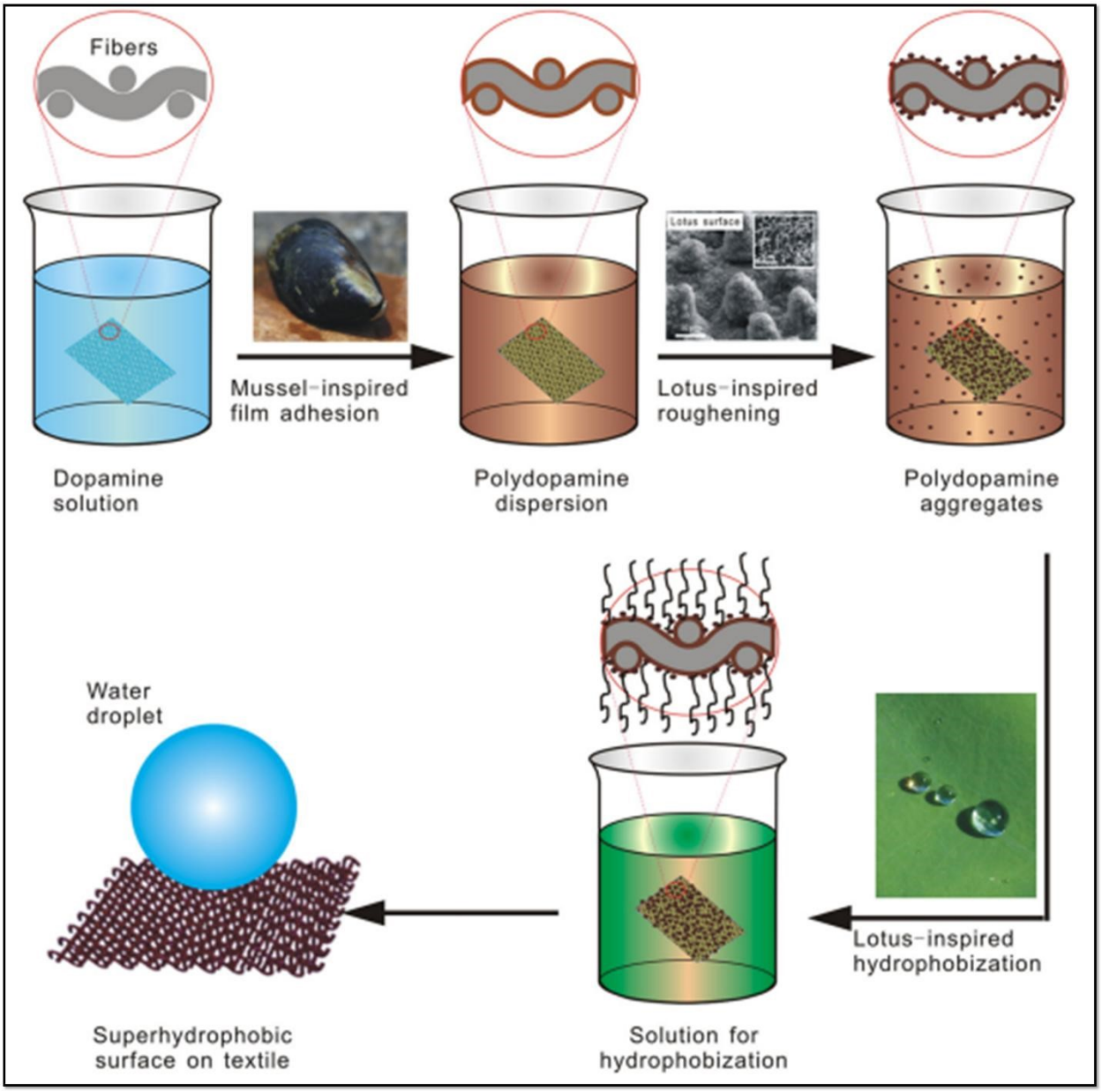

Figure 8 Schematic illustration for the preparation of biomimetic superhydrophobic surfaces by combining mussel-inspired adhesion and lotus-inspired coating. Reproduced with permission from Ref. (92).

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Lu *et al.* employed the polymer fiber fabric (Tetra-copolyester and polypropylene) as a substrate to fabricate hierarchical rough structures to achieve superhydrophobicity by the simple bottom-up approach (93), **Fig.9A.** The fiber fabric was modified through dip coating by the incorporation of SiO_2 NPs (obtained by the hydrolysis of TEOS) into the DA solution, with the successive immersion of coated fabric into the silver ammonia solution containing 0.25 wt% of PVP (stabilizing agent). The incorporation of SiO_2, Ag NPs, and 1H, 1H, 2H, 2H-perfluorodecanethiol (PFDT) endowed the fiber fabric with a micro/nano hierarchical rough structure with low surface energy. The exhibited separation efficiency of above 90% after 20 cycles shows the excellent stability and recyclability of the modified fiber fabric. However, after each cycle, there is a slight change in the WCA and WSA, which further sustain its water-repellency property. Additionally, the evaluated WCA of the modified fabric with the varied particle size of SiO_2 (200, 300, and 400nm) are 147.8°±2.5°, 154.5°±2.2° and 160.1°±2.8° respectively. Thus, the higher value of WCA shows the superhydrophobicity of the modified fabric and it can be increased by increasing the size of SiO_2.

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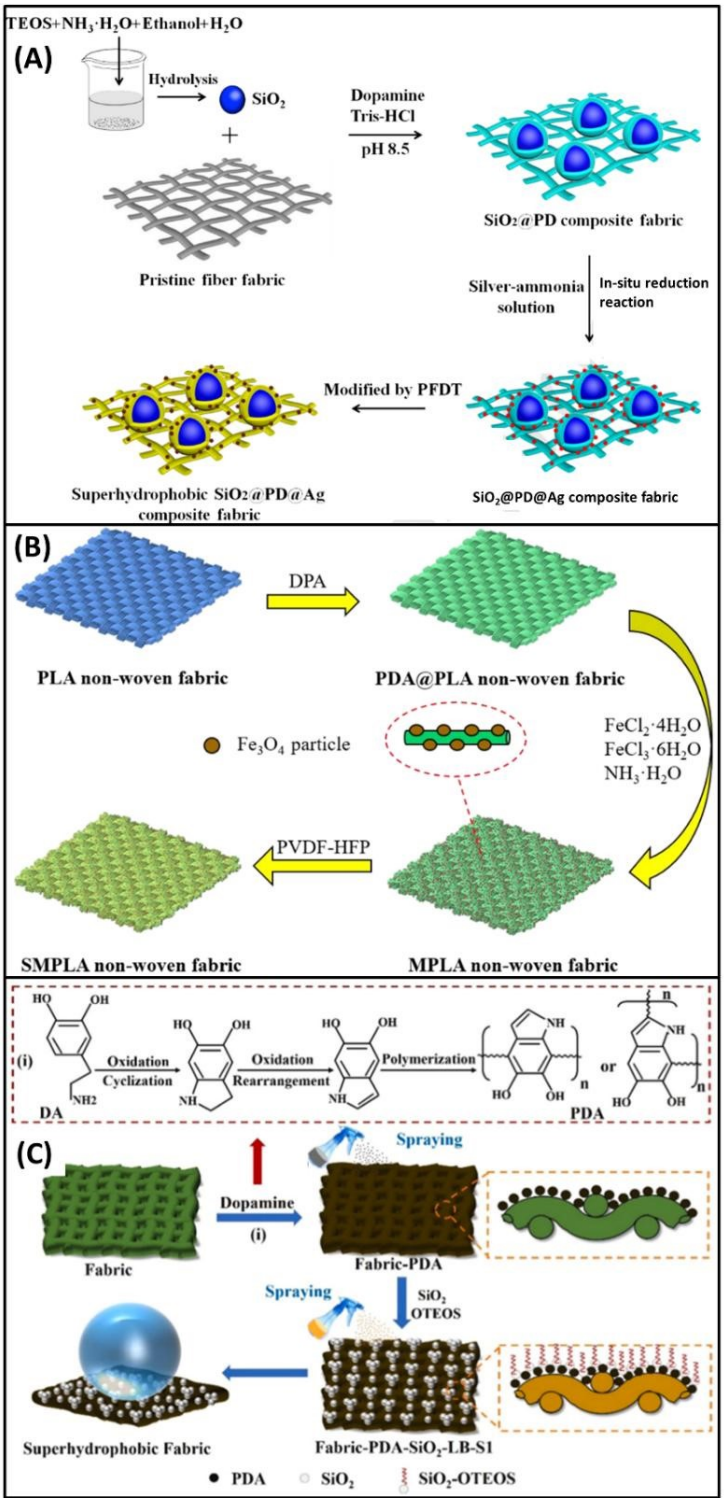

Figure 9 Schematic illustration for the preparation of superhydrophobic fabric (other than cotton) (A) Tetra-copolyester and polypropylene/ SiO2/PD/Ag/PFDT, reproduced with permission from Ref. (93); PLA/PDA/PVDF-HFP (B), reproduced with permission from Ref. (94); other materials/PDA/SiO_2/OTEOS (C), reproduced with permission from Ref. (95).

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Zeng *et al.* reported the fabrication of a bio-degradable superhydrophobic and magnetic poly(lactic acid), (PLA) nonwoven fabric for oil-water separation by solution immersion method (94), **Fig.9B.** PDA was deposited on the PLA nonwoven fabric by an *in situ* polymerization of DA. Then the surface roughness and magnetic property were achieved by the incorporation of ferrous chloride tetrahydrate ($FeCl_2 \cdot 4H_2O$) and ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) in the molar ratio of 1:2 to generate Fe_3O_4 particles on the PDA coated PLA nonwoven fabric. Surface roughness and WCA can be increased by increasing the concentration of $FeCl_3 \cdot 6H_2O$ (150mmol/L). Furthermore, the surface energy was reduced to attain superhydrophobicity by immersing the modified fabric (Fe_3O_4/PDA/PLA) into the solution of poly(vinylidene fluoride-co-hexafluoropro-pylene) (PVDF-HFP) for 4hours and dried at 50°C for 1hour. The modified nonwoven fabric displayed a WCA of 151.7°, which reveals the accomplishment of superhydrophobicity. Surface stability was evaluated by exposing the modified nonwoven fabric to the different organic solvents, aqueous H_2SO_4, aqueous KOH solution, and boiling water for 96hours. The measured WCAs are very near to the initial value, which reveals surface stability of the modified fabric is resistant to acid, alkali, and boiling water. Moreover, the attained WCA of 148.6° even after 20 cycles of abrasion test further confirms the stability of the surface. The separation efficiency of the modified fabric shows more than 97% even after 20 cycles exhibiting excellent recyclability and the absorption capacity shows that the modified nonwoven fabric can absorb organic solvent upto16 times its weight and it reaches 36 times in the case of higher density solvent CCl_4.

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Zhang *et al.* reported the fabrication of a PDA-coated pristine superhydrophobic fabric by the combination of DA polymerization and the sol-gel method (96). Surface roughness was achieved by the incorporation of PDA and SiO_2 nanoparticles and further superhydrophobicity was achieved by the involvement of Polydimethysiloxane (PDMS) to reduce the surface energy. The functionalized fabric showed an enhancement in its thermal stability and excellent oil-water separation efficiency of about 95% after 8 cycles along with a WCA of 155°. Additionally, the functionalized fabric maintains its superhydrophobicity with the WCA ~150°, after immersion in different pH solutions (1 to 13) for 24 hours, organic solvent for 24 hours, washing test for 3 hours, and exposure to UV irradiation for 1 hour.

Yu *et al.* employed various materials like cotton fabric, sponge, concrete, wood, paper, glass, and aluminum sheet to fabricate superhydrophobic material by the scalable two-step spraying method (95), **Fig.9C.** PDA nanoparticles were utilized to attain the surface roughness and SiO_2/Octyltriethoxysilane (OTEOS) was incorporated to reduce the surface energy for the betterment of superhydrophobic properties. All the modified materials exhibited a WCA of 154.2 ± 2° and a WSA of <3°. The results attained from the durability and stability test reveal that the modified material has excellent resistance under mechanical abrasion, acidic/basic condition for 24 hours, organic solvents immersion for 24 hours, UV irradiation for 120 hours, and longtime outdoor environment for 12 months without losing its superhydrophobicity (WCA>151°).

Su et al., (97) developed the pH responsive Nano fibrous polyvinylidene fluoride (PVDF) membrane by the electrospinning process and further modification through solution immersion and metal thiol co-ordination reaction. Surface roughness was attained by the

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incorporation of silver nano particles and successive addition of thiols ($HS(CH_2)_{11}CH_3$/$HS(CH_2)_{10}COOH$) which further modifies the membrane into a pH responsive super wettable membrane. Self-polymerized DA restricts the mobility of silver nanoparticles and thiols which created the microstructure with upright structural stability against harsh mechanical action. The modified membrane showed the efficiency of over 99.2% for light oil/water and 96.5% for heavy oil/water mixture. Mechanism of the wettability switching of pH-responsive membrane as shown in **Fig. 10A** which displays the WCA of 151.5° and UWOCA of 0° for pH=7 and WCA of 0° and UWOCA of 152.5° (pH=13). In addition, it exhibited the energy saving separation efficiency of about 99% for all type of surfactant stabilized emulsions with excellent durability and reusability.

Hatami et al.,(98) utilized the waste woolen felt due to the benefits of fire retardant and self-extinguishing property to prepare oil-water separation material which is inexpensive and bio-degradable on top of that. They employed varied densities (1.165, 1.650,1.682 and 1.747 g/cm^3) of woolen felt as a base matrix which is covered with TiO_2/PDA nano particles followed by silver nano particles through solution immersion and stirring method to attain the rough hierarchical structure on the surface. Further, modified with DDT to reduce the surface energy of the material to prepare the superhydrophobic/superoleophilic material. Among the modified superhydrophobic materials, the woolen felt material with a density of 1.650 g/cm^3 shows the WCA of 155.14° with a superior separation and passage of water at the time of 4:19:120 and 1:37:100 **(Fig. 10B)** which is attributed to the homogeneous distribution of nano particles on the woolen felt which prevents the clusters.

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Li et al., (99) constructed the pH responsive superhydrophobic composite membrane based on PP fibre and the wettability was enhanced through PDA deposition on the substrate. through in-situ polymerization, solution immersion and vacuum assisted filtration method. They used Mxene nano sheets which is a 2D transition metal, due to the attributes of rich active sites, photo thermal properties, ceramic-like stability and polymer-like flexibility. Further to improve the stability and utilize its photo thermal properties **(Fig. 10C)** of Mxene, Polypyrrole (PPy) was deposited on Mxene through in-situ polymerization process. The vacuum assisted filtration method was used to construct the composite membrane, and its outer surface was covered with the transparent pH-responsive polymer PDM through solution immersion process. The prepared composite membrane (PP-MXene/PPy-PDM) at pH=2 exhibits the WCA of 40◦ and the OCA of 130◦, when the pH elevated to 12, the WCA was130◦, and the OCA was 0◦. The above results indicate that the membrane is hydrophilic in acidic environment and becomes hydrophobic in alkali environment. Moreover, it shows the higher separation efficiency ranging from 2.44 to 3.87 kg/m^2.h for the diverse oil-water emulsion with excellent resistance to abrasion, acidic and alkali conditions.

Wang et al., (100) designated the Nylon 56 fabric to fabricate superhydrophobic membrane through eco-friendly method because of its high elasticity, high temperature resistance, wear resistance and environmental protection. They coated this fabric with PDA which can provide reactive sites for other ingredients, followed by the integration of tetraallyl silane (TETRA) and (mercapto) methyl siloxane-dimethyl siloxane copolymer (MMSDC) on the polydopamine coating by thiol−ene click-reaction under ultraviolet light. The hydrophobic reagents of TETRA to MMSDC molar ratio was optimized to 1:3 as it

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shows the SCA of 166°, SA of 7.5° which confirms the effective attainment of low surface energy groups and hydrophobicity on the fabric. Mechanical stability was assessed through sandpaper abrasion for 25 cycles which shows that the WCA dropped from 166° to 151° without losing its superhydrophobicity. Additionally, it exhibits excellent self-cleaning and antifouling properties as it repels the various pollutants of organic dye, saltwater, coffee, milk, cola, and tea.

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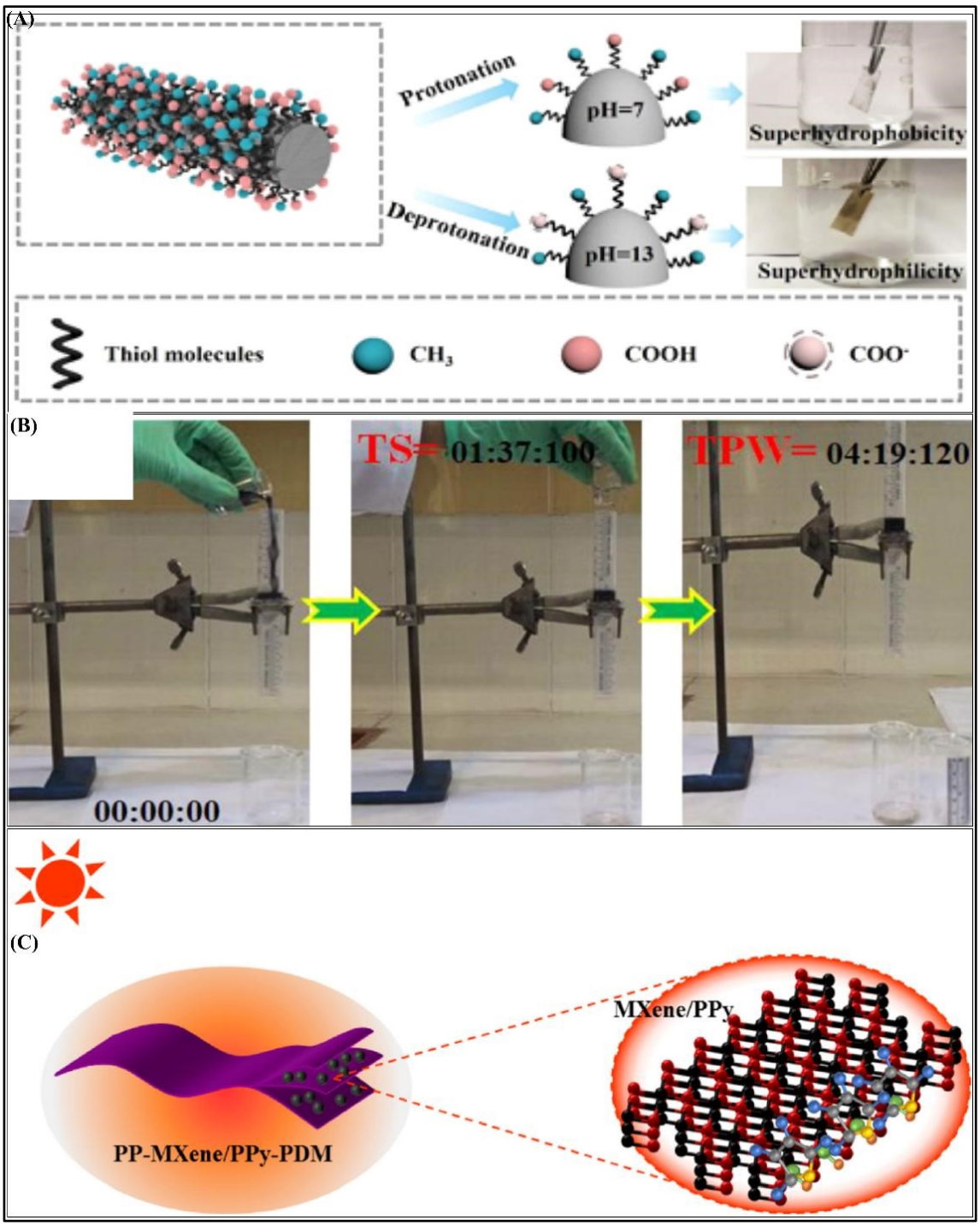

Figure 10 (A) Mechanism illustration of the wettability switching of the pH-responsive membrane at pH 7 and 13, reproduced with permission from Ref. (97); (B) oil-water separation time, reproduced with permission from Ref. (98); (C) Schematic illustration of the photo thermal conversion in the PP-MXene/PPy-PDM membrane, reproduced with permission from Ref. (99).

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4. Superhydrophilic and underwater superoleophobic (SHL/UWSOB) PDA-coated textiles for oil/water separation

SHL/UWSOB textiles are superwettable textiles that display underwater oil contact angles (UWOCAs) above 150º and possess a super affinity for water and will, therefore, remove water from any oil-water mixture while strongly repelling oil (101). SHL/UWSOB textiles can overcome the shortcomings of the oil-removing superhydrophobic textiles which include: (1) inability to remove low-density oils and highly viscous oils from their corresponding oil/water mixtures, and (2) easy fouling of the superhydrophobic textile membrane that usually results in pore blockage and loss of the separation capacity (102). Superhydrophilicity is responsible for an ultra-low fouling capacity that translates to a sustained, efficient, and consistent separation capacity. Though not easy to fabricate, SHL/UWSOB textiles are excellent materials for the easy removal of water from both surfactant-stabilized and non-surfactant-stabilized oil/water mixtures irrespective of their viscosity or density. Many documented fabrication approaches, for SHL/UWSOB materials, involve the use of toxic fluorine-containing particles and the use of substrates sourced from non-renewable materials such as stainless steel meshes, polyvinylidene fluoride (PVDF) membranes, copper meshes, etc. (102). In comparison, woven and nonwoven textiles and fabrics are advantageous as they provide flexibility, porosity, mechanical/ chemical stability, and elasticity that potentially contribute to a scalable process. The surface roughness of the superhydrophilic textiles can be increased by using various nanoparticles. However, the wettability achieved by the incorporation of these nanoparticles is usually short-lived as they are easily dislodged from such surfaces by mechanical abrasion. Therefore, polydopamine has been used in conjunction with

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these nanoparticles to increase the life span of their wettability and decrease the rate of dislodgement and the effect of abrasion. Other polymers such as chitosan, polyethyleneimine, etc. have been used in conjunction with polydopamine to improve the superhydrophilicity already imparted because of the presence of a high number of amine and hydroxyl functional groups on the backbone of these polymers.

4.1. Cotton textiles

Among the substrates used for the fabrication of SHL/UWSOB separation textiles, cotton stands out because it is sourced from readily available and renewable resources, is easily biodegradable, and is relatively inexpensive. Their biodegradability means that they are recyclable which greatly reduces their contribution to the problem of environmental pollution at the end of their life cycle. Just like other textiles, cotton textiles are equally flexible and lend themselves to scalable fabrication processes. One major setback to the use of cotton fabric (CF) as a substrate is its weak adhesion to the NPs and other materials used for imparting surface roughness. Consequently, the ability of the mussel-inspired dopamine to adhere strongly to practically any surface readily solves the challenge of limited adhesion for CF. Additionally, though untreated cotton has abundant hydroxyl groups, its affinity for water is suboptimal due to the presence of non-cellulose components such as pectin, lignin, and wax (103). As a result, pretreatment with ultrasonication or alkali thermal process, or both are undertaken before coating. Micro-dissolution and coagulation have been used as an approach to improve the adhesivity of CF-based material (104). In this approach, the CF is impregnated with a dissolution solution that lightly dissolves the surface of the fabric. The multiple steps involved in this

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approach may prove a disadvantage. Other polymers such as polyurethane (105) and polyethyleneimine (106) have been used to improve adhesivity.

Wang and colleagues developed a SHL/UWSOB cotton fabric by using a combination of PDA and Chitosan (CS), (PDA-CS-CF) (102). They evaluated the effect of each polymer individually and in combination on the UWOCA, and the stability of the hydrogel coat to harsh conditions. The separation efficiency for petroleum ether and soybean oil-water mixtures were above 99.5 % and flux of 41000 and 28000 $L.m^{-2}.h^{-1}$ respectively. These values persisted after 100 filtration cycles and after the (PDA-CS-CF) were subjected to ultrasonic, saltwater, alkaline, and acidic solution treatments. Likewise, Li and coworkers prepared a SHL/UWSOB cotton fabric for the dual removal of heavy metal ions and oil from artificial oily wastewater using the same CS and PDA polymers, but, with a slightly different deposition method (107). Polydopamine was deposited on an ultrasonically cleaned CF in an oxidative polymerization process from a solution of dopamine HCl over 24 hours to yield PDA-CF. Chitosan was subsequently deposited on the dried PDA-CF from a 1 % w/v solution of chitosan in glacial acetic acid to yield PDA-CF-CS after drying. The amino groups in chitosan provided adsorption points for heavy metals and crosslinking points with the quinone structure in the polydopamine coat**. Fig.11A** shows the digital images of the separation process for the copper-contaminated oil-water for the CF, PDA-CF, and PDA-CF-CS. As shown in **Fig.11B**, the lowest water infiltration time was 117s for the superhydrophilic PDA-CF-CS, while the UWOCA was 139.5 °, 147.3 ° and 156.1 ° for the CF, PDA-CF, and PDA-CF-CS respectively. The UWOCA of PDA-CF-CS decreased to 152.6º after 300 abrasion cycles of 40 cm/cycle under a 200g weight.

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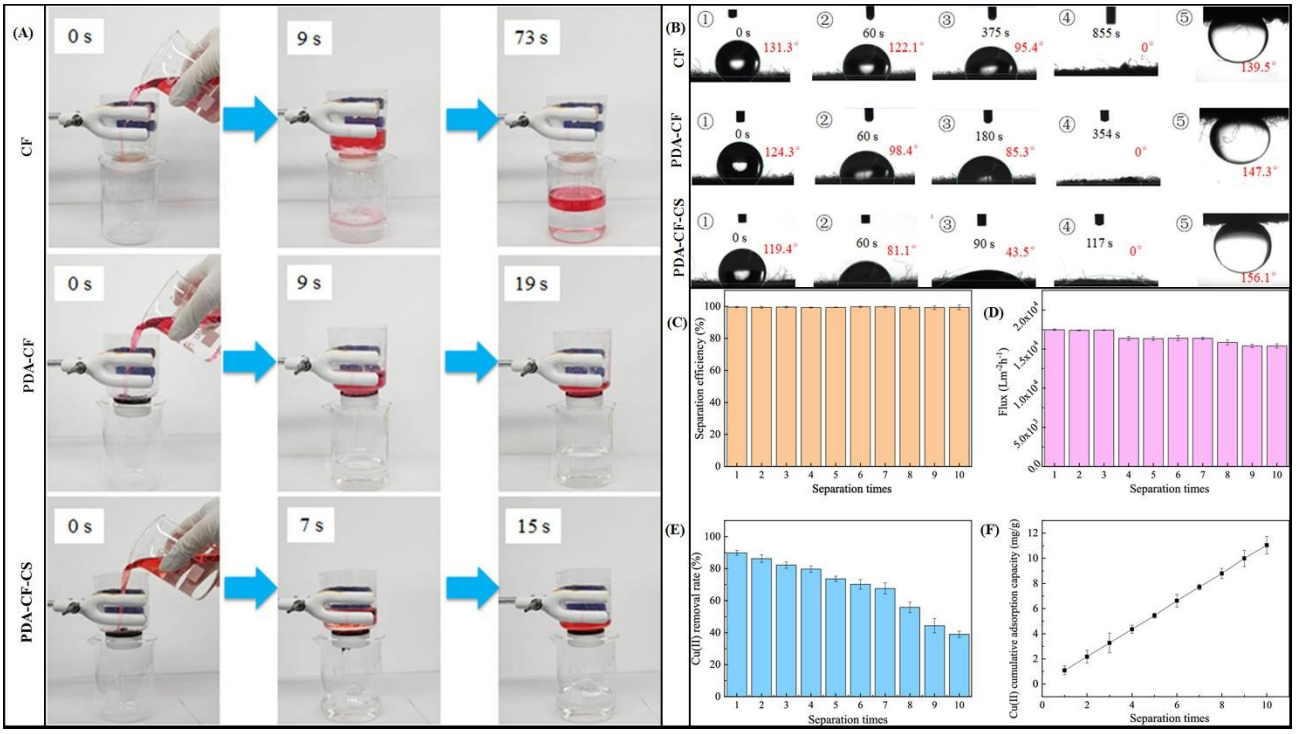

Figure 11 Digital images showing the separation of Cu (II)-oil-water process for CF, PDA-CF, AND PDA-CF-CS (A); WCA and UWOCA for CF, PDA-CF, AND PDA-CF-CS (B); Flux and separation efficiency (C and D); and copper adsorption capacity of the PDA-CF-CS (E and F), Reproduced with permission from Ref. (108).

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The oil-water separation efficiency remained above 99 % after 10 separation cycles while the water flux decreased from 17400 L.m.$^{-2}$h^{-1} to 15400 L m^{-2} h^{-1} by the 10th cycle **(Fig.11 C-D).** The copper removal efficiency was about 89 % which decreased within 2 mins to 67 % during continuous filtration **(Fig.11E-F).** Zhong and coworkers (103) utilized titanium oxide NPs (due to their antibacterial, UV shielding, and hydrophilicity), polydopamine (to improve the adhesion and hydrophilicity), (3-aminopropyl) triethoxysilane (KH550) (to improve the stability of polydopamine in an alkaline environment and additional hydration capacity), in a one-step impregnation process to prepare SHUSO cotton fabric. The TiO_2 NPs and PEG were dispersed in Tris-HCl buffer to which copper sulphate, hydrogen peroxide, KH550, and dopamine were subsequently added. The CF, soaked in sodium hydroxide at 95ºC for 24 hours and ultrasonically cleaned and dried, was soaked in the impregnating solution. Synergistic crosslinking polymerization across the KH550, catechol group of the dopamine, and TiO_2 NPs ensured the firm fixture of the modification on the CF. The water spreading time of the modified CF decreased from 13.45 s for the pristine CF to 0.5 s. The UWOCA for all the oils used was above 169 °. The separation efficiency for an oil-water mixture remained above 99.99 % after 50 filtration cycles and immersion in acidic, alkaline, and salt solutions. A maximum flux of 32000 L.m^{-2} .h^{-1} was obtained that varied slightly after 3 cycles. The separation efficiency of the modified CF remained above 99.99 % after being subjected to 50 abrasion cycles (20 cm/cycle) on 800-grit sandpaper under 2800 pa pressure.

Hu et al., (109) developed the SHL/UWSOB fabric for oil-water separation and removal of dyes by two-step process of dip-coating in poly(sodium 4-styrenesulfonate) (PSS) enveloped carbon nanotube (CNT) dispersion, followed by the subsequent in-situ

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deposition of PDA. Surface roughness (Ra) of virgin cotton was elevated from 27.005 μm to 34.055 μm by the incorporation of PSS and PDA nano particles. The attained fabric shows the separation efficiency of greater than 99.8% with an UWOCA of 160° which clearly reveals its super wetting ability. Besides that, it exhibits about 97% removal rate for cationic dyes especially methylene blue due to the addition of porous CNT which provides the excellent dye adsorption and hydrophilic capability for the modified fabric. Additionally, it displays the UWOCA of >150° after exposed to the harsh conditions viz. sandpaper abrasion, immersion in various organic solvents, 30%NaCl for 7 days, immersion in aqueous solutions of various pH values of 1 to 13. The above outcomes indicated that the modified fabric has an excellent stability and durability which was ascribed to the strong interactions among PDA, CNT, PSS and cotton fabric owing to the presence of hydrogen bonding between them.

Li and his colleagues (110), developed the inexpensive and eco-friendly SHL/UWSOB fabric for oil-water separation via oxidative polymerization of DA on the cotton fabric and SiO2 was integrated through solution immersion process. The attained PDA/SiO2/cotton fabric reveals the UWOCA of 159.7° with the separation efficiency of greater than 98% for crude oil/water and 99% for toluene/water. Additionally, UWOCAs are still greater than 150° after undergone of ultra-sonication (9 hrs.), sandpaper abrasion (500 cycles), strong acid, strong alkali, NaCl solution (immersed in 31days), signifying that the developed cotton fabric has notable mechanical and chemical stability. It is noteworthy to mention that the deionized water can be used easily to remove the crude oil attached to the developed fabric surface which clearly indicates the anti-crude oil fouling property of SiO_2/PDA/CF fabric.

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4.2. Other SHL/UWSOB textiles

Woven fabric and unwoven fibers made from materials other than cotton have been investigated for the fabrication of SHL/UWSOB by different studies. On the forefront of such studies is the electrospun nanofibers from a variety of polymers such as polyacryonitrile (PAN) (111) polyacryonitrile ether nitrile (PEN), polyurethane (PU) (112). Other textiles include nonwoven fabrics such as polyester (113), and nylon (114). Electrospun fibers (EFs) are highly porous substrates with continuously interconnected pores and particularly offer advantages for the separation of surfactant stabilized oil-water mixtures because of their pore and diameter size that can be controlled to range from a few nanometers to micrometers (111). Even though many EFs are superhydrophobic, Zhao and co-workers prepared a polydopamine functionalized SHL/UWSOB fiber using polyacrylonitrile (PAN), a superhydrophilic polymer from acrylonitrile repeating units (111).

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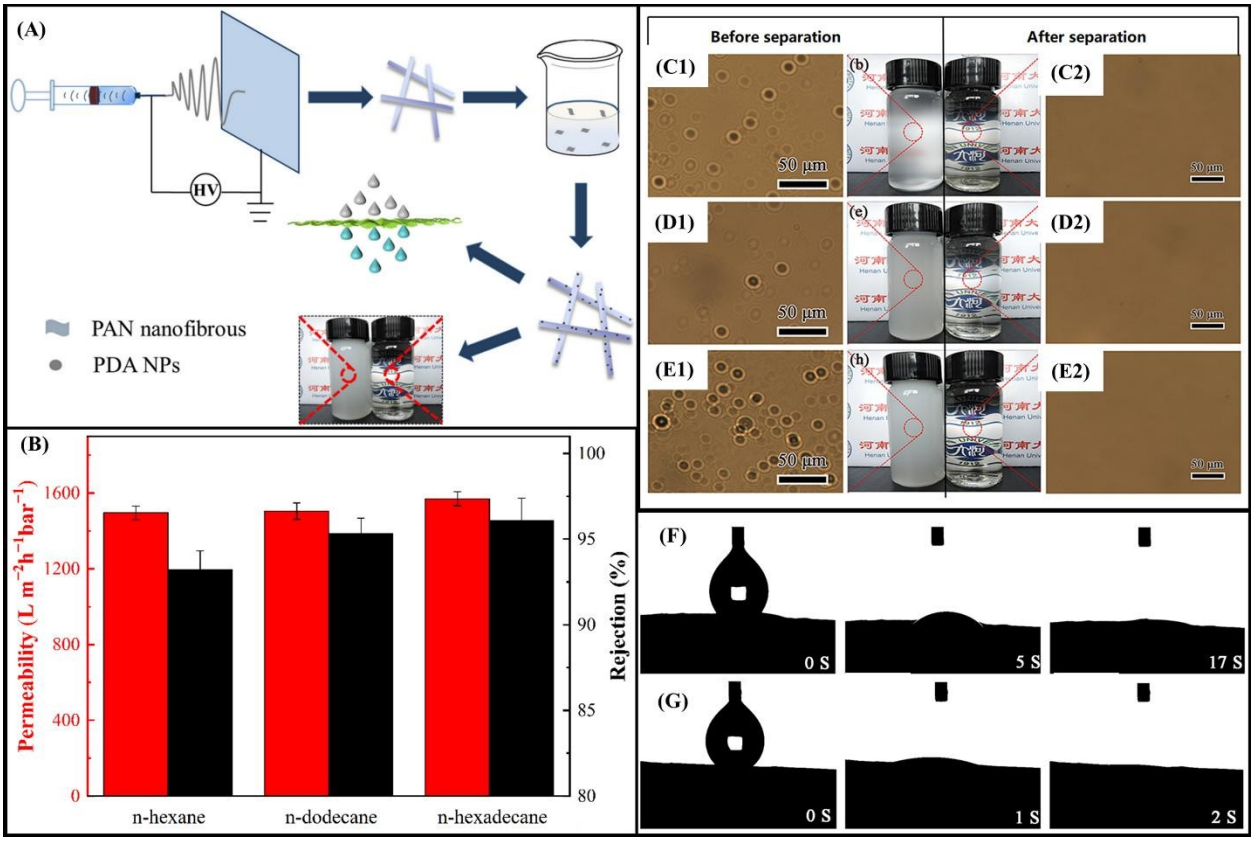

Figure 12. Preparation process (A); permeability and separation efficiency (B); optical micrograph of n-hexane/water (C1), dodecane/water (D1), hexadecane/water (E1) emulsions and their separations (C2, D2, and E2 respectively; and spreading time of water drop on pristine PAN (F) and PAN@PDA (G), Reproduced with permission from Ref. (111).

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As shown in **Fig.12A**, EFs were first prepared using a 10% solution of PAN in DMF and subsequently decorated with PDA NPs by immersing in a Tris-HCl buffer solution of dopamine at pH 8.5 for 7 hours. The permeation flux and the separation efficiency respectively ranged from 1496 $L.m^{-2}.h^{-1}.bar^{-1}$ and 93.2 % for n-hexane/water emulsion to 1570 $L.m^{-2}.h^{-1}.bar^{-1}$ and 96.1 % for hexadecane/water emulsion **(Fig.12B).** The optical micrographs showing the emulsion before and after separation using the PAN@PDA EF for n-hexane (C1/C2), dodecane (D1/D2), and hexadecane (E1/E2) emulsions are shown in **Fig.12C-E**. **Fig.12F-G** depicts the permeation rate for a drop of water on a pristine PAN fiber and a PAN@PDA fiber, respectively.

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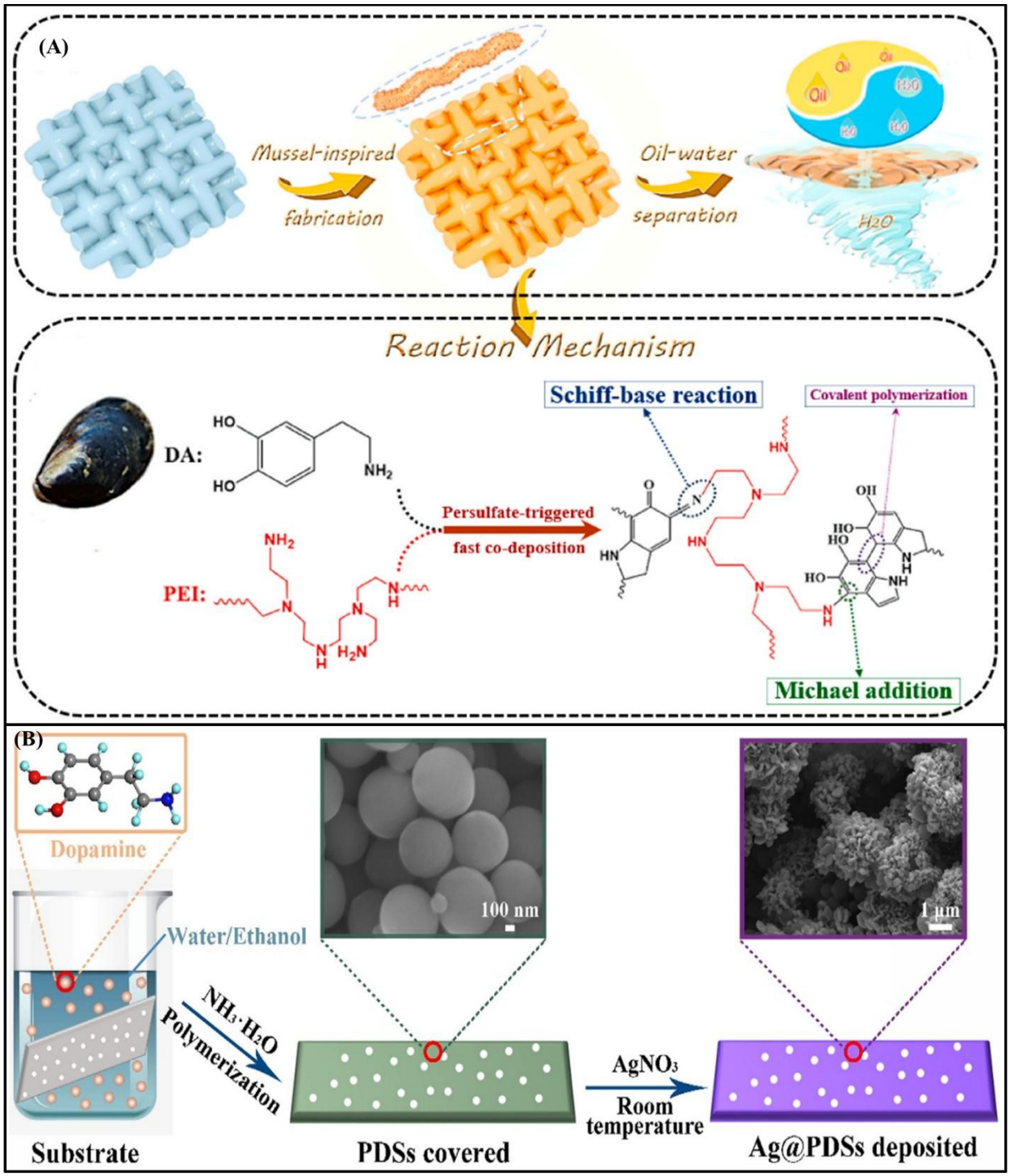

Figure 13 Schematic illustration (A) Mussel-inspired coating for design of superhydrophilic non-woven fabric membranes and its proposed reaction mechanism of fast codeposition of dopamine (DA) and PEI triggered by persulfate via Michael addition and Schiff-base reaction, Reproduced with permission from Ref.(113); (B) Synthesis of Ag@PDSs deposited membrane, Reproduced with permission from Ref. (114)

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Li and colleagues developed a SHL/UWSOB surface on nonwoven polyester fabric by deposition of a polydopamine-polyethyleneimine (PDA-PEI) complex (113), **Fig.13A**. Ammonium persulphate was introduced as a superior oxidizing agent for the co-deposition reaction. The as-prepared fabric showed a UWOCA of 165.4 ±1.1°; a SA of 2.5 ± 0.5°; an artificial oil-water separation efficiency above 99 %; and a flux that ranged from 115000 $L.m^{-2}.h^{-1}$ to 118000 $L.m^{-2}.h^{-1}$ which persisted even after 100 filtration cycles representing capacity for continuous filtration. Furthermore, the stability of the coating to maintain its separation capacity under different harsh conditions was evaluated at pH 2, 7, and 12. The UWOCA of the PDA-PEI coated textile decreased by only 1° under these harsh conditions.

Liu and colleagues reported the fabrication of hierarchical structures composed of silver nanoclusters on polydopamine spheres coated on a nonwoven nylon textile *via* a chemical wet reduction method (114), Fig.13B. The UWOCA was above 150° with a diesel-water emulsion separation efficiency above 98 % that persisted after 12 filtration cycles. The modified SHL/UWSOB membrane equally exhibited antibacterial activity against *E. coli* and demonstrated the catalytic reduction of methylene blue in the oil/water emulsion.

Huang and colleagues prepared a strong, electrically conductive, and stretchable SHL/UWSOB from electrospun polyurethane (PU) by first coating acidified carbon nanotubes on the electrospun PU (112). Subsequently, PDA@PU/ACNTs were prepared by coating PDA unto the PU/ACNT composite as shown in **Fig.14A** The water contact angle of PDA@PU/ACNTs rapidly decreased from 62 ° to 0 ° in less than 30 s with UWOCA above 150 °. The flux ranged from 4108 $L.m^{-2}.h^{-1}$ to 7240 $L.m^{-2}.h^{-1}$ for heptane

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and cyclohexane in water emulsions respectively. The separation efficiency remained above 99.7 % even after 10 cyclic separations though the flux decreased to 3608 $L.m^{-2}.h^{-1}$ (112). Zhang and colleagues exploited the synergy between an electrospun poly (arylene ether nitrile) (PEN) and polydopamine-coated hexagonal boron nitride (h-BN) nanosheets to fabricate a thermo-stable, corrosion-resistant composite SHL/UWSOB double-layered membrane (115). They found that the superhydrophilicity represented by the WCA was associated with the h-BN-PDA content and decreased from 122 ° to 0 ° as the h-BN-PDA content increased to 0.75mg. The UWOCA for different oils evaluated were below 150° and hence below the ideal value for underwater superoleophobicity. The separation flux for hexane-water emulsion was negatively affected by the h-BN-PDA content decreasing from 1894.55 $L.m^{-2}.h^{-1}$ to 1274.51 $L.m^{-2}.h^{-1}$ as the content increased from 0 mg to 0.75mg though the separation efficiency at this point was still above 99 %. Likewise, the flux of the composite membrane decreased to 588 $L.m^{-2}.h^{-1}$ after 10 filtration cycles for the emulsion but remained above 8400 $L.m^{-2}.h^{-1}$ for the oil-water. Overall, the separation efficiency remained above 99 % through the 10 cycles. The resistance of the composite membrane to elevated temperatures in the oven was confirmed by the unchanged flux and separation efficiency of the membrane while it showed a decrease in wettability when exposed to additional corrosive solutions at elevated temperature (65°C) (116).

Wang et al., (117) developed the SHL/UWSOB composite membrane based on micro cellulose ester through in-situ process for synthesizing nano materials and vacuum filtration for membranes **(Fig. 14B)**. Hydrophilicity was achieved by the addition of SiO_2 nano materials on hydroxyapatite (HAP) with the synergistic effect of polydopamine. The

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prepared composite membrane of HAP/SiO2/PDA shows the high-water permeability flux and separation efficiency of 90.90% over a wide range of light to heavy oils. The obtained UWOCA is in between149.75 ± 80.42° to 155.265 ± 1.902° for all the prepared membranes and the WCA was decreased from 27.63 ± 0.997° to 11.07 ± 0.169° due to the influence of TEOS which exhibits the wettability attributes of the composite membranes. Mechanical durability was assessed by the scratch test through knife and finger which exhibits the UWOCA of >150° and a separation efficiency of >98%. Furthermore, it shows the UWOCA of >140° after being immersed separately in corrosive solutions of acidic/ alkali and salt for 3 days which reveals the endurance of underwater super-oleophobic property.

Cui and colleagues (118) fabricated the non-toxic, low cost and environmentally friendly composite membrane based on bacterial cellulose which can decay organic pollutants under ultraviolet light. They employed PDA as an anchoring substance for TiO_2 nano particles on the surface and cross-linked with sodium alginate by vacuum assisted filtration and freeze-drying technique **(Fig. 14C)**. The prepared membrane (bacterial cellulose/PDA/TiO_2) shows the separation efficiency of > 99% for low and heavy viscous oil-water mixtures with the WCA of 0° and UWOCA of 156.5°. It shows the effective attainment of superhydrophilic and under water superoleophobic property due to the addition of TiO_2 and sodium alginate. As the addition of sodium alginate assist in making the membrane with porous structure which increase the water flux of the membrane. However, after repeated separation of oil-water mixture, the WCA was dropped to 125.6° and it was regenerated to 154° with the aid of UV radiation. Additionally, antifouling and

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extended service life of the membrane was attained by the addition TiO_2 which serve as a catalyst in decomposing the organic pollutants under UV radiation.

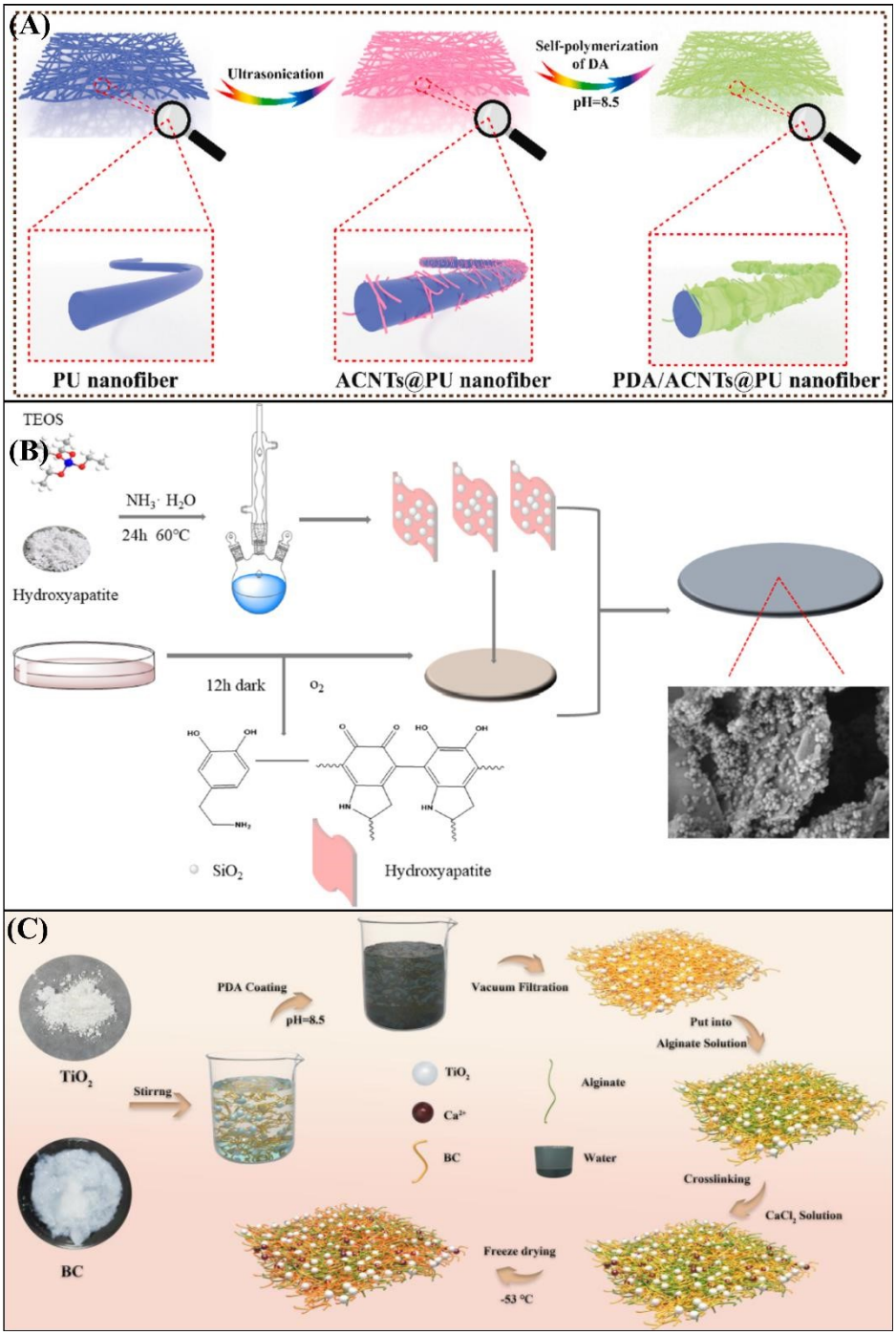

Figure 14 Schematic illustration for preparation of composite membrane (A) PDA@PU/ACNTs, Reproduced with permission from Ref. (112). (B) HAP/SiO_2/PDA, Reproduced with permission from Ref. (117). (C) bacterial cellulose/PDA/TiO_2, Reproduced with permission from Ref. (118)

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5. Emulsion separation:

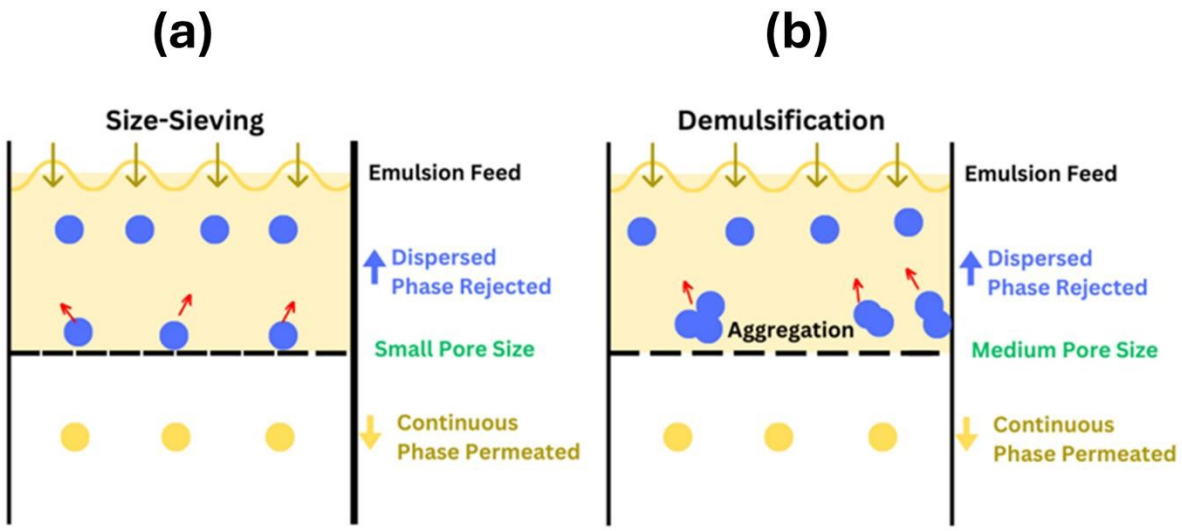

Figure 15 Schematic illustration for the separation mechanisms of emulsified oil/water mixtures (a) size-sieving mechanism, and (b) demulsification mechanism, Reproduced with permission from Ref. (34)

As illustrated in **Fig. 15**, emulsions can be separated through two main mechanisms: size-sieving or demulsification (119,120). In size-sieving, the pore size of the material must be equal to or smaller than the emulsified droplets, (**Fig. 15a)**. However, smaller pores can hinder permeation flux, resulting in lower separation efficiency compared to membranes used for immiscible oil/water separation. On the other hand, membranes designed for demulsification typically have moderate pore sizes, allowing for higher permeation flux and improved separation performance (121,122), (**Fig. 15b)**. In principle, water-in-oil (w/o) emulsions are separated using superhydrophobic membranes, whereas oil-in-water (o/w) emulsions require superhydrophilic and underwater superoleophobic membranes (34). The separation of emulsified oil/water mixtures is still a challenging industrial concern due to the stability of the surfactant-stabilized w/o and/or o/w emulsions as well as their tiny droplet size (< 2 μm) (123,124). Therefore, advanced ultrafiltration (UF) and microfiltration (MF) membranes have been effectively used to separate various types of

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emulsified oil/water mixtures through the size-sieving mechanism (125,126). In contrast, superwetting textiles with macroporous structure (i.e. pore size of hundreds of microns) have been utilized for the separation of emulsified oil/water mixtures through demulsification pathway (26,127–130).

There are limited reports that have utilized polydopamine-coated textiles for the separation of emulsified oil/water mixtures. Li et al. developed a switchable cotton fabric for oil-water separation and water purification (131). To create a rough micro-nano scale surface with high surface energy—allowing selective wetting and repelling of immiscible media—they incorporated polydopamine and $NiFe_2O_4$ nanoparticles. Surface morphology and compositional analysis confirmed a uniform $NiFe_2O_4$ coating on the cotton fabric. The modified fabric exhibited a water contact angle (WCA) of 145° under oil and an oil contact angle (OCA) of 153° under water, demonstrating its super-amphiphilic nature in air and lyophobic properties. When mounted between two vertical glass tubes for emulsion separation, the fabric effectively separated both oil-in-water and water-in-oil emulsions. Optical microscopy confirmed the absence of internal-phase droplets in the collected filtrates, achieving a separation flux of approximately 300 L m^{-2} h^{-1} by gravity with a rejection ratio of about 99%. Similar results were observed for other surfactant-stabilized emulsions, including SDS-stabilized hexadecane-in-water, diesel-in-water, and Span 80-stabilized water-in-hexane and water-in-chloroform. Cyclic performance tests using SDS-stabilized toluene-in-water and water-in-dichloromethane emulsions showed that the fabric maintained stable separation efficiency even after washing with water or dichloromethane. Although oil/water flux gradually decreased over time due to cake formation on the surface, the fabric exhibited antifouling properties, ensuring sustained

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separation performance. Additionally, its porous structure enabled the adsorption of methylene blue, highlighting its potential for water treatment applications.

Gan et al. utilized polydopamine as a binder and reducing agent for the in-situ deposition of silver (Ag) nanoparticles onto a textile surface (132). This was followed by an immersion step in PFOA-Na ethanol solution to achieve a slippery oil-repellent state, creating a superhydrophobic/superhydrophilic (SHB/SHL) Ag/polydopamine-coated textile (Ag/PDA@textile). The incorporation of Ag nanoparticles endowed the textile with photocatalytic and antibacterial properties, enabling its application for the dye's removal from water and the inhibition of bacterial growth. The modified textile exhibited superhydrophilic and superoleophobic characteristics in air, with an oil contact angle (OCA) greater than 150° and a sliding angle (SA) of less than 9°. When wetted with water, it displayed an OCA of 50.1°, allowing oil to flow smoothly across its water-wetted surface due to the water lubricant's fluidity. Notably, even after being submerged in oil for a day, the water-wetted textile retained its oil-repelling properties, demonstrating excellent anti-oil-fouling capabilities. Oil-water separation tests revealed a high-water permeation flux of 1502 L $m^{-2}h^{-1}bar^{-1}$ and a separation efficiency exceeding 99% for toluene-hot water mixtures at 80.5 °C. Comparative microscopic images before and after emulsion separation (Tween 80-stabilized toluene-in-water) further highlighted its effectiveness in separating o/w emulsions. Additionally, Ag/PDA@textile demonstrated the ability to separate immiscible oil/oil mixtures with varying polarities, showing a strong affinity for high-polarity oils while repelling low-polarity oils.

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6. Conclusions and perspectives:

Polydopamine (PDA) coatings are well known for their universal, material-independent application on various solid substrates. They can effectively cover target material surfaces, even in cases of low intermolecular interactions. Over the past decade, PDA has been widely used to fabricate superwetting textiles and fabrics through simple, sustainable, and versatile synthetic pathways. The use of PDA coatings offers several advantages in fabricating superwetting textiles, including inherent hydrophilicity, enhanced adhesion and roughness of the coated surfaces, potential for in-situ post-functionalization (e.g., incorporating nanoparticles using PDA as a reducing agent), good chemical durability, intrinsic antibacterial properties, and biodegradability. Additionally, the strong adhesion properties of PDA coatings facilitate the incorporation of various low-surface-energy materials (e.g., alkyl thiols, alkylamines, and fatty acids) and metal nanoparticles (e.g., silver NPs) through ionic and/or covalent interactions. This improved adhesion contributes to the overall durability and recyclability of superwetting textiles. Furthermore, PDA coatings support the development of smart, multifunctional superwetting textiles with features such as self-healing and UV resistance.

Despite these advantages, conventional PDA coating strategies face challenges such as time consumption and poor controllability. These issues arise from the slow kinetics of DA polymerization and inevitable aggregation, which limit practical manufacturing processes and hinder precise tuning of surface characteristics (133). As shown in **Tables 1 and 2**, the first challenge is related to the fabrication process which is time-consuming. Most reported studies employ in-situ auto-oxidative polymerization using a Tris buffer (pH = 8.5/O_2), requiring long deposition periods (≈24 hours) for PDA coating. The second

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challenge is related to the development of efficient strategies for restricting DA polymerization exclusively to the material's surface, minimizing waste caused by excessive self-polymerization in the solution. The first and the second challenges can be addressed by employing a direct and rapid dip-coating strategy of the textiles/fabrics in pre-polymerized polydopamine dispersions (134). In this approach, nanoparticles can be added to the polydopamine dispersion, or the dispersion itself can be prepared in the presence of the desired nanoparticles. The dip-coating method also helps to reduce the excess PDA waste. However, this proposed technique may influence the final wetting behaviour of the modified textiles due to PDA's inherent hydrophilicity and adhesion properties. To optimize adhesion, the number of dip-coating cycles can be adjusted. Additionally, post-functionalization can still be performed to modify the surface energy properties of polydopamine-coated textiles compared to uncoated ones.

Another solution that enables rapid polydopamine coatings is the use of an effective oxidizing agent and/or auxiliary oxidizing agents (i.e. redox system) that accelerate the rate of oxidative polymerization. As can be inferred from **Tables 1 and 2**, only a limited number of studies have utilized the rapid oxidation procedures during the fabrication process. Hence, this area is an active research area which has not yet been fully explored to accelerate the polydopamine coating and shorten the overall fabrication time. In this respect, there are numerous redox systems that have been reported for the rapid polymerization of polydopamine including Cu^{2+}/H_2O_2 (135,136), Fe^{3+}/H_2O_2 (137), Fe^{2+} (138), ozone (O_3) (139), and sodium periodate ($NaIO_4$) (140). PDA exhibits good anticorrosion resistance in neutral and acidic environments but may partially degrade under alkaline conditions (pH > 12). In practical applications, pretreating water effluent to

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achieve a neutral pH can help extend the lifespan of PDA-coated membranes by avoiding extreme pH conditions. Notably, even state-of-the-art biodegradable superwetting crosslinked membranes degrade easily in alkaline conditions (1 M NaOH) (126). Nevertheless, applying a secondary protective coating (e.g., PDMS) can enhance the chemical durability of PDA-coated textiles, albeit at the expense of increased fabrication cost and time. Additionally, the mechanical durability and relatively high cost of PDA coatings must be carefully considered in future studies (141).

From the perspective of oil/water separation, future research should focus on developing faster, cost-effective, one-step fabrication methods and scaling up the production of PDA-coated superwetting textiles. The use of polydopamine nanoparticles (e.g. synthesized in ethanol/water mixture) is expected to improve the superwetting performance of the coated textiles due to the improved surface roughness (67). Future studies should also emphasize the separation of both stratified and emulsified oil/water mixtures, including viscous crude oils and water-in-oil (w/o) and oil-in-water (o/w) emulsion. Another direction that has emerged is the application of polydopamine-coated materials for water purification and decontamination (e.g. removal of dyes, toxic metal ions, etc.) (142–147). Hence, dopamine-coated fabrics could be utilized for both oil/water separation and decontamination due to the potential adsorption of polydopamine to diverse arrays of toxic organic and inorganic contaminants through different ionic interactions. This can be achieved with/without the presence of photocatalysts immobilized on the surface of the coated textiles (148). Besides the rapid degradation of the organic contaminants, the imbedded photocatalysts are expected to improve the self-cleaning and antifouling performance of the polydopamine-coated textiles.

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Finding:

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Conflicts of interest:

The authors declare that they have no conflict of interest.

Author Contribution:

Conceptualization, Nedal Abu-Thabit; **Visualization**, Abdul Kalam Azad, Mahmoud Abu Elella, and Nedal Abu-Thabit; **Writing—original draft**, Abdul Kalam Azad, Mahmoud Abu Elella, and Nedal Abu-Thabit; **Writing—review & editing**, Nedal Abu-Thabit.

Table 1 PDA-coated superhydrophobic/superoleophilic textiles for oil/water separation

Type of Fabric/ Textile	Modification Materials		Optimum Coating Parameters					Highest Oil Permeation Flux (L. m^{-2}. h^{-1})	Separation efficiency (%) (Recyclability)	Durability Features	Ref.
	Surface Roughness	Surface Energy	DA Conc. (mg/mL)	Buffer [1]	Time (h)	Temp. (ºC)	Other Materials				
PET	PDA NPs	PFDTS	2	T/P	20	35	-	-	-	*Resistance to abrasion and UV radiation	(92)
Blended fabric (35% cotton, 65% polyester)	PDA NPs	ODA	1	T-HCl	24	60	Folic acid as SDA	-	97.1	*Resistance to wear and NaCl solution(3.5Wt.%).	(76)
Cotton fabric, polyester gloves	PDA microcapsules	ODA or ODT	0.5	T-HCl	12	R.T.	-	-	-	*Mechanically induced self-healing superhydrophobicity *Resistance to stretching, compression, friction and mechanical washing.	(77)
Cotton	PDA NPs	-	10mM	T-HCl	24	R.T.	$CuSO_4$	-	-	*Durable antibacterial and sticky superhydrophobic property.	(149)
Cotton fabric	PDA NPs	-	0.02g	TEOS-HCl/HMDS	24	R.T.	SiO_2	4000	99.9	*Excellent mechanical stability *Resistance to abrasion and UV radiation. *Resistance to high temperature boiling water and organic solvent.	(78)
Cotton, polyester, wool fabric	PDA NPs	HDTMS	0.1g/10 mL	-	50	12	-	-	-	*Self-healing ability *Resistance to abrasion and washing (20 cycles) *Coated fabrics achieved 80% transmittance	(150)

Cotton	PDA NPs/PEI	-	60 mg/ml	T-HCl	35	14	Borax, TA	-	>98	*Resistance to high temperature degradation *Resistance abrasion and washing. *Flame retardant and self-extinguishing ability.	(79)
Tetra-copolyester and polypropylene	PDA NPs	PFDT	2	T-HCl	25	12	SiO_2, Ag NO_3	-	>90	*Excellent stability and recyclability *Employed in oil/water separation, smart oil engineering, microfluidic devices,	(93)
Cotton fabric	PDA NPs	ODA	2	Sodium perborate (PBS)	90	50	$FeCl_3$	-	>96	*Resistance to harsh environment *Outstanding recyclable and self-cleaning performance	(84)
Poly lactic acid non-woven fabric	PDA NPs	(PVDF-HFP)	2	T-HCl	20	R.T.	$FeCl_2.4H_2O$, $FeCl_3.6H_2O$	-	99.5	*Excellent recyclability and higher absorption capacity *Higher surface stability and self-cleaning ability *Resistance to abrasion *Ability to separate floated oil due to its magnetic property	(94)
Blended fabric (35% cotton, 65% polyester)	PDA NPs	ODT	8	-	4	25	Sodium periodate	4500	>97	*Resistance to UV irradiation, hot water and acetone *Resistance to abrasion and fouling	(81)
Blended fabric (50% nylon56 and 50% cotton)	PDA NPs	DMPA, POSS	0.3g	T&T-HCl	24	45	MPMDS	-	-	*Excellent mechanical and chemical stability, *Outstanding durability and self-cleaning ability	(80)
PET, cotton, PP and blended fabric of cotton (35%) and PET (65%)	PDA NPs	1H,1H,2H,2H-Perfluorodecanethiol	2.0	T-HCl	2	-	$AgNO_3$	-	-	*Excellent oil-water separation and self-cleaning ability.	(85)

Pristine fabric	PDA NPs	PDMS	80 mg/40 mL	Alkaline	24	R.T.	SiO_2	-	95	*Anti fouling and self-cleaning ability *Resistance to corrosive solution, organic solvent, UV irradiation and washing.	(96)
Other materials	PDA NPs	OTEOS	0.4 g	T-HCl	10	R.T.	SiO_2	-	-	*Excellent durability and mechanical stability *Resistance to harsh environment and UV irradiation	(95)
Cotton	PDA NPs	n-DDT	2.0	25% NH_4OH	24	60	$AgNO_3$, $CuSo_4$	72,172.9 (n-hexane) 14,460.3 (diesel)	>99	*High oil absorption capacity *Stable surface after 100 separation cycles	(83)
Cotton	PDA NPs	DDT	2.0	T-HCl	24	R.T.	$FeCl_3.6H_2O$	-	>98	*Improved thermal stability and excellent photocatalytic activity	(82)
Cotton	PDA NPs	SA	2.0	T	4	25	$CuSO_4.5H_2O$	-	-	*Excellent UV protection and solar-induced rapid self-healing properties *Excellent mechanical and chemical stability	(86)
Polyvinylidene fluoride (PVDF)	PDA NPs	$HS(CH_2)_{11}CH_3$ /$HS(CH_2)_{10}COOH$ (Thiol)	2.0	T-HCl	24	40	$AgNO_3$	11000	99.2% (light oil/water mixture) 96.5% (heavy oil/water mixture)	*Excellent durability and reusability *Rapid stimulus responsibility, superior wettability switching ability, comparable permeability and high separation efficiency *Energy-saving separation of multiphase oil/water mixtures and emulsions.	(97)
Cotton	PDA NPs	PDMS	100 mg/50 mL	T	60	12	$CuSO_4.5H_2O$, ZIF-8 nanomaterials	-	-	*Excellent photothermal antibacterial activity against E. coli and S. aureus under near-infrared (NIR) light irradiation.	(87)

										*Durability and self-cleaning ability	
Cotton	PDA NPs	Hexa decyl tri methoxy silane	2	T-HCl	12hours stirring/24 freeze drying	R.T.	FeCl3.6H2O	8561.02	99.5	*Excellent mechanical and chemical durability *suitable for large scale oil-water separation *Significant photocatalytic properties.	(88)
Woolen felt	PDA NPs	DDT	0.01	T-HCl	14	R.T.	$AgNO_3$, TiO_2	-	-	*Low cost, bio-degradable and self-extinguishing properties	(98)
PP fibre	PDA NPs	3-Aminopropyltriethoxysilane (APTES)	200mg/L	T-HCl	12 hrs reaction/5 hrs immersion	R.T.	MXene nanosheets, Polypyrrole (PPy), pH responsive polymer (PDM)	-	-	*Excellent pH responsive membrane driven by sunlight *Excellent stability, corrosion resistance and recyclability.	(99)
Cotton	PDA NPs	PDMS	2	-	12	R.T.	MWCNT	16.21 m^3 m^{-2} h^{-1}	99	*Resistance to acid, alkali and abrasion test *Self-healing ability on the surface	(89)
Nylon 56	PDA NPs	Trimethylamin omethane, Tetraallyl silane	400mg/200mL	-	0.5	60	3-mercaptopropionic acid, methyl siloxane-dimethyl siloxane copolymer (mercapto)	-	>98	*Excellent self-cleaning and anti-fouling properties *Strong candidate for environmental and industrial applications.	(100)

Abbreviations: T = tris-buffer; P = phosphate buffer; R.T.= Room temperature; PDA NPs= Polydopamine nanoparticles; SDA = structure directing agent; SiO_2 = Silicon dioxide; NH_4OH = Ammonium hydroxide; $CuSO_4$= Copper sulphate; $AgNO_3$ = Silver nitrate; $FeCl_3.6H_2O$ = Ferric chloride.

Table 2 PDA-coated superhydrophilic/underwater superoleophobic textiles for oil/water separation

Type of Fabric/ Textile	Surface Roughness	Optimum Coating Parameters					Water Permeation Flux (L. m^{-2}. h^{-1})	Pressure (MPa)	Separation efficiency (%) (Recyclability)	Durability Features	Ref.
		DA Conc. (mg/mL)	Buffer[1]	Time (h)	Temp. (ºC)	Other Materials					
Cotton	PDA NPs	4.0	T-HCl	24	R.T.	Chitosan	15,100 – 30,000	-	>99	*Outstanding chemical and mechanical resistance	(151)
Polyester fabric	PDA NPs and PEI	2.0	T	7	-	Sodium periodate ($NaIO_4$), Hydrogen peroxide (H_2O_2) and Copper sulfate ($CuSO_4$)	>1,15,000	Gravity	>99.2	*Resistance to extreme environment and long term durability	(113)
Cotton	PDA NPs and TiO_2	120	T-HCl	-	-	(3-aminopropyl) triethoxysilane (KH550, 99%), copper sulfate pentahydrate ($CuSO_4 \cdot 5H_2O$), Polyethylene glycol (PEG)	32,000	Gravity	99.9	*Resistance to salt solution, acids and abrasion	(103)
Poly(arylene ether nitrile) (PEN) nanofibrous mat	PDA NPs	-	T-HCl	4	R.T.	Hexagonal boron nitride (h-BN), $CuSO_4$, H_2O_2	588.1 (Emulsion) 8400 (oil/water mixture)	0.04	>99	*Resistance to harsh environment and antifouling ability	(116)
Thermoplastic polyurethane	PDA NPs	-	T	-	-	Multi-walled carbon nanotubes	4195 – 7240	0.05	99.9	*Enhanced mechanical property, thermal stability and electrical conductivity *Excellent anti oil adhesion properties	(152)

Poly acrylonitrile fibre (PAN)	PDA Nano spheres	40mg	T-HCl	7	25	-	1570 L m^{-2} h^{-1} bar^{-1}	-	96.1	*Good separation efficiency and durability	(153)
Pristine material	PDA Nano spheres	0.5g/10 mL	-	48	R.T.	$AgNO_3$	-	-	99.3	*Outstanding cyclic performance and antibacterial activity	(114)
Polyvinylidene fluoride (PVDF)	PDA NPs	2.0	T-HCl	24	40	$AgNO_3$ and thiols	2500	Gravity	>99	*Superwettability and superior pH responsibility	(97)
Micro cellulose ester membrane (MCEM)	PDA NPs	0.2g/100 mL	T-HCl	12	R.T.	HAP, SiO_2, Tetraethoxysilane(TEOS)	2000 – 7000	-	90.90	*Improved tesnsile strength and mechanical stability *Resistance to acidic/alkali/salt solutions *Separation flux of light oil 3000 L/(m^2·h) *Oil discharge rate of heavy oil can reach 98.95%	(117)
Bacterial cellulose (BC)	PDA NPs	0.46g/150mL	T	24	R.T.	TiO_2, Sodium alginate	7428 – 8774 (heavy oil) 9597 – 10,000 (light oil)	-	>99	*Low cost, non-toxic and eco friendly *Excellent anti-pollution and photo catalytic property	(118)
Cotton	PDA NPs	5mg/mL	T-HCl	1hr sonication/ 4hrs immersion/24	R.T.	Carbon nanotubes	10834	Gravity	>99.8	*Outstanding cationic dye adsorption capacity *Excellent stability and durability under harsh condition	(109)
Cotton	PDA NPs	4	T-HCl	24	R.T.	SiO_2, TEOS	8100 – 34000 (various oils	Gravity	>99	*Ultra-low adhesion towards crude oil *Continuous separation of crude oil/water (15 times) and toluene/water (100 times)	(110)

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