Photoluminescent, Ferromagnetic, and Hydrophobic Sponges for Oil−Water Separation

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ABSTRACT: To find a facile way to produce a hydrophobic sponge that can effectively absorb oils is urgent to resolve the environmental pollution and ecological disaster caused by oil spillage. Here, alkylated carbon dots (C dots) were prepared from pyrolysis of a mixture of dodecylamine and citric acid followed by purification through silica gel column chromatography. Polyurethane sponge was modified by alkylated C dots by a simple dip-coating method, which endows the photoluminescent and hydrophobic sponge with good absorption capacities for various oils and nonpolar organic solvents with high recyclability. The water contact angle of the modified sponge can reach 138.8°. Interestingly, the sponge enables visual absorption under UV irradiation in the dark, which has not been achieved by other carbon-based adsorbents. The sponge was further made ferromagnetic by introducing alkylated Fe₃O₄ nanoparticles into its structure, which allowed controllable oil−water separation.

1. INTRODUCTION

The spillage of oil and the leakage of toxic organic solvents have become a serious problem because of their causing environmental pollution and ecological disaster. Finding means to solve these problems is urgent. The development of materials that can effectively absorb oils has attracted considerable interest in recent years.1−4 Compared to other means such as burning, skimming, bioremediation, and chemical dispersion, absorption is environmentally benign and time-saving. Among various absorbents, sponges are attractive as they are readily available and cheap. In addition, their three-dimensional (3D) networks enable facile further modifications, and their high flexibility makes it possible to recover the absorbed oils by squeezing.

For efficient oil−water separation, the commercially available sponges are usually needed to be modified to get good water repellency and oleophilicity.5 Carbon nanomaterials such as carbon nanotubes and graphene are among the most popular absorbents for oil−water separation.6 They have also been used to modify sponges to get composite materials.5 As a new member of carbon nanomaterials, carbon dots (C dots) have received great attention.7 The investigations are mainly focused on water-soluble C dots, while studies on hydrophobic ones are relatively rare.8 C dots decorated with long alkyl chains have been reported,9−14 and their applications in composite films9 and light-emitting diodes11−13 have been presented. However, connections to the absorbents for oil−water separation have not been well clarified. Recently, Wang et al. prepared a graphene-based holey framework decorated with in situ generated C dots.15 The composite framework, in the form of a film, was used in compressible supercapacitors and oil−water separation. As the C dots are totally mixed with the black graphene, no photoluminescence (PL) properties were reported. In another work, Cheng et al. prepared carbon quantum dots bearing octadecyl chains, which were anchored onto commercial textiles to get superhydrophobic films for high-flux oil−saltwater separation and membrane desalination.16 Similar to Wang’s work, the PL properties of the film have been totally ignored. In addition, since the carbon quantum dots were obtained as precipitates after pyrolysis, they have much larger sizes (38.5 ± 11.7 nm) than traditional C dots (<10 nm) and are thus better described as alkylated carbon nanoparticles. The potential of C dots in oil−water separation needs to be further explored.

In this work, we report a facile way to prepare a new type of hydrophobic sponges for oil−water separation. The strategy relies on the utilization of alkylated C dots to modify the surface of polyurethane (PU) sponge. Compared to the counterparts reported previously, the sponge presented here has high photoluminescence, which enables visual collection of oils in the dark. In addition, we further made the sponge magnetic by introducing alkylated Fe₃O₄ nanoparticles into its...
structure, which allowed the recovery of oils via oil−water separation using a magnetic field.

2. RESULTS AND DISCUSSION

The alkylated C dots were prepared from pyrolysis of a mixture of dodecylamine (DDA) and citric acid (CA, Figure 1a), followed by purification through silica gel column chromatography (for details, see the Section 4). Totally five samples were prepared, where the weight ratio of DDA to CA (W_{\text{DDA}}/W_{\text{CA}}) was varied from 10:1 to 10:5. Statistics on the yields showed that the sample with W_{\text{DDA}}/W_{\text{CA}} = 10:4 gave the highest value of 70.66% (Figure S1). In addition, wettability measurements on glass substrates spin-coated with the C dots revealed that this product also exhibited a slightly higher water contact angle (Figure 1b). It was thus selected for further study. Elemental analysis shows that, after pyrolysis, the contents of H and O decrease (Figure S2), indicating the condensation between −\text{NH}_2 and −\text{COOH}. Thermogravimetric analysis (TGA, Figure S3) shows that the C dots hardly decompose below 200 °C, indicating good thermal stability. When the temperature reaches 500 °C, most of the C dots decompose, leaving only 5.4 wt % residue. Transmission electron microscopy (TEM) observation shows that the C dots have an average diameter of \textasciitilde 3.9 nm (Figure 1c). The Fourier transform infrared (FTIR) (Figure 1d) spectrum of the C dots retains characteristic peaks from both DDA and CA. Compared to DDA, the peak from N−H shows a bathochromic shift by \textasciitilde 29 cm\(^{-1}\), confirming the chemical conversion of the −\text{NH}_2 group during pyrolysis. The peaks from alkyl chains, which locate between 2980 and 2830 cm\(^{-1}\), become broader after pyrolysis, indicating that the alkyl chains could not pack closely after being attached to the carbonized core. Because of this effect, the melting point (m_p) of the C dots (\textasciitilde 26.5 °C, Figure S4) is even lower than that of DDA (\textasciitilde 28.2 °C). The sharp and strong peak at \textasciitilde 1700 cm\(^{-1}\) indicates the presence of carbonyls in the C dots.

The C dots are soluble in a variety of solvents. The solubility depends on both the polarity and the structure of the solvent (Figure S5). The C dots are poorly soluble in highly polar organic solvents and insoluble in water, indicating their hydrophobic character. They exhibit strong absorption in the UV region with a sharp peak at around 361 nm and a shoulder one at 280 nm (Figure 1e), which can be assigned to n−π* and π−π* transitions, respectively. The PL properties of the C dots were investigated in two representative solvents, i.e., chloroform (CHCl_3) and ethanol (EtOH). In both cases, wavelength-dependent emission was observed (Figures 1e and S6). The absolute fluorescence quantum yield was determined to be 44.0% in CHCl_3 and 30.4% in EtOH. Time-resolved fluorescence measurement shows that the C dots undergo a biexponential decay with an average lifetime of 12.26 ns (Figure S7 and Table S1).

Next, the alkylated C dots were used to modify the commercial PU sponge. To prevent the C dots from falling off the surface of the sponge, poly(methyl methacrylate) (PMMA) was used as a binder. The process of modification is illustrated in Figure 2a. After modification, the sponge becomes hydrophobic. The water contact angle slightly depends on the concentration of the C dots, as seen from Figure 2b.

Figure 1. (a) Synthetic route of the alkylated C dots. (b) Contact angles of water droplets on glass substrates spin-coated with C dots (10 mg·mL\(^{-1}\) in CHCl_3, 1000 rpm) obtained by pyrolysis of the DDA/CA mixture with varying W_{\text{DDA}}/W_{\text{CA}}. (c) TEM image of the C dots obtained by pyrolysis of the DDA/CA mixture with W_{\text{DDA}}/W_{\text{CA}} = 10:4. The inset shows the size distribution. (d) FTIR spectra of CA, DDA, and the C dots obtained by pyrolysis of the DDA/CA mixture with W_{\text{DDA}}/W_{\text{CA}} = 10:4. (e) ultraviolet−visible (UV−vis) absorption and PL spectra of the C dots in CHCl_3 (W_{\text{DDA}}/W_{\text{CA}} = 10:4, 2.0 mg·mL\(^{-1}\)) recorded at varying λ\textsubscript{ex} values. The insets show the photos of the C dots under room light (left) and 365 nm UV irradiation (right).
to the good water repellency, the sponge can float on water without sinking (Figure 2c). If the sponge is forced in water by a tweezer, air bubbles will appear on the surface of the sponge, which will isolate the sponge from the bulk water (Figure 2d). Besides pure water, the sponge also shows good repellency toward highly acidic and basic aqueous solutions (Figure 2e). In control experiments, both the unmodified and PMMA-modified sponges sink to the bottom in water (Figure S8). The internal structures of the sponge before and after modification were investigated by scanning electron microscopy (SEM) observations. Before modification, the sponge has a well-defined three-dimensional (3D) network with smooth surfaces (Figure 2f). After modification, the surface of the skeleton becomes rough due to the deposition of C dots and PMMA (Figure 2g).

The performance of the PU sponge modified with alkylated C dots (denoted sponge-C hereafter) in oil–water separation has been investigated in detail. When sponge-C (2.0 cm × 1.0 cm × 1.0 cm) was put on a thick layer of peanut oil on water, the oil was gradually absorbed from the bottom of sponge-C within 20 s (Figure 3a–c and Supporting Movie 1). The absorption capacity at equilibrium \(m_{\text{max}}\) was determined to be 31.8 g·m\(^{-2}\). As sponge-C has high photoluminescence, the absorption process can be visually tracked in the dark under UV irradiation (Figure 3d–f and Supporting Movie 2). Examinations on various oils and nonpolar organic solvents showed that \(m_{\text{max}}\) ranges from ~24.2 to ~36 g·g\(^{-1}\) (Figure 3g). This value is comparable to those obtained for sponges modified by carbon nanotubes,\(^{17}\) carbon spots,\(^{18}\) and carbon fibers.\(^{19}\)

The recyclability of sponge-C has also been examined. Figure 3h gives the results for pump oil. Upon continuous absorption–desorption (by squeezing), \(m_{\text{max}}\) decreases slightly. However, even after 30 cycles, \(m_{\text{max}}\) is still as high as 25.5 g·g\(^{-1}\), which is ~80% of the original value (∼32 g·g\(^{-1}\)). Tests on other oils give similar results (Figures S9 and S10). The kinetics of the absorption was analyzed using peanut oil as an example, which can be construed by the fractal like-linear driving force (FL-LDF) model.\(^{20,21}\) The plot of the absorption capacity \(m\) of sponge-C as a function of time \(t\) is given in Figure 3i, from which one can see that the absorption is fast within the first 20 s, after which the absorption slows down and reaches equilibrium.

For the applications of the absorbents, multiple functions are required. Up to date, absorbents with anticorrosion,\(^{22,23}\) temperature resistance,\(^{24,25}\) and responsiveness triggered by pH\(^{26}\) and light\(^{27,28}\) have been reported. Specifically, absorbents that can respond to magnetic field are important as the absorption process can be facilely controlled.\(^{29–31}\) To impart current system magnetism, alkylated ferroferric oxide (Fe\(_3\)O\(_4\)) nanoparticles were prepared by pyrolysis of iron(III) acetylfenecate (Fe(acac)\(_3\)) in the presence of DFA and 1-dodecanol (for details, see Section 4). The successful preparation of the Fe\(_3\)O\(_4\) nanoparticles was proved by X-ray diffraction (XRD) measurement (Figure S11). TEM observation shows that the sizes of the Fe\(_3\)O\(_4\) nanoparticles are 10–15 nm (Figure 4a). A room-temperature \(M–H\) curve shows that the Fe\(_3\)O\(_4\) nanoparticles are strongly ferromagnetic with a saturation magnetization of ~41 emu·g\(^{-1}\), a coercivity of ~11.8 Oe, and a remanence of ~0.86 emu·g\(^{-1}\) (Figure S12). Magnetic sponge was prepared by dip-coating the pristine PU sponge into a mixture solution containing PMMA, C dots, and Fe\(_3\)O\(_4\) nanoparticles. The sponge co-modified with alkylated C dots and alkylated Fe\(_3\)O\(_4\) nanoparticles (denoted sponge-C-Fe\(_3\)O\(_4\) hereafter) well retains the water repellency, as seen from the photos in Figures 4b,c and S13. When subjected to oil–water separation, the movement of sponge-C-Fe\(_3\)O\(_4\) can be well controlled by a commercial magnet (Figure 4d and Supporting Movie 3). In addition, the introduction of the Fe\(_3\)O\(_4\) nanoparticles did not destroy the PL properties of the sponge, leading to the preservation of the visual tracking of the absorption process in the dark by UV irradiation (Figure 4e and Supporting Movie 4).

### 3. CONCLUSIONS

In summary, we have modified a commercial PU sponge with alkylated C dots via a simple dip-coating method, leading to the formation of a hydrophobic absorbent with high photoluminescence. The sponge shows good absorption capacities for various oils and nonpolar organic solvents with high recyclability. Importantly, the sponge enables visual absorption under UV irradiation in the dark, which has not been achieved by other carbon-based adsorbents. We further introduced alkylated Fe\(_3\)O\(_4\) to the system to prepare a magnetic sponge. Controllable oil–water separation was realized under an external magnetic field, which could also be visually tracked in the dark by UV irradiation. Our work provides the first example of PL carbon-based adsorbents for oil–water separation. Besides, it gives a new opportunity to extend the applications of alkylated C dots.
4. EXPERIMENTAL SECTION

4.1. Preparation of Alkylated C Dots. Alkylated C dots were prepared by the pyrolysis method. In a typical experiment, 10.00 g of DDA and 4.00 g of CA were added to a Teflon reaction tank, which was subjected to mechanical stirring at 70 °C controlled by a water bath for premixing. The tank was then sealed with stainless steel, transferred into an oven, and heated at 240 °C for 1 h. After the mixture was naturally cooled to room temperature, the crude product was purified by silica gel column chromatography using ethanol/ethyl acetate (volume ratio: 1:1) as eluent. The section containing the C dots was gathered, and the organic solvents were removed by a rotary evaporator. The so-obtained C dots were further dried at 45 °C in vacuum for three days for further use.

C dots with other W_{DDA}/W_{CA} values were prepared by following the same procedures. Totally, five types of C dots were obtained with W_{DDA}/W_{CA} values of 10:1, 10:2, 10:3, 10:4, and 10:5, respectively.

4.2. Preparation of Alkylated Fe_{3}O_{4} Nanoparticles. Fe_{3}O_{4} nanoparticles capped with alkyl chains were prepared according to the modified procedures specified in the literature. Typically, Fe(acac)₃ (0.5 mmol) was dissolved in a mixture of DDA (10.0 mL) and 1-dodecanol (2.0 mL). Then, the solution was transferred into a Teflon-lined autoclave, which was sealed and maintained at 240 °C for 2 h in an oven. After the reaction was complete, the autoclave was naturally cooled to room temperature. The black sediment was collected and washed with ethanol followed by centrifugation (4000 rpm) three times. The precipitate was dried at 45 °C in vacuum for three days for further use.

Figure 3. (a) Photo of the peanut oil (stained with Sudan I) floating on the water surface in a plate. (b) Time-dependent absorption of the peanut oil by sponge-C. (c) Photo of the bottom side of sponge-C after absorption of the peanut oil. (d) Photo of the pump oil (stained with 4-methylumbelliferone) floating on the water surface in a plate. (e) Time-dependent absorption of the pump oil by sponge-C. (f) Photo of the bottom side of sponge-C after absorption of the pump oil. Photos in (d)−(f) were taken under 365 nm UV irradiation. (g) Statistics of the equilibrium absorption capability (m_{\text{eq}}) of sponge-C on each type of oil. (h) Tests of the recyclability of sponge-C toward the removal of pump oil. (i) Plot of the absorption capacity (m_{t}) of sponge-C on peanut oil as a function of absorption time.

Figure 4. (a) TEM image of the alkylated Fe_{3}O_{4} nanoparticles. (b) Photo of sponge-C-Fe_{3}O_{4} floating on water. (c) Photos of water with varying pH on sponge-C-Fe_{3}O_{4}. The water droplets were, respectively, stained with methyl orange (pH = 2 and pH = 12) and rhodamine B (pH = 7) for better visibility. (d) Time-dependent absorption of the pump oil (stained with Sudan I) by sponge-C-Fe_{3}O_{4}. (e) Time-dependent absorption of the pump oil (stained with 4-methylumbelliferone) by sponge-C-Fe_{3}O_{4} taken under 365 nm UV irradiation.

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4.3. Treatment of the PU sponges. 4.3.1. Pretreatment of the Sponge. A piece of PU sponge (2 cm × 1 cm × 1 cm) was immersed into a solution containing 5.00 g of CrO₃, 2.77 mL of H₂SO₄ (98 wt %), and 50.0 mL of water for 1 min. The sponge was first washed with a large amount of water and then successively washed three times with ethanol and water under ultrasonication. After that, the sponge was put into an oven and dried at 60 °C for 2 h.

4.3.2. Preparation of the Sponge Modified by C Dots (Sponge-C). A stock solution was prepared by dissolving 0.10 g of PMMA and 0.15 g of C dots in 10 mL of chloroform (CHCl₃). A piece of pre-treated PU sponge was immersed into the aforementioned stock solution, followed by ultrasonication for 10 min to drive away the bubbles inside. Then, the sponge was taken out and dried in an oven at 100 °C for 30 min.

4.3.3. Preparation of the Sponge Modified by C Dots and Fe₃O₄ (Sponge-C-Fe₃O₄). The procedures for the preparation of sponge-C-Fe₃O₄ are the same as those for sponge-C, except that the stock solution contains an additional 0.04 g of alkylated Fe₃O₄ nanoparticles.

4.4. Kinetic Analysis of the Oil Absorption. To analyze the kinetics of the absorption, peanut oil was chosen as an example. In brief, sponge-C was put into the peanut oil and weighed every 4 s. The absorption capacity of sponge-C (mᵣ) as a function of the absorption time (t) can be described by the following fractal-like linear driving force (FL-LDF) model[20,21]

\[ \ln(1 - mᵣ/m_{max}) = -D' t^{\alpha} \]

where \( m_{max} \) is the maximum oil absorption capacity per unit mass of sponge-C, \( D' \) is the observed mass transfer coefficient, and \( \alpha \) is a constant. This model indicates that many pores of different sizes are present in sponge-C.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c00899.

Materials and methods; statistics of the masses and yields of C dots (Figure S1); structural characterization of the C dots including elemental analysis (Figure S2), TGA (Figure S3), DSC (Figure S4), solubility tests (Figure S5), static (Figure S6) and time-resolved (Figure S7) PL properties, and wettability tests of the unmodified and PMMA-modified sponges (Figure S8); tests of recyclability of sponge-C on different oils and organic solvents (Figure S9 and S10); XRD (Figure S11) and the M−H curve (Figure S12) of the alkylated Fe₃O₄ nanoparticles; wettability test of sponge-C-Fe₃O₄ (Figure S13) (PDF)

Oil was gradually absorbed from the bottom of sponge-C within 20 s (AVI)

Sponge-C has high photoluminescence, the absorption process can be visually tracked in the dark under UV irradiation (AVI)

Oil−water separation, the movement of sponge-C-Fe₃O₄ can be well controlled by a commercial magnet (AVI)

Visual tracking of the absorption process in the dark by UV irradiation (AVI)

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Notes

The authors declare no competing financial interest.

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