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# A Capsule-Type Reusable Oil Collector for Oil Spill Recovery for Bridging Material Innovation and Practical Design

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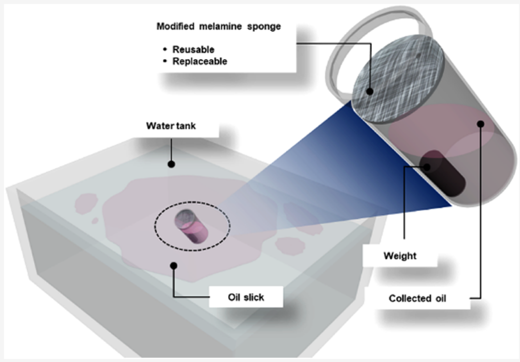
**ABSTRACT:** Oil spills cause severe environmental damage that motivates scientists to explore better and sustainable strategies for cleaner and cheaper remediation techniques. Advances in extreme-wetting materials that selectively absorb oil and yet be water repellent have recently brought about promising results. However, the challenge of practical application persists due to the high costs associated with this technology, the limited oil-absorbing capacity, and the instability of oil retention. An innovative design of capsule-type reusable oil collector (CROC) has been developed to address the current shortcomings of conventional oil collection practices. The CROC can be replaced as a self-contained module and consists of a modified melamine sponge (MMS). The CROC system demonstrated high potential for repeated use (up to 10 times) and achieved an oil collection efficiency of up to 89% during testing in a 1000 L custom-built water pool, with a 0.5 L oil collection per unit when tilted. In our work, we focus on four aspects: (1) MMS integration with the innovative concept of CROC design; (2) theoretical correlation and experimental testing of buoyant force, submergence depth, and tilt angle; (3) performance evaluation with oils of different viscosities in a custom 1000 L water pool; and (4) cyclic reusability of MMS under repeated oil absorption. The combination of MMS with the innovative compact design provides a new route for oil spill cleanup. The technical feasibility of creating a modular CROC unit that can efficiently recover oil is proven through experimental findings, and it also demonstrates its potential for application on an industrial scale. These results indicate that CROC may play an important role in environmental protection by offering a practical, reusable, and scalable solution for oil spill cleanup.




## INTRODUCTION

Oil spillage from either accidents or industrial operations releases massive petroleum discharges into water ecosystems.[1,2] The conventional remediation methods such as mechanical skimmers, chemical dispersants, and *in situ* burning have the common drawback of efficiency, selectivity, or sustainability.[3] Although chemical dispersants are efficient in degrading oil slicks, they have raised environmental concerns due to their toxicity and the potential for secondary contamination.[4] Moreover, *in situ* burning can remove oil quickly but generates harmful by-products and is only feasible under limited circumstances.[5] One of the traditional alternatives of remediation that has proved to have great potential is the absorbent-based methods. Absorbents possess the ability to use surface interactions and capillary effects to physically capture oil on water surfaces. Their parameters of design enable to control their selectivity.[6−11] In recent years, scientists have discovered a variety of materials including natural fibers, synthetic polymers, and nanocomposites to create efficient oil absorbents.[12−20] However, conventional absorbents possess limitations in terms of water uptake and insufficient mechanical strength for reuse. More recently, melamine sponge (MS) has expanded the scope of interest in spill remediation, as the material possesses a distinctive porous architecture, mechanical flexibility, and modifiable surface chemistry.

The commercially available MS is a polymeric foam made of melamine-formaldehyde resin that has a three-dimensional (3D) open-cell structure and a high surface area relative to its mass. Although this material has its merits, it also has disadvantages regarding the selective absorption of oil, as it can absorb both oil and water due to its intrinsic hydrophilicity. To address this shortcoming, intensive research has been conducted on how the surface of MS can be altered to make it superhydrophobic and superoleophilic.[21−23] Among the many available surface modification methods, dip coating has proven to be one of the most promising approaches due to its simplicity, scalability, and low cost.[24−29] The authors have demonstrated exceptional results in oil/water separation by designing surfaces that are both superhydrophobic and

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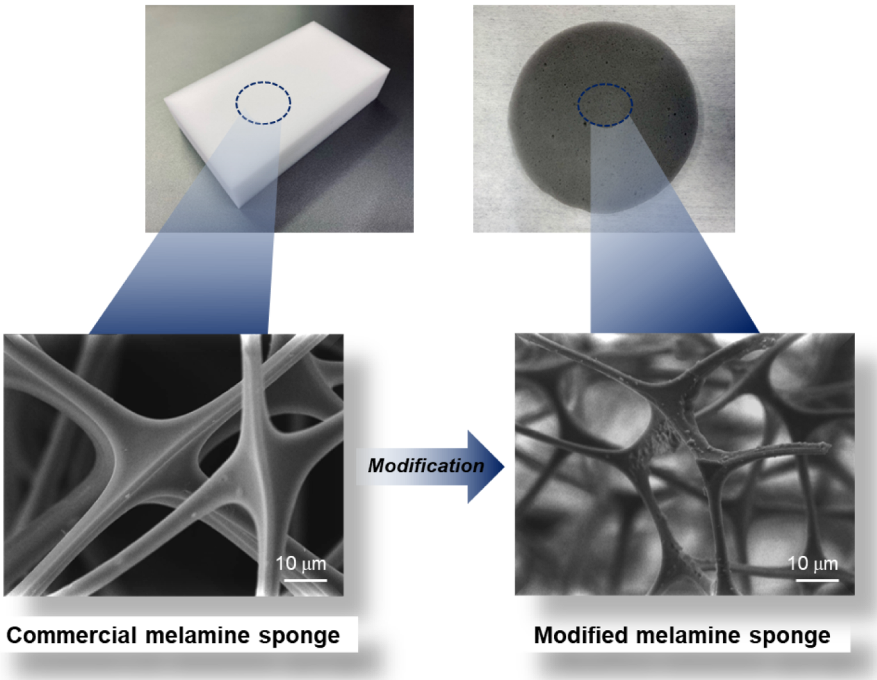


**Figure 1.** Modification process of MS via the facial dip-coating method.

superoleophilic. Superhydrophobicity refers to a surface possessing an outstanding water-proofing property, characterized by a water contact angle of over 150°. Due to this property, water droplets bead and roll off the surface effectively.[30,31] Surface roughness at hierarchical scales, combined with low surface energy achieved through chemical modifications, has been proven instrumental in achieving these properties.

In general, two major approaches can be utilized to create extreme wettability in surfaces: (i) generating hierarchical roughness by means of micro/nano- structuring and (ii) chemical modification of the surface with low surface energy materials, e.g., fluoropolymers, silanes, or polymers.[31−34] The highlighted research on MS-based materials with extreme wettability characteristics has significantly advanced remediation strategies.[35−41] For example, superhydrophobic/superoleophilic melamine sponges were constructed through the impregnation of a deep eutectic solvent (DES) composed of tetraethylammonium chloride and decanoic acid. The sponges demonstrated high oil absorption (40.9−99.8 g $g^{-1}$), low density, high porosity, and excellent reusability. The major innovation lies in the employment of biodegradable, naturally derived DESs, which provide an environmentally friendly, low-cost approach to effectively responding to oil spill cleanup.[26] Moreover, the mechanical recovery of absorbed oil by simple squeezing is possible due to the inherent compressibility of MS, which is essential in determining whether the absorbent is reusable. To achieve multifunctional oil/water separation, Yang et al. synthesized graphene-decorated melamine sponges with opposite wettability. In comparison, the hydrophilic/oleophobic sponge exhibited remarkable water absorption capacities of 72.3−136.5 g $g^{-1}$, while the hydrophobic/oleophilic sponge demonstrated high oil uptake qualities. Bidirectional filtration based on a combination of sponges showed the ability to continuously separate a broad range of oil/water mixtures very efficiently, indicating the usefulness of this system in practical environmental cleanup.[24] Fang and coworkers developed a hydrophobic melamine sponge for oil/water separation by dip-coating it in a solution of urushiol, a compound of natural origin. The urushiol-modified sponge exhibited a large water contact angle of around 139° and an impressive oil absorption volume of up to 181 times its weight. It was also recyclable at a high separation flux and showed great promise as a low-cost and green remedial technique for treating oily wastewater.[26] Although there has been significant progress in the development of extreme wettability substances and the functionalization of MS, the design of a viable and reusable oil collector remains a challenge. First, the mechanical integrity of the MMS may be reduced during multiple absorption-squeezing cycles. Second, it typically lacks an integrated device-level design that facilitates easy deployment, collection, and recovery of the absorbed oil. Addressing these issues requires an in-depth approach involving innovative material adjustments along with practical designs that enable fast application, extraction, and reutilization in realistic oil spillage conditions.

Inspired by the advances in extreme wettability materials, this work aims to bridge the gap between research-level developments and practical applications by presenting a capsule-type reusable oil collector (CROC). CROC is an independent system designed to achieve effective oil/water separation. The basic structural requirement is to develop a portable device that can float on water surfaces and gather oil using its own power without consuming external power sources. The reusable MMS functions as the main component of CROC through a simple dip-coating process and serves as the primary element in combination with a capsule-type collector design. Surface modification enhances the water-repelling ability of MS, enabling selective and efficient oil absorption. The design was guided by the need for a self-contained, modular, deployable system that could operate efficiently at the oil−water interface while being completely


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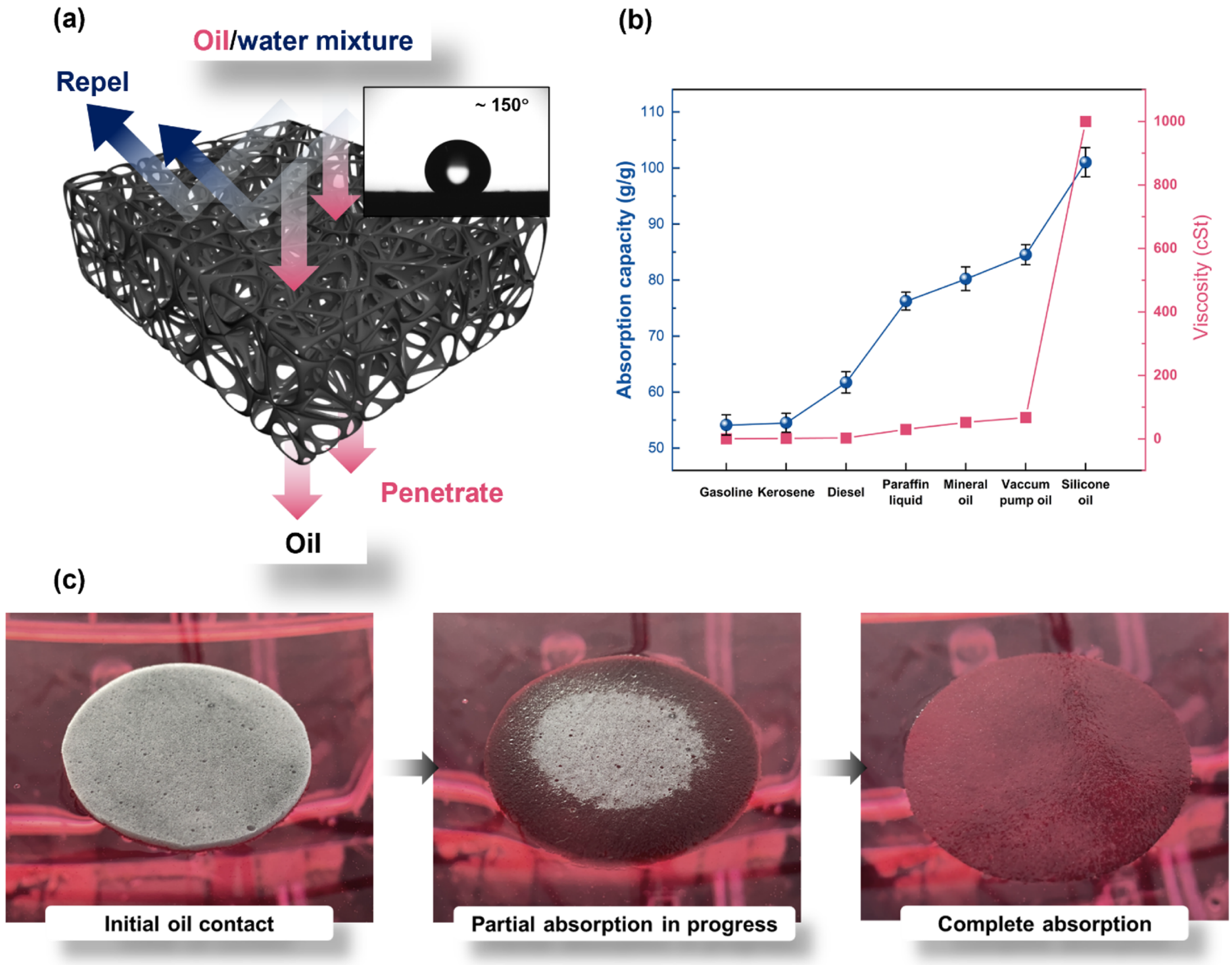


**Figure 2.** (a) Illustration of oil selectivity:oil/water mixture penetration through the MMS structure, (b) correlation between oil viscosity and MMS absorption capacity, and (c) sequential observation of oil absorption behavior in MMS over time.

recoverable and reusable. Accordingly, this study emphasizes the practical utility of CROC as an accessible, cost-effective, environmentally compatible, and easily implementable alternative that addresses key limitations of current oil-spill response solutions.

## ■ EXPERIMENTAL SECTION

**Materials.** The 3D reticular architectural sponge, MMS, was fabricated through a simple dip-coating process that integrated derived carbon with polyvinylidene fluoride (PVDF, Solef) in dimethyl sulfoxide (DMSO, Daejung Chemicals & Metals Co., Ltd.) on the MS scaffold, as shown in Figure 1.[42] Uniform slices of MS (Basotect®) (100 mm in diameter and 2 mm in thickness) were cut. The commercial oils are transparent and difficult to distinguish when spread on water; hence, they were stained with Oil Red O (Daejung Chemicals & Metals Co., Ltd.) to increase their visibility. The dimensions of the profile of the 1L transparent polystyrene oil container are 160 mm × 100 mm in length and diameter. No further physical and/or chemical treatment of any materials was undertaken before tests.

**Fabrication of MMS via a Facile Dip-Coating Method.** The preparation method for MMS was a dip-coating method. The first step involved preparing a PVDF solution (0.5 wt %) by stirring PVDF in DMSO intensively at 60 °C for 8 h in order to ensure the complete dissolution of PVDF. Subsequently, derived carbon powder (0.5 wt %) was added to the PVDF solution, followed by 1 h of stirring to obtain a well-dispersed mixture. Immersing MS slides into the prepared mixture took approximately 10 min to ensure sufficient coating adhered to the slides. Afterward, the samples were allowed to dry in an air oven at 150 °C for 24 h to evaporate excess solvent and form a uniform coated layer. The completed MMS sample was collected for future use after fabrication.

**Evaluation of CROC Performance: Efficiency, Recovery, and Reusability.** Oil collection efficiency can be defined as

$$\text{Oil collection efficiency} = \frac{m_{\text{collected oil}}}{m_{\text{initial oil}}} \times 100\%$$

where $m_{\text{collected oil}}$ is the mass of oil recovered by CROC, and $m_{\text{initial oil}}$ is the mass of oil originally introduced into the tank. The reusability of MMS was measured by several repetitions of absorption−compression cycles to obtain the absorbed oil. The MMS was weighed after every cycle to detect the presence of any permanent oil retention or decrease in absorption capacity. The MMS was weighed after every cycle, and the percentage of permanent oil retention or loss of absorption capacity was measured.

## ■ RESULTS AND DISCUSSION

**Oil Selectivity and Absorption Capacity of MMS.** Figure 1 shows the open-cell structure and pore size of approximately 100 μm in the 3D network porous structure of


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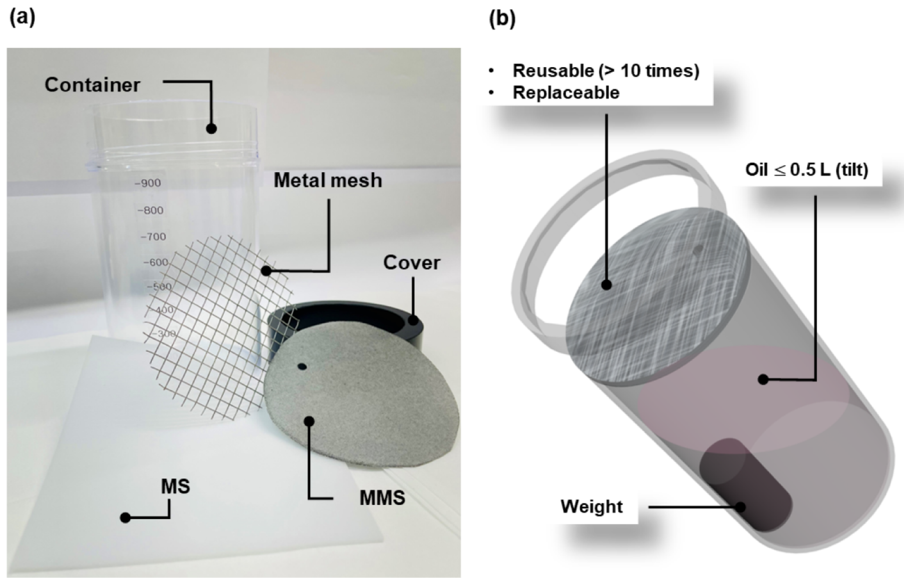


**Figure 3.** (a, b) Capsule-type reusable oil collector and its components.

MS. As observed in SEM (scanning electron microscopy) images, the pristine MS exhibits a 3D structure with a porous nature comprising numerous interconnected networks. PVDF served both as an effective adhesive and a linker, securing the carbon particles uniformly on the MS scaffold. The results demonstrate that the facile dip-coating technique on MS skeletons was effectively utilized for MMS.[42]

The solid–liquid interaction depends on the corresponding surface properties of the structure and its chemical composition. The MMS framework, with open-cell pores, has lower surface energy on the material's surface and functions as both a water repellent and an oil attractant. As a result, Figure 2a shows that the MMS exhibited significantly improved hydrophobicity compared with MS. The water contact angle exceeded 150° on most observations, demonstrating strong water-repelling properties. MMS is attributable to the properties of PVDF and the derived carbon coating, which lower the surface energy and increase oil affinity. The derived carbon particles provide microscale roughness, enhancing the oleophilic nature of MMS, while PVDF contributes additional chemical resistance and hydrophobicity. Moreover, the interconnected pores in the sponge create capillary channels that absorb oil into the MS structure due to the strong affinity between the hydrophobic coating and nonpolar hydrocarbons. This synergistic effect enables MMS to absorb oil quickly while preventing water penetration in spill scenarios.[42]

In order to understand the oil selectivity process, an oil/water mixture was used. The MS and MMS were placed between the funnel and the flask. Oil (stained with Oil Red O) and water (stained with Brilliant Blue) were mixed in a 1:1 ratio by volume. Then, the mixture was poured into the upper funnel through the MS and MMS (Figure S1). MS exhibits amphiphilic properties that absorb both oil and water, which can be clearly observed in the filtrate flask. The hydrophobic-oleophilic property of the MMS developed a stable layer of oil on its textured surface, which formed a three-phase interface, preventing the entry of water into the separation layer. The Cassie and Baxter theory implies that superhydrophobicity can be achieved only through the three-phase interface. During the oil–water separation, the water–oil–solid system also constructed a three-phase interface, which allowed only the oil to pass through while repelling the water off.[43,44]

To determine the oil absorption capacity of MMS, the oil-saturated absorbent was collected after being immersed in oil for 5 min. The mass of the MMS was measured with an electronic analysis scale before and after the absorption test. The initial weight of the MMS (in grams) before oil absorption is represented by $m_0$ whereas the weight of the MMS (in grams) following the absorption of oil is represented by $m_1$ in the equation below. This provides a quantitative measure of the oil absorption capacity of the MMS, offering valuable insight into how effectively the MMS can be used for oil absorption purposes. The absorption capacity of MMS is given by

$$\text{Absorption capacity} = \frac{m_1 - m_0}{m_0}[\text{g g}^{-1}]$$

The physical properties of the commercial oils used in this study, including density and viscosity, are summarized in Table S1. The quantitative mass absorption capacities of MMS for different oil types appear in Figure 2b. The absorption capacities of MMS ranged from 54.1 g $g^{-1}$ to 101.0 g $g^{-1}$, depending primarily on oil viscosity. Oils with lower viscosity penetrate more rapidly into the sponge matrix but tend to yield lower ultimate capacities due to easier drainage, whereas oils with higher viscosity diffuse more slowly yet achieve higher ultimate capacities owing to reduced drainage. Thus, viscosity affects not only the rate of diffusion but also the equilibrium absorption capacity of the MMS. Although both density and viscosity contribute to the absorption capacity, our results indicate that viscosity is the dominant factor, whereas the effect of density is comparatively minor. Furthermore, the sequence of images in Figure 2c records the absorption behavior of the MMS in contact with oil. The initial observation shows oil remaining distinct from the sponge surface, but it eventually starts to penetrate through the outer MMS layer. The interconnected pores in the MMS absorb oil penetration over time, creating noticeable darkening in the affected MMS area. This step highlights the capillary-enhanced oil uptake through MMS due to its oleophilic properties, which enable


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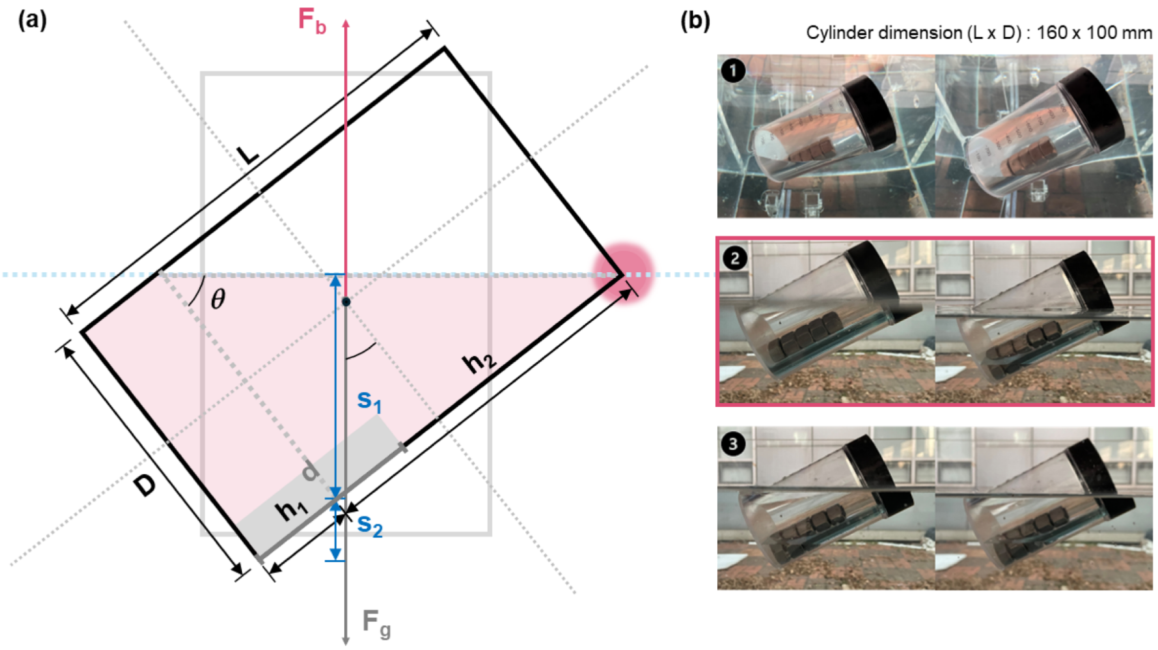


**Figure 4.** (a) Configuration of the geometric profile and buoyant equilibrium of tilted CROC and (b) digital images of tilt angle evaluation.

the sponge to absorb oil. Finally, the MMS becomes completely saturated, demonstrating how effectively the sponge absorbs and stores the oil within its structure. The color change indicates that the oil has permeated the entire internal network of the interconnected pore structure.

**Design of Capsule-Type Reusable Oil Collector.** The capsule-type reusable oil collector shown in Figure 3a,b operates as an independent system to perform efficient oil/water separation through its transparent cylindrical design, along with a metal mesh, lid, and MMS. The transparent container, with a capacity of 1 L, can hold up to 0.5 L of oil in its tilted position, allowing real-time observation of oil absorption. The parts are modularly structured so that they can be easily assembled, disassembled, and replaced. The basic structural requirement is to develop a small, portable device that can float on water surfaces and gather oil under its own power without relying on external power sources. Due to the coating of the MMS with PVDF and derived carbon, the MMS facilitates hydrophobicity and oleophilicity, enabling not only effective absorption but also reusability. The design aims to make the CROC an affordable and sustainable solution for oil spill recovery.

**Tilted Floating CROC: Derivation of Force Balance, Equilibrium Tilt Angle, and Submerged Depth.** In CROC, the system must remain stable enough to avoid sinking or excessive tilting, which could compromise oil absorption. We consider a rigid floating object of length and cross-sectional diameter partially immersed in the surrounding water. CROC has a mass $m$ and tilts by an angle $\theta$ from the horizontal due to buoyant equilibrium. As shown in Figure 4, $V_1 = \left(\frac{\pi D^2}{4}\right)h_1$ is the volume of a fully immersed cylindrical section of height $h_1$, and $V_2 = \frac{1}{2}\left(\frac{\pi D^2}{4}\right)h_2$ is the triangular or wedge-shaped volume of partial immersion $h_2$, the total submerged volume is given by[45]

$$V_{sub} = V_1 + V_2 = \frac{\pi D^2}{4}\left(h_1 + \frac{1}{2}h_2\right)$$

Considering a horizontal cylindrical collector, the buoyancy of the capsule was modeled using Archimedes' principle to ensure stable floating,

$$F_b = \rho_w g V_{sub} = \rho_w g \times \frac{\pi D^2}{4}\left(h_1 + \frac{1}{2}h_2\right)$$

where $\rho_w$ is the density of the surrounding fluid (kg m$^{-3}$), $g$ is the gravitational acceleration (9.81 m s$^{-2}$), $V_{sub}$ is the submerged volume. At equilibrium, the buoyant force balances the weight of the collector:

$$F_b = F_g$$

$$\rho_w g \times \frac{\pi D^2}{4}\left(h_1 + \frac{1}{2}h_2\right) = mg$$

Substitute $h_1 = L - D\tan\theta$ and $h_2 = D\tan\theta$, we have

$$\rho_w g \times \frac{\pi D^2}{4}\left(L - \frac{1}{2}D\tan\theta\right) = mg$$

Thus, the required total weight of the cylinder is

$$m = \rho_w \times \frac{\pi D^2}{4}\left(L - \frac{1}{2}D\tan\theta\right) \quad (1)$$

The submerged depth is conceptually divided into two components, $s_1 = \left(h_1 + h_2 - \frac{d}{2}\right)\cos\theta$, accounts for the height of the main submerged section, adjusted for the tilt and the effective vertical offset of the dominant mass and $s_2 = \frac{d}{2}\cos\theta$, other is based on the geometry of the submerged CROC, where $d$ represents the length of the offset mass (weight block) used to induce tilting.

Substitute $s_1 = \left(h_1 + h_2 - \frac{d}{2}\right)\cos\theta$ and $s_2 = \frac{d}{2}\cos\theta$, we get


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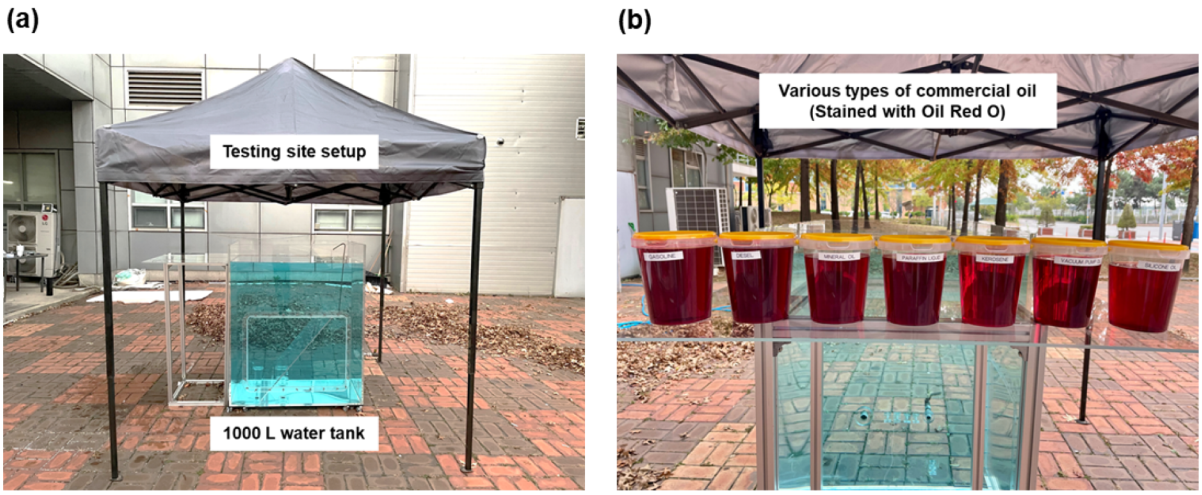


Figure 5. (a) Testing site configuration and (b) various types of commercially stained oil samples.

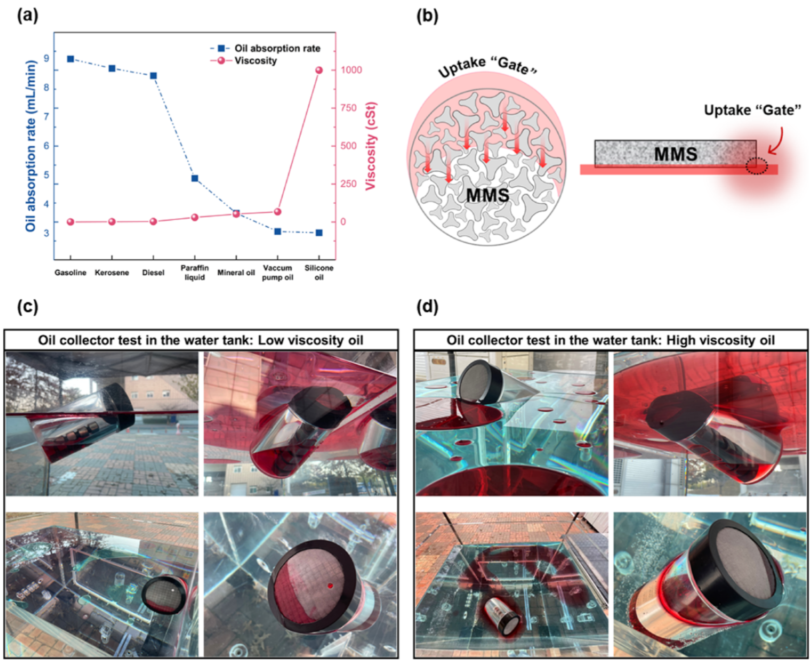


Figure 6. (a) Correlation between oil absorption rate and viscosity, (b) illustration of oil uptake via the oil/water interface, and (c, d) oil collector in the water tank: low-viscosity and high-viscosity oil.

$$h_{sub} = s_1 + s_2 = (h_1 + h_2)\cos\theta = L\cos\theta \quad (2)$$

Achieving the oil−water interface with a stable setup depends strongly on tilted angle adjustment. The floating collector demonstrates that adding adjustment masses enables control and ensures the collector floats with a stable inclination. Consider the cylinder in its tilted position, as shown in Figure 4; the horizontal dashed line represents the original water surface plane. When the cylinder is tilted through the angle $\theta$, the center of buoyancy shifts to $F_b$ as a result of the alteration in the shape of the displaced fluid. The tilted angle $\theta$ of a floating object is determined by the forces acting on it. However, when the collector tilts by an angle $\theta$, the geometry of the submerged section becomes more complex. The tilt angle occurs when the weight inside the cup is not centered.

Figure 4a illustrates the driving forces and tilt angle of the oil collector with respect to the horizontal offset and adjustable mass support. The device is designed to maintain a specific tilt angle during flotation by precisely tuning its center of gravity relative to its center of buoyancy. By controlling the angle of inclination, CROC remains situated at the oil/water interface, where it achieves optimal contact with oil slicks. The dynamic interaction between gravitational and buoyancy forces is further complicated by the absorption of oil. As the MMS absorbs oil, the mass of CROC increases, altering the balance of forces and potentially affecting the tilt angle. Our design anticipates these changes by ensuring that the device maintains stability even as the load varies, as shown in Figure 4b. Based on the derived equations, it reflects the correlation between the buoyancy calculation, submerged height, and tilt angle, and confirms that it satisfies the experimental conditions. In addition, the tilt angle at the equilibrium state was calculated as 59.8°, respectively, which was derived through eq 1. Finally, the submerged height based on the buoyancy eq 2 was


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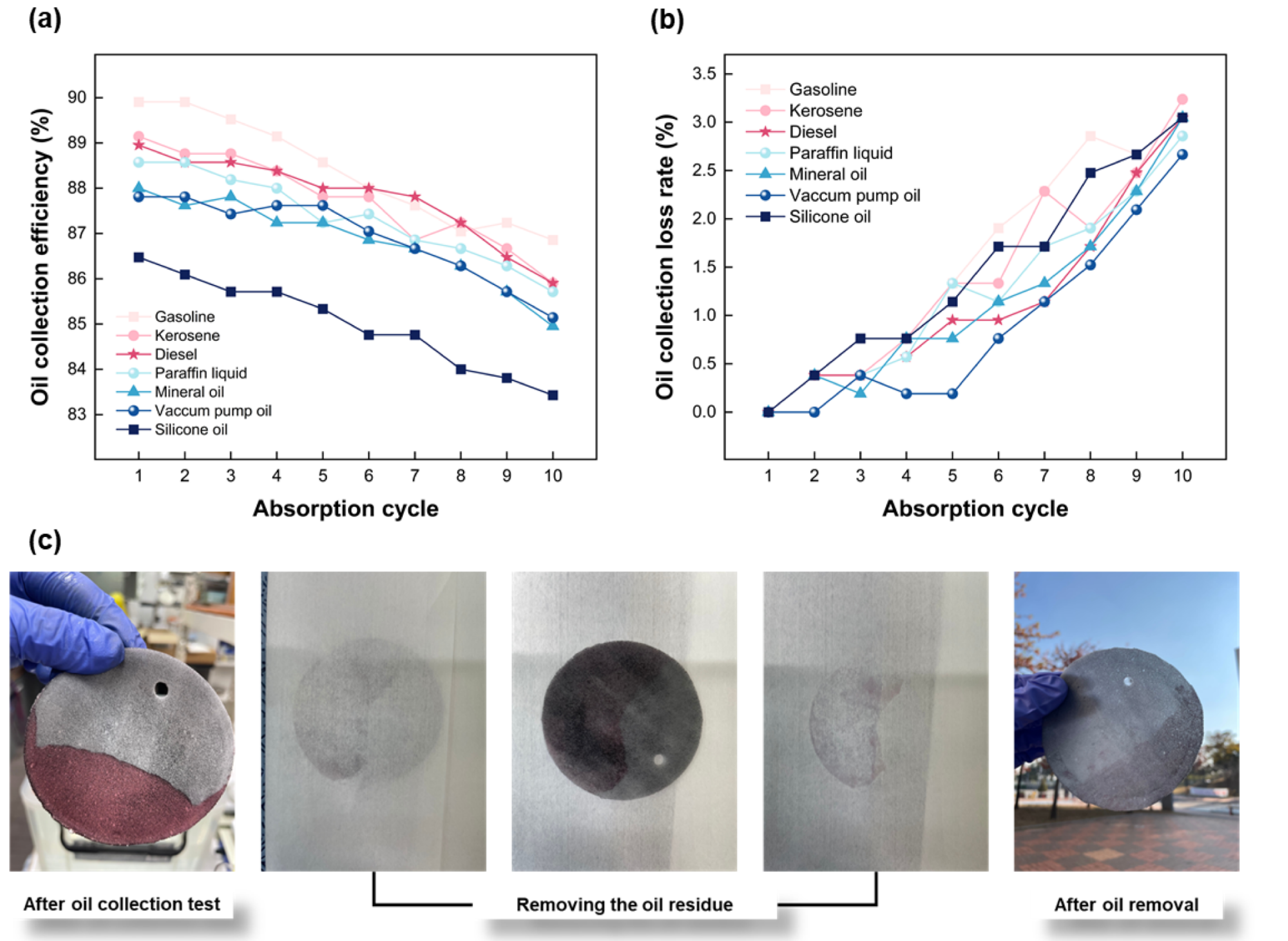


**Figure 7.** (a) Changes in oil collection efficiency, (b) efficiency loss rate according to the recycling cycle of MMS, and (c) reusability of the MMS during the collection process.

calculated as 80.4 mm, which takes into account the volume displacement. This experimental data quantifies the relationship between buoyancy force and submerged height while validating the link between buoyancy and tilt angle for stability evaluations.

**Performance Evaluation of CROC: Practical Testing Site.** Performance evaluations focused on oil/water separation efficiency, absorption capacity, and operational stability under realistic conditions. The protocol used experimental conditions to represent typical pollution events, demonstrating the practical and reliable performance of the oil collector in actual field operations. A testing site was set up using a rectangular, custom-built water tank with a capacity of 1000 L, designed to replicate practical scenarios for realistic performance testing, as shown in Figure 5a. The large tank capacity ensured unrestricted movement of CROC during testing with various types of commercial oil. In Figure 5b, various commercial oils with different physical properties were tested to evaluate the performance of CROC.

CROC is assembled by placing the MMS on the metal mesh inside the PET container and then sealing it with the lid. A known volume of each commercial oil (0.5 L) was gently poured onto the water surface, forming a floating slick. CROC was placed on the surface, and its movement, absorption kinetics, and final oil uptake were monitored. During deployment, the device was gently placed on the water surface of an oil slick. The MMS actively absorbs oil through the mesh into its porous network. As oil accumulates, the capsule tilts, retaining up to 500 mL of oil and preventing overflow or submersion.

CROC performance tests were conducted on low- and high-viscosity oils using the oil collector in a rectangular, custom-built water pool, as shown in Figure 6. In the low-viscosity oil test, a fast absorption rate and stable separation were observed, and efficient operation at the oil/water interface was confirmed. In contrast, in the high-viscosity oil test, the absorption rate of the oil was relatively slow as the viscosity increased, but CROC still absorbed the oil effectively and maintained its structural stability. The reliable performance of CROC across different oil viscosity ranges demonstrates evidence of its practical potential in environmental applications.

The flow of the oil patterns through the open-cell porous structure of MMS and the rapid movement of flows between channels are illustrated in Figure 6b. The oil flow penetrates through the gaps between the MS scaffold, a phenomenon known as capillary imbibition. The wetting effects can have a considerable impact on the dynamics of displacement at the microscale, where capillary forces are dominant. When the oil droplets initially come into contact with the surface, countless pores in MMS immediately pull floating oil into the complex 3D interconnected structure. The kinetic process of oil


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absorption through porous media depends on three factors: capillary forces, viscous resistance, and interfacial tension. Lucas-Washburn theory provides a baseline model for capillary penetration in a single capillary, with the penetration length $x$[45−47]

$$x \propto \sqrt{\frac{\gamma \times r \times t}{\eta}}$$

where $\gamma$ is the interfacial tension, $r$ is the effective pore radius, $t$ is time, and $\eta$ is the fluid viscosity. In complex porous networks, such as MS, deviations from this idealized behavior are predictable. In general, lower-viscosity oils are absorbed more rapidly, whereas oils with higher viscosity yield higher ultimate capacities due to reduced drainage. Although the pore size was not directly varied in this study, the Washburn equation also suggests that it can significantly influence absorption dynamics. From the perspective of capillary flow, a larger pore size may facilitate faster oil uptake due to reduced viscous resistance, while smaller pores may enhance oil-retention stability by providing a higher capillary pressure. Therefore, pore size can be regarded as a structural factor that presents a trade-off between absorption rate and retention stability, highlighting the importance of systematic investigation in future work.

A quantitative measurement characterizing how much oil an absorbent can collect in the case of an oil spill is referred to as oil collection efficiency (%). It is a significant indicator of the ability of an absorbent to remove oil and separate water and oil under specific circumstances. The variation in the rate of oil collection will be represented as a plot in conjunction with the rate of loss during the duration of the reusability test with a series of oils. The relationship between the viscosity of various oils and the oil absorption rate of MMS is depicted in Figure 7a,b. According to the findings, kerosene and gasoline had the highest initial collection efficiencies, at over 89%, and maintained over 87% efficiency after 10 cycles. Over 10 subsequent cycles, the initial collection efficiency of silicone oil dropped from 86% to 82%. This drop results from the high viscosity of silicone oil, which slows regeneration by delaying both absorption and total expulsion. Capillary absorption slows as oil viscosity rises due to increased viscous resistance, which lowers absorption rates. It was confirmed that the absorption capacity of MMS was maintained above 3.03 mL $min^{-1}$ for oils of various viscosities.

MMS achieved 10 cycles of reuse while maintaining high performance in various types of oil, particularly in low-viscosity oils. These findings imply that MMS has the potential to be recycled multiple times in different oil settings and can be deployed as an oil collector, meeting both requirements of reuse and performance. For practical application, MMS should exhibit not only a hydrophobic surface but also mechanical toughness. Mechanical toughness is an important factor in the reusability of MMS. Since a hydrophobic surface alone cannot guarantee the long-term functionality of the material, MMS must possess sufficient mechanical strength to avoid deformation or loss of properties despite repeated use and physical manipulation. Therefore, to continue improving its design and exploring its potential use in actual operating conditions, it is critical to assess the mechanical strength of MMS. One of the core benefits of MMS is its ability to enable mechanical recovery of oil absorption. The sponge can be squeezed or, on large scales, can be passed in rollers to extract the oil, which can then be collected. The reusable material contrasts with single-use absorbents by eliminating waste disposal through burning or landfill that reduces environmental impact. Greater reusability of the material extends its lifespan , which lowers costs and further reduces waste.

The tilt angle of the CROC is governed by the balance among capsule geometry, buoyant force, and submerged depth. Although the present tests were conducted under controlled conditions, the same principle of buoyancy−load balance may also be applicable in ocean environments. While temporary disturbances, such as surface waves, could affect the equilibrium, the modular design of the CROC, with stable flotation and passive self-adjustment, suggests that it may be able to maintain the tilt angle and remain functional under open-ocean conditions.

## ■ CONCLUSIONS

Oil spills present a serious threat that requires breakthrough remediation strategies capable of restoring ecosystems while complying with environmental protocols. In this study, we developed CROC as a remedial solution for oil spills, consisting of the integration of the MMS fabrication technique by employing a dip-coating method utilizing derived carbon/PVDF components and a strategic capsule assembly configuration. The findings highlight several key outcomes:

(i) *Enhanced hydrophobicity and oil selectivity:* MMS achieved a high water contact angle above 150° and demonstrated strong oil selection capabilities, making it effective for oil−water separation.

(ii) *High oil absorption capacity:* MMS exhibited consistently high oil absorption capacity, taking up to 101.0 g $g^{-1}$ of different commercial oils, which can be attributed to the synergistic effects of the porous structure of MS combined with the coated carbon/PVDF.

(iii) *Capsule design for practical deployment:* Stable flotation, simple retrieval, and minimal secondary waste are made possible by the modular and motorless design of CROC. By balancing buoyancy and capacity, the tilting mechanism can hold up to 0.5 L of oil in a 1 L transparent container.

(iv) *Buoyancy and tilt angle optimization:* CROC behaves dynamically stable, as demonstrated by experimental validation and theoretical models based on Archimedes' Principle. The good agreement between calculated and measured tilt angles confirms the robustness of design.

(v) *Performance evaluation of CROC in a 1000 L custom-built water tank:* CROC achieved up to 89% collection efficiency across oils of varying viscosity, while maintaining 80−90% absorption capacity after 10 reuse cycles.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.langmuir.5c04219.

Table S1. Physical properties of the commercial oils. Table S2. Recent research on MS-based materials for oil absorption applications. Table S3. Calculation of equilibrium tilt angle and submerged depth of CROC. Figure S1. Primary oil selectivity test (PDF)

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### Notes

The authors declare no competing financial interest.

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