



## Sawdust-based superhydrophobic pellets for efficient oil-water separation

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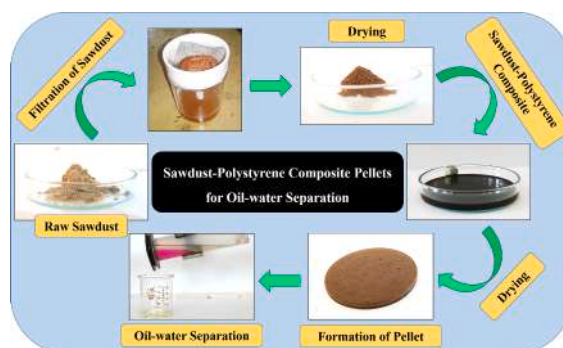
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### HIGHLIGHTS

- Low-cost Sawdust-PS based superhydrophobic pellets prepared for oil-water separation.
- Crack-free, homogeneous and robust superhydrophobic Sawdust pellets were attained.
- Superhydrophobic pellets exhibited oil-water separation efficiency higher than 90%.
- Superhydrophobic pellets revealed good mechanical strength.
- Superhydrophobic pellets could separate oil from muddy as well as warm water.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

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### ABSTRACT

Severe water pollution by means of oil is the major issue worldwide. Emerging materials like superhydrophobic surfaces have shown immense potential to control this issue. Herein we utilized low-cost Sawdust-Polystyrene (SD – PS) composite and developed a facile strategy to prepare a free-standing superhydrophobic pellet for efficient oil-water separation. More importantly, the simple recovery of the absorbed oil is feasible. To achieve crack-free, regular and robust superhydrophobic SD – PS pellet, the concentration of polystyrene, the quantity of sawdust in polymer solution and thickness of the pellet was optimised. The surface morphology analysis confirmed an adequate binding between sawdust and polystyrene in composite structure with formation of micro-voids less than 100  $\mu\text{m}$  that facilitated efficient oil-water separation. The superhydrophobic pellet exhibited oil-water separation efficiency higher than 90% for the oils and organic liquids like hexane, kerosene, diesel and coconut oil with excellent separation cycles around 30. The mechanically durable superhydrophobic SD – PS pellet could separate oil from muddy as well as warm water, which are more suitable for industrial applications.

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## 1. Introduction

Nowadays, oil-spilling is considered as one of the major pollutant in arena of water pollution and become challenging issue to remove the spilled oils [1,2]. Oil-spillage of industrial accidents which contains various oils in the waste water as well as the accident occur during the oil transport through truck or ship can spoil the land, river, and ocean [1, 2]. Likewise, oil-spills in water can cause diverse health issues such as acute and chronic effects to the surrounding population, water creatures, and birds via inhaling the oil vapours in air or consuming the oil-contaminated water or during the capture of the spilled oils [3–5]. Generally, acute effect of oil-spill can cause severe headache, dizziness, eye and skin irritation, nausea, tiredness, and respiratory issues [3–5]. Similarly, chronic effect causes the genotoxic effects, physical disorder, endocrine abnormalities, respiratory disorders [3–5]. Various methods have been used to capture the spilled oils on the water surface such as pressure driven separation boats, gravitational force driven separation, use of powdery materials, sponges and foams, nets and membranes, papers, and various other natural materials [6–12]. All the above mentioned materials might be hydrophobic or sometimes hydrophilic surfaces, whereas, hydrophobic and superhydrophobic properties are much useful for the oil-adsorption and separation [6–12]. The inspiration of superhydrophobic and self-cleaning behaviours of natural lotus as well as other plant leaves and various insects provides a broad perspective of mimicking the surface properties for the development of robust superhydrophobic surface [13–15]. The hierarchical surface morphology covering with a thin layer of low surface energy materials can easily develop the superhydrophobic property on glass or various other substrates such as textile, foam, films, paper, sponge, metal, and wood [16–24]. The applications of superhydrophobic surface are evoking wide interest in various fields of use such as oil-spill capture and separation, anti-icing, anti-fouling, anti-corrosion, anti-staining, biomedical, energy, environmental, micro-fluidic, and aerospace fields due to an excellent water repellent behaviour [6–12]. Most importantly, superhydrophobic surface can be much favourable for the oil-spill capture and separation application due to the complete water-repellent behaviour as well as higher oil loving ability of the superhydrophobic surface which helps for the selective removal of oil from the oil-water mixture [25]. The superhydrophobic property can be easily fabricated on various substrates such as by simple modification with hydrophobic agents for the selective removal and separation of various oils from the oil-water mixture [26]. The pressure-free gravity-driven separation of oil-water mixture is playing an important and most convenient way to separate oils from water [27]. Superhydrophobic surfaces have the drawback of losing the surface property to some extent of contact angle after some cycles of separation [28]. The maintenance of superhydrophobic surface stability on the fabricated substrate is the most important requirement for the continuous oil-spill separation. The drawback of superhydrophobic property in the continuous oil-water separation can be overcome by fabricating a chemically and mechanically stable robust superhydrophobic surface property [29,30].

Apart from conventional superhydrophobic materials, new materials such as candle soot [31–33], corn straw fibers [34], natural leaf mesh [35], waste chicken feathers [36], cigarette filters [37], and sawdust [38–43] are being used for efficient oil-water separation. Nowadays, the bio-degradable sawdust is one of the emerging materials in superhydrophobic oil-water separation applications. The recent research focused on the use of sawdust for oil-water separation adopts the deposition of sawdust particles on metallic mesh [38], sawdust based porous foams [39], merely superhydrophobic sawdust particles [40], modified wood slice [41], activated charcoal from sawdust [42], magnetic and superhydrophobic sawdust particles [43]. Usually, the raw sawdust was filtered through the metallic mesh to procure sawdust particles of desired size. Zulfikar et al. [38] have deposited sieved sawdust particles on adhesive coated surface (glass and SS mesh) followed by dip coating of silicone polymer and again deposited carbon

soot particles through candle flame with subsequent dip coating of silicone. This process improved the superhydrophobicity as well as mechanical durability of the coated surface. The robust superhydrophobic SS (Stain steel) mesh revealed excellent oil separation efficiency (>90%) for light and heavy oils. Tan et al. [39] have prepared light-weight honeycomb-like porous carbon foam and polymer foam from liquified larch sawdust. The prepared foams revealed excellent selectivity, recyclability and oil absorption capacity up to 153 times their own weight due to its interconnected porous microstructure. Zang and researchers [40] have prepared superhydrophobic-superoleophilic sawdust particles by blending sawdust particles in the mixture of OTS-modified silica nanoparticles and polystyrene. The prepared superhydrophobic sawdust particles were put in oil-water mixture which took 90 min to collect 3 mL gasoline. Though the sorption uptake was found comparably high, the separation time was considerably long. The various oils from water-in-oil emulsion was effectively separated with high separation efficiency (>98%) and excellent recyclability by Bai et al. [41] using superhydrophobic wood slice. In typical fabrication of the superhydrophobic-superoleophilic wood slice, the copper hydroxide modified Pine wood slice (thickness ~ 1 mm) was immersed in ethanol solution of dodecanethiol to attain superhydrophobicity. Rajak et al. [42] have synthesized porous activated charcoal from Indian white teak tree sawdust using an impregnation method with various activating agents like phosphoric acid, zinc chloride, and ferrous sulfate heptahydrate. The activated charcoal synthesized using phosphoric acid exhibited higher adsorption capacity to effectively separate oil from oil-in-water emulsion. Gan and researchers [43] have treated the sawdust particles with cobalt ferrite by hydrothermal method and then the obtained magnetic sawdust particles were modified with polysiloxane to attain superhydrophobicity. These magnetic and superhydrophobic sawdust particles were put in oil-water mixture and by utilizing bar magnet, the oil absorbed sawdust particles were collected. Similarly, the sawdust particles can hydrophobically modified by using various hydrophobic agents as well as polymers for enhancing the anti-wetting behaviour, whereas, the surface wettability and water adhesion on the modified sawdust surface depends on various parameters such as kind of hydrophobic modifier, concentration, reaction time, surface roughness, and surface energy. The use of hydrophobic polymer with sawdust particles can enhance the stability and robustness by their strong adhesion and also protect the dust particles comes out from the polymer. Moreover, the low surface energy polymer covers on the sawdust particles might increase the micro-nano hierarchical morphology which is required for developing the anti-wetting surface.

Usually, the limited research carried on the use of sawdust particles for oil-water separation adopted multi-step, multi-chemicals, time consuming and complicated deposition strategies as well as intricate oil-water separation processes. Apart from sawdust used in superhydrophobic mesh, the oil-water separation was carried out by spreading superhydrophobic sawdust particles in oil-water mixture. This strategy is not facile as proper recovery of absorbed oil from superhydrophobic sawdust particles can be critical at both laboratory and industrial scale. To avert the use of merely superhydrophobic sawdust particles for oil-water separation, we have prepared the superhydrophobic sawdust-polystyrene composite pellets for effective separation of oils from oil-water mixtures. To the best of our knowledge, we are reporting for the first time the use of superhydrophobic SD-PS composite pellets for oil-water separation. Our fabricated substrate exhibited excellent superhydrophobic property as well as selective separation of various oils from water, muddy water and warm water by gravitational force driven separation. Moreover, the fabricated substrate also performs excellent stability for continuous separation of various oils which also proved the robust nature of the fabricated pellet.

## 2. Experimental

### 2.1. Materials

Mainly the woods of different types namely *Azadirachta indica* (neem), *Acacia nilotica* (babul) and *Shorea robusta* (sal) were cut in local sawdust mill which was our source of collection of sawdust. Materials such as Stainless steel mesh (pore size  $\sim 125 \mu\text{m}$ , Hengshui Zhongbao Trading Co., Ltd. Hebei, China), Chloroform (Extra Pure AR, Sisco research Laboratory, India), Polystyrene (Average Molecular Weight  $\sim 192,000$ , Sigma-Aldrich, USA) were bought of standard quality and made for laboratory usage.

### 2.2. Preparation of SD-PS composite pellets

At first, the sawdust was washed several times with copious amount of water and then passed through the SS mesh to collect the sawdust particles of size less than  $125 \mu\text{m}$ . The filtered sawdust particles were dried in oven at  $80^\circ\text{C}$  and grinded using agate mortar and pestle to achieve fine powder of sawdust. A  $100 \text{ mg/mL}$  of sawdust was added separately in the solutions of 50, 75, 100 and  $150 \text{ mg/mL}$  concentrations of PS/chloroform under magnetic stirring for 30 min. The total quantity of each mixture was maintained at 15 mL and poured in petri dish of diameter  $\sim 7 \text{ cm}$ . The mixture was kept for drying at room temperature for 24 h. The SD-PS pellets were collected and labeled as SP-1, SP-2, SP-3 and SP-4 for the different concentrations 50, 75, 100 and  $150 \text{ mg/mL}$  of PS in chloroform, respectively. Fig. 1 reveals the detailed experimental procedure of fabrication of SD-PS pellets. We also performed the experiment by changing the concentration of sawdust particles by fixing the PS contents. The prepared pellets showed brittle nature, so, we kept the optimum content of sawdust particles to  $100 \text{ mg/mL}$  and varied the PS amount to anti-wetting surface property.

### 2.3. Characterizations

The prepared SD-PS pellets were characterized by Field Emission Scanning Electron Microscopy (JEOL, JSM-7610F, Japan) for surface morphology analysis, X-ray photoelectron spectroscopy (XPS) (Thermo Scientific Escalab 250Xi, USA) for determination of chemical composition, and Contact angle meter (HO-IAD-CAM-01, Holmarch Opto-Mechatronics Pvt. Ltd. India) to study the wettability. The contact

angle (CA) measurement was performed by placing a water and oil drops ( $\sim 8 \mu\text{L}$ ) at different places on pellets. The average value of contact angles were considered as ultimate contact angle of the pellet. In typical oil-water separation experiments, hexane, kerosene, diesel and coconut oil were used as model oils. To build the oil-water separation setup, the bottom part of transparent plastic measuring cylinder ( $100 \text{ mL}$ ) was cut and SD-PS pellet was attached on it using epoxy resin glue. In between plastic cylinder and pellet, the circular rubber with hole as of plastic cylinder was inserted to insure good contact. This assembly was clamped on retort stand at an angle of nearly  $30^\circ$ . The water was dyed blue, red and pink for easy identification. The oil-water separation efficiency ( $\eta$ ) was calculated based on the following equation [35].

$$\eta (\%) = (1 - m_{\text{out,in}}/m_{\text{in,out}}) \times 100\% \quad (1)$$

Where  $m_{\text{in,out}}$  is the mass of the oil in the initial oil-water mixture and  $m_{\text{out,in}}$  is the mass of the oil within the permeated liquid through the superhydrophobic pellet.

## 3. Result and discussion

### 3.1. Surface morphology and chemical analysis of the pellets

It's been often demonstrated that the superhydrophobic surfaces can be achieved by merely roughening the low surface energy materials. In our case, the surface morphology analysis confirmed that the sawdust particles provided requisite roughness in SD-PS composite pellets. As the raw sawdust was filtered through the SS mesh of pore size  $\sim 125 \mu\text{m}$ , the sawdust particles between  $40$  and  $125 \mu\text{m}$  were observed in Fig. 2a. The binding of sawdust particles by polystyrene was observed in case of SP-1 pellet with irregular gaps between the structures (Fig. 2b), however the composite structure was found to be very delicate and improper handling resulted in the breakage of pellet. This affirms the inadequate concentration of polystyrene in SD-PS composite pellet. For increased concentration of PS, SP-2 pellet revealed rough morphology having aggregated sawdust particles covered by polystyrene with non-uniform micro-voids (size  $\sim 100\text{--}250 \mu\text{m}$ ) (Fig. 2c). The fast evaporation of chloroform could have resulted in the formation of non-uniform micro-voids in the pellet. The rough morphology was also observed in case of SP-3 pellet (Fig. 2d), however with comparably less size of micro-voids (size  $< 100 \mu\text{m}$ ) due to low evaporation rate of chloroform from the composite structure. The higher magnification SEM image of SP-3 pellet

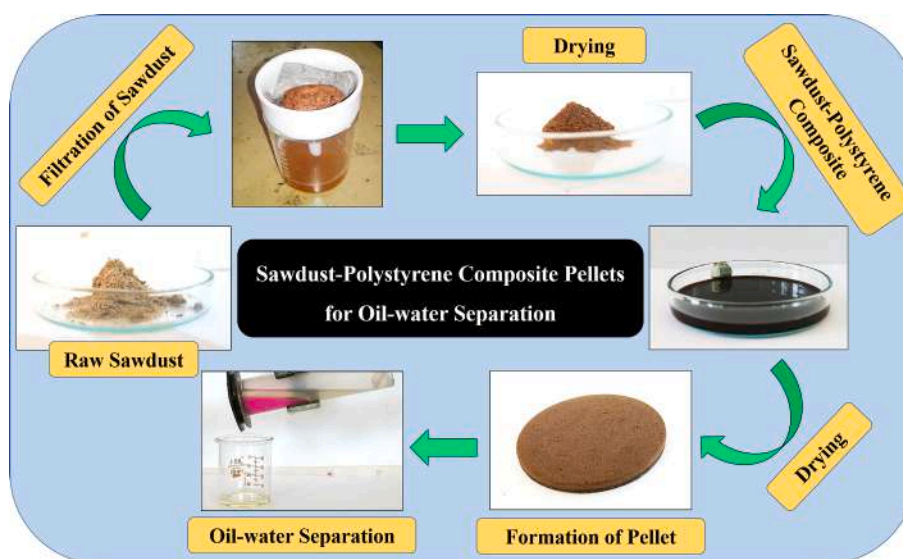
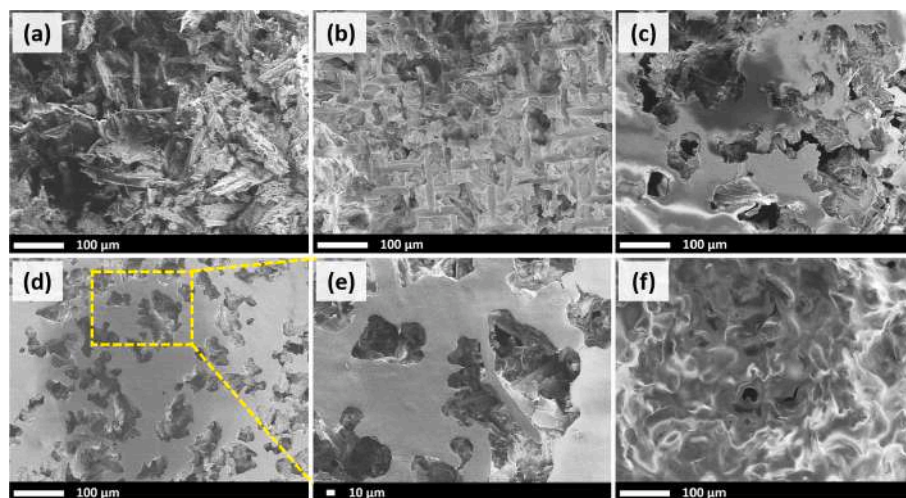


Fig. 1. The detailed experimental procedure of fabrication of SD-PS pellets.



**Fig. 2.** FE-SEM images of (a) filtered raw sawdust, (b) SP-1, (c) SP-2, (d) SP-3, (e) magnified image of SP-3, and (f) SP-4 composite pellet, respectively.

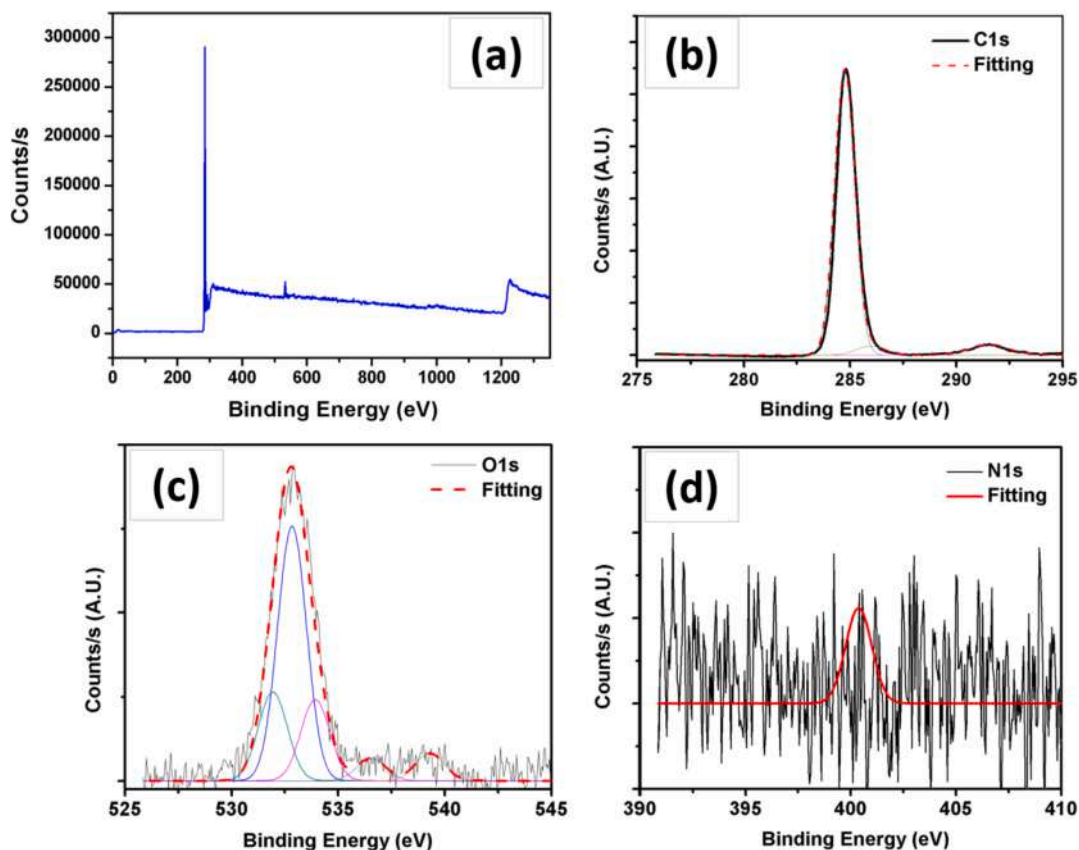
depicts full coverage of polystyrene on sawdust particles forming stable SD-PS composite (Fig. 2e). In case of SP-4 pellet (Fig. 2f), the sawdust particles were completely hidden by the high loading of polystyrene in composite which smoothen the rough structure contributed by the sawdust particles and also suppress the micro-voids.

Fig. 3 shows the broad survey with highly resolute XPS spectra with peaks of C1s, O1s and N1s originating from sample. The C1s peaks confirms the existence of three different kinds of carbon atom in the synthesized composite, having characteristic peaks around 284.17, 285.4, and 288.06 eV. The signatory bonds of C–C and C–H bonds

associated with C1s peak along with low intensity peaks occurring from  $\pi$ - $\pi^*$  transitions in the aromatic ring are present in the spectrum. The O1s peaks affirms the existence of signals related to the carbonyl oxygen (O–C) of methyl group, C=O and O–C=O and the little N1s peaks sustains from the sawdust. There are no additional peaks present in the spectra which indicates the lack of impurities in the sample.

### 3.2. Wettability of the pellets

A sawdust is completely hydrophilic ( $\theta \sim 0^\circ$ ) whereas a thin film



**Fig. 3.** (a) A broad-survey XPS spectrum and high-resolution XPS spectra of peaks (b) C1s, (c) O1s and (d) N1s, generated from sample, respectively. (Vision software obtained from Kratos was used for peak analysis by fitting Gaussian peaks with different parameters with a straight baseline).

prepared by polystyrene (50 mg/mL) is hydrophobic ( $\theta \sim 96^\circ$ ) in nature. Our preliminary experiments were based on the preparation of the SD-PS composite pellets with PS/chloroform concentrations in between 10 and 50 mg/mL. However, the pellets were not formed completely and severe visible cracks were observed in the structure which shows the level of frailness of the sample. The water contact angles measured on these pellets found in the range of  $100^\circ$ – $112^\circ$ . Hence to achieve regular, crack-free and robust SD-PS composite pellet, the concentration of PS/chloroform was further increased above 50 mg/mL. The thickness of the pellets were measured using micrometer screw gauge, where each pellets revealed nearly same thickness. The pellet thickness of 1 mm was optimum and further increase in thickness resulted in substantial cracks in the pellet. On the other hand, the attempt of making pellet thinner than 1 mm caused firm attachment of composite material on petri dish and its forceful detachment lead to complete breakage.

The wetting properties of SP-1, SP-2, SP-3 and SP-4 pellets were investigated by water and oil contact angle measurements (Fig. 4). The shape of SP-1 pellet was observed to be uneven and curvy due to very fast evaporation of chloroform from the composite. All the SD-PS composite pellets exhibited oil contact angle of  $0^\circ$ , however they exhibited different water contact angles. On the SP-1 and SP-2 pellets, the water drops stacked on the surface without rolling with contact angle of  $123^\circ \pm 5^\circ$  and  $135^\circ \pm 3^\circ$ , respectively. The irregular gaps and micro-voids present between the microstructure might act as locking sites for water drop which controls the free movement of the drop (i.e. Wenzel's wetting state). The SP-3 pellet revealed water contact angle of  $152^\circ \pm 1^\circ$  whereas the water drops rolled off at an angle of nearly  $10^\circ$ . The rough surface morphology having micro-voids size less than  $100 \mu\text{m}$  could effectively trap the air pockets underneath the water drop resulting in

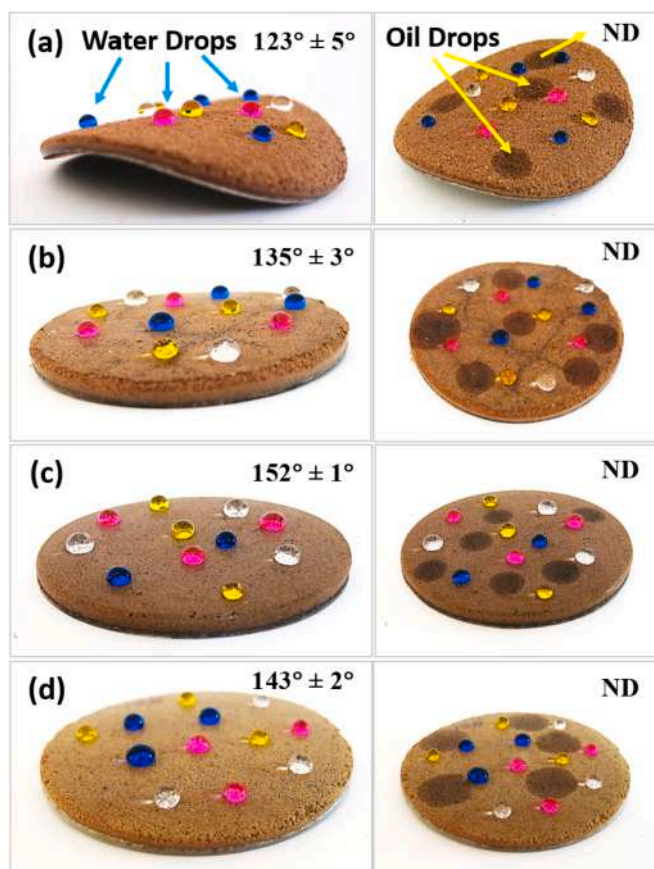


Fig. 4. Photographs of water and oil drops on (a) SP-1, (b) SP-2, (c) SP-3, and (d) SP-4 composite pellets, respectively. Note: ND- Not Detected for oil contact angle.

superhydrophobic and non-sticky nature of SP-3 pellet (i.e. Cassie-Baxter's wetting state). The rough microstructure eventually gets rather smoothed due to addition of high concentration of polystyrene in the composite structure. Therefore, the SP-4 pellet revealed water contact angle of  $143^\circ \pm 2^\circ$  and rolling angle of nearly  $19^\circ$ . Thus the appropriate surface roughness together with low surface free energy of solids can heavily contribute towards superhydrophobicity.

### 3.3. Oil-water separation

We have studied the mechanism of oil-water separation in superhydrophobic SD-PS pellet. As shown in Fig. 5a, SP-3 pellet immersed in the water reveals a shiny plastron layer resulted from the total internal reflection of light from the trapped air in the rough structure of superhydrophobic pellet which further proves the superhydrophobic and non-stick property of the fabricated substrate. A diesel oil was slowly dropped in the beaker (Fig. 5b) which was eventually absorbed by the pellet and subsequently the plastron layer disappears from oil absorbed area releasing air bubbles. This indicates an excellent oil loving behaviour of the fabricated superhydrophobic substrate. As soon as diesel invades pellet, it pushes off the air trapped from the rough structures. The air bubbles were continuously releasing off the pellet surface until the diesel completely gets absorbed in the pellet (Fig. 5b–f). The disappearance of plastron layer is due to replacement of air from solid-air interface by oil. Hence, during the oil-water separation process, oil completely get absorbed in the pellet and pass through making water-oil-solid interface.

The oil-water separation ability of the prepared SD-PS composite pellets were investigated using oil-water separation assembly with various model oils like hexane, kerosene, diesel and coconut oil. The oil-water separation assembly was inclined at an angle of  $30^\circ$  and mixture of water and oil (15 mL each) was poured as shown in Fig. 6. The oil-water separation performance of SP-1 pellet was found to be very poor where both oil and water passed through the pellet in quick succession. The same situation occurs with SP-2 pellet, however oil passes through the pellet very rapidly, whereas water takes time of about 75 s. The irregular gaps and micro-voids present between the microstructure of SP-1 and SP-2 pellets were responsible for easy penetration of both oil and water through the pellets. Also the strong stickiness of water towards pellets could have resulted in easy passage of water through the pellet under gravity. The superhydrophobic SP-3 pellets exhibited efficient oil-water separation as shown in Fig. 6. The various oils like hexane, kerosene, diesel and coconut oil from water were separated effectively. The time required to separate 15 mL of hexane (Fig. 6a), kerosene (Fig. 6b), diesel (Fig. 6c) and coconut oil (Fig. 6d) through SP-3 pellet was 2.35 min, 12 min, 23 min, and 5 h, respectively. The different separation time for

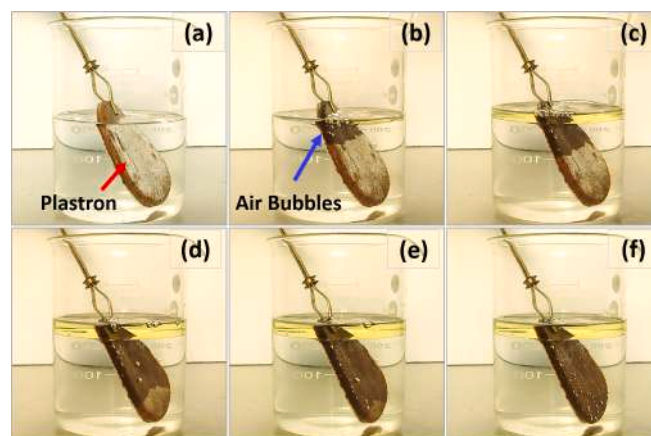


Fig. 5. Photographs of (a) Shiny plastron layer on SP-3 pellet and (b–f) disappearance of plastron layer due to oil-water interface on pellet.

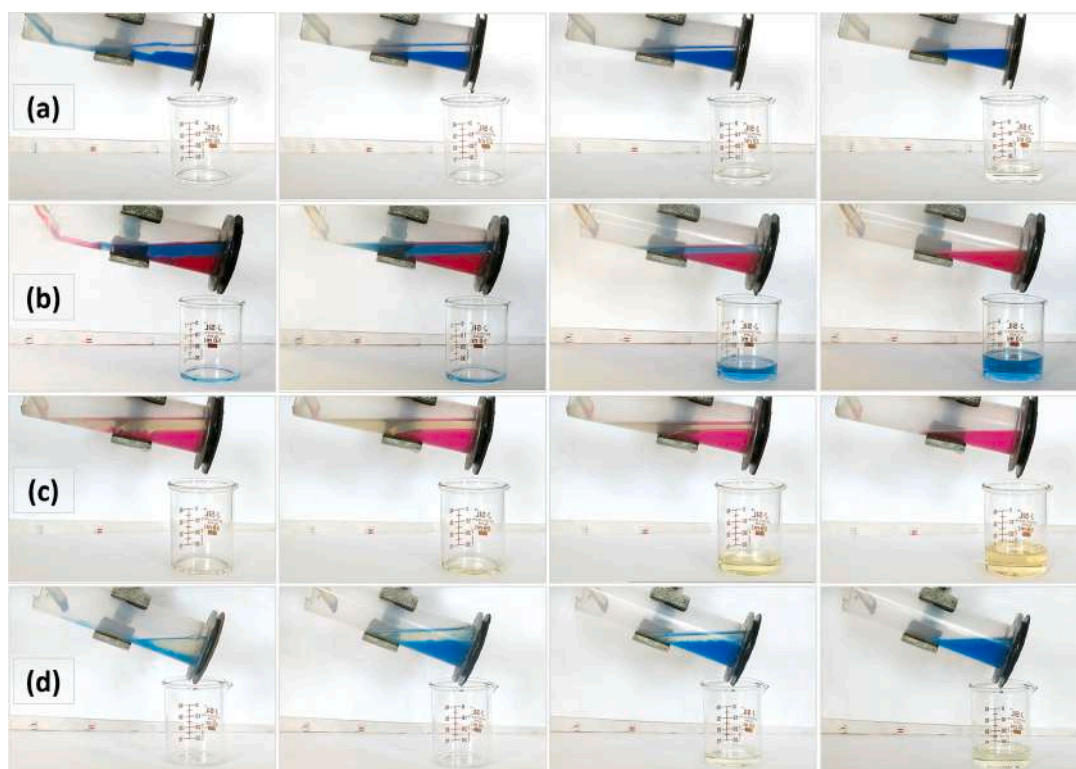


Fig. 6. Oil-water separation by SP-3 pellets, (a) hexane, (b) kerosene, (c) diesel, and (d) coconut oil, respectively.

different liquids is primarily due to their respective absolute viscosities (cP). The hexane, kerosene, diesel and coconut oil have the absolute viscosities of 0.3 cP, 2.0 cP, 2.4 cP, 55 cP, respectively. An appropriate surface roughness and micro-voids less than 100  $\mu\text{m}$  present in the microstructure were responsible for efficient oil-water separation by SP-3 pellets. Though the SP-4 pellets revealed complete oil-water separation, the separation time increased slightly for all the oils. As the high loading of polystyrene in composite suppress the surface roughness as well as the micro-voids, the increased separation time can be appreciated.

The pellet prepared in the absence of PS binder can show brittleness which is unable to use for the oil-water separation. So, we used our optimised condition for checking the separation efficiency and stability of the superhydrophobic SP-3 pellet after performing several numbers of separation cycles with the above oils used in the experiment (Fig. 7). The results expressed that the fabricated superhydrophobic SP-3 pellet can show excellent separation efficiency for various oils from water. However, the present work designates better separation efficiency ( $98 \pm 1\%$ ) for hexane than the other oils used for the separation experiment as it maintained over 98% of separation efficiency even around 36 repeated cycles of hexane separation from water. In addition, kerosene ( $96.8 \pm 1.3\%$ ) and diesel ( $95 \pm 1.2\%$ ) also performed better separation efficiency and stability as like hexane. On the other hand, coconut oil showed around  $88 \pm 2.7\%$  of separation efficiency due to high viscosity of the oil which take much time to separate than the other oils. The obtained results also illustrate the low viscosity oil (hexane) can show much faster and efficient separation as well as excellent separation efficiency than the high viscosity oils used for the separation. This is due to easier penetration of low viscosity oil through the voids in the superhydrophobic SP-3 pellet which induces the faster separation of low viscosity oils. The stable separation of various oils in the repeated cycles highlight an excellent stability of superhydrophobic (SP-3) pellet for oil-water separation. The prepared superhydrophobic pellet also have the ability to absorb the oils from water, whereas, our primary focus on the

use of the prepared superhydrophobic pellet for oil-water separation application as well as study the robustness of the pellet. So, we studied the stability of the prepared superhydrophobic pellet at various conditions.

Industrial oily wastewater is normally expelled in rivers. The separation of oils from muddy water is one of the important task. We also checked further practical use of the fabricated superhydrophobic SP-3 pellet in the model of oil-water separation for industrial separation application. We prepared a model solution of muddy water (15 mL) mixed with kerosene oil (15 mL) and performed similar operation of the use of superhydrophobic SP-3 pellet in industrial separation (Fig. 8a). The muddy water mixed kerosene oil showed faster ( $\sim 12$  min) and selective separation of kerosene oil from the muddy water in the absence of any

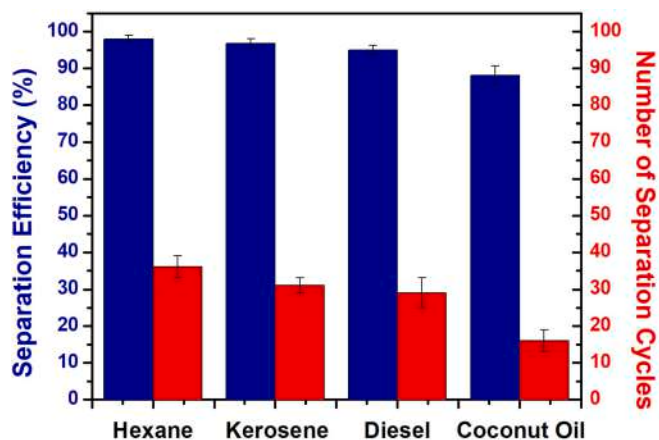
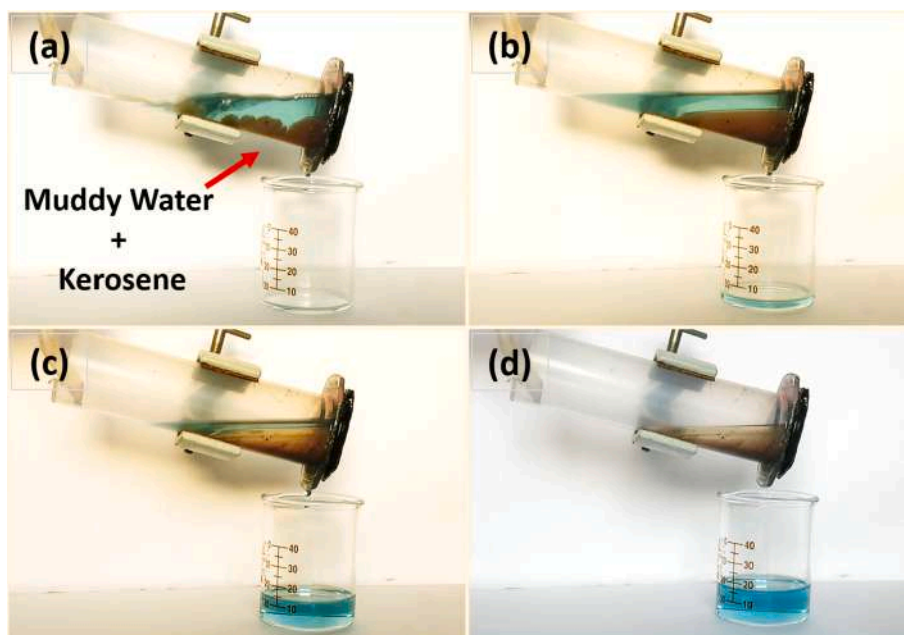


Fig. 7. Separation efficiency and recycle ability of various oil-water mixtures by the superhydrophobic SP-3 pellet.



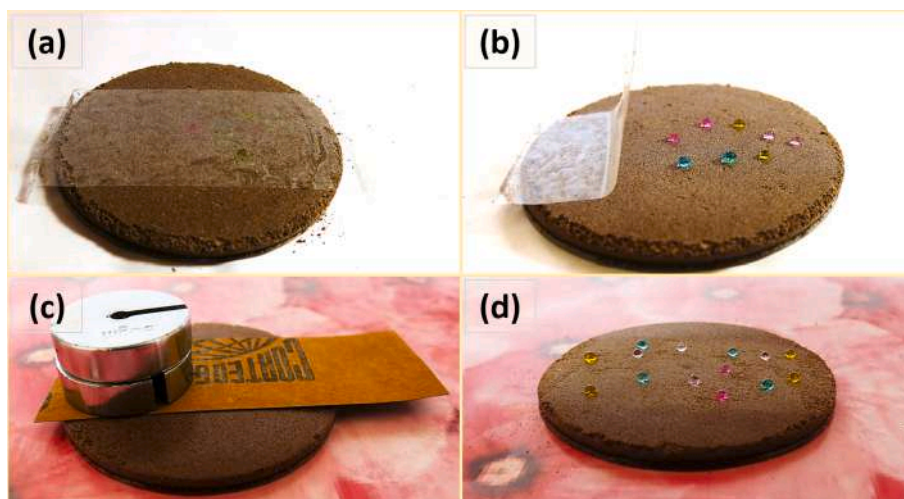
**Fig. 8.** Muddy water - kerosene separation by superhydrophobic SP-3 pellet at before separation (a), after 3 min of separation (b), after 8 min of separation (c), and after 12 min of separation (d).

pressure-driven separation (Fig. 8b–d). These results outcome the faster separation occurred by the gravitational force induced separation of various oils. The selective separation of kerosene from the muddy water mixture suggest the much possibility to use the fabricated superhydrophobic SP-3 pellet for real application in industrial need of oil-water separation.

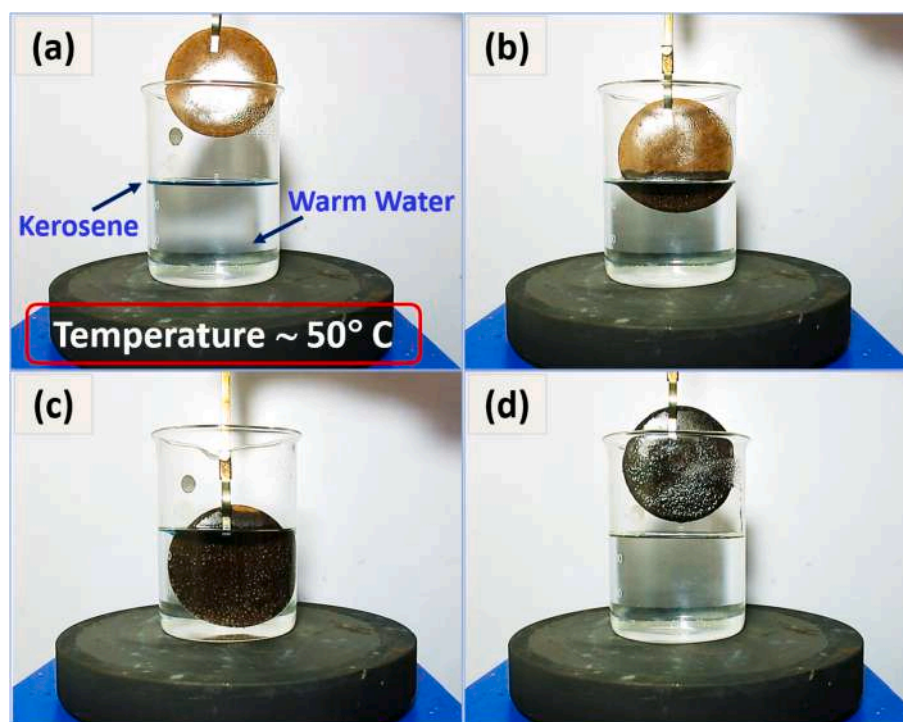
#### 3.4. Durability tests of superhydrophobic pellet

The durability of the superhydrophobic SP-3 pellet was checked further by adhesive tape and sand paper abrasion methods (Fig. 9). First, we checked the durability of superhydrophobic SP-3 pellet using adhesive tape test method by fixing an adhesive tape on the superhydrophobic SP-3 pellet surface and gently peeling-off the adhesive tape followed by checking the superhydrophobic SP-3 pellet surface wettability on the surface (Fig. 9a). The superhydrophobic SP-3 pellet

maintained the excellent superhydrophobic stability even peel-off the adhesive tape for more than 100 times. Moreover, no traces of superhydrophobic SP-3 pellet were found on the adhesive tape surface which also illustrates an excellent stability and durability as well as robustness of the fabricated substrate (Fig. 9b). The durability of the superhydrophobic SP-3 pellet was confirmed further by checking via sandpaper abrasion method (Fig. 9c and d). A piece of sand paper was kept on the surface of the superhydrophobic SP-3 pellet followed by placing a load (100 g) and the sandpaper was gently moved for 50 cycles from left to right and right to left and checked the surface wettability on the surface (Fig. 9c). The superhydrophobic SP-3 pellet also expressed the excellent superhydrophobic stability and durability on the fabricated substrate for sandpaper abrasion test apart from few scratches on the pellet (Fig. 9d). These results clearly demonstrate the excellent durability of our fabricated superhydrophobic SP-3 pellet which is much helpful for the continuous separation of larger quantity of oil-water



**Fig. 9.** Adhesive tape test at (a) before and (b) after peeling the tape. Similarly, sandpaper abrasion test at (c) before and (d) after 50 cycles on superhydrophobic SP-3 pellet.



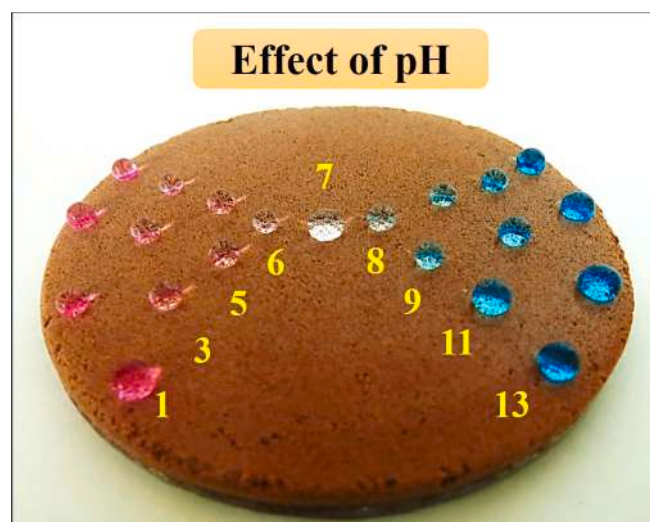
**Fig. 10.** Oil separation from warm water ( $\sim 50\text{ }^{\circ}\text{C}$ ) by superhydrophobic SP-3 pellet at (a) before absorption, (b and c) partial and complete immersion, (d) after removal from the mixture of warm water and kerosene oil.

separation in the real-time application.

Additionally, we checked the oil-water separation ability of the superhydrophobic SP-3 pellet from a mixture of warm water and kerosene oil (Fig. 10). The temperature of warm water used in the experiment was around  $50\text{ }^{\circ}\text{C}$ . The superhydrophobic SP-3 pellet was immersed gently in the warm water-kerosene mixture and checked the sorption ability of the kerosene oil by the superhydrophobic SP-3 pellet. The immersion of superhydrophobic SP-3 pellet can demonstrate clearly the oil-loving nature of superhydrophobic SP-3 pellet and faster sorption ability of kerosene oil. The obtained results proved the excellent usability of our fabricated superhydrophobic SP-3 pellet for in room and warm temperature oil-water separation. This superhydrophobic pellet can be helpful in kitchen hood, where the oily vapours and steam from the cooking process are removed out by exhaust and hence the oil from vapours and steam can be separated by the superhydrophobic pellet. Moreover, we hope that, our results demonstrate an excellent practical applicability of the fabricated superhydrophobic SP-3 pellet for large quantity of oil-water separation. Previously, Li et al. [44] have reported the separation of kerosene from hot water ( $\sim 92\text{ }^{\circ}\text{C}$ ) using the superhydrophobic SS mesh coated with candle soot and hydrophobic silica nanoparticles. Both the candle soot and silica nanoparticles are thermally stable at relatively higher temperatures, however in our case, the polystyrene has very low glass transition temperature ( $>100\text{ }^{\circ}\text{C}$ ). The effect of different pH solutions on the wetting properties of the superhydrophobic SP-3 pellet is shown in Fig. 11. The drops of various aqueous solutions with pH ranging from 1 to 13 were placed on the superhydrophobic pellet and every drop revealed the contact angle around  $150^{\circ}$ , confirming its durability against the liquids having different pH values. We have compared the oil separation efficiency and recycle ability of various SD based superhydrophobic surfaces with our present results (Table 1).

#### 4. Conclusions

Sawdust is quite abundantly and cheaply available in sawmills. We made use of sawdust and polystyrene to fabricate a facile



**Fig. 11.** Effect of different pH liquids on wettability of the superhydrophobic SP-3 pellet (Numbers show the pH values of the liquids).

superhydrophobic pellets for efficient oil-water separation. Such pellets can be fabricated at large scale for industrial use due to its very low fabrication cost compared to available superhydrophobic materials. Our fabricated superhydrophobic SP-3 pellet perform excellent separation of various oils from the oil-water mixture with higher separation efficiency. Moreover, the fabricated substrate maintains an excellent stability for the continuous recycling cycles of oil-water separation and also illustrate an excellent durability by maintaining the superhydrophobic property even after carrying-out the pH, adhesive tape and sandpaper abrasion tests. In addition, we also checked the real practical applicability of the superhydrophobic SP-3 pellet with muddy water-kerosene oil separation. The obtained results demonstrate excellent separation of kerosene oil even mixed in the muddy and warm water. In addition,



**Table 1**  
Comparison of separation efficiency and recycle ability of SD based superhydrophobic surfaces.

Materials	Fabrication Method	Oil/organic solvent used for separation	Separation Efficiency	Recycle Ability	Ref.
SD particles, polystyrene	Pellet formation	Hexane, kerosene, diesel	>96%	36	Present Study
SD particles, polychloroprene adhesive, carbon soot and silicon polymer	Brush and Dip Coating on SS mesh	Toluene, n-hexane, chloroform, and dichloromethane	>90%	~5	[38]
Liquefied-SD-based polymer and carbon foam	Wet Chemical Method	Methanol, octadecane, heptane, isopropanol, N,N-dimethylformamide, tetrachloromethane, n-hexane, olive oil, epoxidized soybean oil, dimethyl silicone oil, liquid paraffin, diesel, and gasoline	~95%	~5	[39]
SD particles, OTS-modified silica particles, and polystyrene	Blending	Gasoline, n-hexane, diesel oil, crude oil, and engine oil	~96%	Not Reported	[40]
Wood slice and dodecanethiol	Solution Immersion	Water-in-oil emulsions of kerosene, diesel, hexane, heptane and petroleum ether	~98%	~6	[41]
Activated charcoal from SD particles using phosphoric acid, zinc chloride, and ferrous sulfate heptahydrate	Impregnation Method	Oil-in-water emulsion of crude oil	~98%	Not Reported	[42]
SD particles, cobalt ferrite, vinyltriethoxysilane	Hydrothermal Method	Lubricant oil	~96%	>10	[43]

an excellent separation behaviour of hexane, diesel, and coconut oil highlight the important superhydrophobic SP-3 pellet for various oil-water separation. Owing to pellets microstructure and its thickness of nearly 1 mm, oil separation time was found slightly higher. However, further research using various polymers and methods can be carried out to overcome this issue.

#### Declaration of competing interest

There are no conflicts of interest to declare.

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