Facile synthesis of superhydrophobic MS/TiO₂/PDMS sponge for efficient oil–water separation

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ABSTRACT

To obtain a kind of superhydrophobic sponge with high oil and water selectivity, the MS/TiO₂/PDMS sponge was prepared via a two-step hydrophobic fabrication based on the melamine sponge (MS), tetrabutyl titanate (TBOT), and polydimethylsiloxane (PDMS). The effects of modification time, the concentrations of TBOT and PDMS on the properties of the MS/TiO₂/PDMS sponge were studied, and the separation mechanism was also discussed based on the interaction between the oil and the surface of the MS/TiO₂/PDMS sponge. The results suggest that under optimal conditions, the MS/TiO₂/PDMS sponge show superhydrophobicity. The contact angle and adsorption capacity for oil of the MS/TiO₂/PDMS sponge are 149.2° and 98.5 g·g⁻¹, respectively, and they can be recycled for about 25 cycles after oil–water separation test. This study prepares a new composite material with high oil–water selectivity, which is a good foundation for the development and research of new oil adsorbents.

Key words: impregnation method, melamine sponge, oil–water separation, silanization, superhydrophobicity

HIGHLIGHTS

- The MS/TiO₂/PDMS sponge was prepared via a two-step hydrophobic fabrication.
- The contact angle of the MS/TiO₂/PDMS sponge was 149.2°.
- The adsorption capacity for oil of the MS/TiO₂/PDMS sponge are 98.5 g·g⁻¹.
- The MS/TiO₂/PDMS sponge can be recycled about 25 cycles after oil–water separation test.

1. INTRODUCTION

With the growth of industry and marine transportation in recent few decades, rivers and oceans have become increasingly polluted, which is mainly due to the leakage of crude oil, the uncontrolled disposal of waste water containing heavy metal ions, and the leakage of chemical reagents (Varela et al. 2013; Fan et al. 2015; Zou et al. 2015). Pollutants in oil-bearing wastewater have significant impacts on air, surface, and underground water, so their removal is essential because of their harmful nature and serious environmental risks. Therefore, it is necessary to treat the oil pollution to achieve water purification. The conventional oil spill methods mainly include mechanical collection, physical adsorption, in situ burning, chemical dispersion, bioremediation, and oil–water separation techniques (Yu et al. 2015). Although these methods are simple and convenient, they often cause secondary pollution during the clean-up process, and thus it is necessary to seek new methods to realize the goal of environmental protection. Compared with those commonly used methods mentioned above, the physical adsorption method has many advantages, such as low cost, simple operation, low secondary pollution, and good recyclability (Raju et al. 2017). The method is helpful for extensive research and application. However, the synthesis of adsorbent materials used in physical adsorption is still a challenge in that the adsorbent materials need to have the properties of low cost, superior adsorption capacity, good selectivity, and good reusability.

Familiar sorbents and dispersants such as sponge, mineral products (Adebajo et al. 2003), chemical dispersants (Kleindienst et al. 2015), polymers (Ge et al. 2016), textiles/fibers (Seddighi & Hejazi 2015; Hu et al. 2016), metallic meshes (Wang et al. 2015), carbon materials (Gupta & Tai 2016), and membranes (Sadiq et al. 2020) have been used...
widely to separate oil from water. Among them, the three-dimensional porous structure materials play an important role in the field of oil removal from water due to their low cost, they are easily mass-produced, they have high mechanical strength, excellent adsorption capacity, and are suitable for recycling (Guan et al. 2019). Melamine sponges (MS) are frequently used in oil–water separation field because of their advantages of low density, large porosity, large specific surface area, high thermal stability, corrosion resistance, and flame retardancy (Qiu et al. 2015). However, the MS without modification are naturally hydrophilic and cannot selectively adsorb oils and organic solvents from water, which greatly affects their oil–water removal efficiency and restricts its application to oil–water treatment (Lei et al. 2017a, 2017b). A summary for the adsorption capacities of oils solvents by different MS-based adsorbents is shown in Table 1. To satisfy the demand of application in water oils separation, it is necessary to exploit an efficient and facile method to tune the opposite surface of sponge. Recent reports have focused on the superhydrophobic modification of the sponges by various methods, including dip coating (Qiang et al. 2017), chemical vapor deposition (Khosravi & Azizian 2015; Zhang & Seeger 2015), in situ chemical reaction (Chung et al. 2018), carbonization (Yao et al. 2017), and other methods (Lei et al. 2017a, 2017b).

Studies reported that hydrophilic surfaces can be converted to hydrophobic surfaces by silanization (Xu et al. 2015; Yujing et al. 2019; Yan et al. 2020; Shi et al. 2020; Yin et al. 2020; Chen et al. 2021), carbonization (Feng & Yao 2018; Peng et al. 2019), and fluorination (Ruan et al. 2014; Gurav 2015; Li & Guo 2017). The silanization treatment has more application prospects because fluoration has certain toxicity and will cause secondary pollution (Chen et al. 2016). Polydimethylsiloxane (PDMS) is considered to be one of the most commonly used hydrophobic reagents for the modification of oily material adsorbents, not only because of their hydrophobicity, but also because of their stability and mechanical flexibility (Ge et al. 2016). It can firmly bond onto the sponge skeleton surface and does not fall off easily (Chen et al. 2016). Meanwhile, hydrophobic nanoparticles, such as TiO2 (Chen et al. 2016), SiO2 (Yeom & Kim 2016), Ag (Qin et al. 2018), ZnO (Tian & Lee 2017), Fe3O4 (Li et al. 2018), and attapulgite particles have been used to modify the surface of sponges. In particular oleophilic TiO2 nanoparticles, which are widely used as a functional material with good chemical stability, were adsorbed onto the surface of the MS’ skeleton to enhance the surface roughness of the sponge and the adsorption performance for oil spill cleanup (Padervand et al. 2011; Ge et al. 2014; Li et al. 2020). The nanoscale protrusion of hydrophobic TiO2 forms a rough surface similar to lotus leaf, which enhances their superhydrophobicity. The incorporated TiO2 nanoparticles produce rough structures, and PDMS is available for design of hydrophobic surface. Although good progress has been made in improving the lipophilicity and hydrophobicity of the MS, these materials still have drawbacks such as poor oil–water selectivity and poor practical application.

Inspired by these achievements (Shi et al. 2014; Li et al. 2015; Wei et al. 2018), this study developed a two-step hydrophobic fabrication of MS/TiO2/PDMS sponge with low cost, simple operation, high adsorption capacity, and high oil and water selectivity. The MS/TiO2 sponge was first prepared using the sol-gel method with the MS and tetrabutyl titanate (TBOT) as the matrix and precursor, and the MS/TiO2/PDMS sponge was finally synthesized by using dipping method with PDMS as the matrix. During this process, the TiO2 nanoparticles can be uniformly grown in situ on the MS sponge framework under the water atmosphere, and no other operation is required. This is followed by surface modification with PDMS to take advantage of its hydrophobic siloxane in an effort to enhance the hydrophobicity of the sponge.

2. EXPERIMENTAL SECTIONS

2.1. Materials

The MS was purchased from Zhengzhou Fengtai Nanomaterials Co. Ltd. TBOT (AR, ≥ 99.0%) was purchased from Tianjin Kemiou Chemical Reagent Co. Ltd. PDMS (40cst, HO-(C2H6OSi)n-H) was purchased from Shanghai Maclean Biochemical Technology Co. Ltd. The oil used in this study is an edible blend oil and sesame oil for golden arowana. All chemicals used in this study were of commercially available analytical grade.

2.2. Preparation of MS/TiO2/PDMS sponge

At room temperature, the pretreated melamine sponge was first immersed in 5%TBOT/absolute ethanol solution for 3.5 h, and then hydrolytic polycondensation occurred using the sol-gel method in a moist closed system to obtain MS/TiO2 sponge. The prepared materials were immersed in 0.5% PDMS/n-hexane solution for 3.5 h, followed by washing and drying. A new functional composite material of MS/TiO2/PDMS sponge was then obtained.
During the experiment process, the volume concentrations of TBOT absolute ethanol solution selected were 10%, 15%, 20%, and 25%, and the volume concentration of PDMS/n-hexane solution selected were 1%, 2%, and 4%.

### 2.3. Oil-water separation tests

The adsorption capacity of MS, MS/TiO₂, and MS/TiO₂/PDMS sponges for oils was determined with a weighing method. At room temperature, the sponge was placed in a beaker containing 50 mL of oil for 30 min and weighed before and after...
adsorption. The preparation process is shown in Figure 1. When the sponge stopped dripping water (oil), it was put it in a clean beaker and weighed it to calculate the water (oil) adsorption capacity. The adsorption capacity can be calculated according to the following formula:

\[ Q = \frac{m_1 - m_0}{m_0} \]

where \( Q \) refers to the water (oil) adsorption capacity of the sponge, g·g\(^{-1}\); \( m_1 \) the mass after the sponge adsorbs water (oil) saturation, g; \( m_0 \) the mass when the sponge does not adsorb water (oil), g. During the above oil adsorption test, oil was measured three times, and the final water (oil) adsorption capacity was averaged.

2.4. Characterization

The surface functional groups of MS, and MS/TiO\(_2\), MS/TiO\(_2\)/PDMS sponges were analyzed using an attenuated total reflection–Fourier transform infrared spectroscopy (ATR-FTIR, PerkinElmer, Spectrum Two) (Mohammed et al. 2014; Khalaf et al. 2021). The surface morphology was observed using scanning electron microscopy (SEM, Hitachi Model S-3400N, with gold spray). The contact angle was measured using a video optical contact angle tester (OCA20, Dataphysics). A 4 \( \mu \)L water sample was dropped as an indicator onto the surface of the sponges by sessile drop, and the measurements were performed three times on each sample and the mean contact angle values were obtained.

2.5. Reuse test of MS/TiO\(_2\)/PDMS sponge in oil–water separation

The modified MS/TiO\(_2\)/PDMS sponge was subjected to an oil (water) adsorption test, and the oil (water) adsorption capacity was calculated. After the test, the oil (water) was removed by squeezing and then the MS/TiO\(_2\)/PDMS sponge was washed with anhydrous ethanol and dried. The sponge that had been washed was subjected to repeated oil (water) adsorption experiments, and reusability capacity was finally measured.

3. RESULTS AND DISCUSSION

3.1. Morphology of materials

To study the effect of modification methods on the surface morphology of MS, the surface morphology of the MS before and after modification were monitored by SEM (as shown in Figure 2). It can be seen that the unmodified MS presents a 3D porous network structure which allows the sponge to have a large specific surface area, while the skeleton is particularly smooth (Figure 2(e)). In comparison, the surface of the MS/TiO\(_2\) composite has adherents, and the surface roughness increases in Figure 2(b)–2(d). With an enlarged surface of MS/TiO\(_2\) sponge, it is clearly shown that there are small nanoparticles on the MS/TiO\(_2\) when the concentration of TBOT solution was 5% (Figure 2(f)). TiO\(_2\) nanoparticles on the skeleton of...
MS/TiO₂ sponge increase with the concentration of TBOT solution (Figure 2(g)); however, the laminated TiO₂ adhered on the skeleton of the MS/TiO₂ sponge when TBOT concentration was 25%. When the MS is treated by TBOT solution and then placed in a moist atmosphere, the reaction of hydrolytic polycondensation of the TBOT happened on the skeleton of MS, and TiO₂ nanoparticles were generated (Wang et al. 2007). The surface roughness of MS/TiO₂ increased because the TiO₂ nanoparticles were loaded on the skeleton of MS.

Figure 3 shows SEM images of different concentrations of PDMS-modified MS/TiO₂ under the following conditions: 20% TBOT concentration and modification time of 3.5 h. From the enlarged surface of the MS/TiO₂/PDMS sponge
(Figure 3(d)–3(f), it can be seen that the MS/TiO$_2$/PDMS sponge retained a 3D porous network compared to the unmodified MS (Figure 2(c)) and MS/TiO$_2$ (Figure 2(d)). The MS/TiO$_2$ sponge presents a coarser skeleton structure and it can be clearly observed that the surface of MS/TiO$_2$ sponge is covered with a layer of adherent material. However, it is also observed from Figure 3(d)–3(f) that the dispersion of PDMS is heterogeneous, and is mainly distributed at the junction of the sponge skeleton, and the amount of sediment on the surface increases with the increase of PDMS concentration. Furthermore, deposit sediment (PDMS: 4%) shows little change when the concentrations of PDMS exceed 2%. In addition, by comparing to Figures 2 and 3, it can be clearly seen that the pore size remains the same.

### 3.2. Chemical composition of materials

The chemical composition of unmodified MS, MS/TiO$_2$ sponge, and MS/TiO$_2$/PDMS sponge were investigated by infrared (IR)-ATR analysis, as shown in Figures 4 and 5. Figure 4 shows the IR-ATR image of unmodified MS (Figure 4(a)) and modified MS with different concentrations of TBOT (Figure 4(b) and 4(c)) at 3.5 h. The spectrum of the MS shows absorption bands at 807 cm$^{-1}$, 1,540 cm$^{-1}$, and 3,309 cm$^{-1}$, which are assigned to triazine ring bending, C–N stretching, and N–H stretching, respectively. Furthermore, bands around 1,325 cm$^{-1}$ and 1,467 cm$^{-1}$ are indicative of –CH– bending. Moreover, two small peaks at 2,800–2,900 cm$^{-1}$ are attributed to –CH$_2$ and –CH$_3$ stretching. These groups of absorption bands verify the chemical composition of the MS (Figure 4(a)). When compared to unmodified MS, the weak stretching vibration absorption peak of Ti–O bond appears at 679 cm$^{-1}$ (see Figure 4(b) and 4(c)), which confirms that the TiO$_2$ particles were successfully loaded on the surface of MS. At the same time, the intensity of the stretching vibration peak of the hydrophilic group –CO at 1,140 cm$^{-1}$ and 1,120 cm$^{-1}$, and the absorption peak intensity of the triazine ring bending vibration at 1,325 cm$^{-1}$ and 807 cm$^{-1}$ gradually decrease or even disappear (Figure 4(b) and 4(c)) with the increasing concentration of TBOT. According to Ge’s report (Ge et al. 2016), the surface of TiO$_2$ has a certain interaction force with the hydrophilic group. The hydrophilic groups (–NH, –C=O and –CO) of the surface of MS and that of the TiO$_2$ interact via a secondary bond. The force makes the TiO$_2$ nanoparticles tightly attach to the surface of the MS skeleton. The functional group of the MS surface has changed, and the surface of the sponge changes from a hydrophilic surface to a hydrophobic surface. The analysis results showed that the surface composition of the modified MS was changed and the hydrophilic group was relatively weakened. In other words, the MS/TiO$_2$ sponge gradually changed from hydrophilic to hydrophobic after treatment with the TBOT solution.

The IR-ATR images of the MS/TiO$_2$/PDMS sponge with different concentrations of PDMS-modified MS/TiO$_2$ (TBOT: 20%) are shown in Figure 5. It can be seen that the stretching vibration peaks of the Si–O bond are observed at 1,011 cm$^{-1}$ and 1,084 cm$^{-1}$, the stretching vibration peak of the Si–C bond is observed at 866 cm$^{-1}$ and 795 cm$^{-1}$, and –CH$_3$ groups are shown at the absorption peak at 2,962 cm$^{-1}$. These peaks belong to the characteristic absorption peaks

![Figure 4](https://example.com/figure4.png)

**Figure 4** | IR-ATR image of MS (a) and MS/TiO$_2$ prepared in different TBOT concentrations (TBOT concentrations: 15% (b), 20% (c) respectively).
of PDMS. Meanwhile, the intensity of the absorption peak at 2,962 cm\(^{-1}\) of \(\text{-CH}_3\) groups, the stretching vibrational absorption peak of Si-O bond and the stretching vibration absorption peak of Si-C increases gradually as the concentration of PDMS increases. The peak intensity is the maximum when the PDMS concentration is 2%, while the peak intensity decreases as the concentration of PDMS is 4%. The above results indicate that the concentration of PDMS has a certain effect on the hydrophobicity of the MS. This result is consistent with the SEM image.

3.3. Surface wettability of materials

3.3.1 Effect of TBOT concentration on the contact angles of MS/TiO\(_2\) sponge

Figure 6 shows the contact angles of unmodified MS and MS/TiO\(_2\) with different concentrations of TBOT at 3.5 h. From Figure 6(a), it can be observed that the unmodified MS has particularly good wettability, water droplets are adsorbed in less than 1 s, and the contact angle is 0°. To make the MS have the selectivity of oil and water, the surface of MS was modified...
with different concentrations of TBOT solution. Compared to the unmodified MS, the MS/TiO$_2$ manifests a certain hydrophobicity (see Figure 6). The results of the measurement of the contact angle are depicted in Figure 6. From Figure 6, it can be seen that the contact angle of the MS/TiO$_2$ gradually increases as TBOT concentration increases. When the TBOT concentration is 20%, the contact angle reaches a maximum value of 142.2°. Nevertheless, the contact angle decreases to 138.2° when the TBOT concentration increases 25%. The contact angle increases first and then decreases with the increase of TBOT concentration. The nanoscale protrusion of the hydrophobic TiO$_2$ combined with microporous structure of the original sponge forms a double rough surface, which is similar to the structure of lotus leaf, resulting in an enhanced superhydrophobicity (Barthlott & Neinhuis 1997; Cho et al. 2016). The surface roughness caused by TiO$_2$ nanoparticles from TBOT hydrolysis increases with the increase of the concentration of TBOT solution, and the contact angle increase with surface roughness, leading to the gradual increase of hydrophobicity. When excess TBOT was hydrolyzed, the large amount of TiO$_2$ particles obtained aggregated on the MS framework and had a laminaceous structure (see Figure 2(h)), which resulted in macro-scale roughness, but a decrease in the micro-scale roughness. As a result, hydrophobicity is reduced. Furthermore, the sponge modified with solid particles showed a decrease in oil adsorption capacity due to the increase of the sponge density to some extent, which indicates the contact angle decrease (Zhang et al. 2020a, 2020b). The result was consistent with the SEM. Therefore, the optimal concentration of TBOT modified MS is 20%.

3.3.2. Effect of modification times on the contact angles of MS/TiO$_2$/PDMS sponge

Figure 7 examines the effects of different modification times on the properties of modified MS when TBOT concentration was 20% and PDMS concentration was 2%. Figure 7 shows that the immersion time has a great effect on the contact angle of the MS/TiO$_2$/PDMS sponge. The contact angle of the MS/TiO$_2$/PDMS sponge gradually increases from 136.7° to 149.2° as the immersion time increases from 2 h to 3.5 h. However, the contact angle decreases to 147.2° when the immersion time increased to 4 h. During the experiment, white powder appeared on the surface of the MS/TiO$_2$/PDMS sponge when the immersion time reached 4 h. The reason may be that the MS/TiO$_2$/PDMS sponge pores were filled with a large amount of white hydrolyzed TiO$_2$ powders after drying, and this white powder easily falls off because there was no force between TiO$_2$ particles. With the increase in modification time, the contact angle of MS/TiO$_2$/PDMS sponge increased first and then decreased mainly due to the long immersion time and the TiO$_2$ nanoparticles blocking the pores of the sponge. The TiO$_2$ nanoparticles and the siloxane cannot continue to adhere to the surface of the sponge skeleton, resulting in the decrease of contact angle of MS/TiO$_2$/PDMS sponge. Therefore, the optimal modification time of MS/TiO$_2$/PDMS sponge is 3.5 h.

3.3.3. Effect of PDMS concentrations on the contact angle of MS/TiO$_2$/PDMS sponge

Figure 8 shows the effect of PDMS concentrations on the contact angle of the MS/TiO$_2$/PDMS sponge with different concentrations of PDMS solution when the modification time of a given concentration (20%) of TBOT was 3.5 h. Compared
to the unmodified sponge (see Figure 6(a)), it can be observed that the MS/TiO₂/PDMS sponge (see Figure 8) have superior hydrophobicity. Figure 8 shows that the concentration of PDMS solution has a significant effect on the contact angle of MS/TiO₂/PDMS sponge. The contact angle of the MS/TiO₂/PDMS sponge increases rapidly as the concentration of PDMS solution increases. When the concentration of PDMS is 2%, the contact angle reaches a maximum value of 149.2°. The contact angle is reduced as the concentration of PDMS is over 2%. From the analysis of the experimental process, the roughness and the contact angle of the MS/TiO₂/PDMS sponge decreases because the high concentration of 4% PDMS solution were used, and the porous structure of the MS/TiO₂/PDMS sponge was clogged during the impregnation process, which hindered the further adhesion of the PDMS to the surface of the MS/TiO₂ composite. The optimum concentration of PDMS is 2%.

3.4. Oil adsorption capacity of the sponge
3.4.1. Oil-water adsorption capacity of the sponge before and after modification
To test oil (or water) adsorption capacity of the sponge after modification, the MS/TiO₂/PDMS sponge was prepared using a 2% PDMS concentration, and 20% of modified TBOT was used for 3.5 h. Figure 9 demonstrates the MS and MS/TiO₂/PDMS sponge immersed in water and oil. Compared to the unmodified MS, the MS/TiO₂/PDMS sponge seems to be plated with a bright silver film on the surface (see Figure 9(a)) after being immersed in water, due to the superhydrophobicity of the
MS/TiO$_2$/PDMS sponge. When the MS/TiO$_2$/PDMS sponge is squeezed and immersed in water under external force, a dense layer of air bubbles adheres to the surface of the modified sponge, and this layer of bubbles reflects incidental light and produces a silver film, which demonstrates the hydrophobicity of the modified sponge. When the pressure stops, the MS/TiO$_2$/PDMS sponge will immediately emerge from the water, and will not adsorb water for several hours. (Figure 9(b) shows the sponge on the water surface. The quality of the sponge will also not change significantly before and after immersing in the water. The Cassie–Baxter nonwetting behavior leads to this phenomenon (Cassie & Baxter 1944; Pham & Dickerson 2014). Figure 9(c) and 9(d) are the unmodified MS and the modified MS/TiO$_2$/PDMS sponge immersed in oil, respectively. It can be observed that when they are immersed in oil, they both rapidly adsorb oil and sink. The experimental results show that the modified sponge has lipophilic and hydrophobic properties, it is not wetted in the water, and it floats on the water surface; however, it quickly adsorbs oil and sinks in the oil. It has a selectivity for oil and water. The unmodified sponge has lipophilic and hydrophilic properties. It quickly adsorbs water, and sinks in the water, and it also adsorbs oil and sinks in the oil. This means it does not have oil–water selectivity. Therefore, MS/TiO$_2$/PDMS sponge can be used as oil–water adsorptive separation materials.

Figure 10 shows the process of the MS/TiO$_2$/PDMS sponge selective adsorption of oil in water. It can be observed from the image that the modified MS/TiO$_2$/PDMS sponge can quickly and selectively adsorb oil in water and achieve the effect of oil–water separation and purification of water, which shows a potential use for the separation of oil–water mixtures in real life.

### 3.4.2. Effect of TBOT concentrations on the oil–water adsorption capacity of MS/TiO$_2$/PDMS sponge

Figure 11 shows the oil–water adsorption capacity of MS/TiO$_2$/PDMS sponge prepared with different concentrations of TBOT solution when the modification time of a given concentration (2%) of PDMS was 3.5 h. It can be observed from

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**Figure 10** | Photographs of MS/TiO$_2$/PDMS sponge selective adsorption of edible blend oil from water.
the image that the adsorption capacity of the unmodified sponge to water and oil were 136 g·g⁻¹ and 123 g·g⁻¹, respectively, which are much higher than that of MS/TiO₂/PDMS sponge. In the unmodified MS, the condensation product structure of melamine and formaldehyde contains both hydrophilic polar group (imine) and hydrophobic group (methylene). MS therefore has a certain hydrophilic and hydrophobic property (see Figure 11). However, the water adsorption capacity of the MS/TiO₂/PDMS sponge gradually decreases as the concentration of the TBOT solution gradually increases, i.e. the hydrophobicity of MS/TiO₂/PDMS sponge increases. When the TBOT concentration is 20%, the adsorption capacity of MS/TiO₂/PDMS sponge reaches a maximum of 98.5 g·g⁻¹; however, the hydrophobicity of MS/TiO₂/PDMS sponge decreases when the TBOT concentration is over 20%. This is consistent with the measurement of the contact angle of MS/TiO₂/PDMS sponge. There is no significant change in oil adsorption capacity before and after MS modification. That is because although the MS/TiO₂/PDMS sponge has an increased roughness compared to the unmodified sponge, the porosity of the modified MS/TiO₂/PDMS sponge decreases, resulting in no increase in adsorption capacity. Compared with the adsorption capacity of MS@SiO₂@VTMS sponge to soybean oil (71 g·g⁻¹) (Gao et al. 2018), SiO₂-DTMS-MS to peanut oil (46 g·g⁻¹) (Yujing et al. 2019), Ag/PDA/MS to soybean oil (90 g·g⁻¹) (Xu et al. 2015), S-PDA-Fe₃O₄-Ag-ODA to edible oil (91 g·g⁻¹) (Chen et al. 2021), CW-coated Fe₃O₄@MS to bean oil (78 g·g⁻¹) (Yin et al. 2020) (see Table 1), the adsorption capacity of MS/TiO₂/PDMS sponge far exceeds the adsorption capacity reported in the literature, and the preparation process is simpler. The results show that the MS/TiO₂/PDMS sponge has higher adsorption capacity and can be used in practice.

3.4.3. Effect of PDMS concentrations on the oil–water adsorption capacity of the MS/TiO₂/PDMS sponge

Figure 12 shows the oil–water adsorption capacity of the MS/TiO₂/PDMS sponge prepared with different concentrations of PDMS solution when the modification time of a given concentration (20%) of TBOT was 3.5 h. It can be observed from the image that the adsorption capacity of the modified MS/TiO₂/PDMS sponge gradually increases as the concentration of the PDMS solution gradually increases, i.e. the hydrophobicity of MS/TiO₂/PDMS sponge increases. When the PDMS concentration is 2%, the adsorption capacity of MS/TiO₂/PDMS sponge reaches a maximum of 98.5 g·g⁻¹; however, the adsorption capacity of the MS/TiO₂/PDMS sponge decreases when the PDMS concentration is over 2%. This is consistent with the measurement of the contact angle of MS/TiO₂/PDMS sponge (see Figure 8). The possible reason is that when an appropriate amount of PDMS is added, PDMS completely interacts with TiO₂ and remains multistage structural pore of the sponge. When excessive PDMS is added, the excess PDMS clogs the porous structure of the MS/TiO₂/PDMS sponge, resulting in declining surface hydrophobicity, and the oil absorption ability is reduced.

![Figure 11](image-url) Edible blend oil and water adsorption capacity of MS/TiO₂/PDMS sponge with different TBOT concentrations when the modification time of a given concentration (2%) of PDMS was 3.5 h.
3.4.4. Effect of pH on the oil–water adsorption capacity of MS/TiO₂/PDMS sponge

To investigate the effects of solution pH on adsorption capacity, MS/TiO₂/PDMS was immersed in oily wastewater (prepared using HCl, distilled water, and NaOH) with pH values of 4–10. Figure 13 shows that the adsorption capacity of the MS/TiO₂/PDMS sponge at different pH when the modification time of a given concentration (2%) of PDMS and (20%) of TBOT was 3.5 h. Under acidic conditions, the adsorption capacity remains constant with increasing pH. When the pH was 7, the adsorption capacity of MS/TiO₂/PDMS sponge reached a maximum of 98.5 g·g⁻¹. When the pH value continued to rise and the solution became alkaline, the adsorption capacity decreased. The above results indicate that for MS/TiO₂/PDMS sponges, both alkaline and acidic environments are not suitable for the adsorption, and that the optimum pH for adsorbing oil by MS/TiO₂/PDMS sponge was 7. The possible reasons are, on the one hand, under alkaline condition, due to partial saponification of vegetable oil, the charge changed from ester group to carboxylate, which enhanced the hydrophilicity of modified sponge; on the other hand, partial hydrolysis of PDMS occurs under strong alkaline conditions, resulting in the decrease of the lipophilicity of the adsorbent. The results were consistent with the reported in the literature (Cai et al. 2019; Wang et al.

![Figure 12](image1.png)

Figure 12 | Edible blend oil and water adsorption capacity of MS/TiO₂/PDMS sponge with different PDMS concentrations when the modification time of a given concentration (20%) of TBOT was 3.5 h.

![Figure 13](image2.png)

Figure 13 | Edible blend oil and water adsorption capacity of MS/TiO₂/PDMS sponge with different pH when the modification time of a given concentration (2%) of PDMS and (20%) of TBOT was 3.5 h.
2019; Wang et al. 2021), which the contact angle of modified MS firstly increases and then decreases with the increase of solution pH, and reaches the maximum value when pH = 7.

3.4.5. Reusability of MS/TiO2/PDMS sponge

Figure 14 shows the results of repeated use of the unmodified and modified sponges prepared was chosen, when the concentration of TBOT solution was 20%, the concentration of PDMS solution was 2%, and modification time was 3.5 h. It can be seen that MS has high adsorption capacity of not only water (123 g·g⁻¹) but also of oil (117 g·g⁻¹) (see Figure 14) due to MS having a hydrophilic polar group (imine) and hydrophobic group (methylene). However, the water adsorption capacity of the original MS decreases continuously with an increase in cycle times, and the oil adsorption capacity of MS is basically stable after reusing 10 times. For the MS/TiO2/PDMS sponge, with the increase of the number of cycles, the oil adsorption capacity is stable, while the water adsorption capacity increases slowly. The increase of water adsorption capacity of the MS/TiO2/PDMS sponge is because the TiO2 nanoparticles and PDMS molecule on the surface of the modified sponge skeleton is removed when continuously squeezed. From the experiments of reusing, the modified MS/TiO2/PDMS sponge can be recycled more than 10 times. This result shows that the MS/TiO2/PDMS sponge has better oil–water selectivity and reusability.

3.5. The oil–water separation mechanism with MS/TiO2/PDMS sponge

According to the wettability theory, high selective adsorption of oil or organic solvent can be achieved on the sponge by constructing the rough surface morphology or modifying the material with low surface energy (Zhou et al. 2019). The MS/TiO2/PDMS sponge indicated an excellent superhydrophilic and porous structure, which prompted a further study of the mechanism of oil–water separation. Figure 15 shows the schematic of the oil–water separation mechanism with the MS/TiO2/PDMS sponge. Before modification, the condensation product structure of melamine and formaldehyde contains both hydrophilic polar group (imine) and hydrophobic group (methylene); therefore, MS has a certain hydrophilic and hydrophobic (see Figure 11). When MS was immersed in TBOT/absolute ethanol solution, the TiO2 nanoparticles were uniformly grown in situ on the MS sponge framework by the sol-gel method. The nanoscale protrusion of the hydrophobic TiO2 combined with microporous structure of the original sponge forms a double rough surface, which is similar to the structure of lotus leaf (Barthlott & Neinhuis 1997; Cho et al. 2016). Furthermore, TiO2 nanoparticles with certain hydrophobicity and high surface-to-volume ratio were supported on the porous wall of the MS, and the rough structure strengthened its hydrophobicity. After further modification by hydrophobic PDMS, the hydrophobicity of the sponge wall was further strengthened, so that the sponge formed rich hydrophobic pores with multistage structure. The hydrophobicity of MS/TiO2/PDMS sponge was significantly enhanced, which enhanced its oil–water separation efficiency. Furthermore, under the action of capillary
force, the oil is easily adsorbed into the sponge, while water is completely excluded from the hydrophobic surface due to the high surface tension, indicating that MS/TiO$_2$/PDMS sponge has the ability of oil–water separation. Due to oleophilic silanization of PDMS on the sponge’s interconnected skeleton, the adsorbed oil rapidly diffuses into the interior space of the sponge. The oil is then stored inside the porous MS/TiO$_2$/PDMS sponge, leading to its high oil adsorption capacity (Gao et al. 2018). In a word, the oil adsorption capacity of the MS/TiO$_2$/PDMS sponge after hydrophobicity modification is determined by the double factors of multistage structural pore and hydrophobic pore wall.

4. CONCLUSION

In this paper, the MS was chosen as the raw material to prepare the MS/TiO$_2$ sponge via the sol-gel process. The superhydrophobic MS/TiO$_2$/PDMS sponges were then prepared by dipping MS/TiO$_2$ sponge in PDMS solution. These fabricated MS/TiO$_2$/PDMS sponges displayed excellent hydrophobicity, and also higher oil–water selectivity and adsorption capacity for different kinds of oils. The results showed that the optimal hydrophobicity of the prepared MS/TiO$_2$/PDMS sponge was achieved when the TBOT concentration was 20%, the PDMS concentration was 2%, and the modification time was 3.5 h. The contact angle of the MS/TiO$_2$/PDMS sponge reached 149.2°, while the adsorption capacity was 98.5 g·g$^{-1}$. After the oil–water separation test, the MS/TiO$_2$/PDMS sponge was recycled more than 25 times. The super oil adsorption capacity of the MS/TiO$_2$/PDMS sponge after hydrophobicity modification is determined by the double factors of multistage structural pore and hydrophobic pore wall. These conclusions suggest that by changing the chemical composition of the surface of the 3D porous material, it is possible to prepare oil-adsorbing materials that have super oil–water selectivity properties, and are convenient and environmentally friendly to use. These materials have potential to be widely used in oil–water separation processes.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

Adebajo, M. O., Frost, R. L., Kloprogge, J. T., Carmody, O. & Kokot, S. 2003 Porous materials for oil spill cleanup: a review of synthesis and adsorbing properties. Journal of Porous Materials 10, 159–170.

Barthlott, W. & Neinhuis, C. 1997 Purity of the sacred lotus, or escape from contamination in biological surfaces. Planta 202, 1–8.
Cai, L., Zhang, Y., Zhou, Y., Zhang, X., Ji, L., Song, W., Zhang, H. & Liu, J. 2019 Effective adsorption of diesel oil by crab-shell-derived biochar nanomaterials. *Materials* **12**, 236.

Cassie, A. B. & Baxter, S. 1944 Wettability of porous surfaces. *Transactions of the Faraday Society* **40**, 546–551.

Chen, X., Weibel, J. A. & Garimella, S. V. 2016 Continuous oil–water separation using polydimethylsiloxane-functionalized melamine sponge. *Industrial & Engineering Chemistry Research* **55** (12), 3596–3602.

Chen, T., Zhou, S., Hu, Z., Fu, X., Liu, Z., Su, B., Wan, H., Du, X. & Gao, Z. 2021 A multifunctional superhydrophobic melamine sponge decorated with Fe3O4/Ag nanocomposites for high efficient oil-water separation and antibacterial application. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **626** (5), 127041–127051.

Cho, E.-C., Chang-Jian, C.-W., Hsiao, Y.-S., Lee, K.-C. & Huang, J.-H. 2016 Interfacial engineering of melamine sponges using hydrophobic TiO2 nanoparticles for effective oil/water separation. *Journal of the Taiwan Institute of Chemical Engineers* **67**, 476–483.

Chung, C. H., Liu, W. C. & Hong, J. L. 2018 Superhydrophobic melamine sponge modified by cross-linked urea network as recyclable oil absorbent materials. *Industrial and Engineering Chemistry Research* **57**, 8449–8459.

Duman, O., Diker, C. Ö. & Tun, S. 2021 Development of highly hydrophobic and superoleophilic fluoroo organothiol-coated carbonized melamine sponge/rGO composite absorbent material for the efficient and selective absorption of oily substances from aqueous environments. *Journal of Environmental Chemical Engineering* **9**, 105093–105107.

Ejeta, D. D., Wang, C.-F., Lin, C.-H., Kuo, S.-W., Chen, J.-K., Tsai, H.-C., Hung, W.-S., Hu, C.-C. & Lai, J.-Y. 2021 Preparation of a main-chain-type polybenzoxazine-modified melamine sponge via non-solvent-induced phase inversion for oil absorption and very-high-flux separation of water-in-oil emulsions. *Separation and Purification Technology* **265**, 118387–118396.

Fan, Y., Ma, W., Han, D., Gan, S., Dong, X. & Niu, L. 2015 Convenient recycling of 3D AgX/graphene aerogels (X = Br, Cl) for efficient photocatalytic degradation of water pollutants. *Advanced Materials* **27** (25), 3767–3773.

Feng, Y. & Yao, J. 2018 Design of melamine sponge-based three-dimensional porous materials toward applications. *Industrial and Engineering Chemistry Research* **57**, 7322–7330.

Gao, H., Sun, P., Zhang, Y., Zeng, X., Wang, D., Zhang, Y., Wang, W. & Wu, J. 2018 A two-step hydrophobic fabrication of melamine sponge for oil absorption and oil/water separation. *Surface & Coatings Technology* **339**, 147–154.

Ge, J., Ye, Y. D., Yao, H. B., Zhu, X., Wang, X., Wu, L., Wang, J.-L., Ding, H., Yong, N., He, L.-H. & Yu, S.-H. 2014 Pumping through porous hydrophobic/oleophilic materials: an alternative technology for oil spill remediation. *Angewandte Chemie International Edition* **53** (14), 3612–3616.

Ge, J., Zhao, H. Y., Zhu, H. W., Huang, J., Shi, L.-A. & Yu, S.-H. 2016 Advanced sorbents for oil-spill cleanup: recent advances and future perspectives. *Advanced Materials* **28** (47), 10459–10490.

Guan, Y. H., Cheng, F. Q. & Pan, Z. H. 2019 Superwetting polymeric three dimensional (3D) porous materials for oil/water separation: a review. *Polymers* **11**, 806–839.

Gupta, S. & Tai, N. H. 2016 Carbon materials as oil sorbents: a review on the synthesis and performance. *Journal of Materials Chemistry A* **4**, 1550–1565.

Gurav, A. B. 2015 Superhydrophobic/superoleophilic magnetic polyurethane sponge for oil/water separation. *RSC Advances* **5**, 68293–68298.

Hu, D., Li, L., Li, Y. & Yang, C. 2016 Fibrous coalescer for the treatment of hydrometallurgical oil dispersions. *Industrial and Engineering Chemistry Research* **55**, 11809–11817.

Ke, Q., Jin, Y., Jiang, P. & Yu, Y. 2014 Oil/water separation performances of superhydrophobic and superoleophilic sponges. *Langmuir* **30**, 13137–13142.

Khalaf, I. H., Al-Sudani, F. T., Abdul Razak, A. A., Aldahri, T. & Rohani, S. 2021 Optimization of Congo red dye adsorption from wastewater by a modified commercial zeolite catalyst using response surface modeling approach. *Water Science and Technology* **83** (6), 1369–1383.

Khosravi, M. & Azizian, S. 2015 Synthesis of a novel highly oleophilic and highly hydrophobic approach for rapid oil spill cleanup. *ACS Applied Materials & Interfaces* **7**, 25326–25333.

Kleindienst, S., Paul, J. H. & Joyce, S. B. 2015 Using dispersants after oil spills: impacts on the composition and activity of microbial communities. *Nature Reviews Microbiology* **13**, 388–396.

Lei, Z., Zhang, G., Ouyang, Y., Liang, Y., Deng, Y. & Wang, C. 2017a Simple fabrication of multi-functional melamine sponges. *Materials Letters* **190**, 119–122.

Lei, Z., Zhang, G., Deng, Y. & Wang, C. 2017b Thermoresponsive melamine sponges with switchable wettability by interface-initiated atom transfer radical polymerization for oil/water separation. *ACS Applied Materials & Interfaces* **9**, 8967–8974.

Li, B., Liu, X., Zhang, X., Zou, J., Chai, W. & Lou, Y. 2015 Rapid adsorption for oil using superhydrophobic and superoleophilic polyurethane sponge. *Journal of Chemical Technology & Biotechnology* **90**, 2106–2112.

Li, D. K. & Guo, Z. G. 2017 Versatile superamphiphobic cotton fabrics fabricated by coating with SiO2/FOTS. *Applications of Surface Science* **426**, 271–278.

Li, Z. T., Lin, B., Jiang, L. W., Lin, E. C., Chen, J., Zhang, S. J., Tang, Y. W., He, F. A. & Li, D. H. 2018 Effective preparation of magnetic superhydrophobic Fe3O4/PU sponge for oil-water separation. *Applications of Surface Science* **427**, 56–64.

Li, F., Kong, W., Zhao, X. & Pan, Y. 2020 Multifunctional TiO2-based superoleophobic/superhydrophilic coating for oil-water separation and oil purification. *ACS Applied Materials & Interfaces* **12** (15), 18074–18083.
Shi, X., Lan, Y. & Peng, S. 2020 Green fabrication of a multifunctional sponge as an absorbent for highly efficient oil/water separation. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 567, 128–138.

Liu, W., Huang, X., Peng, K., Xiong, Y., Zhang, J., Lu, L., Liu, J. & Li, S. 2021 PDA-PEI copolymerized highly hydrophobic sponge for oil-in-water emulsion separation via oil adsorption and water filtration. *Surface & Coatings Technology* 406, 126743.

Mohammed, M. I., Abdul Razak, A. A. & Hussein Al-Timimi, D. A. 2014 Modified multiwalled carbon nanotubes for treatment of some organic dyes in wastewater. *Advances in Materials Science and Engineering* 1, 1–10. doi:10.1155/2014/201052.

Nguyen, D. D., Tai, N. H., Lee, S. B. & Kuo, W. S. 2012 Superhydrophobic and superoleophilic properties of graphene-based sponges fabricated using a facile dip coating method. *Energy & Environmental Science* 5 (7), 7908–7912.

Padervand, M., Tasviri, M. & Gholami, M. R. 2011 Effective photocatalytic degradation of an azo dye over nanosized Ag/AgBr-modified TiO2 loaded on zeolite. *Chemical Papers* 65, 280–288.

Peng, M., Zhu, Y., Li, H., He, K., Zeng, G., Chen, A., Huang, Z., Huang, T., Yuan, L. & Chen, G. 2019 Synthesis and application of modified commercial sponges for oil-water separation. *Chemical Engineering Journal* 373, 213–226.

Pham, V. H. & Dickerson, J. H. 2014 Superhydrophobic silanized melamine sponges as high efficiency oil absorbent materials. *ACS Applied Materials & Interfaces* 6, 14181–14188.

Qiang, F., Hu, L. L., Gong, L. X., Zhao, L., Li, S. N. & Tang, L. C. 2017 Facile synthesis of superhydrophobic, electrically conductive and mechanically flexible functionalized graphene nanoribbons/polyurethane sponge for efficient oil/water separation at static and dynamic states. *Chemical Engineering Journal* 334, 2154–2166.

Qiu, S., Jiang, B., Zheng, X., Zheng, J., Zhu, C. & Wu, M. 2015 Hydrophobic and fire-resistant carbon monolith from melamine sponge: a recyclable sorbent for oil-water separation. *Carbohydrate Polymers* 144, 551–559.

Qin, L., Zeng, G., Lai, C., Huang, D., Xu, P., Zhang, C., Cheng, M., Liu, X., Liu, S., Li, B. & Yi, H. 2018 ‘Gold rush’ in modern science: fabrication strategies and typical advanced applications of gold nanoparticles in sensing. *Coordination Chemistry Reviews* 359, 1–31.

Raju, K. G., Gary, J. D., Matt, W. E. & Atsushi, H. 2017 Oil/water separation techniques: a review of recent progresses and future directions. *Journal of Materials Chemistry A* 5, 16025–16058.

Ruan, C., Ai, K., Li, X. & Lu, L. 2014 A superhydrophobic sponge with excellent absorbency and flame retardancy. *Angewandte Chemie International Edition* 53, 5556.

Sadiq, A. J., Shabeec, K. M., Khalil, B. I. & Alsalsha, Q. F. 2020 Effect of embedding MWCNT-g-GO with PVC on the performance of PVC membranes for oily wastewater treatment. *Chemical Engineering Communications* 207 (6), 733–750.

Seddighi, M. & Hejazi, S. M. 2015 Water-oil separation performance of technical textiles used for marine pollution disasters. *Marine Pollution Bulletin* 96, 286–293.

Shi, H., Shi, D., Yin, L., Yin, L., Yang, Z., Luan, S., Gao, J., Zha, J., Yin, J. & Li, R. K. Y. 2014 Ultrasonication assisted preparation of carbonaceous nanoparticles modified polyurethane foam with good conductivity and high oil absorption properties. *Nanoscale* 6, 13748–13753.

Shi, X., Lan, Y. & Peng, S. 2020 Green fabrication of a multifunctional sponge as an absorbent for highly efficient and ultrafast oil – water separation. *ACS Omega* 5, 14232–14241.

Tran, V. T. & Lee, B. K. 2017 Novel fabrication of a robust superhydrophobic PU@ZnO@Fe3O4@SA sponge and its application in oil-water separations. *Scientific Reports* 7, 15702.

Varela, A., Oliveira, G., Souza, F. G. S., Rodrigues, C. H. M. & Costa, M. A. S. 2013 New petroleum absorbers based on cardanol-furfuraldehyde magnetic nanocomposites. *Polymer Engineering & Science* 53 (1), 44–51.

Wang, N. & Deng, Z. 2019 Synthesis of magnetic, durable and superhydrophobic carbon sponges for oil/water separation. *Materials Research Bulletin* 115, 19–26.

Wang, X., Daodao Hu, A. & Yang, J. 2007 Synthesis of PAM/TiO2 composite microspheres with hierarchical surface morphologies. *Chemistry of Materials* 19 (10), 2610–2621.

Wang, B., Liang, W., Guo, Z. & Liu, W. 2015 Biomimetic superlyophobic and super-lyophilic materials applied for oil/water separation: a new strategy beyond nature. *Chemical Society Reviews* 44, 336–361.

Wang, Y., Chen, A. & Peng, M. 2019 Preparation and characterization of a fluorized kaoline modified melamine sponge as an absorbent for efficient and rapid oil/water separation. *Journal of Cleaner Production* 217, 508–316.

Wang, H., Gao, F., Ren, R., Wang, Z., Yue, R., Wei, J., Wang, X., Kong, Z., Zhang, H. & Zhang, X. 2021 Caffeic acid polymer rapidly modified sponge with excellent anti-oil-adhesion property and efficient separation of oil-in-water emulsions. *Journal of Hazardous Materials* 404, 124197–124210.

Wei, Q., Oribayo, O., Peng, X., Rempel, G. L. & Pan, Q. 2018 Synthesis of polyurethane foams loaded with TiO2 nanoparticles and their modification for enhanced performance in oil spill cleanup. *Industrial and Engineering Chemistry Research* 57, 8918–8926.

Xu, Z., Miyazaki, K. & Hori, T. 2015 Dopamine-induced superhydrophobic melamine foam for oil/water separation. *Advanced Materials Interfaces* 2, 1500255–1500263.

Yan, S., Li, Y., Xie, F., Wu, J., Jia, X., Yang, J., Song, H. & Zhang, Z. 2020 Environmentally safe and porous MS@TiO2@PPy monoliths with superior visible-light photocatalytic properties for rapid oil – water separation and water purification. *ACS Sustainable Chemistry & Engineering* 8, 5347–5359.

Yao, Q., Zhao, P., Li, R., Li, C., Luo, Y., Zhou, G. & Yang, M. 2017 Fabrication of recyclable carbonized asphalt-melamine sponges with high oil-absorption capability. *Journal of Chemical Technology & Biotechnology* 92, 1415–1420.
Yecom, C. & Kim, Y. 2016 Purification of oily seawater/wastewater using superhydrophobic nano-silica coated mesh and sponge. *Journal of Industrial and Engineering Chemistry* 40, 47–53.

Yin, Z., Li, Y. & Song, T. 2020 An environmentally benign approach to prepare superhydrophobic magnetic melamine sponge for effective oil/water separation. *Separation and Purification Technology* 236, 116308.

Yu, L., Hao, G., Liang, Q. & Jiang, W. 2015 Fabrication of magnetic porous silica submicroparticles for oil removal from water. *Industrial & Engineering Chemistry Research* 54 (38), 9440–9449.

Yujing, L., Ning, L., Yannan, J., Yujing, L., Ning, L., Yannan, J., Jiang, X., Yu, L. & Yan, X. 2019 Surface design of durable and recyclable superhydrophobic materials for oil/water separation. *Colloids and Surfaces A* 567, 128–138.

Zeng, Z.-w. S. & Taylor, S. E. 2020 Facile preparation of superhydrophobic melamine sponge for efficient underwater oil-water separation. *Separation and Purification Technology* 247, 116996–117003.

Zhang, J. & Seeger, S. 2015 Polyester materials with superwetting silicone nanofilaments for oil/water separation and selective oil absorption. *Advanced Functional Materials* 21, 4699–4704.

Zhang, N., Qi, Y., Zhang, Y., Luo, J., Cui, P. & Jiang, W. 2020a A review on oil/water mixture separation material. *Industrial and Engineering Chemistry Research* 59, 14546–14568.

Zhang, Z. Y., Liu, H. & Qiao, W. C. 2020b Reduced graphene-based superhydrophobic sponges modified by hexadecyltrimethoxysilane for oil adsorption. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 589, 124433.

Zhou, Y., Zhang, N., Zhou, X., Hu, Y. B., Hao, G., Li, X. D. & Jiang, W. 2019 Design of recyclable superhydrophobic PU@Fe3O4@PS sponge for removing oily contaminants from water. *Industrial and Engineering Chemistry Research* 58 (8), 3249.

Zou, W., Lei, Z., Liu, L., Wang, X., Sun, J., Wu, S., Deng, Y., Tang, C., Gao, F. & Dong, L. 2015 Engineering the Cu2O-reduced graphene oxide interface to enhance photocatalytic degradation of organic pollutants under visible light. *Applied Catalysis B Environmental* 181, 495–503.

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