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Synergistic dual-modification of polyurethane sponge with MIL-53(Al) and dimethylsilicone oil for efficient oil–water separation of oily sludge thermal washing wastewater

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26 **Abstract:**

27 Developing highly efficient adsorbents is one of the most feasible strategies to
28 achieve oil-water separation. Conventional sponge adsorbents often suffer from limited
29 capacity, poor cyclic stability, and low selectivity. To overcome these limitations, in this
30 study, we synergistically modified a commercial polyurethane sponge using MIL-
31 53(Al) and dimethylsilicone oil (DMS) via a facile dip-coating strategy, resulting in the
32 M/Si-6@PU sponge. Through the optimization of the synthesis process, the dual-
33 modified sponge evinced a water contact angle of 140° and an oil contact angle of 0°
34 in air. This super-wetting property significantly accelerated the oil absorption process,
35 reducing the absorption time by 48 % (2.6 s compared to 5.0 s for the pristine
36 polyurethane (PU) sponge). Meanwhile, this sponge exhibited outstanding adsorption
37 capacities for various oils, including toluene (43 g·g⁻¹), pump oil (40 g·g⁻¹), diesel (39
38 g·g⁻¹), vegetable oil (35 g·g⁻¹), soybean oil (33 g·g⁻¹), and real oily wastewater (31 g·g⁻¹).
39 Furthermore, the sponge maintained a high separation efficiency (> 99.0 %) for oily
40 sludge thermal washing wastewater over 8 cycles, with the efficiency remaining robust
41 at 93.6 % after 30 cycles. The composite also exhibited excellent chemical stability,
42 retaining its hydrophobicity after 1 h of immersion in extreme pH environments (pH =
43 1 and 12). Therefore, the synergistic combination of hierarchical roughness and low
44 surface energy renders the M/Si-6@PU sponge a highly promising candidate for
45 practical and harsh industrial oily wastewater treatments.

46 **Keywords:** Polyurethane sponge · MIL-53(Al) · Dimethylsilicone oil · Hydrophobic ·

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1. Introduction

Oily sludge, a hazardous by-product abundantly generated during petroleum extraction, transportation, and refining processes, poses significant environmental and operational challenges due to its complex composition with high oil and water content (Duan et al., 2019; Hu et al., 2013; Madbouly et al., 2024; Velasquez et al., 2026; Xiao et al., 2025). While thermal washing and solvent extraction have emerged as relatively efficient approaches for recovering oil from oily sludge, these methods inevitably produce large volumes of oily wastewater, which is rich in emulsified oil droplets, surfactants, and residual contaminants (Hu et al., 2021; Liang et al., 2017). To solve this oil-relevant pollution problem, various oil-water treatment methods have been developed, including combustion, gravity separation, chemical reaction, biodegradation and adsorption separation (Bai et al., 2025; Bullock et al., 2019; Guo et al., 2024; Wang et al., 2022; Zhao et al., 2022; Zhu et al., 2022). Among them, the combustion method generates harmful substances and leads to the waste of petroleum resources; the chemical method is prone to secondary pollution and involves complex treatment processes; the biodegradation method is environmentally friendly but suffers from a slow degradation rate; the adsorption separation method features simple operation, low secondary pollution, and high separation efficiency, and is thus considered a promising approach. Therefore, there is an urgent need for adsorbent materials with good hydrophobicity and high oil adsorption capacity (Hailan et al., 2023; Liu et al., 2024a).

At present, adsorbents can be primarily categorized into 2D (Al-Maas et al., 2021; Rasouli et al., 2021; Ren et al., 2018; Wei et al., 2018) or 3D (Duan et al., 2021; He et al., 2022; Zhu et al., 2021) structural types based on their spatial dimensions. Conventional 2D adsorbents, exemplified by filtration membranes, are highly vulnerable to severe fouling, as the accumulation of oil droplets forms a pore-blocking polarized layer that drastically attenuates permeation flux and compromises separation characteristics (Li et al., 2020; Sutrisna et al., 2022; Zhang et al., 2026). By contrast, 3D macro-porous materials, notably polyurethane (PU) sponges, have demonstrated profound potential as substrates for oil- water separation. This is primarily attributed to their innate hierarchical porosity, excellent mechanical robustness, and highly tunable surface chemistry (Shi et al., 2021). Nevertheless, pristine PU sponges inherently lack the requisite hydrophobicity and selectivity for complex wastewater remediation (Chen et al., 2023; Hosseini et al., 2025). Consequently, surface modification with functional groups or materials has emerged as a crucial strategy to augment their oil–water separation efficacy (Liu et al., 2024b).

Endowed with an intrinsically flexible framework structure (Furukawa et al., 2010; López-Olvera et al., 2021; Tomar and Singh, 2021), MIL-53(Al) features distinctive one-dimensional diamond-shaped channels constructed by its organic ligands; these channels provide ideal accommodation sites for adsorbate molecules (Yuan et al., 2019). Furthermore, the exceptional thermal and hydrolytic stability, along with the tailorable porosity of MIL-53(Al), render it an extremely attractive candidate for the development

of advanced superhydrophobic materials. In this study, the loading of MIL-53(Al) on the macroscopic PU sponge scaffold, simultaneously grates a continuous pore network and a hierarchical micro/mesoporous structure. The modification further enhances the specific surface area and surface roughness of the PU sponge, thereby strengthening the physical capture of oil molecules, while facing a shortcoming of simultaneous improvement water-absorbing performance.

Most oils and water-insoluble organic solvents have surface tensions between 20 and 35 $\text{mN}\cdot\text{m}^{-1}$, whereas water has a surface tension of 72.75 $\text{mN}\cdot\text{m}^{-1}$ (Zhou et al., 2025). Therefore, the surface energy of modified PU must fall within this range to facilitate oil absorption while efficiently repelling water (Zhou et al., 2025). Silane treatment can effectively reduce the surface energy of the sponge and strengthen the hydrophobic property. Dimethylsilicone oil is identified as an ideal silane modification reagent, owing to its exceptionally low surface tension (15.9 $\text{mN}\cdot\text{m}^{-1}$), remarkable cost-effectiveness, and eco-friendly nature. The $-\text{Si}-\text{CH}_3$ functional groups effectively reduce the surface energy of the PU sponge, endowing it with excellent hydrophobic-lipophilic properties. Meanwhile, the continuous silicon-oxygen network forms a dense molecular-level shielding layer, effectively maintaining the coordination integrity of MIL-53(Al) and ensuring efficient oil/water selective separation even in chemically corrosive environment. Moreover, surface roughness can enhance the hydrophobicity, particularly when the material's surface energy is lower than that of water (Zhou et al., 2025).

Herein, we proposed to fabricate a hydrophobic/oleophilic composite sponge (M/Si-6@PU) by integrating the MIL-53(Al) with dimethylsilicone oil onto a PU sponge scaffold via a facile dip-coating approach. The coupling of the physical micro/nano-roughness provided by MIL-53(Al) and the intrinsic low surface energy endowed by dimethylsilicone oil jointly facilitated the hydrophobic transformation of the pristine PU sponge. This study systematically investigates the microscopic morphology, sorption capacities toward various oils/organic solvents, and acid-alkali resistance of M/Si-6@PU, as well as the effect of synthesis parameters on its surface wettability. Furthermore, the separation performance for real oily sludge thermal washing wastewater was comprehensively evaluated, and the underlying separation mechanism was preliminarily elucidated. This facile, cost-effective, and scalable strategy broadens the application composite materials in industrial oily wastewater treatment, thereby paving a viable new pathway for sustainable and practical oil pollution remediation.

2. Materials and methods

2.1 Materials

Oily sludge was supplied by Sinopec Co., Ltd. Saponin powder and pristine PU sponges were purchased from local market. Ethyl acetate (EA), methanol, N,N-dimethylformamide (DMF), ethanol, $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, terephthalic acid (TPA) and dimethylsilicone oil (DMS) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. All chemicals were of analytical grade and used without further

purification. Deionized (DI) water was used throughout all experiments.

2.2 Oily sludge thermal washing wastewater

Initially, 10 g of the raw oily sludge was placed into a beaker and mixed with a specific concentration of saponin aqueous solution (Zhang et al., 2025). The mixture was stirred for 15 min to ensure a homogeneous dispersion. Subsequently, it was subjected to three cycles of ultrasonication (30 min per cycle). Finally, the mixture was centrifuged, and the resulting supernatant was collected as the oily sludge thermal washing wastewater.

2.3 The synthesis of MIL-53(Al)

As shown in **Fig. S1**, it was prepared according to the reference elsewhere (Kazemzadeh et al., 2022). Based on the molar ratio (x) of $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and TPA, the obtained products were denoted as MIL-53(Al- x), where $x = 1.0, 1.1, 1.2, 1.3, 1.4$ and 1.5 . It is noted that if no special reminder, MIL-53(Al) refers to MIL-53(Al-1.3).

2.4 Preparation of M/Si- y @PU sponge

The pristine PU sponges cubes ($1 \times 1 \times 1 \text{ cm}^3$) were ultrasonically washed with DI water and ethanol three times to remove surface impurities, followed by drying in an oven at 80°C for 12 h. To systematically investigate the effect of silicone oil concentration during the dual-modification process, a series of coating suspensions were prepared. Specifically, various volumes of DMS (1, 2, 4, 6, 8, and 10 mL) were dispersed into 100 mL of ethanol under ultrasonication for 30 min to achieve homogeneous solutions with corresponding volume concentrations of 1 %, 2 %, 4 %, 6 %, 8 %, and 10 %.

6 %, 8 %, and 10 % (v/v), respectively. Subsequently, the synthesized MIL-53(Al) powder (0.2 g) was added to each of the above mixtures under an additional 30 min of ultrasonication. The pre-cleaned PU sponges were then immersed in these specific suspensions for 2 h and subsequently dried at 80 °C for 12 h. This dip-coating process was repeated twice. Based on the respective concentration of DMS used (y %, where y represents 1, 2, 4, 6, 8 and 10, respectively), the as-prepared composite sponges were designated as M/Si-y@PU.

2.5 Adsorption capacity

The oil adsorption capacity of the sponges was determined using the weight difference method. First, a clean M/Si-y@PU sponge was weighed three times, and the average initial mass was recorded as m_1 . The sponge was then completely immersed in a beaker containing various oils or organic solvents until adsorption saturation was reached. The mass of the saturated sponge was promptly weighed and recorded as m_2 . The adsorption capacity of the sponge was calculated using the difference method according to **Equation (1)** (Kutenaei et al., 2026; Xiao et al., 2025):

$$\text{Adsorption capacity (g} \cdot \text{g}^{-1}) = \frac{m_2 - m_1}{m_1} \quad (1)$$

For recyclability tests, the saturated sponges were first mechanically squeezed to remove excess liquid, then washed three times with ethanol, and finally dried in an oven at 80 °C for 2 h before proceeding to the next adsorption cycle.

2.6 Oil-water separation efficiency

The oil-water separation efficiency was evaluated using a custom-built separation

setup (**Fig. S2**). First, the initial oil content of the original oily wastewater was measured and denoted as η_1 . The wastewater was then slowly poured into the syringe. Due to the selective wettability of the sponge, the oil phase was rapidly adsorbed and retained by the M/Si-y@PU sponge, while the water phase permeated through and dripped into a collection beaker below. The oil content in the collected effluent (η_2) was measured, and the separation efficiency (%) was calculated according to **Equation (2)** (Bairagi et al., 2023; Xiao et al., 2025; Yu et al., 2025):

$$\text{Separation efficiency (\%)} = \left(1 - \frac{\eta_2}{\eta_1}\right) \times 100 \% \quad (2)$$

2.7 Characterizations

The crystalline phase of the MIL-53(Al) particles was characterized by X-ray powder diffraction (XRD, D8 Advance, Bruker). The surface morphology and microstructure of the prepared sponges were observed using a field emission scanning electron microscope (FE-SEM, GeminiSEM500). Thermogravimetric analysis (TGA, STA200) was conducted to investigate the thermal stability of the materials from 25 °C to 800 °C at a heating rate of 10 °C·min⁻¹ under a nitrogen atmosphere. Fourier transform infrared spectroscopy (FT-IR, CCR-1, Thermo Nicolet) was utilized to identify the functional groups of PU sponges before and after modification in the range of 400 - 4000 cm⁻¹, as well as thermal washing wastewater before and after treatment. The wettability of sponges were evaluated using a contact angle surface tension meter (SDC 350KS, Kunshan Shengding).

3. Results and discussion

3.1 Structural characterizations

As shown in **Fig. S3**, the crystal arrangement of the synthesized MIL-53(Al) can be reflected by the characteristic peaks at 2θ values of 9.3° , 12.4° , and 17.4° , which correspond to the diffraction from the (200), (011), and (002) crystal planes of MIL-53(Al), respectively. These peaks suggest the formation of highly pure MIL-53(Al) crystals, thereby confirming the successful synthesis of the target MOF phase (Salahshoori et al., 2023). In particular, the sharp and intense diffraction peak at 9.3° characterizes the unique crystal orientation of the material, while the peaks at 12.4° and 17.4° verify that the organic ligands and metal nodes within the framework are arranged in a highly ordered manner (Riyal et al., 2023).

As shown in **Fig. S4(a)**, the characteristic symmetric and asymmetric C=O stretching vibrations of the pure MIL-53(Al) are located at 1409 cm^{-1} and 1590 cm^{-1} , respectively (Choi et al., 2025). The presence of the TPA organic framework is further substantiated by the aromatic C-H out-of-plane bending vibration the $750\text{-}800\text{ cm}^{-1}$ region (Baghani et al., 2025), while the sharp peak at 600 cm^{-1} is attributed to the Al-O stretching vibration. The pure DMS (**Fig. S4b**) exhibits prominent Si-CH₃ stretching vibrations at $2962\text{ and }2905\text{ cm}^{-1}$, along with strong Si-O-Si stretching bands at $1071\text{ and }812\text{ cm}^{-1}$ (Felemban et al., 2023). For the pristine PU sponge (**Fig. S4c**), the absorption bands at 1690 cm^{-1} and 1547 cm^{-1} conform to the C=O stretching vibrations of the amide and urea groups (Hao et al., 2018), while the peaks at 1287 cm^{-1} and 810

cm⁻¹ correspond to the aliphatic C-O stretching and aromatic C-H bending modes, respectively (Yu et al., 2020). Following the dual-modification process, the FT-IR spectrum of the resulting M/Si-6@PU sponge (**Fig. S4d**) reveals several crucial changes that verify the successful integration of the components. Notably, the aromatic C-H bending vibration exhibits a slight red-shift from 810 cm⁻¹ to 795 cm⁻¹, which can be ascribed to the altered local electron cloud density and molecular environment induced by the synergistic surface coatings. Moreover, the characteristic Si-CH₃ (1260 cm⁻¹) and Si-O-Si (1020 cm⁻¹) peaks emerge distinctly, providing unequivocal evidence for the successful grafting of DMS. Importantly, the persistent C=O stretching vibrations (1590 cm⁻¹ and 1418 cm⁻¹) related to the MOF signify that the structural integrity of MIL-53(Al) is well-preserved and firmly immobilized onto the PU skeleton. These combined spectral features firmly establish the successful construction of the dual-component modification layer.

Furthermore, the TGA curve of modified MIL-53(Al) (**Fig. S5**) displays an obvious weight loss step in the range of 260-360 °C, which is attributed to the thermal decomposition of the grafted DMS (Cao, 2023; Riyal et al., 2023). Meanwhile, within the decomposition temperature range of MIL-53(Al) (450-650 °C) (Hajjashrafi and Kazemi, 2019), the weight losses of both samples are nearly identical. These results further confirmed that the DMS successfully adhered to the surface of MIL-53(Al), and did not affect its thermal stability.

As illustrated in **Fig. S6**, alongside the inherent C, N, Al, and O elements of MIL-

53(Al), a uniform distribution of Si elements was clearly detected across the surface. No obvious agglomeration or localized deposition of the silicone oil was observed. This confirms that the DMS was successfully and homogeneously anchored onto the MIL-53(Al) surface, providing a robust chemical foundation for hydrophobicity enhancement.

The surface morphologies are illustrated in **Fig. 1**. The MIL-53(Al) material is composed of spherical particles with different sizes stacked together, exhibiting a rough overall surface with obvious interparticle gaps (**Fig. 1a-b**). After modification with DMS (**Fig. 1c-d**), the morphology remained intact, and the surface roughness showed no significant change, indicating that the modification process had a negligible impact on the structure of MIL-53(Al). **Fig. 1e-f**, display the porous structure and the smooth, flat skeleton of the pristine PU sponge. In contrast, after dual-modification, a large number of particulate structures were attached onto the sponge skeleton, and the surface become rougher, with an obvious hierarchical structure (**Fig. 1g-h**). The increased roughness acts as one of the key factors responsible for excellent hydrophobic properties of the sponges.

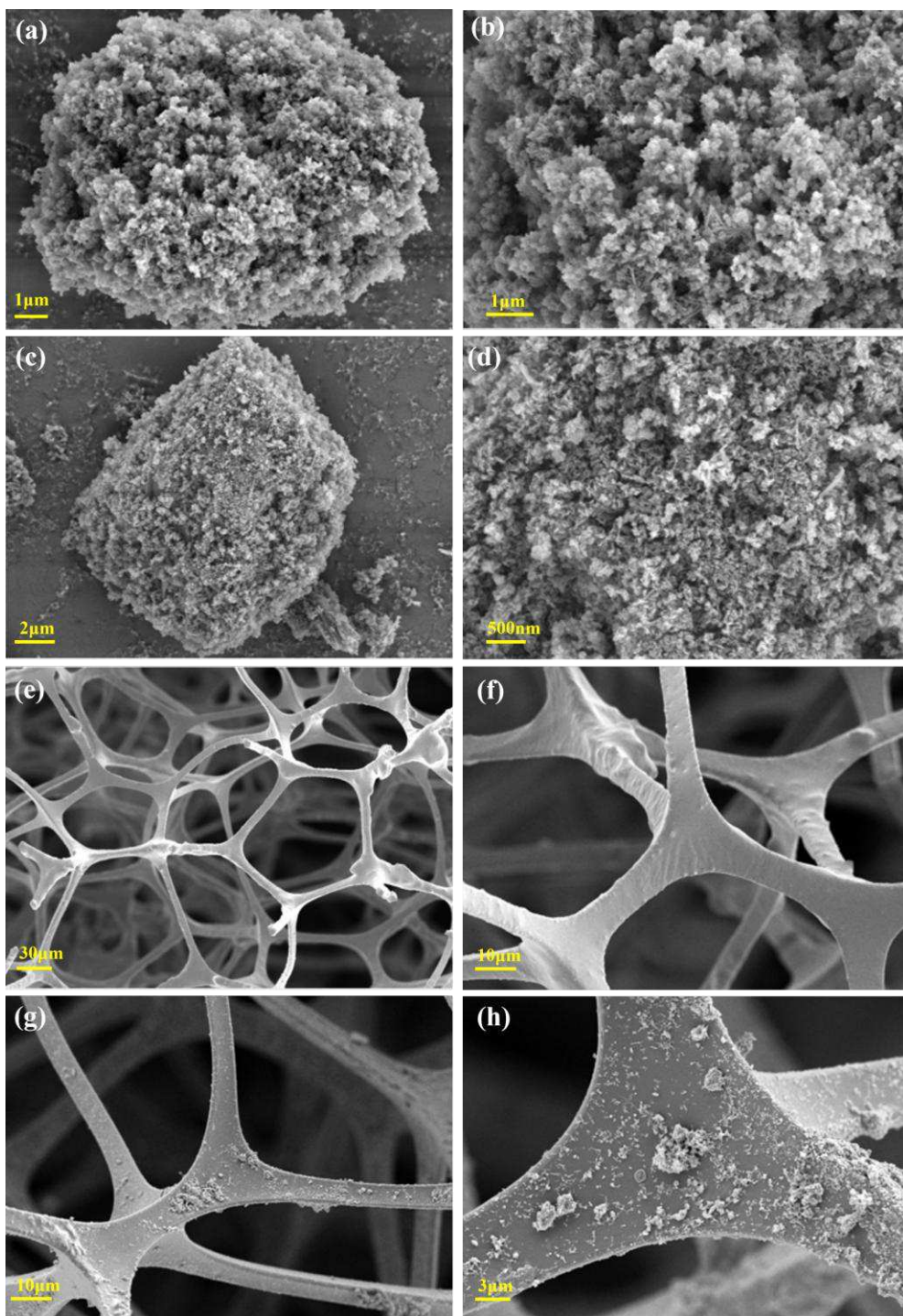


Fig. 1 SEM of (a-b) MIL-53(Al), (c-d) MIL-53(Al) modified by DMS, (e-f) pristine PU sponge, (g-h) M/Si-6@PU sponge, respectively

3.2 Pore structure and wettability

Adsorption kinetics and capacities are intrinsically linked to the pore structure characteristics, such as specific surface area and pore size distribution (Hu et al., 2024).

As illustrated in **Fig. 2a**, the MIL-53(Al) exhibits a typical Type IV isotherm with a narrow hysteresis loop ($P/P_0 = 0.01-1.0$), confirming a bimodal micro-mesoporous structure (Zhang et al., 2022). The corresponding BJH pore size distribution (**Fig. 2b**) reveals a peak situated at 10-20 nm. Increasing the molar ratio of $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ to TPA from 1:1 to 1:1.3 significantly elevated the BET surface area from $890 \text{ m}^2 \cdot \text{g}^{-1}$ to $1199 \text{ m}^2 \cdot \text{g}^{-1}$ and expanded the average pore diameter from 10.8 nm to 17.5 nm (**Fig. 2c** and **Table S1**). This expanded mesoporosity is highly beneficial for the diffusion and capture of viscous, macromolecular components typically presented in thermal washing wastewater. Conversely, further increasing the ratio to 1:1.5 drastically diminished the surface area to $523 \text{ m}^2 \cdot \text{g}^{-1}$, indicating that excessive TPA induces framework defects (Li et al., 2023). Thus, the 1:1.3 ratio was established as the optimal formulation.

The wettability of the sponge is a pivotal aspect of effective oil-water separation. The influence of DMS concentration on the wetting property of the sponge based on MIL-53(Al) was investigated. As shown in **Fig. 3**, initially, the water contact angle (WCA) of the modified sponge gradually increased with increasing concentration of DMS, reaching a maximum of 140° at a concentration of 6 %, which corresponds to a quasi-superhydrophobic state. This robust repellency to water facilitates highly selective oil adsorption. However, further increasing the DMS concentration to 8 % and 10 % resulted in a decline of the WCA, which is likely attributed to the non-uniform aggregation of excessive silicone oil that obscures the MOF-induced roughness.

As presented in **Fig. 2d-e**, following optimization of the synthetic conditions,

dynamic contact angle analysis was performed on the M/Si-6@PU sponge and the pristine PU sponge. When an oil droplet was deposited onto the pristine PU sponge, it required approximately 5.0 s to be completely absorbed. In contrast, the M/Si-6@PU sponge completed the entire oil absorption process in just 2.6 s, indicating a 48 % acceleration in the absorption rate. The fundamental reason for this dramatic enhancement lies in the synergistic combination of the MIL-53(Al)-derived hierarchical roughness and the low-surface-energy from DMS coating, which promotes the rapid expandability and permeability of oil droplets while maintaining an ultralow adhesion to water.

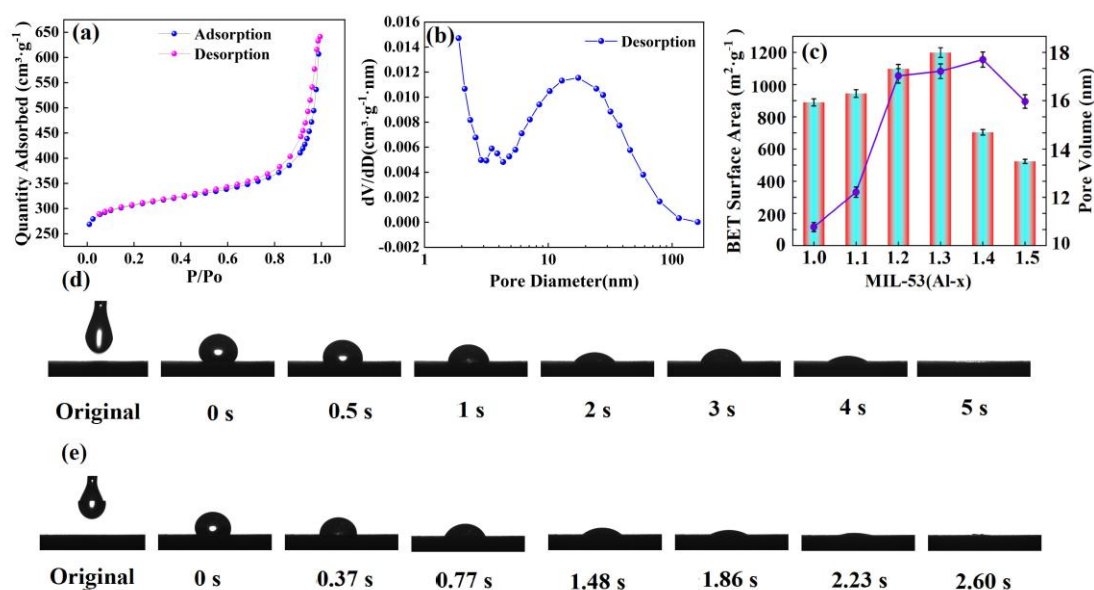


Fig. 2 (a) N₂ adsorption–desorption isotherm of MIL-53(Al) at 77 K, (b) pore size distribution curve of MIL-53(Al), (c) specific surface area and average pore diameter distribution of MIL-53(Al-1.x), dynamic contact angles of (d) pristine PU sponge and (e) M/Si-6@PU sponge

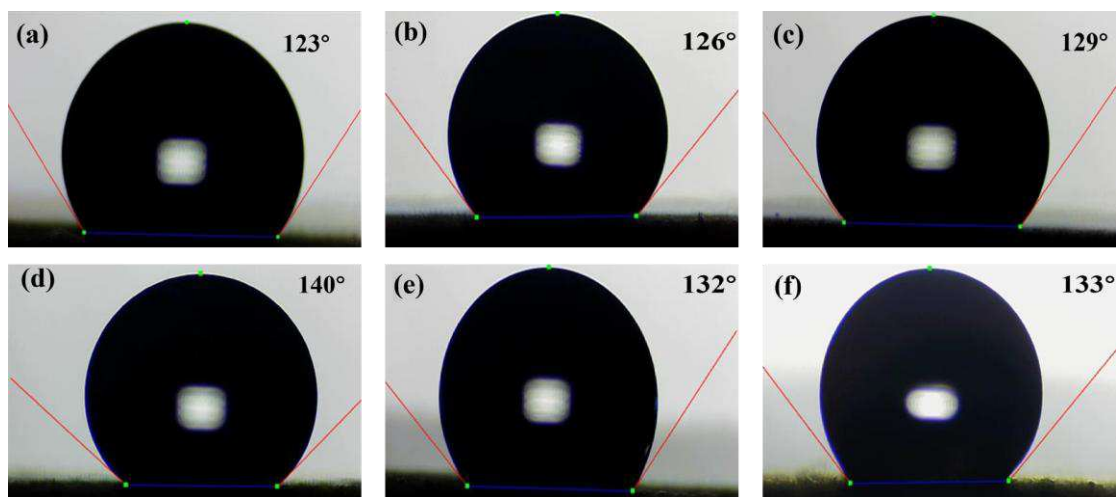


Fig. 3 The water contact angles of (a) M/Si-1@PU, (b) M/Si-2@PU, (c) M/Si-4@PU, (d) M/Si-6@PU, (e) M/Si-8@PU, (f) M/Si-10@PU, respectively

3.3 Absorption property and cyclic stability

As shown in **Fig. 4a**, M/Si-6@PU sponge exhibited the excellent adsorption capacity for toluene ($43 \text{ g} \cdot \text{g}^{-1}$), pump oil ($40 \text{ g} \cdot \text{g}^{-1}$), diesel ($39 \text{ g} \cdot \text{g}^{-1}$), vegetable oil ($35 \text{ g} \cdot \text{g}^{-1}$), and soybean oil ($33 \text{ g} \cdot \text{g}^{-1}$), respectively. The variations in sorption capacity are primarily attributed to the differences in the viscosities and polarities of the various oils (Aier and Purkayastha, 2025). Remarkably, despite the intricate oil composition and the presence of various impurities in the actual thermal washing wastewater, the sponge exhibited a high adsorption capacity of $31 \text{ g} \cdot \text{g}^{-1}$, lightly lower than those of the pure oils. The cyclic stability of the M/Si-6@PU sponge for treating actual oily wastewater was evaluated over 40 cycles (**Fig. 4b**). Even after 30 successive cycles, an outstanding separation efficiency was well-maintained (with a marginal decrease from the initial 99.0 % to 93.6 %), validating the superior cyclic stability of M/Si-6@PU in real-world scenarios.

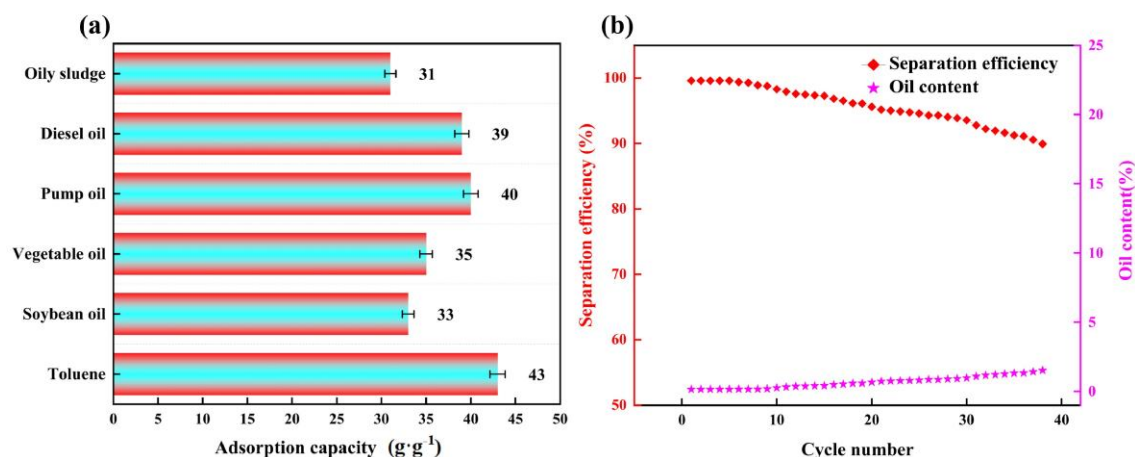


Fig. 4 (a) Adsorption capacity of M/Si-6@PU for different oils, (b) cycling test of M/Si-6@PU

3.4 Acid and alkali resistance test

In practical oil spill remediations, absorbent materials often face complex and harsh environmental conditions. To assess the chemical durability of the M/Si-6@PU sponge, stability tests were conducted by exposing the sponges to extreme acidic (pH = 1), alkaline (pH = 12) and neutral (pH = 7) environments. As shown in **Fig. S7**, the sponges exhibited excellent initial hydrophobicity across all pH values, floating stably on the liquid surfaces. After continuous immersion for 1 h, the sponge in the neutral solution remained completely unwetted. In the strongly acidic (pH = 1) and alkaline (pH = 12) environments, partial wetting (approximately 50 % and 70 % of the volume, respectively) was observed, indicating that aggressive ions gradually induce local damage to the siloxane networks and MOF coordination bonds. Complete loss of hydrophobicity and submersion occurred until an extended immersion of 3 h. These findings verify that the M/Si-6@PU sponge possesses a sufficient operational time window to execute highly efficient oil-water separation even in highly corrosive

environments.

3.5 Separation of thermal washing wastewater

The separation of emulsified industrial wastewater has long been a laborious encumbrance. Conventional separation materials capably remove buoyed-up oils but struggle to eliminate stable oil droplets and dissolved organics. To visually and chemically validate its practical effectiveness, the M/Si-6@PU sponge was utilized to treat actual thermal washing wastewater. The original wastewater appeared dark and turbid (**Fig. S8a**), containing abundant floating oil slicks and suspended solid emulsions. Following a single pass through the M/Si-6@PU sponge setup (**Fig. S8b**), the effluent became remarkably clear and transparent, signifying the efficient demulsification and capture of the oil phase.

FT-IR analysis was performed on the water samples before and after the aforementioned separation, and the results are shown in **Fig. S9**. For the original wastewater, the broad absorption peaks at 3557, 3478, and 3417 cm^{-1} (O-H and N-H stretching) designated the presence of water-soluble organics and amides. The distinctive peaks at 3039 cm^{-1} (aromatic C-H) (Arazo et al., 2021), 2925 and 2854 cm^{-1} (aliphatic C-H) (Sun et al., 2023), and 1457 cm^{-1} (benzene skeleton C=C) confirmed the massive presence of petroleum hydrocarbons, alkanes, and fatty acids (Emenike et al., 2025). Furthermore, the peak at 1030 cm^{-1} (aliphatic ether C-O-C) indicated oxygen-containing polar contaminants (Ma et al., 2021; Ren et al., 2022). Strikingly, in the spectrum of the treated effluent, all these contaminant-associated absorption peaks

were either completely eliminated or drastically weakened. This profound spectroscopic evidence demonstrates the exceptional capability of the M/Si-6@PU sponge to not only separate free oil but also deeply adsorb emulsified and dissolved organic pollutants.

3.7 Mechanisms

The exceptional oil–water separation capability and deep purification mechanism of the M/Si-6@PU sponge can be elucidated through a highly synergistic, multi-step process, which is systematically illustrated in **Fig. 5**. Firstly, the foundation of the separation performance relies on the meticulously engineered "macro-meso-micro" multi-scale architecture (**Fig. 5a**). The macroscopic PU sponge skeleton provides a continuous network of large macro-pores that serve as efficient fluid transport channels. Onto this skeleton, the in-situ incorporation of MIL-53(Al) introduces a highly porous micro-mesoporous hierarchy. This endows the composite with an ultra-high specific surface area and an average pore size, providing abundant physical storage sites for oil molecules. Finally, the conformal coating of DMS constructs a low-surface-energy protection layer rich in Si-CH₃ groups, completing the structural and chemical functionalization of the composite.

Secondly, the outstanding separation selectivity is governed by the quasi-superhydrophobic/superoleophilic interfacial mechanism (**Fig. 5b**). The synergistic combination of the MIL-53(Al)-induced hierarchical roughness and the low-surface-energy DMS layer traps microscopic air pockets within the rough structures,

establishing a stable "air cushion" characteristic of the Cassie-Baxter state (Karimdoost Yasuri, 2026; Pan et al., 2019). According to the Young-Laplace equation (Zhang and Guo, 2023), this highly hydrophobic interface generates a robust capillary intrusion pressure (Δp) that effectively prevents water entry. Conversely, the oleophilic nature of the coating allows oil droplets to rapidly wet and penetrate the porous network in less than 2.6 s. Furthermore, the robust siloxane protection layer shields the underlying MIL-53(Al) framework, granting the composite remarkable chemical stability and ensuring long-lasting quasi-superhydrophobicity even in highly corrosive environments (pH = 1 and pH = 12).

Lastly, the deep purification of the complex oily sludge wastewater is achieved through a cooperative physical and chemical treatment process (**Fig. 5c**). When raw wastewater-comprising free oil, oil droplets stabilized by surfactant shells, and solid particles-contacts the sponge, the sharp micro-nano hierarchical structures physically pierce and disrupt the surfactant shells, inducing rapid physical demulsification. Subsequently, capillary forces drive the free oil to transport deeply into the macroscopic channels. At the micro-scale, strong π - π stacking interactions (Prajapati et al., 2022) between the aromatic terephthalic acid ligands of the MIL-53(Al) framework and the aromatic/asphaltene components of the crude oil facilitate robust chemical anchoring. This intense chemical affinity selectively extracts and retains the complex oil fractions while simultaneously excluding water and solid particulates, ultimately yielding a highly purified filtrate with a removal efficiency exceeding 99.0 %.

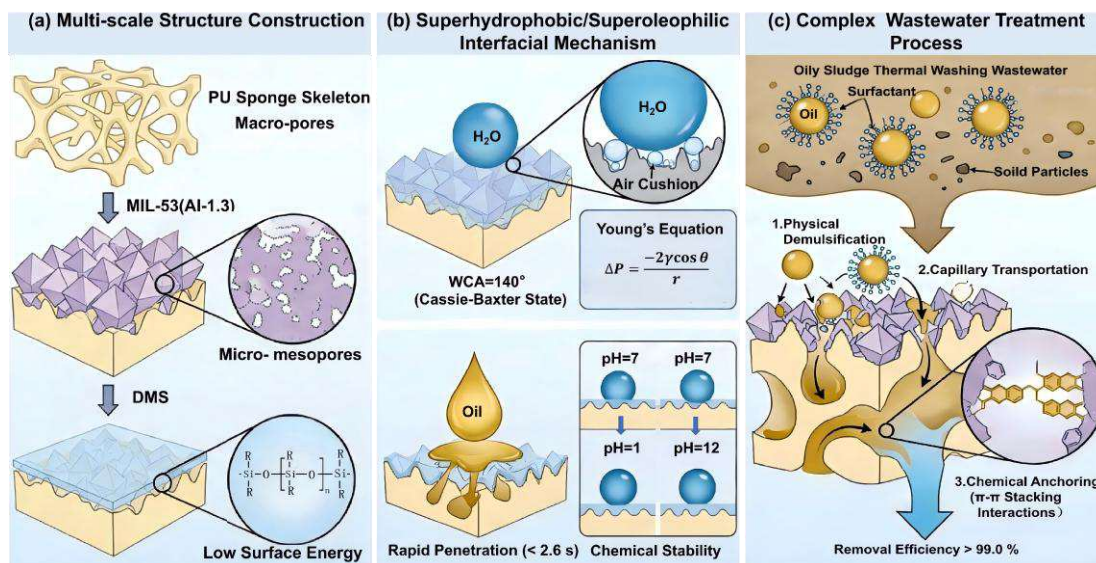


Fig. 5 Schematic illustration of the synergistic oil/water separation mechanism of M/Si-6@PU sponge

4. Conclusions

In summary, a robust "macro-meso-micro" hierarchical architecture was constructed by synergistically integrating MIL-53(Al) with a low-surface-energy DMS coating onto a 3D PU skeleton, endowing the composite sponge (M/Si-6@PU) with highly efficient, dual-functionalized quasi-superhydrophobic/superoleophilic properties for the advanced treatment of complex oily sludge thermal washing wastewater. Specifically, the conformal coating of 6 % DMS and MIL-53(Al) established a robust Cassie-Baxter anti-wetting interface (WCA = 140°), which not only endowed the composite with exceptional quasi-superhydrophobicity but also dramatically accelerated the oil absorption kinetics, reducing the penetration time to a mere 2.6 s. Driven by the synergistic effects of physical demulsification and chemical anchoring, the M/Si-6@PU composite exhibited remarkable adsorption capacities for

various oils (up to 43 g·g⁻¹ for toluene) and achieved an impressive capacity of 31 g·g⁻¹ for highly emulsified actual thermal washing wastewater. Furthermore, the composite demonstrated outstanding durability and chemical resilience, maintaining a separation efficiency of >99.0 % over the initial 8 cycles and >93.5 % after 30 cycles, while retaining its superwettability even after prolonged exposure to highly corrosive acidic (pH = 1) and alkaline (pH = 12) environments. Ultimately, the rational integration of a multi-scale structural design with customized interfacial chemistry presents a highly scalable and innovative strategy, making the M/Si-6@PU sponge a highly promising and durable platform for the practical treatment of harsh industrial oily wastewater and marine oil spills.

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Author Contributors

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Data Availability

Data will be made available on request.

Declarations

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Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical Approval

All the authors have checked and agreed to publish this manuscript into water, air, and soil pollution.

Consent for Publication

The authors are well aware and sure that the used data in this study is not previously published.

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