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Fabrication of superwetting gelatin/phytic acid/cellulose composites with antibacterial characteristics for separating complex contaminants from wastewater

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ABSTRACT

The treatment of complex contaminants in wastewater has become a serious worldwide challenge at present due to the speedy growth of industrial activities and chemical discharges. Consequently, there is a strong demand for the development of materials with superwetting properties from inexpensive materials to treat oil/water mixtures, emulsions and dye solutions. In this study, cellulose materials including raw cotton and cotton fabric were modified using gelatin (Ge) and phytic acid (PA) to prepare superwetting composites through an eco-friendly method without consuming harmful chemical compounds. The resulting Ge/PA-modified cellulose materials exhibited superamphiphilic and underwater superoleophobic properties. We used Ge/PA-modified cotton fabric (GPCF) to separate oil/water mixtures that exhibited exceptionally high fluxes (reaching up to 83,730 L m^{-2} h^{-1}) and outstanding separation efficiency (> 99.99 %). The Ge/PA-modified raw cotton (GPRC) can be used for gravity-driven surfactant-stabilized oil-in-water emulsions (SOWEs) separations showing very-high fluxes (up to 7080 L m^{-2} h^{-1}), and separation efficiencies reaching over 99.1 %. Furthermore, the GPRC can be employed in treating aqueous dye solutions and emulsions containing dyes, and it was found out that the GPRC possesses good antibacterial characteristics. Due to their outstanding oil/water separation abilities and eco-friendly fabrication approach, our Ge/PA-modified cellulose materials demonstrate strong potential for practical applications.

1. Introduction

The rapid growth of industries, paired with recurring oily wastewater discharges, dye-containing effluents, and chemical leaks, has induced significant ecological impairment and environmental problems. Industrial oily wastewater, originating from sectors like food, metal processing, mining, textile, oil and gas, and chemical industries, contains varying concentrations of oil ranging from 10 to 200,000 ppm. The treatment of this wastewater is crucial to meet regulatory standards for discharging oily wastewater [1–3]. Traditional oily wastewater treatment methods including in-situ burning of oil, skimmers, and flotation technologies, often struggle from high energy consumption and low efficiency. Thus, the utilization of economical materials for efficiently separating oil from water with high efficiency is essential. Recently, numerous studies have focused on constructing materials with superwetting characteristics for oil/water separation [4–9]. Zheng et al. developed a superwetting coating with liquid-like surface to modify commercial polytetrafluoroethylene (PTFE) membranes for anti-fouling and effectively separating viscous water-in-oil emulsions. [10]. Zhang and coworkers used acrylate functionalized β-cyclodextrin with acrylamide to prepare a polyacrylamide cyclodextrin hydrogel to modify stainless steel meshes. The modified stainless steel meshes could be

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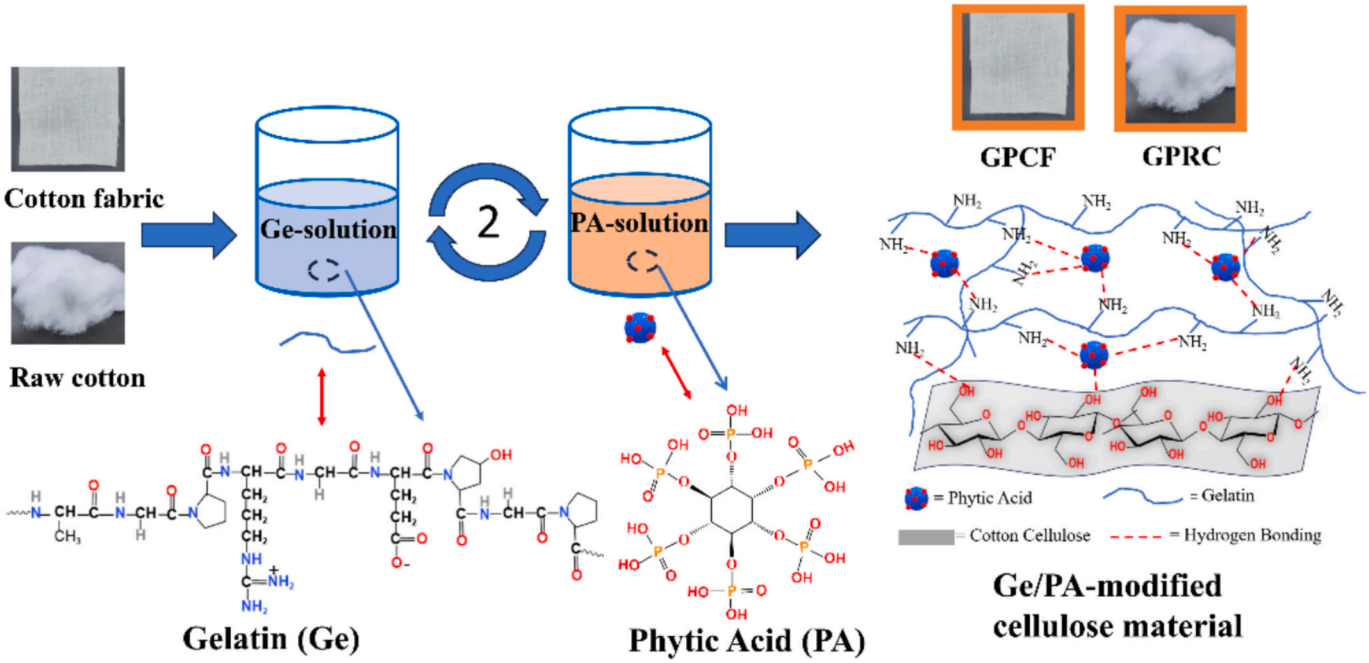

Fig. 1. Preparation of Ge/PA-modified cellulose materials.

employed in separating oil/water mixtures with high separation efficiencies [11]. Chen et al. applied metal–organic frameworks, polymethyl methacrylate, and polydimethylsiloxane to modify cotton fabric and prepared superwetting fabric. Their superwetting fabric showed superhydrophobic property and can be utilized in water-in-oil emulsions separation. Their materials also exhibited outstanding anti-icing performance [12].

Furthermore, the release of dye-containing wastewater is indeed a significant concern due to its hazardous nature and its widespread generation by industries like printing, textile, and semiconductor manufacturing [13,14]. Traditional wastewater treatment methods often struggle to effectively remove dye pollutants due to their complex chemical compositions and resistance to degradation. Moreover, the presence of various other contaminants in wastewater further complicates the treatment process [15]. Addressing this challenge requires the development of innovative materials and technologies capable of efficiently removing multiple pollutants from wastewater simultaneously.

The development of multifunctional materials capable of addressing both oil/water emulsions and dye contaminants represents a promising approach tackling complex wastewater treatment challenges, but has been rarely reported [5,16,17]. Li et al. fabricated a Janus PET textile embedded with Ag nano-particles via polydopamine coatings. The modified PET textile exhibited separation capabilities for different organic contaminants including water insoluble oil and soluble dyes [18]. Wang et al. developed a superwetting membrane through the hydrothermal treatment of ZnO/Ag nanoparticle onto a PVDF membrane. The membrane demonstrated good oil-water emulsion separation and dye removal by photodegradation and adsorption [19]. Zhang et al. fabricated a superhydrophilic-underwater superoleophobic (PVDF/CS@DA) membrane by co-depositing chitosan and dopamine onto a PVDF membrane in an acidic condition. The membrane exhibited simultaneous oil-in-water emulsion removal and anionic dye adsorption capability [20]. Wu et al. prepared a switchable underwater-superoleophobic and underoil-superhydrophobic pine pieces powder-coated PVDF membrane via a one-step deposition method. The fabricated membrane demonstrated highly efficient separation of various emulsions and dye adsorption [21]. However, most of the above-mentioned materials are photocatalyst-embedded materials that face limitations such as complex fabrication processes, using expensive materials, slow degradation rates, and reliance on UV light. Therefore, there is a strong need to develop new ecofriendly multifunctional materials using cost-effective and facile methods for the efficient separation of oil-water emulsions and organic dyes simultaneously.

Gelatin (Ge) is a natural polymer that can be derived from collagen through hydrolytic degradation. It can be used in fabricating hydrogels, nanofibers, and cell transplantation carriers [22,23]. On the other hand, phytic acid (PA) is a naturally occurring, non-toxic organophosphorus substance that can be extracted from plant seeds [24]. It exhibits strong coordination ability to metal ions because of its six phosphate groups. Scientists utilized PA metal complexes to prepare superhydrophilic coatings on both inorganic and organic substrates [25]. Building on these findings, researchers have recently constructed hydrogels from Ge and PA through hydrogen bondings for various applications such as flame retardant coatings, thermal insulators and skin tissue scaffolds [26–28]. In this research, we prepared Ge/PA modified cellulose composites (including cotton fabrics and raw cotton) with superwetting properties by simple eco-friendly layer-by-layer (LBL) procedures without employing organic solvents. The as-synthetized Ge/PA-modified cellulose composites showed superamphiphilic and underwater superoleophobic characteristics. Our Ge/PA-modified cellulose composites can be divided into Ge/PA-modified cotton fabrics (GPCF) and Ge/PA-modified raw cotton (GPRC). GPCF and GPRC can be employed for the separation of oil/water mixture and surfactant-stabilized oil-in-water emulsions (SOWE), respectively, demonstrating excellent separation performance. In the gravity-driven SOWE separation experiments, the permeation flux reached up to 7080 L m^{-2} h^{-1}, while the separation efficiency was over 99.1 %. Furthermore, the GPRC also exhibited selective cationic dye absorption ability possessing a separation efficiency >99 %. In addition, we successfully applied GPRC to treat dye-containing oil-in-water emulsions. The Ge/PA-modified cellulose composites hold great potential for real-life applications due to their eco-friendly fabrication procedure, outstanding wastewater treatment capabilities and feasibility for mass production.

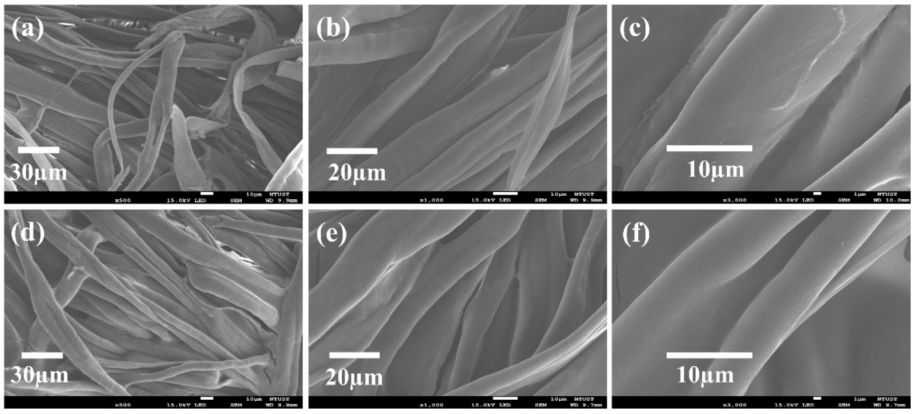

Fig. 2. SEM images of (a), (b), (c) PCF and (d), (e), (f) GPCF.

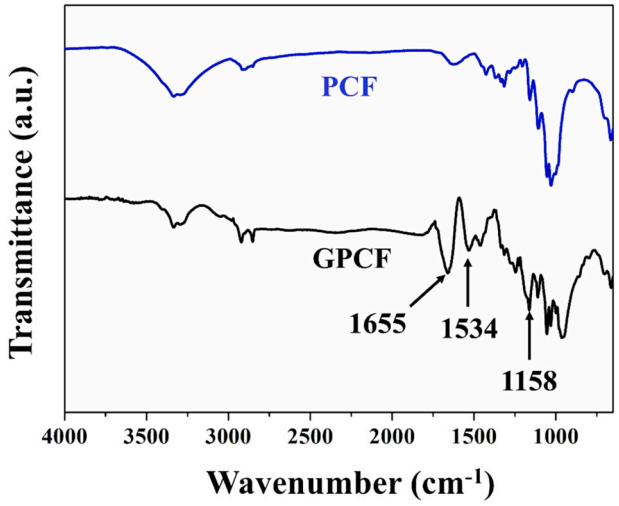

Fig. 3. FTIR spectra of the PCF and GPCF.

2. Experimental section

2.1. Materials

Comprehensive details regarding the materials can be found in the Supporting Information file.

2.2. Fabrication of the Ge/PA-modified cellulose composites

The Ge/PA-modified cellulose composites were fabricated via an eco-friendly LBL procedure (Fig. 1). Firstly, 1 g of Ge was added to 100 mL of deionized water and was stirred and dissolved at 50 °C to get a homogeneous and clear solution. PA solution of 20.0 mg/mL was also prepared. Then, cleaned cotton fabrics or raw cotton were soaked in Ge solution for 10 min. After being removed from the solution, they underwent a squeezing procedure to ensure that there is no residual solution, and were dried at 50 °C. The resulting gelatin-coated fabric/cotton was then immersed in PA solution for 5 min to form a Ge/PA double layer. The same treatment was repeated to obtain cellulose materials with two Ge/PA double layers. Finally, Ge/PA-modified cellulose composites were washed multiple times with deionized water to ensure that there are no excess reactants.

2.3. Preparation of surfactant-stabilized oil-in-water emulsion (SOWE)

To prepare SOWEs, 0.03 g of Tween 80 was added and dissolved in 100 mL of water. Then, 2 mL of oils or organic solvents was added to the solution, followed by stirring for 3 h.

2.4. SOWE separation experiments

In the gravity-driven tests for emulsion separation, compressed GPRC with a thickness of 2.7 cm was set inside a funnel and initially wetted with water to promote the facilitate the separation of SOWEs. A 25-mL emulsion was used to evaluate the flux, which represents the amount of liquid passing through an effective membrane area per unit time. The separation efficiencies for both oil/water mixtures and emulsions can be calculated based on the following formula [4]:

$$\text{Eff.}(\%) = \left(1 - \frac{C_p}{C_o}\right) \text{x } 100\% \quad (1)$$

where Eff. (%) represents the separation efficiency, C_o denotes the oil content in the feed oil/water mixtures or emulsions, and C_p corresponds to filtrates' oil content.

2.5. Dye adsorption tests

In the dye adsorption tests, 25 mg/L solution of methylene blue and Alcian blue dyes were employed as representative cationic dyes, where a total volume of 25 mL was filtered through a compressed GPRC under gravity to evaluate its dye adsorption property. Other anionic dyes like Congo red and methyl blue were also subjected to separation tests using the material. The dye adsorption efficiency was then evaluated using the following formula [8]:

$$R\ (\%) = \left(\frac{C_f - C_p}{C_f}\right) \times 100\% \quad (2)$$

where R (%) represents the dye adsorption efficiency, and C_f and C_p stand for the concentrations of the dye solution in the feed and permeate, respectively.

2.6. Antibacterial experiments

Either raw cotton or GPRC (20 mg) was introduced into 1 mL of *E. coli* O157:H7 suspended in LB broth (10^3 CFUs/mL), followed by incubation at 37 °C with constant shaking. After 24 h of incubation, the bacterial suspension was collected, and its optical density (OD) was measured using a microplate reader at an absorbance of 600 nm. The bacterial survival rate was then calculated using the following equation [29]:

$$\text{Bacterial survival rate} = (\text{OD600}_{\text{treatment}})/(\text{OD600}_{\text{control}}) \times 100\%$$

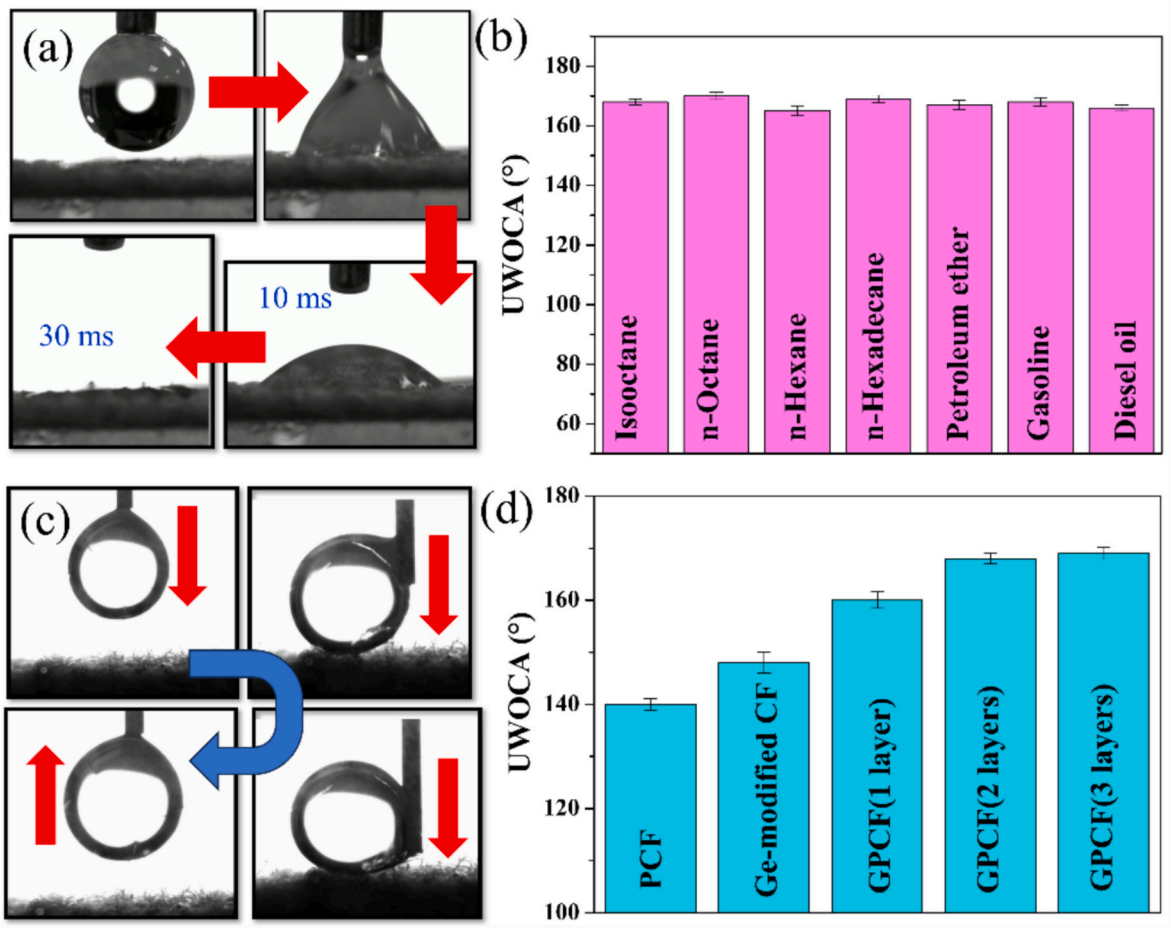

Fig. 4. (a) Approach and contact of a water droplet with respect to the GPCF. (b) UWOCAs of the GPCF. (c) Investigation of the underwater anti-oil properties of the GPCF. (d) The UWOCAs of different samples.

2.7. Instruments and characterization

Comprehensive details regarding the characterization procedures and instruments used can be found in the Supporting Information file.

3. Results and discussion

This study underscores the potential for environmentally friendly approaches in modifying cellulose materials. We fabricated Ge/PA-modified cellulose composites (both raw cotton and fabrics) through an ecofriendly LBL methodology without the use of any harmful chemical compounds (Fig. 1). First, we immersed cellulose materials in Ge aqueous solution to deposit Ge through the formation of hydrogen bondings between Ge and cellulose materials. Then, we immersed the Ge-modified cellulose materials in PA aqueous solution to form a Ge/PA composite layer on the cellulose materials through the formation of hydrogen bondings between PA and Ge [25].

We employed scanning electron microscopy (SEM) to study the morphology of the pristine cellulose materials and the Ge/PA-modified cellulose composites. In Fig. 2, the SEM images of the cellulose materials before and after Ge/PA modification were provided. Both the pristine cotton fabric (PCF) and the GPCF exhibited smooth morphology, the Ge/PA-modified cellulose composite had a morphology similar to that of the pristine material. Hence, we have limited the gelatin solution to 10 mg/mL to prevent the formation of an aggregate layer on the cellulose materials.

Energy-dispersive X-ray spectroscopy (EDS) was used to further study the surface chemical compositions of pristine cotton fabric (PCF) and GPCF. The main component of cotton fabric is cellulose, which contains the carbon and oxygen elements. Fig. S1 shows the EDS data of the PCF, revealing that the main elements of the sample are carbon and oxygen, which is in good agreement with previous findings. After the modification with Ge/PA surface coating, besides carbon and oxygen, we also found nitrogen (from Ge) and phosphorous (from PA) on the GPCF. This indicates that the PCF was successfully modified by the Ge/PA coating. Furthermore, the EDS mapping results show that the cotton fabric was uniformly coated with Ge/PA composites.

Attenuated total reflectance Fourier-transform infrared spectroscopy (ATR-FTIR) was utilized to investigate the chemical compositions (functional groups) of the PCF and GPCF. As it is indicated in Fig. 3, the ATR-FTIR spectra of the pristine cellulose fabrics show typical signals at 3300, 2900, 1162, and 1060 cm^{-1} which could be attributed to –OH stretching vibration, C—H stretching, asymmetric C-O-C stretching, and C—O stretching, respectively. Moreover, the peaks at 1030 and 895 cm^{-1} correspond to the C—H deformation, and C—H out of plane bending stretching, respectively. The peak around 1640 cm^{-1} represents the absorbance of water molecules [30]. The GPCF exhibits a new absorption band at 1655 cm^{-1}, corresponding to C═O stretching vibration, and a peak at 1534 cm^{-1} corresponding to N—H bending vibrations. These two new peaks were from the amide functional groups in gelatin. Additionally, the GPCF depicts a peak at 1158 cm^{-1} which is ascribed to the P═O group (from phytic acid) [26,27,31]. These findings indicate that a Ge-PA modified cellulose material has been successfully prepared.

To explore the wetting characteristics of the Ge/PA modified cellulose composites (GPCF and GPRC) towards water and oil, we conducted

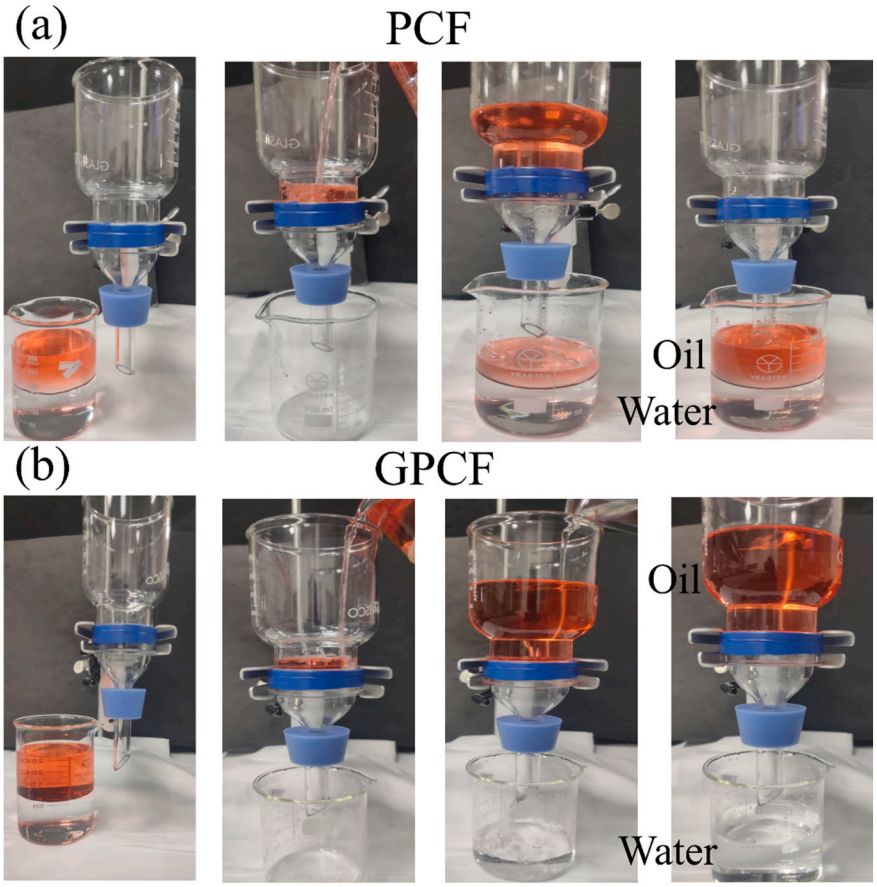

Fig. 5. Oil/water mixtures separation by using (a) the PCF and (b) the GPCF.

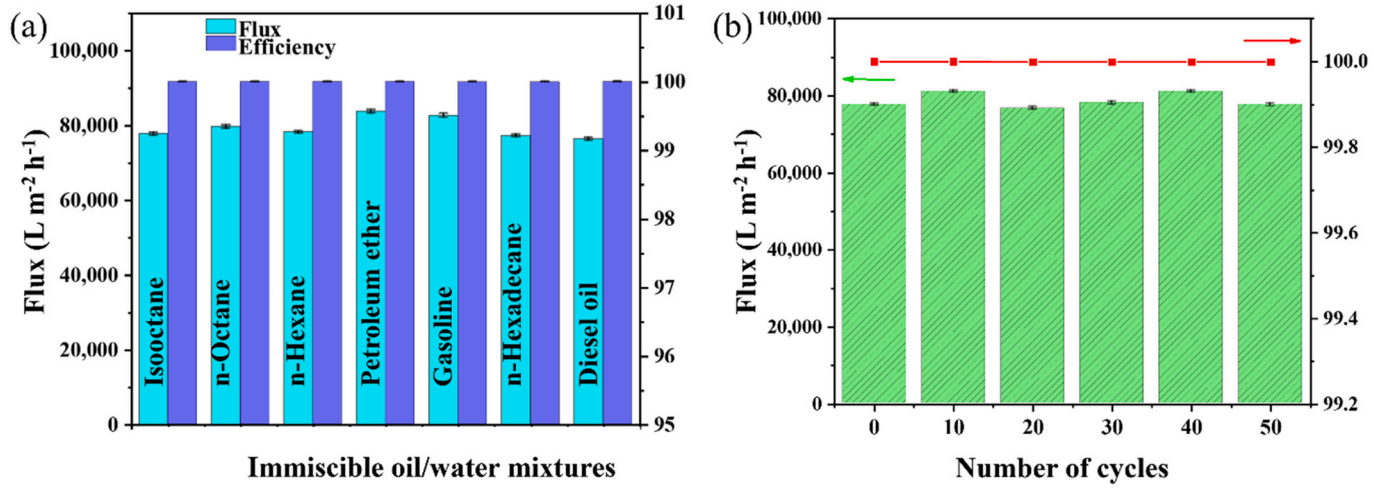

Fig. 6. (a) GPCF's separation performance against different oil/water mixtures. (b) GPCF's flux and separation efficiency during the oil/water mixture separation cycles.

experiments where their droplets' spreading behavior were monitored using a contact angle goniometer with a built-in camera system. The Ge/PA modified cellulose composites showed superhydrophilicity with a water contact angle close to 0^{o} (Fig. 4a). Similarly, when various oil droplets such as n-hexane, n-octane, isooctane, diesel, and petroleum ether were tested, comparable outcomes were observed with oil contact angles approaching 0°. These observations indicate that the Ge/PA modified cellulose composites exhibit both superoleophilic and superhydrophilic properties, demonstrating super amphiphilicity in air. Additionally, the underwater wettability of the Ge/PA modified cellulose composites were assessed. When we immersed the Ge/PA modified cellulose composites in water, the water covered their microstructures; consequently, when oil was brought in, an oil/water/solid composite interface was formed, resulting in underwater superoleophobic property. Fig. 4 shows the underwater oil contact angles (UWOCAs) for the numerous tested oils on the GPCF. All UWOCAs in Fig. 4b were greater than 150^{o}, confirming the underwater superoleophobic characteristic of the GPCF. The anti-oil adhesion properties of the Ge-PA modified

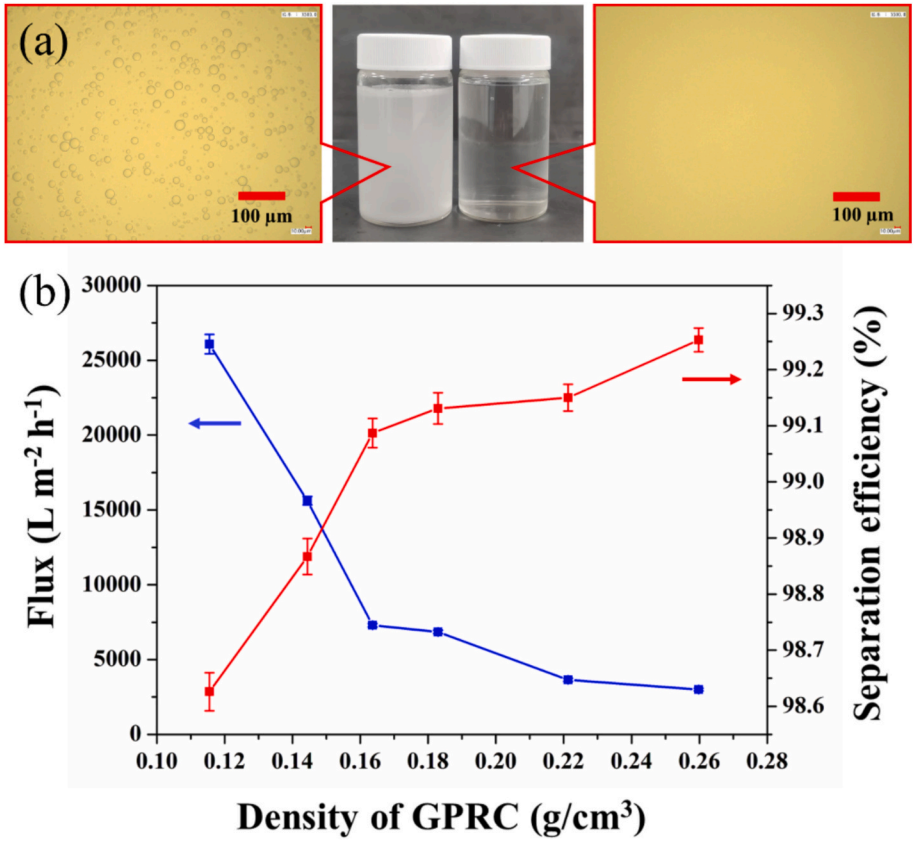

Fig. 7. (a) OM images and photographs of the SOWE separation. (b) The effect of varying the GPRC density on the flux and separation efficiency during SOWE separation.

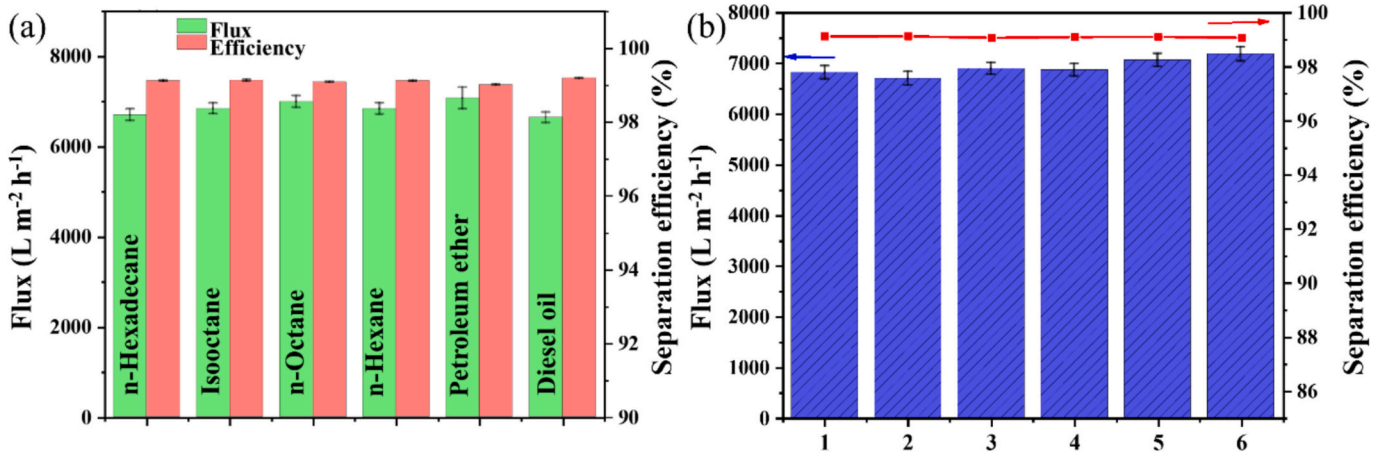

Fig. 8. (a) Fluxes and separation efficiency of SOWEs. (b) Reusability of GPRC for SOWEs separations.

cellulose materials were also examined (Fig. 4c). In this test, a suspended chloroform droplet was brought into contact with the material's surface underwater, following a sequence of approach, contact, deformation, and withdrawal. The results demonstrated that the oil droplet detached easily and completely from the GPRC surface, even when subjected to considerable compression during the underwater interaction. As shown in Fig. 4d, the UWOCA of PCF is 140°. The UWOCA of PCF can be increased to 148° through the modification of gelatin. The UWOCA of PCF can be further enhanced by Ge/PA coatings; the UWOCAs of single Ge/PA double-layer-coated fabric, double Ge/PA double-layer-coated fabric, and triple Ge/PA double-layer-coated fabric were 159, 168° and 169°, respectively. The GPCF prepared from the double Ge/PA double-layer-coated fabric was selected for the subsequent experiments throughout in this research.

The GPCF's self-cleaning and anti-fouling capabilities were also studied. It can be observed from Fig. S2 that we prewetted PCF and GPCF by water, then immersed them in n-hexadecane. Finally, we rinsed the polluted samples by water. Fig. S2a depicts that when the pristine fabric was first prewetted by water, followed by the immersion in n-hexadecane, the fabric surface will absorb the oil which could not be removed from the polluted fabric just by rinsing with water. However, the oil contaminant was observed to be easily removed from the water-prewetted GPCF (Fig. S2b), through a water-rinsing process, which indicate that the water-prewetted GPCF possesses an excellent anti-

Table 1
Comparison of different superwetting materials used for separating SOWEs.

Materials	Oil-in-water emulsion separation		Reference
	Flux (L m^{-2} h^{-1})	Efficiency (%)	
Biomimetic hydrophilic membrane	2100	>99.6	[33]
sodium alginate/chitosan-Ag modified membrane	874	>99.30	[34]
cellulose hydrogel-coated PVDF membrane	2675.2	>99	[35]
Silica-decorated microfiltration membranes	1431	>99	[36]
Cu/$Fe(OH)_3$/α-FeOOH Membrane	2598.4	>98.5	[37]
Ag/EGCG modified PVDF membrane	735	>95.4	[38]
polydopamine/PEI/CFO composite membrane	232.2	>99.5	[39]
Gelatin/phytic acid-modified raw cotton	7080	>99.1	This work

fouling capability.

GPCF has been observed to facilitate efficient gravity-driven oil/water mixture separations because of its underwater superoleophobic and superhydrophilic properties. A mixture of *n*-hexadecane/water was poured onto a pre-wetted PCF or GPCF, which was placed between two vertical glass tubes. Oil/water separation experiments were done as presented in Fig. 5. It was observed that when the oil/water mixture got into contact with the water-prewetted PCF, both water and oil layers passed through the fabric indicating that the PCF could not be employed in oil/water separation (Fig. 5a). Fig. 5b shows the process of oil/water separation, wherein water permeates through the water-presoaked GPCF quickly. On the other hand, the oil just stayed above the GPCF, indicating its oil/water separation ability.

The separation performance of the prewetted GPCF across different oil/water mixtures was further evaluated. We mixed various oils (such as n-hexadecane, isooctane, n-octane, n-hexane, petroleum ether, gasoline, and diesel oil) with water at a 1:1 *v*/v ratio. It was found that the separation fluxes achieved under gravity ranged from 77,260, 77,720, 79,600, 78,190, 83,730, 76,350, 82,560 L m^{-2} h^{-1}, respectively for n-hexadecane, isooctane, n-octane, n-hexane, petroleum ether, diesel, and gasoline oil-water mixtures, demonstrating the GPCF's outstanding separation performance (Fig. 6a). Furthermore, separation experiments revealed that the separation efficiency consistently exceeded 99.99 %, underscoring the water-prewetted GPCF's superior performance, as depicted in Fig. 6a. Additionally, the reusability of the prewetted GPCF was assessed by performing cyclic tests employing isooctane-water mixture as the feed. Even after subjecting the GPCF to 50 cycles of

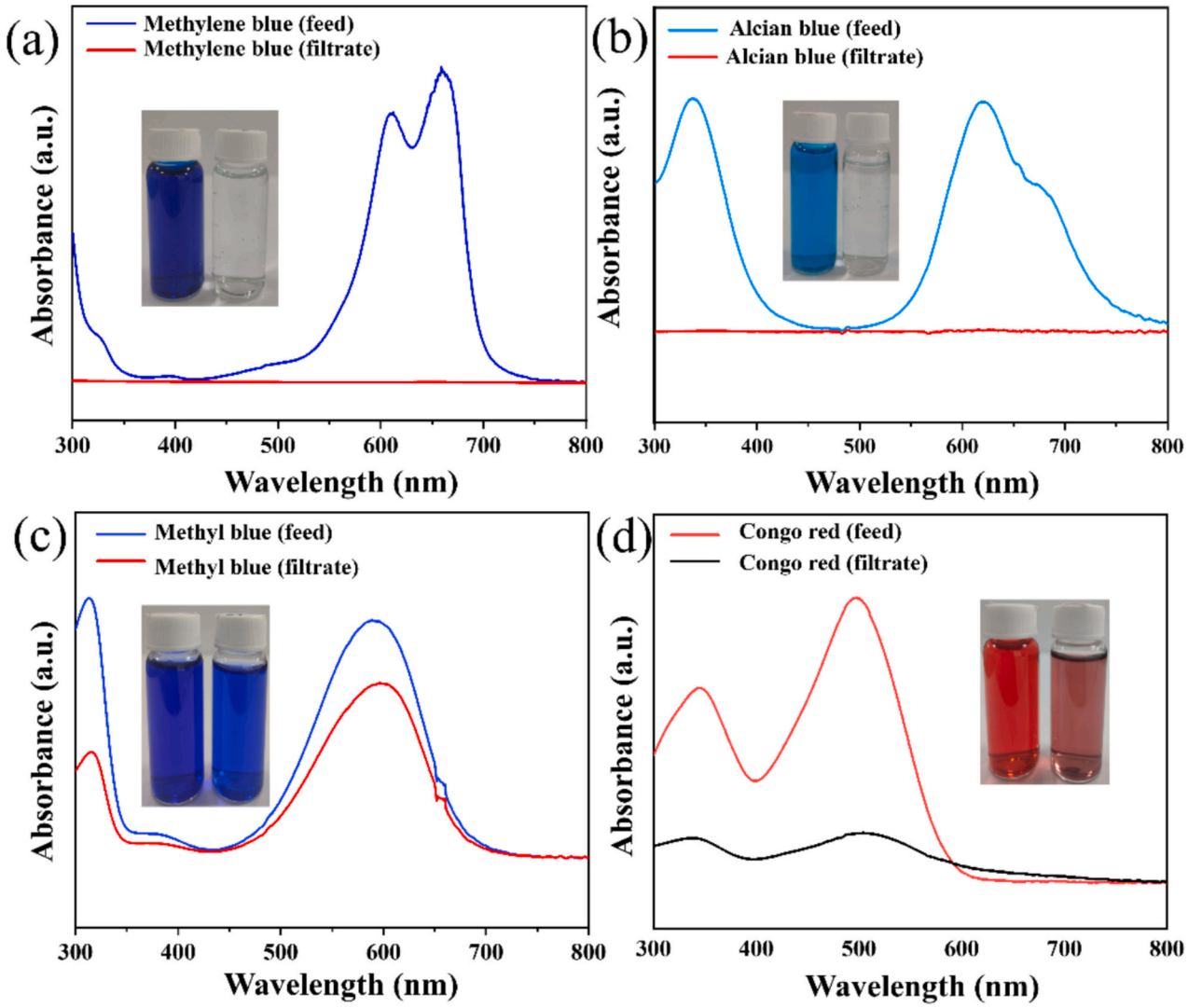

Fig. 9. Photographs and UV–vis spectra of the feed and filtrate of the cationic dyes, (a) methylene blue and (b) Alcian blue; and the anionic dyes, (c) methyl blue and (d) Congo red. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

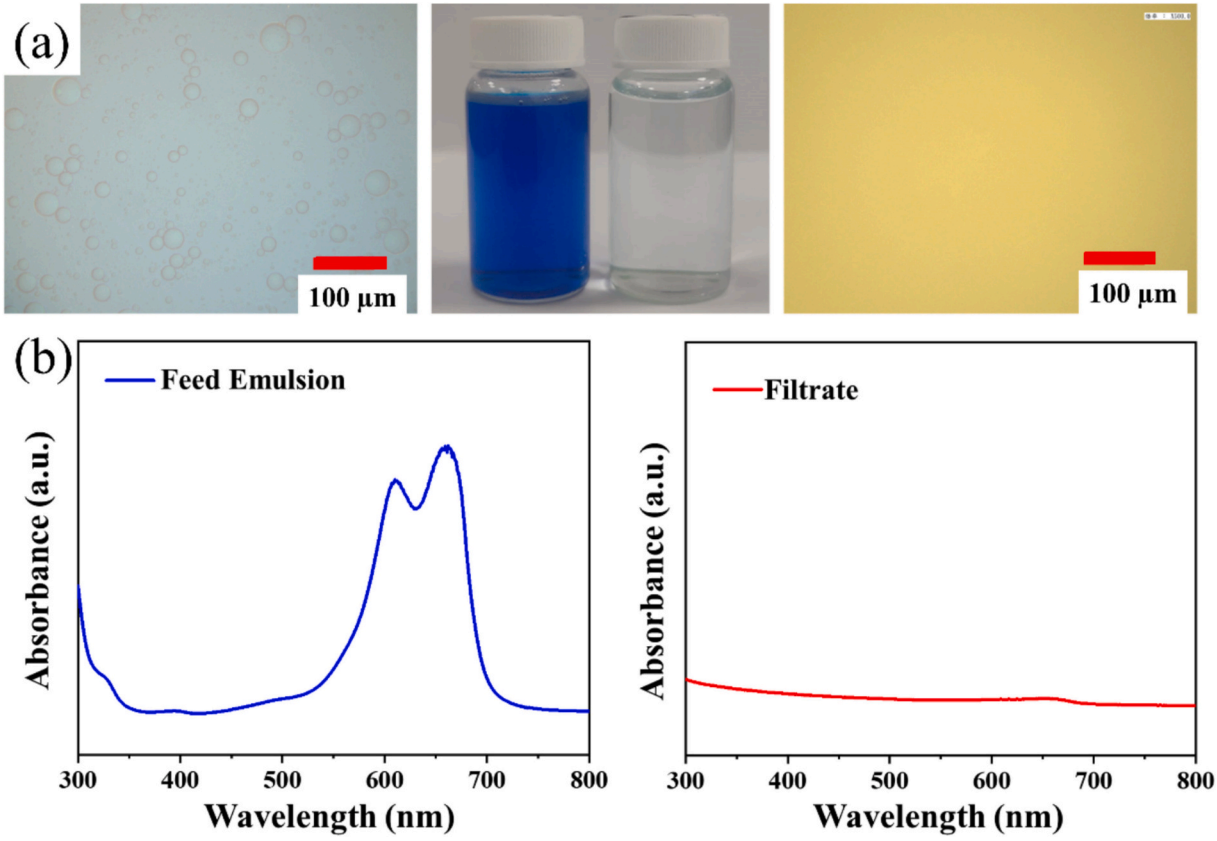

Fig. 10. (a) OM images and photographs, and (b) UV–Vis spectra of the methylene blue-containing SOWE, captured before and after the separation procedure. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

separation test, it maintained a flux of over 77,700 L m^{-2} h^{-1} and sustained a separation efficiency exceeding 99.99 %, highlighting its exceptional reusability (Fig. 6b).

Since conventional techniques for oil/water separation do not effectively treat wastewater comprising emulsified oil/water mixtures having small particle sizes, there has been a growing interest in cotton-based materials with exceptional wetting characteristics for the efficient separation of these emulsions [8,32]. Because of its underwater-superoleophobicity properties, the GPRC possesses a potential for separating SOWEs. In this study, the compressed GPRC was employed in treating SOWEs. The process for surfactant-stabilized emulsion separation using GPRC involves tightly packing the as-prepared cotton into a cylindrical funnel. Then the compressed GPRC was fully pre-wetted by water. Finally, the milky-white emulsion is poured from the top, which then allows the permeate to flow through by gravity. The photographs and optical microscopy (OM) images of the feed surfactant-stabilized isooctane-in-water emulsion and its respective filtrate are displayed in Fig. 7a. The initial emulsion feed contained numerous oil droplets with micro-nano scale diameters. During the separation process, a resulting transparent filtrate indicates that these emulsified oil droplets were effectively removed. The particle size distributions of the emulsion before and after separation were measured through dynamic light scattering (DLS). The droplet size changed from 2581 nm to 140 nm (Fig. S3). Above results highlights the successful removal of oil from the isooctane-in-water-emulsion, confirming the effectiveness of the separation method using the GPRC. The optimized density of compressed GPRC with respect to the separation performance was achieved by employing varying mass of GPRC packed into the funnel base with fixed thickness and testing their separation ability. In the isooctane-in-water emulsion separation tests, increasing the density of the compressed GPRC from 0.11 to 0.26 g cm^{-3} resulted in a flux decline from 26,090 L m^{-2} h to 3000 L m^{-2} h. Correspondingly, the separation efficiency rose from 98.6 % to 99.3 %. For the gravity-driven SOWE separations, we selected a compressed GPRC with a 0.18 g/cm^3 density because of its moderate flux and separation efficiency (Fig. 7b).

The mechanism of SOWE separation by compressed GPRC was showed in Fig. S4. When the SOWE was poured into the compressed GPRC surface, the stability of emulsion could be dissevered due to the superwetting properties of GPRC. The water was absorbed and permeated through the compressed GPRC rapidly. In the meantime, various oil droplets joined gradually with each other to form larger droplets, thereby being effectively rejected by the compressed GPRC. Above circumstances induced to the outstanding performance and very high flux for SOWEs separations of the compressed GPRC. For the separation of SOWEs prepared by n-hexadecane, isooctane, n-octane, n-hexane, petroleum ether, and diesel the fluxes are 6700 ± 130, 6845 ± 120, 7000 ± 240, 6840 ± 170, 7080 ± 250 and 6645 ± 115 L m^{-2} h^{-1}, respectively (Fig. 8a). Moreover, their separation efficiencies were determined to be greater than 99.1 %, revealing a promising oil rejection capacity of the compressed GPRC. Moreover, cyclic separation tests were performed against an emulsion to assess the recyclability of the GPRC, which is a crucial factor in emulsion separation processes. After each filtration, the GPRC was washed by ethanol, dried and re-fitted into the separating funnel. In this test, isooctane was utilized to formulate the SOWE. Fig. 8b presents that the fluxes remained relatively stable throughout the 6-cycle test. Additionally, the SOWE separation efficiencies consistently exceeded 99 % for all tests. These findings demonstrate the superior antifouling ability of the GPRC, emphasizing its potential as a highly effective material for practical emulsion separation applications. Compared with other superwetting materials utilized for SSOIWEs separation, GPRC exhibited relatively high fluxes and good separation efficiencies (Table 1) [33–39].

The dye adsorption capabilities of the GPRC were studied by employing two cationic dyes (methylene blue and Alcian blue) and two

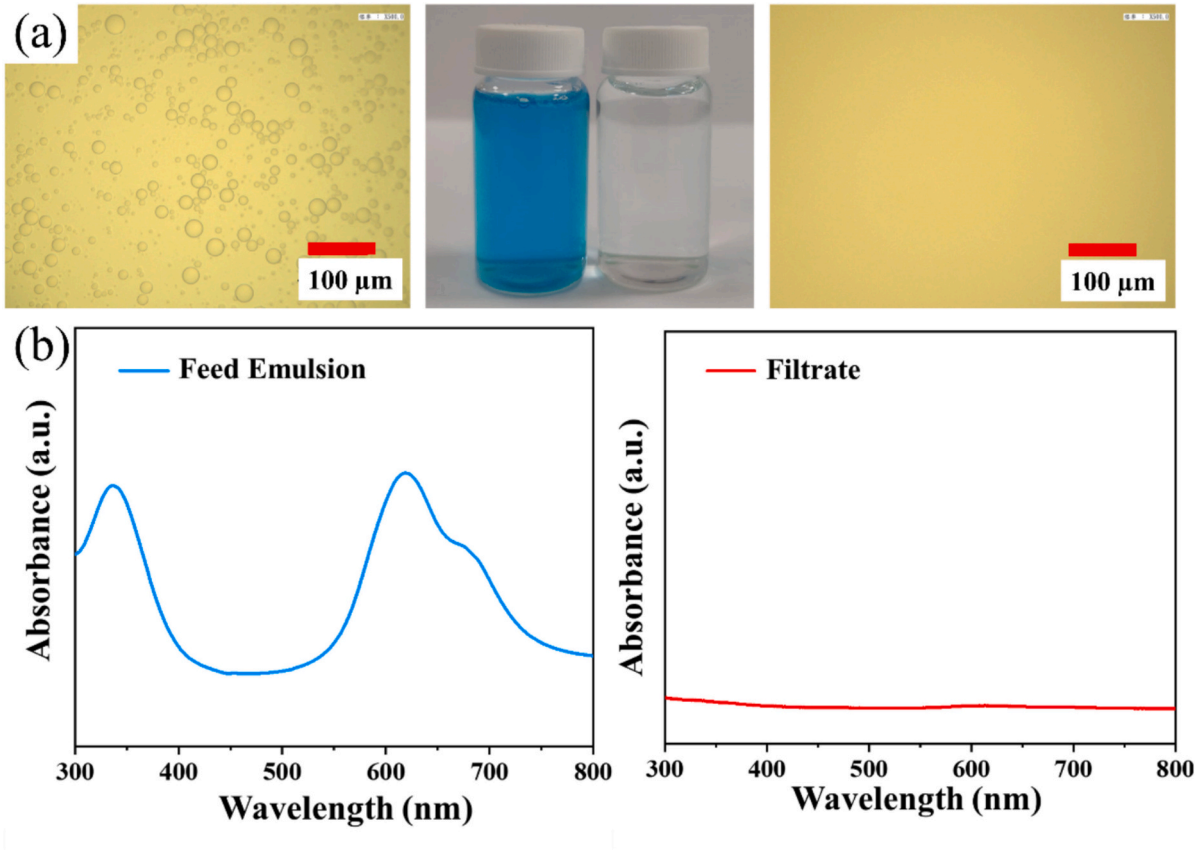

Fig. 11. (a) OM and photographic images, and (b) UV–Vis spectra of the Alcian blue-containing SOWE, captured before and after the separation procedure. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

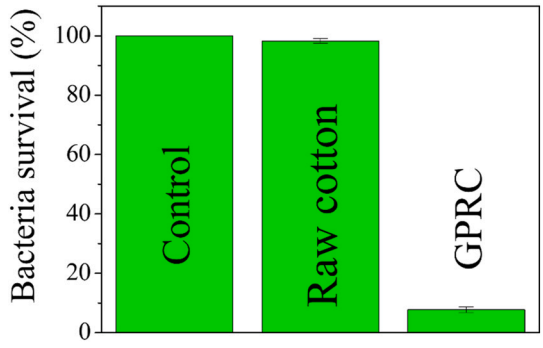

Fig. 12. The viability of *E. coli* O157:H7 cells after 24 h of incubation to the raw cotton, GPRC, and a control group.

anionic dyes (Congo red and methyl blue). Dye separation tests were conducted by using a dead-end filtration setup, and the filtrates were tested using UV–Vis analysis to evaluate the GPRC's separation efficiency. Fig. 9 illustrates that the distinctive absorption peaks corresponding to methylene blue and Alcian blue are absent in the filtrates' spectra (Fig. 9a and b), indicating successful adsorption of the positive dyes by the GPRC. The dye removal efficiencies of methylene blue and Alcian blue were 99.18 %, and 99.59 %, respectively. For the dye adsorption potential of the GPRC against anionic dye, it can be observed from Fig. 9c and d that the UV–vis spectra of the filtrates still contain the absorption peaks of methyl blue and Congo red, and the filtrates still retained the colour of the original dye solution. The selective dye adsorption abilities of the GPRC can be attributed to the presence of numerous phosphate groups in the GPRC which endow the GPRC surface with negatively charged active sites to absorb positively-charged dye molecules through electrostatic attractions [40–42].

Furthermore, the multifunctionality of the GPRC was investigated by performing separation tests of surfactant stabilized isooctane-in-water emulsions containing 25 ppm positively charged dyes (methylene blue or Alcian blue). As demonstrated in Figs. 10 and 11, the compressed GPRC shows effective removal of contaminants from complex mixtures. The feed emulsions are colorful and opaque, but upon filtration, the complex mixtures transform into colorless and transparent filtrates. OM images revealed that the presence of emulsified oil droplets are effectively removed through the filtration process and the TOC test results revealed that the emulsion separation efficiencies for methylene blue- and Alcian blue-containing SOWEs were higher than 99.1 %. Moreover, both the original emulsions and their filtrates further showed the effective rejection of the dyes from the emulsions as proven by their UV–Vis spectra where the dye removal efficiencies of methylene blue- and Alcian blue-containing SOWEs were 99.89 % and 99.93 %, respectively. These results highlight the compressed GPRC's capability for highly efficient removal of dye contaminants and emulsified oil from wastewater.

To evaluate the antibacterial activity, the optical density at 600 nm (OD_{600}) of the *E. coli* O157:H7 suspension was measured after 24 h of incubation with the cotton samples. As presented in Fig. 12, the untreated cotton exhibited minimal antibacterial impact. In contrast, the GPRC demonstrated strong antibacterial performance, achieving over 92 % reduction in bacterial population when 20 mg of GPRC was added to 1 mL of the bacterial suspension. These findings confirm the effective antibacterial nature of the CMRC material.

4. Conclusions

Separating complex contaminants from wastewater is typically challenging, causing a serious environmental concern. Herein, we reveal an eco-friendly method to fabricate cellulose materials with superwetting properties using gelatin, phytic acid and cotton-based materials without consuming any organic solvents. The Ge/PA-modified cellulose materials possessed superamphiphilic and underwater superoleophobic characteristics. In addition, GPCF was effectively used for separating oil/water mixtures, exhibiting exceptionally high permeation fluxes (reaching 83,730 L m^{-2} h^{-1}), with outstanding separation efficiencies (exceeding 99.99 %). Furthermore, the compressed GPRC presented an excellent emulsion separation ability during the gravity-driven tests for SOWEs, with extremely high fluxes (reaching 7080 L m^{-2} h^{-1}), paired with excellent separation efficiencies (exceeding 99.1 %). Additionally, we successfully treated dye solutions and dye-containing SOWEs by using the compressed GPRC. Furthermore, the GPRC also showed good antibacterial properties. The Ge/PA-modified cellulose composites have considerable possibility for practical applications through their eco-friendly construction procedure, outstanding wastewater treatment ability and feasibility for mass generation.

CRediT authorship contribution statement

Guyita Berako Belachew: Writing – original draft, Methodology, Investigation, Conceptualization. **Chien-Chieh Hu:** Supervision, Resources, Conceptualization. **Chih-Feng Wang:** Writing – review & editing, Supervision, Resources, Conceptualization. **Mei-Yi Liao:** Resources, Formal analysis. **Wei-Song Hung:** Resources, Formal analysis. **Ching-Hsuan Lin:** Resources, Formal analysis. **Shiao-Wei Kuo:** Resources, Formal analysis. **Juin-Yih Lai:** Supervision, Resources, Funding acquisition.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jwpe.2025.108658.

Data availability

Data will be made available on request.

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