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Design and construction of CuS functionalized superhydrophobic cellulose paper with effective oil/water separation, and photothermally boosted antimicrobial and anti-/deicing performances

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Abstract:

This work involved the rational design and successful fabrication of a superhydrophobic cellulose paper functionalized with copper sulfide (CuS), which combined effective continuous separation of oil/water mixtures, photothermally enhanced antibacterial activity, anti-icing, and deicing properties. Dopamine was used to create a polydopamine (PDA) coating on cellulose fibers via spontaneous oxidative polymerization in order to address the crucial problem of weak interfacial contact between CuS and cellulose paper substrates. The PDA-modified paper surface was then in situ coated with CuS particles. The resulting functional paper possessed water contact angle (WCA) as high as 156.4° and a virtually zero oil contact angle (OCA), revealing its extraordinary superhydrophobicity and superoleophilicity. In addition to favorable wettability, the CuS-decorated superhydrophobic cellulose paper exhibited outstanding chemical stability, reliable mechanical durability, excellent antifouling and self-cleaning capabilities, as well as high-efficiency continuous separation for diverse oil/water mixtures. These desirable properties render the composite paper highly promising

for realistic industrial applications. Given that CuS are well-known for their excellent photothermal conversion capability, the photothermal performance of the modified paper was evaluated. Specifically, the maximum surface temperature of the modified paper could reach up to approximately 78.6 °C within 60 s under one standard solar irradiation (1.0 kW m⁻²). More importantly, it maintained a stable surface temperature of about 77.8 °C even after 10 cycles of light on-off tests, indicating excellent photothermal stability and recyclability. Leveraging its outstanding photothermal performance, the modified paper further exhibited remarkable photothermal antibacterial efficacy, which could efficiently suppress the growth of both Gram-positive and Gram-negative bacteria. In addition to the antibacterial property, the excellent photothermal conversion efficiency also endowed the modified paper with integrated anti-icing and deicing functionalities. Consequently, the unique photothermal-driven feature provides a novel and promising pathway for superhydrophobic paper to realize efficient absorption of spilled oil even under harsh low-temperature conditions upon solar irradiation. The findings validated that the as-fabricated CuS-functionalized superhydrophobic cellulose paper possessed versatile and superior functionalities, including efficient oil/water separation, antifouling, self-cleaning, antibacterial activity, and anti-icing/deicing properties. The versatile designed paper holds enormous potential for widespread and reliable application in actual industrial applications.

1. Introduction

Frequent oil spills and uncontrolled discharges of oily wastewater pose escalating environmental threats amidst rapid industrialization, endangering ecosystem integrity and human health through persistent contamination ^[1–5]. High-profile disasters, exemplified by the 2010 Deepwater Horizon spill and the 2011 Bohai Bay incident, released vast quantities of crude oil into coastal waters,

inflicting catastrophic and long-term damage on marine ecosystems and biodiversity. These events underscore the critical need for eco-efficient oily wastewater treatment technologies. Conventional approaches—such as in situ combustion, bioremediation, skimming, air flotation, and chemical dispersion, remain widely deployed^[6-8]. However, inherent drawbacks like operational complexity, high energy demands, and limited scalability hinder their practical effectiveness for large-scale applications^[9, 10]. Consequently, research is urgently directed toward innovative strategies utilizing cost-effective, scalable, and high-performance materials. Among promising solutions, engineered superhydrophobic surfaces (SHSs) have garnered significant interest due to their unique physicochemical properties^[11, 12]. Inspired by natural models like the lotus leaf and water striders, SHSs exhibit exceptional water repellency, defined by a water contact angle (WCA) exceeding 150° and a contact angle hysteresis (CAH) below 10°. This behavior stems from the synergistic interplay between low surface energy chemistries and multiscale micro/nanostructures. Such surfaces offer substantial potential for environmental remediation, particularly in applications demanding self-cleaning, anti-biofouling, corrosion resistance, anti-icing, and selective oil/water separation.

In oil/water separation research, two distinct classes of functional materials have been rationally devised based on wettability principles. The first comprises surfaces exhibiting superhydrophilicity and underwater superoleophobicity. These materials enable selective water permeation: an aqueous layer stably bound to the substrate permits water passage while effectively blocking oil penetration. This mechanism is commonly termed “aqueous phase extraction” technology. The second category of materials exhibits superhydrophobic and superoleophilic properties, which can efficiently reject water molecules and drive the absorption and permeation of oil via capillary action, thereby enabling the recovery and transportation of oil products. This mechanism is commonly referred to “oil phase

harvesting” technology^[13, 14]. Despite their considerable promise for large-scale oil/water separation applications, the large-scale application of these two types of materials remains constrained by limitations including insufficient mechanical stability and high fabrication costs. In the process of fabricating SHS, numerous researchers have intentionally fabricated or adhered hydrophobic layers on different substrate materials to achieve excellent oil/water separation performance. The commonly employed substrates include filter paper, fabrics, metallic foils, sponges, polymeric membranes^[15–19]. Among these candidates, cellulose-based paper stands out as the most eco-friendly alternative, owing to its abundance as a natural resource. Cellulose is considered to be one of the most attractive renewable resources towards fabricating advanced functional materials due to its ubiquitous presence in nature, very low density, and high biodegradability^[20, 21]. However, the inherent abundance of surface hydroxyl groups in cellulose has significantly impeded its practical implementation in the development of renewable superhydrophobic paper, thereby facilitating tremendous efforts devoted to functionalizing the superhydrophobic properties of cellulose paper to address this major issue. Drawing inspiration from the classical lotus effect, the wetting performance of engineered surfaces is essentially dominated by the synergistic contribution of topographical architecture and chemical composition. It is widely acknowledged that a robust superhydrophobic surface can be rationally achieved by simultaneously introducing micro/nanoscale hierarchical roughness and decorating low-surface-energy compositions. These two factors are recognized as the indispensable prerequisites for constructing superhydrophobic surfaces with exceptional water-repellent characteristics^[22].

For the practical realization of such superhydrophobicity, particularly for paper-based substrates, various nanomaterials have been strategically selected to decorate their microstructured fibers,

including titanium dioxide (TiO_2) nanoparticles, polytetrafluoroethylene nanoparticles, hydroxyapatite nanowires, Fe_3O_4 , SiO_2 nanoparticles, Ag nanoparticles, metal–organic framework (MOF) nanoparticles, carbon nanotubes, SiC, and ZnO nanoparticles [23, 24]. Recently, strategies that integrate photothermal properties into hydrophobic surfaces have been widely reported, which considerably enhance their overall performance and thus extended the scope of their application in different industries. Various strategies have been reported for the integration of different types of photothermal components into hydrophobic surfaces by utilizing their photothermal characteristics. During the fabrication process, these photothermal compositions can be embedded within the material matrix. Representative photothermal absorbers include carbon-based nanomaterials (e.g., graphene, carbon nanotubes) [25, 26], metal oxides such as Fe_3O_4 and CuO [27], polymers including polydopamine and polypyrrole [28, 29], MXenes [30], and metal sulfides (e.g., copper sulfide, molybdenum disulfide) [31, 32].

In particular, copper sulfide (CuS), a typical p-type semiconductor, has shown great promise for photothermal conversion applications, owing to its excellent optical absorption capacity, fast photoresponse originating from d–d* transitions, localized surface plasmon resonance, and outstanding chemical and thermal stability. Furthermore, CuS possesses inherent advantages of negligible toxicity, low cost (due to its composition of earth-abundant elements), simple synthesis process, and strong chemical/thermal stability, thus being extensively investigated and applied in the fields of biomedicine and energy storage. Up to now, CuS has been extensively utilized in solar cells, photocatalysis, and photothermal therapy [33–35]. However, pure CuS particles tend to peel off from the substrates, which impairs their photothermal performance. Therefore, it is necessary to design durable superhydrophobic materials for such environmental remediation scenarios to ensure

their high performance during service life and practical applicability.

To address the critical issue of CuS peeling and enhance material durability, polydopamine (PDA), inspired by mussel adhesive proteins, was introduced to reinforce the adhesion between particle and cellulosic microfibers, thereby boosting the durability and recyclability of superhydrophobic materials in oil/water mixture separation. Furthermore, PDA can also harvest light and generate heat, which remarkably enhances the photothermal performance of the nanoparticles [36, 37]. Consequently, PDA serves as a promising multifunctional linker to anchor CuS onto cellulose fibers, effectively mitigating the practical application limitations of CuS. Considering these aspects, we attempt to fabricate a multifunctional superhydrophobic cellulose paper featuring efficient oil/water separation and excellent photothermal performance.

Herein, CuS-functionalized superhydrophobic cellulose paper was successfully fabricated. Inspired by the natural adhesion mechanism of mussels, PDA was employed as an intermediary to establish strong interactions with CuS. Through self-polymerization of dopamine, CuS particles were firmly attached to cellulose substrates, which effectively improved the interfacial adhesion bonding between CuS and cellulose fibers. The CuS particles provided micro/nano hierarchical roughness to the cellulose paper, while octadecylamine (ODA) molecules containing long-chain hydrocarbons lowered the surface energy through providing hydrophobic groups. Therefore, the combined effects of hierarchical roughness and low surface energy endowed the paper substrates with superhydrophobicity and high photothermal performance. The resultant superhydrophobic cellulose paper exhibited high efficiency for oil/water mixture separation, and its separation performance remained nearly unchanged during repeated cycling tests. Furthermore, the modified paper demonstrated outstanding durability, maintaining superhydrophobicity even in challenging

conditions like chemical corrosion, mechanical abrasion, UV irradiation, thermal stress, and long-term usage. The exceptional superhydrophobic performance of the modified paper greatly increased its capacity for self-cleaning, thereby broadening its range of useful applications. Water rinsing effectively removes surface impurities without the use of chemical detergents or physical abrasion. More significantly, the CuS anchored on the cellulose substrate showed exceptional photothermal behavior, converting light energy into heat quickly and efficiently. The paper's unique functionality made it extremely promising for photothermal bacterial inactivation (such as water disinfection) and anti-icing/de-icing applications. It is especially valuable for treating water purification and ecological sustainability issues in low-temperature environments. The functionalized cellulose paper could be a versatile, high-performance platform for water purification and environmental remediation due to its excellent stability and the synergistic integration of superhydrophobicity and photothermal responsiveness.

2. Materials and methods

2.1 Materials

The cellulose papers (CP) were purchased from Grace Co., Ltd., which located at Zhuji, China. Copper chloride dihydrate ($\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$), thiourea ($\text{CH}_4\text{N}_2\text{S}$), dopamine (DA), tris(hydroxymethyl) aminomethane (Tris), and n-octadecylamine (ODA) were acquired from Shanghai Macklin Biochemical Co., Ltd. Meanwhile, ethylene glycol (EG), hydrochloric acid (HCl), sodium hydroxide (NaOH), sodium chloride (NaCl), Oil Red O, methylene blue (MB), methyl orange (MO), petroleum ether, n-hexane, cyclohexane, ethyl acetate, dichloromethane, chloroform, and carbon tetrachloride were provided by Sinopharm Group Co., Ltd. (China). Gasoline and lubricating oil were obtained from China Petrochemical Corporation, whereas grape juice and cola were purchased

from a local supermarket. Notably, all chemicals and materials were used as received without any additional purification procedures, which ensures the consistency and reliability of the experimental results

2.2 Construction of CuS powders

The synthesis of CuS powders was conducted as follows: 1.7 g of $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ was dissolved in 60 mL of EG and magnetically stirred in a water bath at 90 °C for 30 min. Subsequently, 3.04 g of $\text{CH}_4\text{N}_2\text{S}$ was dissolved in 50 mL of EG, and the resulting solution was mixed with the aforementioned $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ solution, followed by continuous stirring for 30 min to form a transparent homogeneous solution. This mixed solution was then transferred into a Teflon-lined autoclave, which was sealed and maintained in an oven at 170 °C for 5 h. After the reaction, the autoclave was naturally cooled to room temperature, and the resultant product was collected via centrifugation at 10,000 rpm for 10 min, followed by thorough washing (more than three times). Finally, the obtained CuS powders were dried in an oven at 90 °C for 4 h.

2.3 Construction of CP/PDA/CuS

Initially, 0.2 g of dopamine hydrochloride was fully dissolved in 100 mL of Tris-HCl buffer solution (pH = 8.5). Polydopamine (PDA) deposition onto the cellulose paper was achieved by immersing CP in the prepared dopamine hydrochloride solution for 24 h; the modified CP was then rinsed with deionized water and dried in an oven at 40 °C for 2 h. Subsequently, 0.2 g of CuS was dispersed in 100 mL of Tris-HCl buffer solution (pH = 8.5) via ultrasonic dispersion for 1 h, followed by magnetic stirring at room temperature for 12 h. The CP/PDA/CuS composite was finally obtained after rinsing with deionized water and drying at 40 °C for 2 h.

2.4 Construction of superhydrophobic CP/PDA/CuS/ODA

The superhydrophobic functionalization of CP/PDA/CuS composites was successfully realized by a facile immersion method. Specifically, the as-prepared CP/PDA/CuS was immersed into an ODA-ethanol solution for 12 h. During this immersion period, ODA molecules were gradually absorbed onto the surface of CP/PDA/CuS, initiating the formation of a hydrophobic coating. After the modification, the samples were retrieved from the ODA-ethanol solution and dried in a constant-temperature oven at 50 °C for an extended period. The drying process guaranteed the complete evaporation of ethanol and the stable immobilization of the ODA hydrophobic layer on the substrate surface. Through the above-mentioned modification procedure, superhydrophobic CP/PDA/CuS/ODA composites were successfully fabricated, and the schematic diagram of the entire modification process is presented in Figure 1.

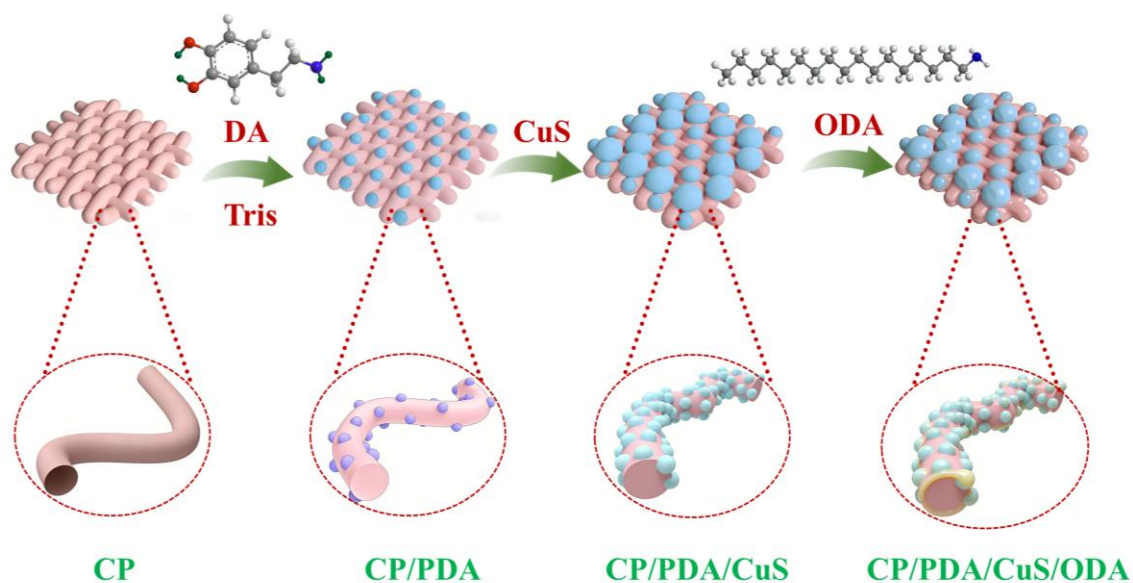


Figure 1 Schematic diagram of the entire modification process

2.5 Characterization

A scanning electron microscope (SEM, Quanta FEG 250, FEI Company, USA) was employed to characterize the morphological and microstructural features of the prepared samples. The crystal structures of the samples were determined by X-ray powder diffraction (XRD, SmartLab, Rigaku,

Japan) with Cu-K α radiation ($\lambda = 1.54 \text{ \AA}$) under optimized operating conditions: a scan rate of $10^\circ/\text{min}$, an applied current of 40 mA, and a 2θ scanning range of $5\text{--}90^\circ$. Fourier Transform Infrared Spectroscopy (FT-IR, Nicolet iS50, Thermo Scientific, USA) was used to detect the chemical composition and variations in functional groups. The surface wetting behaviors were assessed by measuring the water contact angles (WCAs) and oil contact angles (OCAs) using XG-CAMB optical contact angle instrument. A sunlight simulator (BBZM-I, Bobei Optics, China) was utilized to provide simulated sunlight for photothermal measurements, and an infrared camera (K20, Hikmicro, China) was used to capture infrared thermal images and track the temperature evolution of samples during tests.

2.6 Evaluation of selective oil removal performance and oil absorption capacity

Gasoline was used as a representative light oil and chloroform as a representative heavy oil, respectively, the selective oil removal performance of CP/PDA/CuS/ODA was assessed. Oil Red O was used to stain both oil phases in order to improve visual clarity during observation. The CP/PDA/CuS/ODA sample was typically placed at the oil-aqueous phase interface and removed from the water phase with tweezers. To clearly depict the oil absorption behavior, a digital camera was used to record the entire oil removal process and pictures were taken at various points. The oil absorption capacity of the CP/PDA/CuS/ODA was investigated by immersing the samples in various oils and organic solvents, including petroleum ether, n-hexane, cyclohexane, ethyl acetate, gasoline, lubricating oil, dichloromethane, chloroform and carbon tetrachloride. All tests were performed at room temperature with an immersion time of 5 min. After reaching full absorption saturation, the samples were removed, and excess surface liquid was gently wiped off prior to weighing. The oil absorption capacity K (g/g) was calculated according to Eq. (1):

where the mass (g) of the sample before and after absorption, respectively.

$$K = (m_1 - m_0) / m_0 \times 100\% \quad (1)$$

Where m_0 represents the initial mass of the dried sample, and m_1 represents the mass of the sample after complete saturation.

The reusability of CP/PDA/CuS/ODA was evaluated by repeated absorption–desorption cycles. After each oil absorption test, the absorbed oil and organic solvents were removed by mechanical compression. The absorption–desorption process was performed for 20 cycles to evaluate the long-term durability of the sample under cyclic usage.

2.7 Continuous gravity-driven oil/water separation

The continuous gravity-driven oil/water separation performance of CP/PDA/CuS/ODA was evaluated using a home-made separation device consisting of two connected funnels. The CP/PDA/CuS/ODA sample was fixed between the two funnels, and an oil/water mixture was poured into the upper funnel. During separation, the oil phase selectively permeated through the paper and was collected in a collection vessel, while water was retained above the sponge.

The employed oil phases included cyclohexane, petroleum ether, n-hexane, dichloromethane, chloroform and carbon tetrachloride. The separation efficiency (η) was calculated using Eq. (2):

$$\eta = (V_1 / V_0) \times 100\% \quad (2)$$

where V_0 represents the volume of the original oil before separation, V_1 represents the the volume of recovered oil after separation.

The oil permeation flux was determined according to Eq. (3):

$$Flux = V_1 / (S \cdot \Delta t) \quad (3)$$

Where V_1 (L) represents the volume of collected oil, Δt (h) represents the separation time, and S

(m²) represents the effective area of the sample.

The reusability of CP/PDA/CuS/ODA was evaluated over 20 successive separation trials. After each trial, the employed paper was washed thoroughly with ethanol to eliminate any remaining oil and contaminants, then dried in an oven at 50 °C until the ethanol was completely removed. The resulting dried paper was then employed for the next separation trial.

2.8 Durability evaluation

A series of mechanical and environmental durability tests was investigated, which were designed to replicate its practical operational conditions. The tests included the tape peeling test, folding test, abrasion test, chemical and saltwater resistance tests, UV irradiation test, thermal aging test, and long-term storage stability test.

2.8.1 Tape peeling test

For the tape peeling test, the sample of CP/PDA/CuS/ODA was tightly adhered onto an scotch tape and then peeled off, during which a consistent adhesion force was guaranteed by applied new tapes for each 20 peeling cycles. The WCAs on the sample surface were measured periodically during the peeling process to track changes in its superhydrophobicity.

2.8.2 Folding endurance test

Cyclic bending tests were performed on CP/PDA/CuS/ODA samples for determining their durability under repetitive deformation. In the tests, the composite samples were repeatedly folded to a 180° angle and then unfolded back to their original planar state, with one folding-unfolding sequence defined as a single cycle. Extra pressure was avoided during the folding process to prevent damage to the superhydrophobic surface. The surface hydrophobicity was analyzed by measuring the WCA on the surface at each 20th cycle for monitoring the degradation behavior of

superhydrophobicity.

2.8.3 Abrasion durability test

The abrasion durability of the CP/PDA/CuS/ODA sample was evaluated via a sandpaper abrasion test method. The sample was placed upside-down on sandpaper with an 800-grit abrasive surface, and a 10 g weight was loaded onto the upper surface to maintain consistent pressure during the test. The sample was moved horizontally across a fixed 10 cm linear path at a constant speed, with forward-backward reciprocating motion applied repeatedly. To assess the durability of hydrophobicity, the WCA was measured after every two rubbing cycles.

2.8.4 Chemical durability

Chemical stability of the samples was tested by immersing the samples in aqueous solutions with pH ranging from 1 to 12 overnight, to evaluate their resistance to highly corrosive media. After the immersion treatment, the samples were rinsed with ethanol and deionized water to remove residual liquid from the sample surfaces. The samples were then dried in an oven at 50 °C for 4 h, and WCA measurements were performed to assess the retention of the superhydrophobic property.

2.8.5 Salt resistance test

The salt resistance of the CP/PDA/CuS/ODA samples were evaluated through static immersion tests. Specifically, the samples were fully immersed in NaCl solutions with gradient mass concentrations (1 wt%, 2 wt%, 3 wt%, 4 wt%, and 5 wt%) for 12 h at room temperature. After immersion treatment, all specimens were taken out from the solutions and rinsed by deionized water to remove residual NaCl, and then dried thoroughly. The effect of the surface wettability was measured by contact angle before and after being exposed to saline water, respectively.

2.8.6 UV resistance test

The UV irradiation resistance of the CP/PDA/CuS/ODA samples was evaluated by placing the samples vertically under an ultraviolet lamp of 20 W power with a peak emission wavelength of 254 nm, and a fixed distance of 8 cm was maintained between the sample surface and the lamp. The WCA of the samples was measured at regular intervals during continuous irradiation to track the evolution of surface superhydrophobicity.

2.8.7 Thermal durability test

The durability under thermal treatment was evaluated in a thermostatic chamber set at a series of gradient temperatures (20 °C, 40 °C, 60 °C, 80 °C, and 100 °C) for 12h. After thermal treatment, the samples were allowed to cool naturally to room temperature to avoid surface property changes induced by thermal stress. Subsequently, the WCA measurement was carried out to examine the retention of superhydrophobicity after high-temperature exposure.

2.8.8 Long-term storage stability

The long-term stability of the superhydrophobic property is a critical metric for evaluating the practical application potential of materials, particularly for their performance stability during storage and transportation. Herein, the as-prepared CP/PDA/CuS/ODA samples were stored under ambient laboratory conditions (25 ± 2 °C, $50 \pm 5\%$ relative humidity, RH) throughout the test. The WCA of the samples was measured at predetermined intervals (1 week, 2 weeks, 3 weeks, 4 weeks, and 5 weeks). The long-term storage stability of the samples was evaluated according to the evolution of WCA values over the storage period.

2.9 Antifouling and self-cleaning performance

The antifouling performance of the sample was evaluated, as fouling resistance against typical aqueous contaminants is a critical prerequisite for its long-term in-situ service in practical oil/water

separation applications. Four typical aqueous foulants, including grape juice, soda water, methylene blue solution, and methyl orange solution, were selected as the test media for the assessment. The samples were individually immersed into four typical liquid contaminants. After immersion, the paper specimens were gently pulled out vertically. The surface of each paper specimen was then carefully observed and recorded with a digital camera to check for the presence of residual stains, discoloration, or contaminant adhesion. The antifouling performance of the CP/PDA/CuS/ODA was ultimately evaluated based on stain residue on the paper surface.

To further validate the practical applicability of the CP/PDA/CuS/ODA, its self-cleaning performance was assessed. A piece of CP/PDA/CuS/ODA paper sample was mounted on a glass slide and secured in a Petri dish at a certain inclination angle. Chalk powder, used as a model contaminant, was uniformly spread onto the inclined sample surface. Deionized water stained with methylene blue (MB) was subsequently dispensed via a syringe to mimic a rinsing process, and the removal behavior of the chalk powder was closely observed and recorded to evaluate the self-cleaning efficiency.

2.10 Photothermal performance

2.10.1 Photothermal conversion test

To assess the photothermal conversion performance of CP/PDA/CuS/ODA, the sample was irradiated under a xenon lamp-based simulated solar source. Real-time thermal images and surface temperature variations were captured and recorded using an infrared thermal imager. The photothermal behavior was further investigated under four different light intensities (0.5 Sun, 1 Sun, 2 Sun, and 3 Sun). After each irradiation test, the solar simulator was turned off, and the sponge sample was allowed to cool naturally until its temperature reached thermal equilibrium with the

ambient environment. The sample was then exposed again to simulated sunlight at an intensity of 1 sun to initiate a new heating-cooling cycle. The cyclic procedure was carried out 10 times to evaluate the long-term durability and reversibility of the photothermal response.

2.10.2 Photothermal antimicrobial test

The photothermal antimicrobial activity of the CP/PDA/CuS/ODA was evaluated by selecting *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) as model strains, representing Gram-positive and Gram-negative bacteria, respectively. Original CP and CP/PDA/CuS/ODA samples (1 cm × 1 cm in size) were subjected to sterilization and placed into sterile centrifuge tubes. Two parallel groups were set up: one group was exposed to xenon lamp irradiation for 20 min, while the control group was incubated in darkness for an equivalent period. Subsequently, all samples were submerged in 3 mL of bacterial suspension (10^7 CFU/mL) and incubated in a temperature-controlled shaker at 37 °C with a rotational speed of 200 rpm for 24 h. After incubation, the bacterial suspension was subjected to 10-fold serial dilution with sterile water. A portion of the diluted suspension (20 µL) was spread onto LB agar plates and further incubated at 37 °C for 24 h to assess the photothermal bactericidal efficacy.

2.10.3 Anti-icing and deicing test

The anti-icing performance of CP/PDA/CuS/ODA was systematically investigated by simulating a cryogenic environment at −10 °C to induce water freezing. Prior to the test, the cooling stage was pre-cooled until its surface temperature stabilized at the preset target value. A water droplet was carefully deposited onto the surface of the CP/PDA/CuS/ODA composite, and the entire icing process was dynamically recorded using a camera under two distinct conditions: simulated sunlight irradiation and dark environment.

For the evaluation of de-icing performance, pre-formed ice droplets were first fabricated in a low-temperature chamber and subsequently transferred onto the surface of CP/PDA/CuS/ODA. The ice-melting behavior of the composite was then monitored and recorded in detail under two control conditions: in the absence of light irradiation and under simulated sunlight irradiation with an intensity of 1.0 kW/m².

3. Results and discussion

3.1 Surface morphology and structural characterizations

The surface morphology of the original CP and modified CP at each modification stage were characterized using SEM. As illustrated in Figure 2a, the original CP consisted of microscale cellulose fibers, where each individual fiber featured a relatively smooth surface. After immersion in the dopamine solution, a substantial number of granular particles were formed on the surface of the cellulose fibers (Figure 2b), confirming that dopamine was successfully adsorbed onto the paper surface through oxidative polymerization to form PDA. To further improve the surface roughness of the paper, sequential deposition of PDA and CuS was performed on its surface. As observed in Figure 2c, the paper surface was fully covered by randomly dispersed PDA/CuS clusters, along with the formation of irregular aggregates distributed uniformly across the entire surface. As a result, the abundant microclusters remarkably enhanced the surface roughness of the paper, laying a solid foundation for the subsequent fabrication of superhydrophobicity during the modification process. As clearly presented in Figure 2d, following surface hydrophobic modification with ODA (octadecylamine), the boundaries of the microclusters became obscure, which indicates the successful coating of an ODA layer on the paper surface.

Figure 2e displays the XRD patterns of the original cellulose paper (CP) and its modified

derivatives at each stage of the modification process. The original CP showed distinct characteristic peaks at $2\theta = 14.9^\circ$, 16.8° , and 23.1° , which are assigned to the (1-10), (110), and (200) crystal planes of cellulose I in the cellulosic matrix, respectively [38, 39]. When CP was modified with PDA to form the CP/PDA, the characteristic diffraction peaks of cellulose were well-retained without any new peaks appearing, demonstrating that the introduction of PDA had no impact on the crystalline structure of original CP. In contrast, the XRD pattern of pure CuS exhibited prominent diffraction peaks at $2\theta = 27.8^\circ$, 32.1° , and 46.1° , which correspond to the (102), (103), and (110) crystal planes of CuS (JCDPS No. 06-0464), confirming the successful synthesis of CuS nanoparticles [40-42]. Subsequently, when CuS was anchored onto the CP/PDA to fabricate CP/PDA/CuS, the XRD pattern integrated the characteristic diffraction peaks of both CP and CuS, confirming the successful decoration of CuS nanoparticles onto the cellulosic fiber surfaces. Further modification with ODA verified the structural stability of the composite, no new diffraction peaks were induced, suggesting that the crystalline structures of both CP and CuS remained intact and unaltered by the presence of ODA.

Figure 2f illustrates the FTIR spectra of the original CP and its modified derivatives at each modification stage. The FTIR spectrum of original CP exhibited typical characteristic bands corresponding to cellulosic fibers. Specifically, the absorption bands centered at 3334 cm^{-1} and 2886 cm^{-1} were assigned to the O-H stretching vibration and C-H symmetric stretching vibration, respectively. The band observed at 1641 cm^{-1} was associated with the bending vibration of water absorbed on the surface of cellulose paper. Additionally, the bands at 1016 cm^{-1} and 897 cm^{-1} were ascribed to the C-O stretching vibration and β -glycosidic linkages in the cellulose backbone, respectively [43-45]. After modification with PDA, several new absorption bands emerged in the FTIR

spectrum of the CP/PDA composite. The peak at 1621 cm^{-1} was due to the overlap of the C=C skeletal vibrations of aromatic rings and N-H deformation vibrations, while the peak at 1510 cm^{-1} was derived from N-H shearing vibration. The band at 1423 cm^{-1} was assigned to the -C=O group. Furthermore, the bands at 1367 and 1337 cm^{-1} corresponded to the stretching vibration of the aromatic C=C bond and the C-O stretching vibrations in phenolic structures, respectively. A weak band at 1276 cm^{-1} resulted from the combination of the deformation vibrations of O-H in phenol and cellulose, and the band at 1111 cm^{-1} was attributed to the tensile vibration of the C-O bond. The appearance of all these new absorption bands confirmed the successful deposition of PDA on the cellulose paper surface ^[46-48]. For the spectrum of CP/PDA/CuS, the absorption band at 1108 cm^{-1} 612 cm^{-1} was assigned to the Cu=S bond stretching vibration, confirming the successful decoration of CuS onto the CP/PDA surface ^[49]. After coating with ODA, a new band appearing at 1633 cm^{-1} was attributed to the C=N stretching vibration, which originated from the Schiff-base reaction between PDA and ODA. Furthermore, the bands at 2850 , and 2917 cm^{-1} were assigned to the C-H stretching vibrations of $\text{-CH}_2\text{-}$ and $\text{-CH}_3\text{-}$ groups in the long alkyl chains of ODA, respectively ^[50]. The emergence of these characteristic bands confirms the successful grafting of the low-surface-energy ODA coating onto the CP/PDA/CuS composite surface.

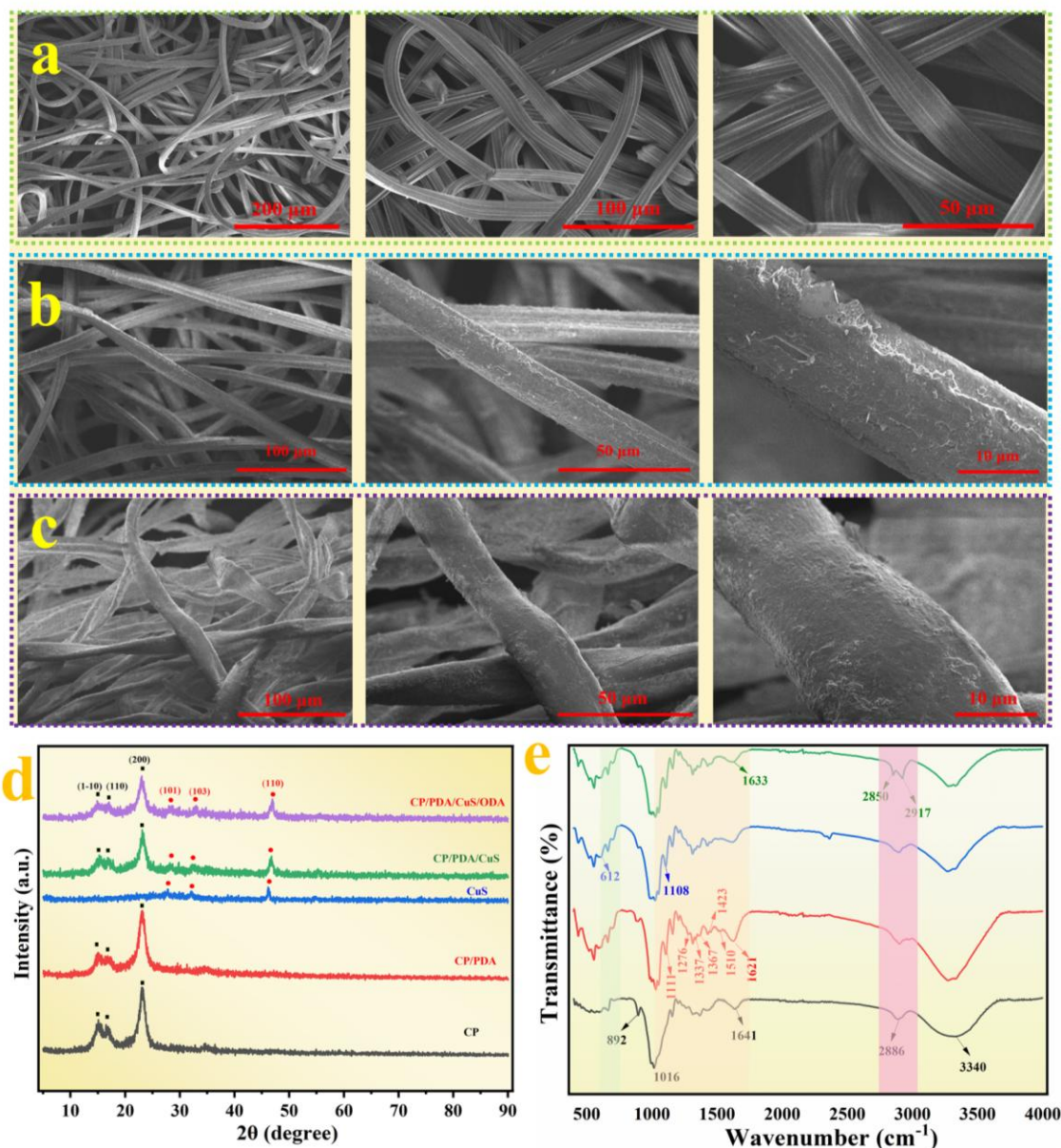


Figure 2 SEM images for original CP (a), CP/PDA (b), CP/PDA/CuS (c), CP/PDA/CuS/ODA (d); FTIR spectra (e), and XRD patterns (f) of the original CP and its modified derivatives at each stage of the modification process.

3.2 Surface wetting characterization

To investigate the wettability of the as-prepared superhydrophobic CP/PDA/CuS/ODA, a series of wettability tests were carried out. As anticipated, the synergistic effect of chemical composition and hierarchical structure remarkably altered the wettability of the modified cellulose paper. Figs. 3a-3f illustrate the wetting behaviors of the pristine CP and the superhydrophobic

CP/PDA/CuS/ODA.

The original CP exhibited hydrophilicity, rapidly absorbing water upon contact and completely submerging to the bottom of the water (Figure 3a). In contrast, the CP/PDA/CuS/ODA showed no water absorption and merely floated on the water surface. Even when pressed below the water surface by external force using a pair of tweezers, the CP/PDA/CuS/ODA quickly returned to the water surface (Figure 3b). Notably, the contact interface between the CP/PDA/CuS/ODA and water exhibited a silver-mirror-like reflection, which is attributed to the uniform air layer formed between the superhydrophobic surface of the CP/PDA/CuS/ODA and water, which is consistent with the non-wetting Cassie-Baxter model. As shown in Figs. 3c and 3d, the surface of original CP was easily wetted by both water and oil droplets, with both water and oil contact angle of 0° , indicating its amphiphilic nature (hydrophilic and lipophilic). In contrast, the modified CP/PDA/CuS/ODA exhibited significantly enhanced hydrophobicity. As illustrated in Figure 3e, the dark blue water droplets maintained spherical shape, while the red oil droplets were rapidly absorbed by the CP/PDA/CuS/ODA, confirming the transformation of the paper from amphiphilic to hydrophobic after modification. Additionally, the water contact angle of the modified composite was significantly increased to 156.4° , as presented in Figure 3f.

3.3 Evaluation of selective oil removal performance and oil absorption capacity

Based on the outstanding superhydrophobic and superoleophilic properties of the as-prepared CP/PDA/CuS/ODA, the capacity for selective oil removal and oil absorption were evaluated, with the associated findings depicted in Figure 3g-Figure 3j. In contrast to the original unmodified CP, which exhibited non-selective absorption towards both oil and water (Figure 3c), the as-prepared CP/PDA/CuS/ODA exhibited rapid oil absorption performance coupled with excellent resistance to

water penetration. The coexistence of superhydrophobicity and superoleophilicity enables the functionalized paper with exceptional selectivity and efficiency in extracting oils from oil/water mixtures. For the absorption of floating light oil, gasoline stained with Oil Red O was dispensed onto the water surface to generate a uniform oil film, and the CP/PDA/CuS/ODA was brought into contact with the gasoline/water mixture (Figure 3g). The CP/PDA/CuS/ODA quickly and thoroughly captured the floating gasoline in a matter of seconds, leaving a clean water surface. During the absorption process, the CP/PDA/CuS/ODA maintained entirety of the water repellency. In order to absorb heavy oil, the CP/PDA/CuS/ODA was submerged in a beaker filled with water that contained chloroform (stained with Oil Red O). Given that chloroform is denser than water, it formed a distinct lower layer. In a similar manner, the CP/PDA/CuS/ODA completely absorbed the chloroform in a matter of seconds (Figure 3h). These findings demonstrated the selective oil absorption capacity of the CP/PDA/CuS/ODA for both floating light oils and underwater heavy oils.

Quantitative measurements were conducted to determine the saturated absorption capacities for a range of commonly utilized oils and organic solvents. The absorption capacities of the CP/PDA/CuS/ODA ranged from 7.68 g/g to 21.92 g/g, as shown in Figure 3i. Carbon tetrachloride possessed the highest value (21.92 g/g), while petroleum ether possessed the lowest value (7.68 g/g). The low surface energy of the superhydrophobic coating, well-tuned pore size, and the intrinsic interconnected porous structure of cellulose substrate are primarily responsible for the remarkable oil absorption capacity. These factors greatly increased the capillary driving force for oil uptake and retention within the porous network. The findings showed that the CP/PDA/CuS/ODA exhibited broad compatibility with a variety of hydrocarbons and organic solvents as well as a high absorption capacity.

Excellent reusability is essential for the practical application of oil absorbent materials, particularly in treating industrial oily wastewater and remediating marine oil spills. As shown in Figure 3j, the CP/PDA/CuS/ODA exhibited nearly unchanged absorption capacity with no significant degradation after 20 consecutive absorption-desorption cycles. These results validated that the CP/PDA/CuS/ODA showed outstanding recyclability and high practical applicability, thereby presenting great potential for realistic industrial implementation in oil–water separation and organic solvent treatment.

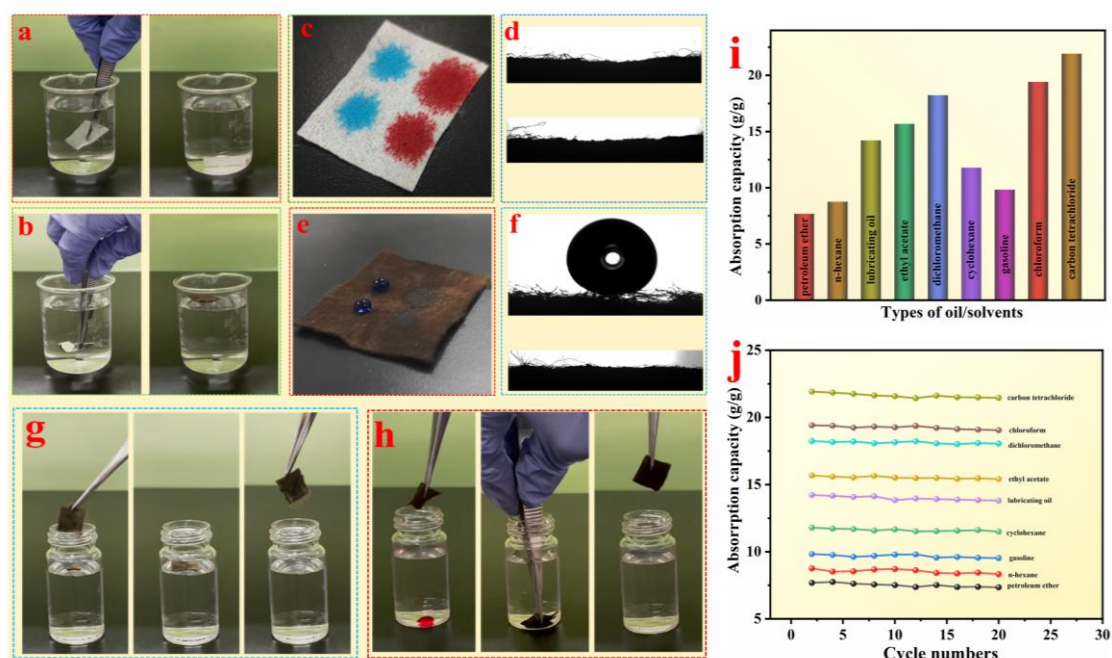


Figure 3 The original CP submerging in water (a), the superhydrophobic CP/PDA/CuS/ODA floating on the water surface (b), WCA and OCA for the original CP (c), oil and water droplets depositing on the surface of original CP (d), oil and water droplets depositing on the surface of superhydrophobic CP/PDA/CuS/ODA (e), WCA and OCA for the superhydrophobic CP/PDA/CuS/ODA (f), selective oil removal performance for both light (g) and heavy oils (h), absorption capacity of the CP/PDA/CuS/ODA for different oils and solvents (i), recyclability of the CP/PDA/CuS/ODA over 20 absorption-desorption cycles (j)

3.4 Continuous gravity-driven oil/water separation

The continuous oil-water separation performance of the as-prepared superhydrophobic CP/PDA/CuS/ODA was evaluated by gravity-driven apparatus. Figure 4 shows the separation device for oil-water mixtures, and the entire separation process was driven solely by gravity. In this work, the superhydrophobic paper was applied to separate various types of oil-water mixtures, such as cyclohexane/water (Figure 4a), petroleum ether/water (Figure 4b), n-hexane/water (Figure 4c), dichloromethane/water (Figure 4d), chloroform/water (Figure 4e), and carbon tetrachloride-water (Figure 4f). When the oil/water mixture, with oil dyed by oil Red O and water dyed by methylene blue, was poured into a glass funnel fitted with the superhydrophobic CP/PDA/CuS/ODA as a filter, the red oil was rapidly absorbed by the paper, then permeated through the funnel and dripped into the collection beaker. In contrast, the blue water was retained in the funnel due to the superhydrophobicity of the CP/PDA/CuS/ODA.

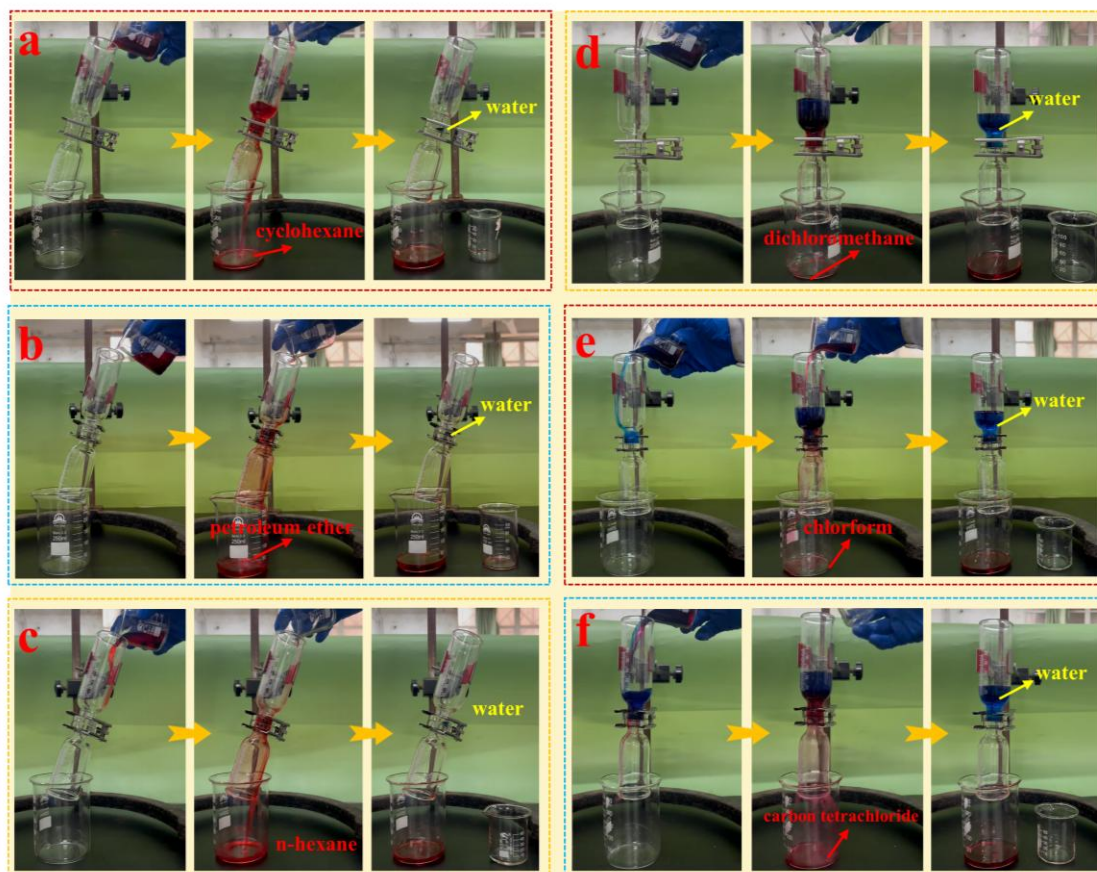


Figure 4 Continuous oil-water separation process for cyclohexane/water (Figure 4a), petroleum ether/water (b), n-hexane/water (c), dichloromethane/water (d), chloroform/water (e), and carbon tetrachloride/water (f)

To quantify the separation performance, the final separation efficiency and flux were calculated using Eq. (2) and Eq. (3), respectively. As illustrated in Figure 5a, the separation efficiencies of all tested oil-water mixtures exceeded 99.35%, with the highest value of 99.83 % achieved for carbon tetrachloride and the lowest of 99.35% for petroleum ether. Figure 5b showed that the prepared superhydrophobic CP/PDA/CuS/ODA paper had a high separation flux for all the tested oil-water mixtures, including cyclohexane-water ($9554.14 \text{ L m}^{-2} \text{ h}^{-1}$), petroleum ether-water ($10034.19 \text{ L m}^{-2} \text{ h}^{-1}$), n-hexane-water ($8443.31 \text{ L m}^{-2} \text{ h}^{-1}$), dichloromethane-water ($8819.21 \text{ L m}^{-2} \text{ h}^{-1}$), chloroform-water ($8944.09 \text{ L m}^{-2} \text{ h}^{-1}$), and carbon tetrachloride-water ($9096.43 \text{ L m}^{-2} \text{ h}^{-1}$). Experimental results demonstrated that the synthesized paper possessed superior separation efficiency and permeate flux, a distinct advantage in that these flux values were achieved exclusively through gravitational driving force, without the need for external pressure supplementation.

Beyond static separation performance, recyclability and reusability are indispensable prerequisites for advancing superhydrophobic materials toward cost-effective practical engineering applications, as they play a decisive role in determining the long-term economic viability and large-scale scalability of such materials. The CP/PDA/CuS/ODA exhibited outstanding operational stability and consistent performance across 10 successive separation runs (Figure 5c). The separation efficiencies for carbon tetrachloride and petroleum ether were consistently maintained above 99.37% and 99.02%, respectively, while the corresponding permeate fluxes remained stable at 8903.72 and $10034.19 \text{ L m}^{-2} \text{ h}^{-1}$ (Figure 5d). The minimal decline in separation performance after repeated cycling confirmed the superior reusability of the paper, laying a solid foundation for its

future practical application in oily wastewater remediation and oil/water separation processes.

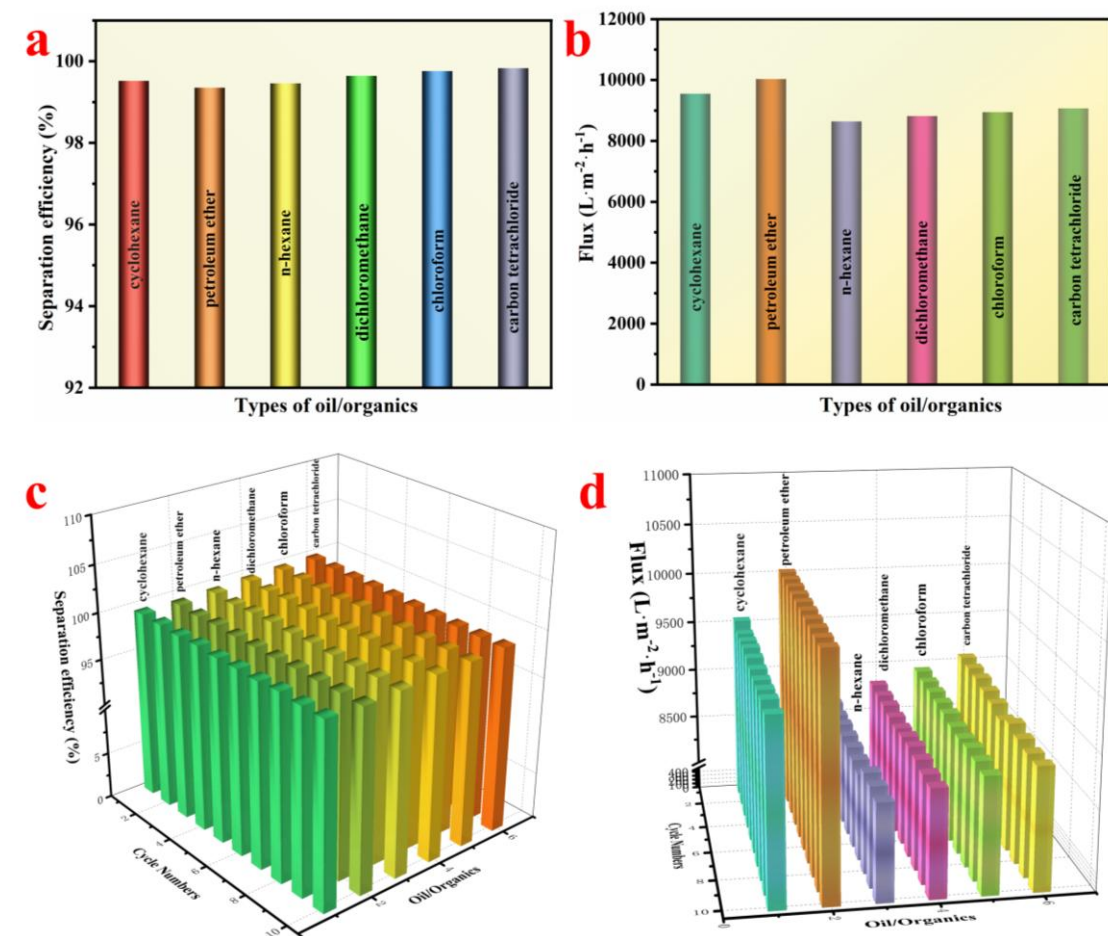


Figure 5 Separation efficiency for various oil-water mixtures (a), separation flux for various oil-water mixtures (b), separation efficiency for various oil-water mixtures during ten cycles (c), separation flux for various oil-water mixtures during ten cycles (d)

3.5 Fundamental theories for oil/water separation performance

The interfacial wettability behavior between the solid substrate, ambient air, water, and oil phases largely determines the oil/water separation performance^[51-53]. Young's equation (Eq. (4)) can be used to determine the contact angle of a liquid droplet resting on an ideal smooth solid surface in an air atmosphere (Figure 6a):

$$\cos\theta_0 = (\gamma_{SA} - \gamma_{SW}) / \gamma_{WA} \quad (4)$$

The formula provides a basic theoretical framework for examining the intrinsic wetting behaviors in oil/water separation systems by establishing a relationship between the Young contact angle with the surface and interfacial tensions among the coexisting phases. Here, θ_0 refers to the equilibrium contact angle on an ideal solid surface; γ_{SA} and γ_{WA} represent the solid/air and liquid/air interfacial tensions, respectively; and γ_{SW} denotes the solid/liquid interfacial tension.

However, Young's equation presents significant restrictions, as it operates only under conditions where surfaces maintain both complete smoothness and chemically homogeneous composition. The Wenzel model (Eq. (5)) and Cassie-Baxter model (Eq. (6)) can be applied to measure the wetting properties of liquid droplets which appear on genuine rough surfaces. For water droplets, the apparent contact angle measured on rough surfaces can be expressed by the Wenzel equation:

$$\cos\theta_w = r \cos\theta_0 \quad (5)$$

In Eq. (4), r ($r > 1$) denotes the roughness ratio (or topological enhancement factor) of the actual interface, which is defined as the ratio of the actual surface area to the apparent projected area. As a result, surface morphology (represented by r) and intrinsic wettability (represented by θ_0) both influence the measured contact angle. The Wenzel model shows that surface roughness effectively amplifies the material's intrinsic wetting properties in the fully wetted state (Figure 6b), where the

fluid fully penetrates the surface roughness. In particular, in this state, a hydrophilic surface becomes more hydrophilic and a hydrophobic surface becomes more hydrophobic. On the other hand, air pockets confined beneath the droplet usually form a solid-water/air contact system in the heterogeneous wetting state (Figure 6c). For this scenario, the apparent contact angle (θ_{CB}) is described by the Cassie-Baxter equation:

$$\cos\theta_{CB}=r_f f_{SW} \cos\theta_0 + f_{SW} - 1 \quad (6)$$

Herein, f_{SW} represents the fraction of the contact area covered by the solid-water interface. Unlike the total roughness factor r , r_f is defined as the roughness coefficient corresponding to the solid-water contact region.

To further explore the oil-water separation mechanism of the prepared cellulose paper, its interfacial contact state was simulated. The prepared paper shows superhydrophobic and superoleophilic characteristics in air, indicating that oil droplets can pass through the paper while water droplets are blocked, thus promoting the oil-water separation process. The oil contact angles on the paper surface are less than 10° , while the water contact angle is more than 150° .

The intrusion pressure of the paper can be described by the intrusion pressure equation:

$$\Delta P = 2\gamma/R = -\gamma l \cos\theta / A \quad (7)$$

In Eq. (7), ΔP is the intrusion pressure; γ represents the water-oil interfacial tension; R is the radius of the curved liquid surface; l denotes the perimeter of the pores; θ is the water contact

angle (WCA) or oil contact angle on the surface of the superhydrophobic cellulose paper; and A is the pore diameter of the cellulose paper. Simulation results show that when the contact angle θ is greater than 90° , the breakthrough pressure $\Delta P > 0$. Under gravity, oil droplets can penetrate the paper while water droplets are intercepted (Figure 6d and Figure 6e), thus realizing efficient separation of oil-water mixtures (Figure 6f).

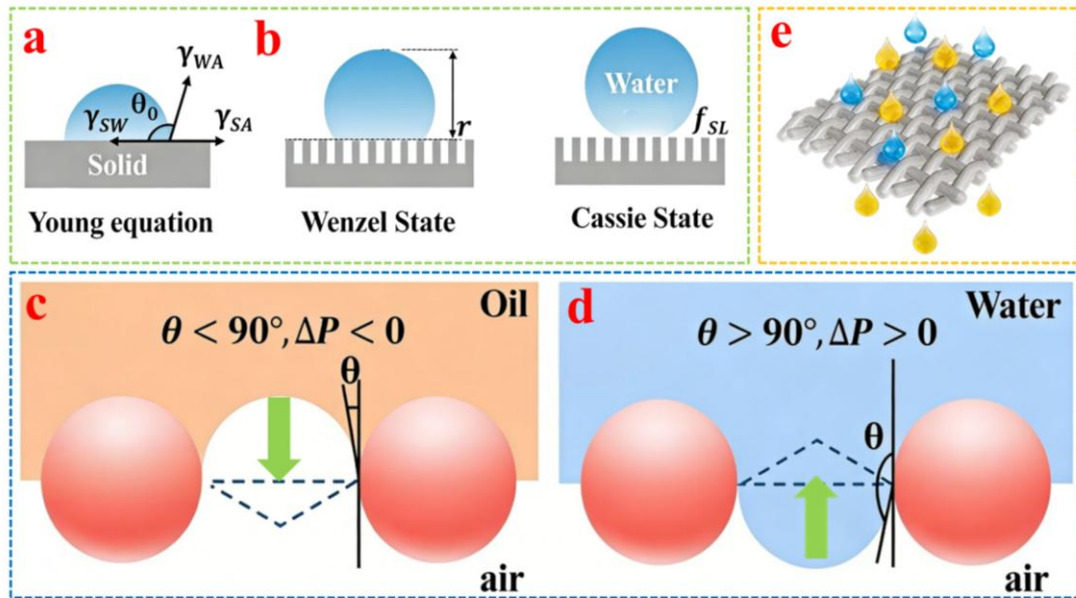


Figure 6 Schematic illustrations of Young's Equation (a), Wenzel State (b), Cassie State (c), wetting models for the oil–water separation process (when $\theta < 90^\circ$ (d) and $\theta > 90^\circ$ (e)), Schematic diagram of separation of oil-water mixtures (f)

3.6 Durability evaluation

Durability is a critical prerequisite for superhydrophobic surfaces, as these materials inevitably encounter external mechanical impacts and environmental perturbations in practical applications. The delicate micro/nano hierarchical structures of superhydrophobic surfaces break down when they

experience severe environmental conditions which lead to their wettability performance degradation. The resistance to mechanical stress and environmental stability of the CP/PDA/CuS/ODA were evaluated. The WCA was measured to evaluate resistance to mechanical stress which involved tape peeling and cyclic folding and sandpaper abrasion. In the tape-peeling test, a sticking double-sided adhesive tape was applied to the sample surface before they started the peeling process, which was defined one test cycle. The tape was replaced after every 20 cycles to maintain stable adhesive bond strength throughout the test. The results of Figure 7a showed that the WCA stayed constant through 100 peel tests, which means the surface demonstrated excellent resistance to adhesion. The folding stability of CP/PDA/CuS/ODA superhydrophobic paper was tested by performing multiple folding tests, which involved moving the paper from -90° to 90° for each cycle. The WCA was conducted at intervals of every 20 cycles, as shown in Figure 7b. The WCA stayed above 150° through 100 folding tests, suggesting the material showed outstanding resistance to repeated folding stress.

To assess its abrasion resistance, the sample was subjected to linear sliding against 800-grit sandpaper under a constant load of 10 g in order . One abrasion cycle was equivalent to a 10 cm sliding distance. Every two cycles, the WCA measurements were obtained. due to the fact that the hierarchical surface topography was gradually destroyed, the WCA showed a progressive decline with increasing abrasion cycles, as displayed in Figure 7c. However, after ten abrasion cycles, the WCA remained high at about 152.6° , indicating favorable abrasion stability.

Superhydrophobic materials need to demonstrate sufficient environmental durability, as their operational environments regularly expose them to severe conditions, including corrosive media, saltwater, UV irradiation, and high temperatures. To assess chemical stability, the samples were immersed in various acidic and alkaline aqueous solutions for 24 h. As depicted in Figure 7d and

Figure 7e, all samples maintained their superhydrophobic performance because their WCAs stayed above 150° , although the values showed a slight decrease. Similarly, after immersion in 1-5 wt% NaCl solutions for 12 h, all samples still maintained superhydrophobicity with WCAs higher than 150° , revealing the ability to resist saltwater immersion at elevated concentrations. UV durability was evaluated by vertically placing samples beneath a 20 W UV lamp (254 nm, 8 cm distance), with WCA measured at varying exposure intervals. (Figure 7g). The WCAs values gradually decreased with prolonged irradiation, likely due to UV-induced photodegradation of surface compositions. However, WCA remained above 150° after 12 h. Given the lower UV intensity of natural sunlight, the paper is expected to exhibit superior UV durability in real conditions. Figure 7h shows the modified paper's temperature resistance after 12 h exposure to 20– 100°C ; WCA exceeded 150° under all conditions, demonstrating excellent thermal stability. For long-term storage, samples were exposed to ambient conditions ($25 \pm 2^\circ\text{C}$, $50 \pm 5\%$ RH) for five weeks (Figure 7i). WCA remained stable above 150° , as ambient conditions did not damage the surface structure, confirming stable superhydrophobicity during storage. Nevertheless, the WCA still remained greater than 150° after 12 h of UV irradiation. Given that the UV intensity of natural sunlight is much lower than that used in this test, it can be anticipated that the superhydrophobic paper will exhibit superior UV durability under actual natural conditions. Figure 7h illustrates the temperature resistance of the modified paper when exposed to different temperatures (100/80/60/40/ 20°C) for 12 h. As clearly shown in Figure 7h, the water contact angle of the modified paper was measured to be higher than 150° under all tested temperature conditions, manifesting excellent thermal stability. In addition, the superhydrophobic paper may be exposed to ambient environments during long-term storage and transportation. Figure 7i presents the variations in WCA of the superhydrophobic paper after

exposure to ambient conditions (25 ± 2 °C, $50 \pm 5\%$ relative humidity) for five weeks. The WCA remained stabilized above 150° , which is attributed to the fact that the room temperature and moderate humidity conditions did not cause obvious damage to the surface structure of the paper samples. This result indicates that the superhydrophobic paper possesses stable superhydrophobicity during long-term storage.

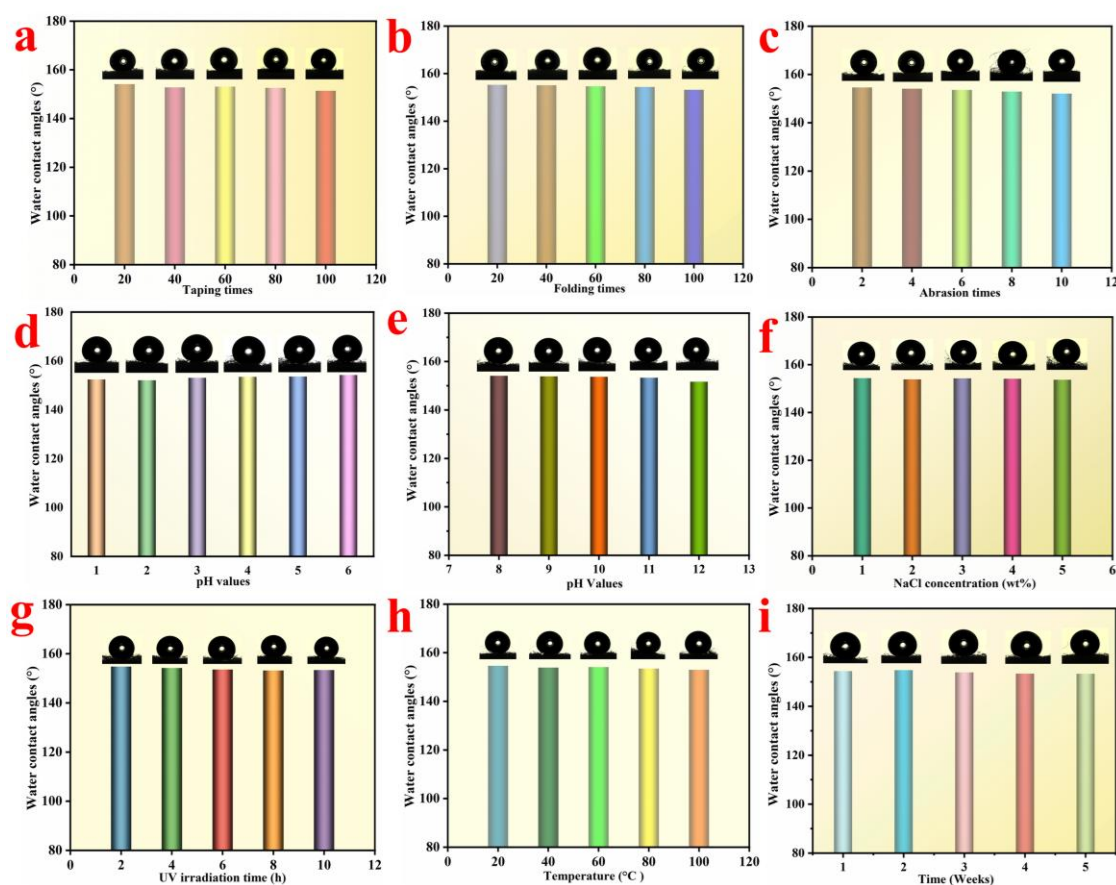


Figure 7 Durability evaluation under taping (a), folding (b), abrasion (c), acidic solutions (d), alkaline solutions (e), salty solutions (f), UV irradiation (g), high temperature (h), and long-term storage (i)

3.7 Antifouling and self-cleaning performance

Antifouling and self-cleaning abilities are highly significant for superhydrophobic paper, as they can effectively avoid surface contamination in practical use. The antifouling characteristic of the superhydrophobic CP/PDA/CuS/ODA sample was comprehensively evaluated, with the original CP

serving as the control group. Specifically, the two kinds of samples (original CP and superhydrophobic CP/PDA/CuS/ODA) were respectively immersed in four typical liquid pollutants, namely grape juice, Coca-Cola, methylene blue (MB) colored water, and methyl orange (MO) colored water. It was found that the original CP was easily contaminated by all four liquid pollutants, as shown in Figure 7a (grape juice), Figure 7b (Coca-Cola), Figure 7c (MB colored water), and Figure 7d (MO colored water). In comparison, after being removed from the above - mentioned liquid pollutants, the superhydrophobic CP/PDA/CuS/ODA sample still kept a clean and dry surface without any obvious contamination, which was in line with the observations in Figure 7e (grape juice), Figure 7f (Coca-Cola), Figure 7g (MB colored water), and Figure 7h (MO colored water). This excellent performance fully validates the superior antifouling capacity of the prepared superhydrophobic CP/PDA/CuS/ODA composite paper.

The antifouling mechanism of the superhydrophobic CP/PDA/CuS/ODA paper could be ascribed to the synergistic effect of its surface chemical composition and micro-nano structure. The hydrophobic long alkyl chains of ODA grafted on the paper surface significantly reduce its surface free energy, thereby hindering the wetting of liquid contaminants. This is attributed to the weakened intermolecular forces between contaminants and the sample surface, which effectively prevents contaminant spreading and adhesion. Additionally, the composite coating of PDA and CuS constructs a stable micro-nano hierarchical structure on the original CP surface. This structure traps air to form an air film, which further isolates contaminants from the paper's solid surface. Consequently, liquid contaminants exist as spherical droplets that roll off easily with minimal external force, leaving no contamination traces. Moreover, the PDA layer acts as a bridging agent to enhance the binding force between CuS particles, ODA, and the CP matrix, ensuring the stability

of the superhydrophobic structure during testing. Its weak adhesion to organic and inorganic contaminants also further enhances antifouling performance. In summary, the excellent antifouling ability of the CP/PDA/CuS/ODA stems from the synergistic effect of low surface energy of ODA and the micro-nano hierarchical structure constructed by PDA and CuS.

Moreover, the self-cleaning property of the prepared superhydrophobic composite paper was evaluated by using chalk powder as a solid pollutant. The chalk powder was evenly spread on the surfaces of both the original CP and the CP/PDA/CuS/ODA specimen, and then rinsed with dyed water using a syringe. As shown in Figure 7i, water drops were rapidly absorbed by the original CP when in contact, leading to the remaining adhesion of chalk powder on its surface. On the contrary, water drops maintained a spherical shape on the CP/PDA/CuS/ODA sample's surface, rolled smoothly over the surface, and removed nearly all the chalk powder marks (Figure 7j). A schematic diagram of the self-cleaning mechanism is shown in Figure 7k. In line with its antifouling mechanism, the outstanding hydrophobicity of the modified composite paper, resulting from the low surface free energy of ODA and the PDA/CuS micro-nano structure, allows water drops to roll quickly without being absorbed; while rolling, the drops take away surface contaminants, thereby giving the sample remarkable self-cleaning performance.

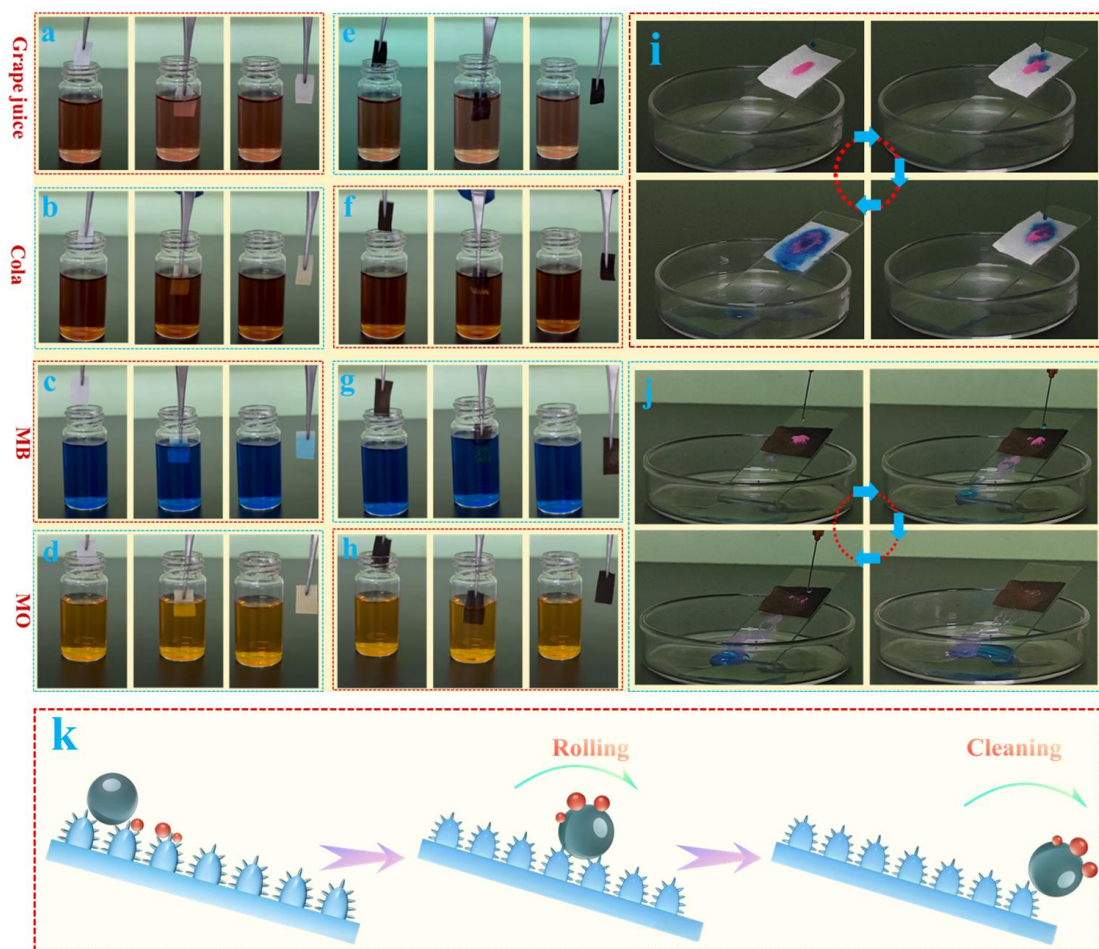


Figure 7 Anti-fouling behavior of the original CP against grape juice (a), Coca-Cola (b), MB colored water (c), and MO colored water (d), anti-fouling behavior of the CP/PDA/CuS/ODA against grape juice (e), Coca-Cola (f), MB colored water (g), and MO colored water (h), self-cleaning behavior of the original CP (i), self-cleaning behavior of the CP/PDA/CuS/ODA (j), schematic illustration of the self-cleaning mechanism (k)

3.8 Photothermal performance

3.8.1 Photothermal conversion test

The photothermal effect has attracted significant research interest because of its improved functionality in the areas of anti-icing, deicing, and antibacterial applications. To comprehensively assess the photothermal conversion ability of the fabricated samples, the temperature change of different specimens (i.e., CP, CP/PDA, CP/PDA/CuS, and CP/PDA/CuS/ODA) was dynamically

monitored as a function of irradiation time under simulated solar light (1 sun, $1.0 \text{ kW} \cdot \text{m}^{-2}$). As shown in Figure 8a, the maximum surface temperature of the pure CP reached a stable value of $18.0 \text{ }^{\circ}\text{C}$, which clearly shows the absence of effective photothermal conversion capacity. On the contrary, the maximum surface temperatures of CP/PDA and CP/PDA/CuS rose to $61.8 \text{ }^{\circ}\text{C}$ and $73.1 \text{ }^{\circ}\text{C}$ respectively. These results confirm that the combination of PDA and CuS brings about a synergistic photothermal effect under solar irradiation. Subsequent modification with ODA led to a maximum surface temperature of $74.3 \text{ }^{\circ}\text{C}$ for CP/PDA/CuS/ODA, indicating that the ODA coating does not have a significant impact on the composite's photothermal conversion performance. Moreover, the maximum surface temperature of CP/PDA/CuS/ODA was measured under different solar irradiation intensities (i.e., 0.5, 1.0, 2.0, and 3.0 sun), reaching 67.8 , 74.8 , 79.7 , and $95.3 \text{ }^{\circ}\text{C}$ respectively (Figure 7b). The thermal stability of CP/PDA/CuS/ODA was further confirmed through 10 cyclic photothermal tests under solar irradiation (1 sun); the sample showed a quick thermal response within a few seconds when exposed to light, followed by a thermal retention phase and a final cooling phase when the light was switched off. Additionally, the maximum surface temperature of the sample increased as the light intensity went up. Notably, the surface temperature of CP/PDA/CuS/ODA stayed above $70.0 \text{ }^{\circ}\text{C}$ after 10 cyclic tests (Figure 7c), verifying its excellent cyclic photothermal stability. Overall, the photothermal and superhydrophobic CP/PDA/CuS/ODA composite can be stably used in various environments with different sunlight intensities.

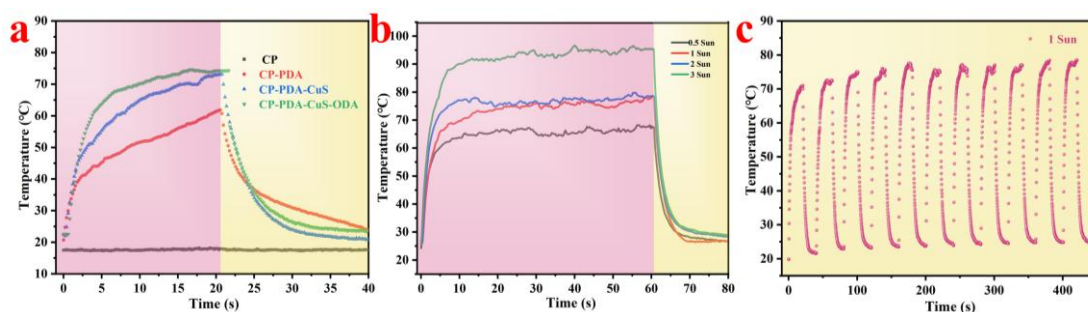


Figure 8 Photothermal conversion performance of different samples under simulated solar light (a), photothermal conversion performance of CP/PDA/CuS/ODA under different solar irradiation intensities (b), cyclic photothermal performance of CP/PDA/CuS/ODA through 10 cyclic photothermal tests under solar irradiation (c)

3.8.2 Photothermal antimicrobial test

Considering the outstanding photothermal conversion capability and excellent cyclic stability of the CP/PDA/CuS/ODA, it exhibits considerable potential for application as a photothermal antimicrobial applications to treat bacterial infections. The antimicrobial performance of the CP/PDA/CuS/ODA was evaluated under different conditions. Two key bacterial strains (*E. coli* and *S. aureus*), major contributors to most infectious diseases, were chosen for this evaluation. As depicted in Figure 9a, for the control group, *E. coli* and *S. aureus* bacterial colonies exhibited robust growth on agar plates irrespective of light irradiation. These observations demonstrated that light irradiation exerted a relatively insignificant impact on the bacterial viability of the original CP.

For the CP/PDA/CuS/ODA composite, no obvious difference was observed relative to the control groups without irradiation. In contrast, nearly all bacteria on the samples under illumination were inactivated compared with those kept in dark (Figure 9b), demonstrating that the as-prepared CP/PDA/CuS/ODA exhibits remarkable photothermal bactericidal activity and can efficiently inactivate bacteria under visible-light irradiation.

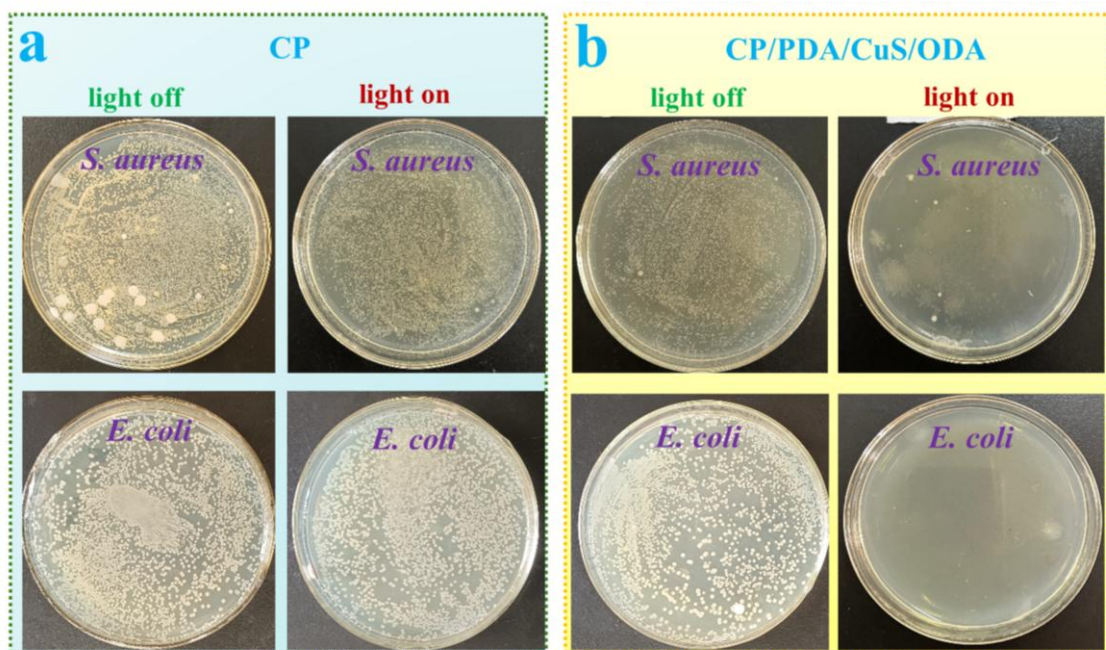


Figure 9 Photothermal antimicrobial test for original CP (a) and CP/PDA/CuS/ODA (b) without irradiation and with irradiation

3.8.3 Anti-icing and deicing test

Ice accretion is a common natural occurrence across various scenarios, and inhibiting ice formation on exposed surfaces is of critical importance for diverse applications, spanning from household settings to large-scale industrial processes. A promising approach to mitigate icing is the construction of superhydrophobic surfaces, which can significantly delay the freezing process of water droplets. This delayed freezing behavior mainly arises from the reduced solid–liquid contact area endowed by hierarchical micro/nanostructures and the superior thermal insulation provided by the air pockets trapped within the surface structures. Nevertheless, in practical outdoor environments, exposed surfaces, including conventional icephobic surfaces, inevitably suffer from ice nucleation and accumulation with prolonged exposure and decreasing temperatures. To overcome this drawback, integrating photothermal conversion function into superhydrophobic surfaces has become an effective method. Under sunlight, these photothermal superhydrophobic

surfaces show notably improved anti-icing performance compared to traditional superhydrophobic surfaces.

As shown in Figure 10a, a 15 μL water droplet was placed on the CP/PDA/CuS/ODA surface, with the substrate temperature kept at $-5\text{ }^{\circ}\text{C}$. Without sunlight irradiation, the water droplet on the CP/PDA/CuS/ODA surface needed 13 min 02 s to freeze entirely. When sunlight irradiation (1 Sun) was used for passive anti-icing, the freezing time was notably extended to 29 min 22 s (Figure 10b), showing an increase of about 124.7% compared to the non-irradiation situation. This substantial extension of freezing time showed the remarkable improvement in passive anti-icing performance caused by sunlight irradiation. Apart from its passive anti-icing performance, the CP/PDA/CuS/ODA surface also showed excellent active photothermal deicing ability. To assess the deicing efficiency of the CP/PDA/CuS/ODA surface, the melting processes of ice droplets were observed and recorded. Without sunlight irradiation, the ice droplet on the CP/PDA/CuS/ODA surface took 18 min 08 s to melt fully (Figure 10c). In contrast, under sunlight irradiation (1 Sun), the melting time was greatly decreased to 3 min 34 s (Figure 10d), corresponding to a reduction of approximately 80.8% compared with the non-irradiation condition. The experimental result clearly verified that the CP/PDA/CuS/ODA surface has superior active deicing performance.

The hierarchical micro/nanostructures on the surface trap a large number of air pockets, which not only decrease the solid-liquid contact area and endow superior superhydrophobicity, but also provide remarkable thermal insulation to delay the nucleation and freezing of water droplets. When exposed to solar irradiation, the photothermal components embedded in the surface efficiently convert light energy into thermal energy, elevating the local surface temperature and melting the initial ice or preventing ice formation and accumulation. As a result, compared to traditional

superhydrophobic surfaces under outdoor conditions, the photothermal superhydrophobic surface shows synergistically enhanced passive anti-icing and active deicing properties.

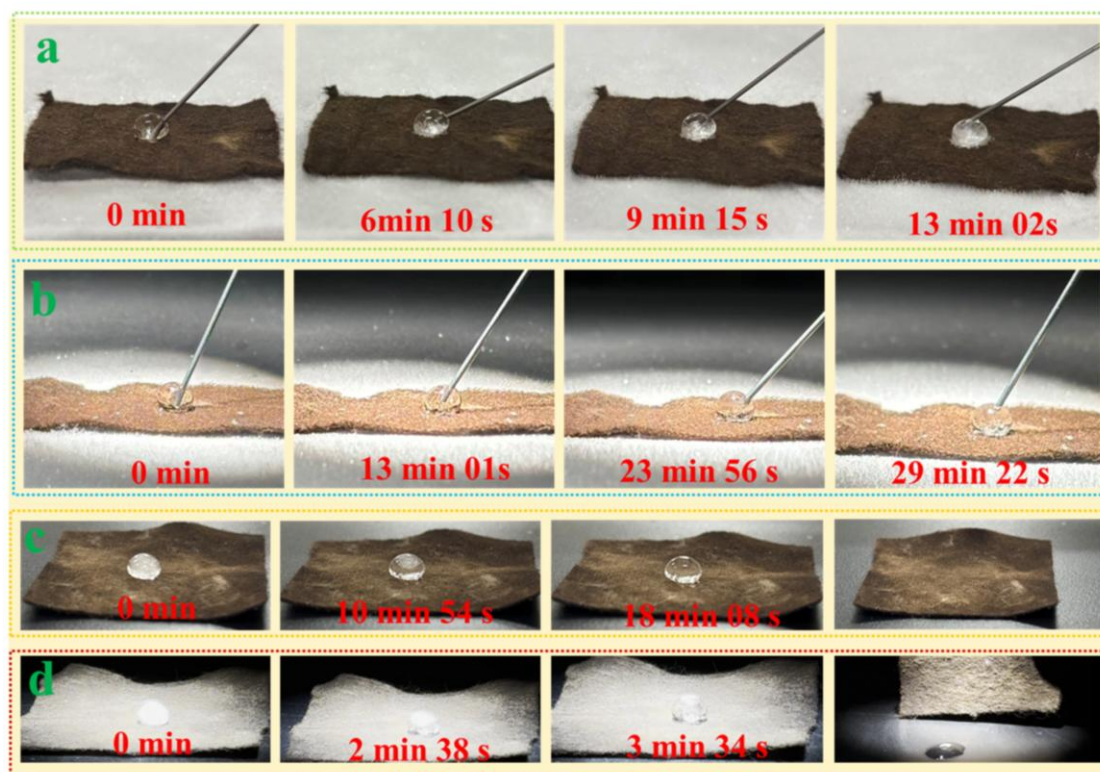


Figure 10 Passive anti-icing performance of the CP/PDA/CuS/ODA without sunlight irradiation (a) and with sunlight irradiation (b), active deicing performance of the CP/PDA/CuS/ODA without sunlight irradiation (c) and with sunlight irradiation (d)

Conclusion

Herein, a superhydrophobic photothermal cellulose paper was rationally designed and successfully fabricated through a tailored modification approach. The resultant modified cellulose paper showed a high water contact angle (WCA) of 156.4° and an oil contact angle (OCA) of 0° , manifesting exceptional water-repellent and oil affinity characteristics. Owing to the strong chemical bonding interactions and highly cross-linked surface architecture, the modified paper retained its superhydrophobicity even after being subjected to a series of harsh environmental and mechanical stresses, such as chemical corrosion, salt spray erosion, ultraviolet (UV) exposure, high-

temperature treatment, 100 tape-peeling cycles, 100 bending cycles, and 10 sandpaper abrasion cycles. Moreover, the modified cellulose paper exhibited prominent self-cleaning and antifouling capacities, coupled with efficient oil-water separation performance. It is worth noting that the oil-water separation efficiency remained over 99% and the separation flux exceeded $8900 \text{ L m}^{-2} \text{ h}^{-1}$ even after 10 reuse cycles, suggesting excellent long-term durability for practical application scenarios. Furthermore, the CP/PDA/CuS/ODA demonstrated superior comprehensive performance, attributed to its excellent photothermal properties, which not only enhanced the antimicrobial activity against *E. coli* and *S. aureus* under light irradiation, but also exhibited remarkable anti-icing and deicing capabilities. The results highlighted the promising application prospects of the novel reinforced cellulose-based composite paper for addressing pressing environmental issues, especially oil spill remediation and water purification.

Notes

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