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Research paper

Superhydrophobic kaolinite modified graphene oxide-melamine sponge with excellent properties for oil-water separation

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ABSTRACT

In this work, a facile and cost-effective method was reported to fabricate a novel, robust, and superhydrophobic kaolinite modified graphene oxide-melamine sponge (K-GOMS). The GO sheets were used to increase the roughness of the sponge smooth surface. The hydrophobic kaolinite adhered on GO sheets significantly enhanced the superhydrophobicity of the K-GOMS surface with a water contact angle of 152.3°. Several organic solvents and oils were selected to test the adsorption performance of the obtained adsorbents. The results revealed that the superhydrophobic K-GOMS exhibited not only superior adsorption capacity for various organic solvents and oils, from 60 to 113 times of its own weight, but also outstanding recyclability with an adsorption capacity retention higher than 94% after 30 cycles of diesel oil adsorption, and excellent environmental tolerance in wide range of pH values and temperatures. Therefore, this study provides an effective way for constructing facile, low-cost, and large-scale production of superhydrophobic adsorbents for oil-water separation.

1. Introduction

The rapid development of modern industrialization inevitably released various types of contaminants, such as dyes, (Deng et al. 2013; Gong et al. 2009) organic compounds, (Chen et al. 2015; Chen et al. 2016b; Cheng et al. 2016; Ren et al. 2017) and heavy metals, (Hu et al. 2017: Liang et al. 2017: Tang et al. 2014: Wan et al. 2017: Xu et al. 2012b; Zeng et al. 2017) even resulted in oil spill, (Tjandra et al. 2015; Wang et al. 2017; Zhang et al. 2017) which gave rise to a continuously increasing worldwide concern for water pollution.(Long et al. 2011; Tan et al. 2015; Xu et al. 2012a) Among these pollutions, oil spill, an unavoidable problem from the exploration, production, and transportation of offshore oil, has become a serious threat to environment and ecology.(Allan et al. 2012; Qiang 2013; Zhai 2014) Cleaning up oil spill and oil slick from water surface and coastline is a challenge work. (Pham and Dickerson 2014) To date, several methods such as in situ burning,(Aurell and Gullett 2010) skimming,(Broje and Keller 2007) bioremediation,(Zahed et al. 2010)chemical dispersion,(Kujawinski et al. 2011) and adsorption(Adebajo et al. 2003; Choi and Cloud 1992) have been applied to reduce the adverse influences of oil spill in environment. Among these methods, adsorption attracted great attention because of its convenient operation, low-cost and easy design.(Lü et al.

2016; Pham and Dickerson 2014) The traditional porous adsorbents such as exfoliated graphite,(Wang et al. 2010) wool,(Radetić et al. 2003) and vegetable fibers(Annunciado et al. 2005) generally have drawbacks including low adsorption capacity, poor recyclability and environmental adaption, thereby limiting their application.(Lü et al. 2016; Wu et al. 2017; Zhang et al. 2015b) Recently, sponge adsorbents have aroused considerable interest in the field of oil-water separation due to their low density, good flexibility, high porosity, and three dimensional skeleton structure.(Zhou et al. 2016; Zhu et al. 2011) However, sponge is naturally hydrophilic and oleophilic, which restricts the separation of oils and organic solvents from water.(Wang et al. 2017)

With the purpose of sponge application for oil-water separation, modifications are necessary to transform the hydrophilic-oleophilic sponge surface to hydrophobic-oleophilic surface as well as increase the surface roughness for enhancing the adsorption capacity of organic solvents and oils. For example, TiO_2 nanoparticles were used to modify the melamine sponge surface, which could significantly enhance the hydrophobicity of melamine sponge.(Cho et al. 2016) A melamine sponge incorporated with Fe₃O₄ nanoparticles and lignin not only exhibited excellent hydrophobicity and self-cleaning performance, but also could be magnetically driven to absorb the oils.(Lei et al. 2017) More recently, graphene oxide (GO), composed of two-dimensional sp^2

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carbon network, with one-atom thickness honeycomb lattice structure, can greatly enhance the pollutant removal performance due to its intriguing structure and unique physicochemical properties.(He et al. 2017; Li et al. 2010; Wang and Chen 2015; Zhang et al. 2015b) For example, (Szabó et al. 2013) prepared TiO₂-GO nanocomposites for phenol photooxidation. The presence of GO induced an accelerate sedimentation of catalyst slurry to completely separate the composites from the phenol solution in a short time as compared to pure TiO₂. Besides, GO has been reported as good adsorbent for the removal of a wide range of organic solvents. (Dékány et al. 1998) Moreover, GO coated on the skeletons of sponge could significantly increase the roughness of the sponge surface and improve the adsorption capacity of the modified sponge. Rahmani et al. (2017) immersed polyurethane (PU) sponge into GO solution to obtain GO/PU sponge. And it is confirmed that the coating process does not have a profound impact on the porous structure of the PU sponge. Even, the presence of GO gives the roughness of the surface and improved the adsorption capacity and recyclability. However, GO shows hydrophilic property due to its abundant oxygen-containing functional groups,(Lerf et al. 2006; Tamás Szabó et al. 2015) which limits its application of oil-water separation. Therefore, there is a need to explore a feasible approach to enhance the hydrophobicity of graphene oxide sponge for oil-water separation. It is well known that the chemical reduction of GO is a facile method to alter the hydrophilicity of GO. The commonly used reducing agents including N₂H₄, (Szabó et al. 2005) NaBH₄, hydroquinone, and so on. (Athanasios B. Bourlinos et al. 2003) Although the reduction of GO is considered to be an effective approach to enhance the hydrophobicity of the fabricated graphene oxide sponge, (Gao et al. 2009; Liu et al. 2015; Tjandra et al. 2015; Zhou et al. 2016) the reducing agents commonly used for the reduction of GO are either toxic or hazardous. Besides, the preparation methods were relatively complex. These restrict the large-scale preparation of graphene-based sponge.(Li et al. 2010) To our knowledge, various hydrophobic materials have been used for oil-water separation. (Cho et al. 2016; Lei et al. 2017) However, it is still an open question whether the combination of graphene oxide sponge with hydrophobic materials will enhance the oils removal performance. Thus, it is worthwhile to explore the potential of hydrophobic materials modified graphene oxide sponge in oil-water separation.

Clay mineral materials have received great attention due to their low-cost, easy availability, and environmental friendly.(Huang et al. 2016a) Among those materials, kaolinite, is abundant in kaolin, has been widely used for different applications because of its rich reserves, chemical stabilization, and easy modification.(Huang et al. 2016b) The modified kaolinite composites have been regarded as efficient adsorbents for the removal of dyes,(Huang et al. 2016a; Nandi et al. 2009) and heavy metals.(Huang et al. 2016b) Whereas, the application of kaolinite composites for oil-water separation is vacant. It has been reported that kaolinite can be diversely fabricated to the hydrophobic, oleophobic-hydrophilic, and amphiphobic materials, respectively.(Qu et al. 2016) Based on this property, the hydrophobic kaolinite was used to increase the hydrophobicity of graphene oxide melamine sponge for oil-water separation.

In this work, the main objective was to fabricate a superhydrophobic kaolinite modified graphene oxide-melamine sponge (K-GOMS) for oil-water separation. As a comparison, superhydrophobic kaolinite modified melamine sponge (K-MS) was prepared to test the effect of GO sheets on K-GOMS. Moreover, adsorption experiments were carried out to compare the adsorption performance of K-MS and K-GOMS for the removal of various organic solvents and oils. The results indicated that the prepared K-GOMS exhibited high adsorption capacity, outstanding recyclability and excellent environmental adaption. It is believed that this study can provide a novel and practical methodology for further constructing facile, low-cost, and high quality superhydrophobic adsorbents for oil spill treatment in environment.

2. Materials and methods

2.1. Materials

Graphite powder (C, \geq 99.85) was obtained from Sinopharm Chemical Reagent Co., Ltd. Commercial melamine sponges were purchased from Chengdu Xuyang Sponge Product Co., Ltd. Kaolin, the major composition is kaolinite (Al₂Si₂O₉H₄), stearic acid, K₂S₂O₈, P₂O₅, 3-aminopropyltriethoxysilane (APTES), and all other chemicals were purchased from Shanghai First Reagent Co., China.

2.2. Preparation of GOMS

GO was synthesized from natural graphite powder via a modified Hummers Method. (Wang and Chen 2015) A commercially available melamine sponge (MS) was cut into $2.0 \times 2.0 \times 1.0 \text{ cm}^3$ cubes and cleaned by DI water and ethanol for three times, followed by drying at 60 °C for 24 h. 25 mg GO was dispersed in 50 mL DI water under ultrasonic shock for 2 h. Then, the melamine sponge was dipped into GO solution for 1.5 h and dried in a vacuum oven at 60 °C to obtain graphene oxide-melamine sponges (GOMS).

2.3. Synthesis of K-MS and K-GOMS

Kaolin was dried at 120 °C for 5 h to remove the water and treated with stearic acid firstly.(Qu et al. 2016) Briefly, stearic acid (0.5 g) was added into 24 mL of ethanol under magnetic stirring. While dissolved completely, kaolin (10 g) was added to the solution and ultrasonically dispersed for 30 min. Then, the solution was transferred into a preheated water bath (95 °C), refluxed for 2 h and finally dried at 120 °C for another 6 h to obtain hydrophobic kaolinite. Afterwards, 100 mg hydrophobic kaolinite was first dispersed in 100 mL of ethanol by probe sonication for 2 h. Then, 2 mL of APTES was added into the solution under stirring for 10 min. After that, GOMS and MS were impregnated with the solution for 1 h. Then, the sponges were raised and washed with ethanol for three times, and dried in a vacuum oven at 60 °C to obtain K-MS and K-GOMS.

2.4. Characterization

The sponge materials were characterized by attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR, NICOLET 5700 FT-IR Spectrometer). The elemental composition of the surface of the sponge materials was determined by X-ray photoelectron spectroscopy (XPS), performed on ESCALAB 250Xi (Thermo Fisher Scientific, USA). The surface morphology was characterized using a JSM-6700F Field Emission Scanning Electron Microscope (FE-SEM) (JEOL, Japan). X-ray analysis was done by the powder X-ray diffractometer (D8 ADVANCE, Bruker Axs). Water contact angle measurements were conducted by an OCA 15Pro goniometer (Dataphysics). Raman spectra were recorded from 200 to 4000 cm⁻¹ on Raman Microprobe (Labram-010).

2.5. Adsorption experiments of K-MS and K-GOMS

The K-MS and K-GOMS were dipped into various organic solvents (*N*, *N*-dimethyl formamide (DMF), ethanol, methanol, and n-hexane) and oils (diesel oil, kerosene, motor oil, and liquid paraffin) for 1 min. The adsorption capacity Q_e (g/g) was calculated according to Eq. (1).

$$Qe(g/g) = (m1 - m0)/m0$$
 (1)

where m_0 and m_1 are the weight of the sponge before and after adsorption, respectively.

The recyclability of sponge was evaluated by repeated adsorption-squeezing processes. The adsorption-squeezing was performed by immersing the sponge into DMF and diesel oil, until the



Scheme 1. Schematic illustration of the fabrication procedure and mechanism of K-GOMS.

sponge became saturation, then manually squeezing the sponge to extract absorbed oils, repeating 30 times to test the recyclable utilization performance of K-MS and K-GOMS.

3. Results and discussions

3.1. Synthesis of superhydrophobic K-MS and K-GOMS

The mechanism and procedure for the fabrication of K-GOMS are illustrated in Scheme 1. The melamine sponge can act as a template due to its 3D porous structure, high mechanical, chemical and environmental stability. The white sponge was immersed in GO dispersion and then dried in an oven to produce the yellow monolith GOMS. The GO sheets were adhered to the skeleton of the sponge through the interaction between NH- on melamine sponge and -COOH or -OH on GO. (Liu et al. 2015) Before using, kaolinite was treated with stearic acid to remove the hydroxyl of the kaolinite surface via the esterification reaction, which made the kaolinite surrounded with abundant hydrophobic groups of -CH2- and -CH3, decreased the surface energy of kaolinite, and then formed the hydrophobic surface. (Qu et al. 2016) In order to fabricate the K-GOMS, APTES was used as a linkage to bond kaolinite to GO sheets. APTES could be immobilized on kaolinite through Si-O bond, and its terminal -NH2 group could react with the -COOH and -OH of the edge of GO sheets. Besides, the terminal -NH2 group of APTES could interact with the residual -COOH and -OH through hydrogen bonds.(Zhang et al. 2015a) Therefore, the kaolinite could be successfully adhered on GO sheets. Specifically, it is insufficient to form a superhydrophobic sponge surface through roughing the sponge surface with GO sheets, kaolinite is the key role to endow sponge a superhydrophobic surface. Kaolinite coated on GOMS not only react with GO to remove the oxygen-containing functional groups on GO, which decrease the hydrophilicity of GO, but also could significantly decrease the surface energy of modified sponge. To evaluate the role of the GO sheets in this work, as a comparison, the melamine sponge was directly immersed into hydrophobic kaolinite solution to obtain K-MS, and APTES was used as a linkage as well. Both K-GOMS and K-MS were used to investigate their adsorption performance.

3.2. Characterization of materials

3.2.1. Characterization of kaolin and GO

Kaolinite, one of the well-known low cost clay mineral materials, has been widely used as an economical adsorbent. The chemical composition of kaolinite was (by mass %) $Al_2O_3:39.5\%$, $SiO_2: 46.5\%$ and $H_2O: 14\%$. The surface morphology of the kaolin was observed by SEM (Fig. 1a), it showed that the kaolin has flaky shaped particles and the particle size was inhomogeneous.(He et al. 2018) Fig. 1b showed the



Fig. 1. SEM images of kaolin (a), and GO (b).



Fig. 2. XRD patterns of kaolin (a), and GO (b).

morphology of GO, it has a thin layer and full of wrinkles. The X-ray diffraction of kaolin indicated that the major constituent of kaolin was kaolinite (PDF 2004 14-0164), as shown in Fig. 2a. Fig. 2b showed a typical sharp peak at 10.9°, which was a characteristic peak of GO.

3.2.2. Characteristic of sponge materials

To better understand the formation of hydrophobic kaolinite and GO on sponge coating, the four sponge materials, including MS, GOMS, K-MS, and K-GOMS, were characterized by ATR-FTIR, XPS and Raman. Fig. 3a showed the ATR-FTIR spectra of the sponges. The spectrum of MS showed prominent peaks at 810, 1147, 1540 and 3394 cm⁻¹, which attributed to triazine ring bending, C–O stretching, C=N stretching, and the stretching vibrations of secondary amine (N–H) on the surface of MS, respectively.(Pham and Dickerson 2014) The peaks centered at 974, 1327, and 1471 cm⁻¹ were assigned to C–H bending.(Merline et al. 2013) Moreover, two small peaks at 2814 and 2887 cm⁻¹ were indicative of C–H stretching.(Arkles 1977) In contrast, for K-MS and K-GOMS, the intensity of peak centered at 1004 cm⁻¹ strengthened significantly with slightly red-shift due to the overlapping of the C–H bending peak (974 cm⁻¹) and a new peak at 1076 cm⁻¹ that was attributed to Si-O-Si stretching of kaolinite.(Zhang et al. 2017)

The XPS was applied to further confirm the kaolinite modification of the MS and GOMS. The XPS survey spectra in Fig. 3b showed four elements, including C, N, O, and Si. For MS, three peaks centered at



Fig. 3. ATR-FTIR (a), XPS (b) and Raman (c) spectra of the various materials.

284.97, 398.97, and 531.97 eV were attributed to the bonding energies of C1s, N1 s, and O1s, respectively, which are chemical components in melamine. After GO sheets coating, the intensities of C1s and O1s increased significantly indicating the functionalization of GO on the melamine sponge. When kaolinite was attached to the MS and GOMS, a new element Si at 101.97 eV (Si2p) and 152.97 eV (Si2s) emerged, (Chen et al. 2016a) which confirmed that the MS and GOMS skeletons

Table 1

Element composition, and atom ratios of adsorbents.

Samples	Atom percentage (%)			
	C1s	N1s	01s	Si2p
MS	62.649	22.082	15.269	-
GOMS	66.455	11.334	22.211	-
K-MS	53.458	11.722	25.13	9.691
K-GOMS	56.73	10.797	21.982	10.491

were successfully modified with kaolinite. Furthermore, the element contents of MS and modified sponge based on XPS analysis are listed in Table 1. After GO sheets coating, the content of C and O increased to 66.455% and 22.211%, respectively. When kaolinite was introduced, the content of Si increased to 9.691% and 10.491%, respectively. These results indicated that the sponge was successfully modified with GO and kaolinite.

The Raman spectrum (Fig. 3c) of the untreated melamine sponge presented two intense peaks at 977 and 2970 cm⁻¹, which were dedicated to the three-dimensional hierarchical porous structure of melamine skeleton. The peak at 977 cm^{-1} was ascribed to the breathing modes of the triazine ring of melamine.(Scheepers et al. 1995) Two broad and weak bands at $600-800 \text{ cm}^{-1}$ and $1300-1600 \text{ cm}^{-1}$ were the characteristic of melamine sponge.(Ji et al. 2017; Wang et al. 2008) After GO coating, the 2970 cm⁻¹ peak vanished and two new peaks were assigned to the carbon D-band (1330 cm^{-1}) and G-band (1600 cm⁻¹), which showed that part of skeletal pores of GOMS and K-GOMS were covered by GO sheets. Compared with GOMS, K-GOMS presented significantly strengthened D-band and G-band, and a new peak at 980 cm⁻¹ was indicative of the overlapping of the characteristic peak of kaolinite (944 cm⁻¹) and the Al-O-H vibration peak $(986 \text{ cm}^{-1}), (Qiao 2003)$ indicating the modification of kaolinite on the surface of GOMS. Compared with MS, the intensity of peak at 977 cm^{-1} increased slightly in K-MS, due to the overlapping of the melamine sponge skeleton and Al-O-H vibration, which indicated that the modification of kaolinite on the skeletons of sponge.

3.3. Morphology and superhydrophobicity of the sponge materials

Morphology of the four melamine sponge materials before and after



Fig. 4. SEM images of MS (a), GOMS (b), K-MS (c), and K-GOMS (d).



Fig. 5. (a) Photograph of various sponges after being placed on water. (b) Photograph of K-GOMS immersed in water by force. (c) Water contact angle of K-MS. (d) Water contact angle of K-GOMS.

modification was examined by SEM as shown in Fig. 4. The pristine MS is composed of three-dimensional porous network structure (Fig. 4a). After GO sheets coating, part of the pores of MS were overspread by GO sheets (Fig. 4b). The surface morphology of K-GOMS was presented in Fig. 4d, large amounts of interlaced-wrinkles were found, and the kaolinite was successfully adhered tightly on the GO sheets surface, which could significantly enhance the roughness of the sponge skeletons. As a comparison, the K-MS was observed in Fig. 4c. It could be observed clearly that the kaolinite was successfully attached to the skeletons of the sponge and conspicuously improved the roughness of sponge skeletons.

The hydrophobicity of the four melamine sponge materials was also tested. As shown in Fig. 5a, the K-MS and K-GOMS floated on the water surface while MS and GOMS sank to the bottom, indicating that just GO sheets coated on the sponge surface would not transform the hydrophilicity of sponge surface to hydrophobicity. However, the sponge switched from hydrophilic surface to hydrophobic surface when kaolinite was introduced. It confirmed that GO sheets enhanced the roughness of the melamine sponge surface, and the kaolinite endowed melamine sponge a superhydrophobic surface. In Fig. 5b, while K-GOMS was immersed in water by an external force, silver mirror-like surface appeared due to the Cassie-Baxter nonwetting behavior.(And et al. 2008; Zhou et al. 2013; Zhu et al. 2010) The two facts indicated that the K-GOMS displayed hydrophobic surface. To further demonstrate the hydrophobicity of K-GOMS and K-MS, water contact angle measurement was carried out. As shown in Fig. 5c and d, the water contact angles of K-MS and K-GOMS reached 156.5° and 152.3°, respectively, which indicated the superhydrophobic characteristics of the functionalized melamine sponges. The water contact angle of K-GOMS was lower than that of K-MS due to the residual oxygen-containing functional groups on the GO sheets which have not been completely occupied by the hydrophobic kaolinite. All results indicated that the asprepared sponges possessed excellent superhydrophobicity and could serve as promising candidate adsorbent for oil-water separation.



Fig. 6. Adsorption of diesel oil (dyed with Sudan III) from water with K-MS (a) and K-GOMS (b).

3.4. Application of K-MS and K-GOMS as adsorbents

The three-dimensional hierarchical porous structure and superhydrophobic surface are crucial properties of adsorption material, which make the material an ideal adsorbent for removal of various organic solvents and oils from water. As shown in Fig. 6, when K-MS and K-GOMS contacted with the diesel oil (dyed with Sudan III) on the water, the diesel oil was quickly absorbed into sponges within a few seconds, and the absorbed oil could be easily gathered by the squeezing process.

To further investigate the adsorption performance, the rate of adsorption was also tested. As seen in Fig. 7a, the adsorption capacity of K-GOMS was higher than that of K-MS, and both sponges achieved saturation rapidly within 10s. Furthermore, adsorption experiments were conducted in various types of organic solvents and oils without water. Some common oils were selected, such as diesel oil, motor oil, liquid paraffin, and kerosene, which are usual pollutants in aqueous environment. The other kinds of organic solvents, such as DMF, ethanol, methanol, and n-hexane, were also tested. Although both K-MS and K-GOMS showed excellent adsorption capacity, K-GOMS exhibited higher adsorption capacity than K-MS for all organic liquids mentioned above as seen in Fig. 7b. The results demonstrated that GO sheets could significantly improve the adsorption capacity of the sponge. Importantly, the K-MS and K-GOMS showed much higher adsorption ability than other reported adsorption materials (Table 2). For example, magnetic exfoliated graphite exhibited adsorption ability for diesel oil of 33 g/g and engine oil of 49 g/g, (Wang et al. 2010) silanized graphene PU sponge showed adsorption ability for diesel oil and n-hexane of 26 g/g,(Zhou et al. 2016) and polymethylsilsesquioxane coated melamine sponge absorbed 78 g/g for diesel oil and 70 g/g for gasoline. (Zhang et al. 2017) Furthermore, the adsorption capacities of the K-MS and K-GOMS are also compared with other adsorbent materials with



Fig. 7. (a) Instant adsorption capacity of K-MS and K-GOMS for DMF. (b) Adsorption capacity of K-MS and K-GOMS for various organic solvents and oils.

high adsorption ability. For example, reduced graphene oxide (rGO) melamine sponge adsorption capacity for diesel oil of 99 g/g,(Liu et al. 2015) and graphene coated melamine sponge exhibited high adsorption capacity of pump oil (100 g/g) and diesel oil (108 g/g).(Zhao et al. 2016) However, the facile fabrication process and cheap raw material of K-MS and K-GOMS enable the sponge a potential and efficient adsorbent material for various organic solvents and oils removal from aqueous environment.

Reusability is an important indicator to evaluate the performance of the adsorbent materials. In this study, DMF and diesel oil were selected on behalf of organic solvents and oils respectively, to test the recycling capacity of K-MS and K-GOMS by adsorption/squeezing process. Fig. 8 revealed the adsorption capacity of the superhydrophobic K-MS and K-GOMS after 30 consecutive cycles for DMF (Fig. 8a) and diesel oil (Fig. 8b). The K-MS remained 90% adsorption capacity of DMF and 92% of diesel oil, and K-GOMS remained 96% of DMF and 94% of diesel oil. It clearly showed that adsorption capacity of both sponges were no apparent decrease over 30 cycles, and K-GOMS exhibited higher stability and recycling performance than that of K-MS.

Furthermore, the stability of the superhydrophobic K-GOMS was investigated in a wide range of pH values (pH: 2–12) and different temperatures (-20-90 °C). The stability of sponge was examined by adsorption capacity, which normalized by the initial weight gain. To test the impact of pH on sponge, the K-GOMS was immersed sequentially 12 h into each aqueous solution of pH 2–12. The results clearly showed that the sponge remained higher than 95% adsorption capacity, as shown in Fig. 9a. The influence of temperature on the adsorption of K-GOMS was also tested. The organic solvent was cooled or heated at

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a

Table 2

Comparison of various sponge adsorbent materials.

Adsorbent materials	Absorbed substance	Adsorption capacity (g/g)	Ref.
Magnetic exfoliated graphite	Diesel oil	33	Wang et al. (2010)
	Engine oil	49	
Silanized graphene PU sponge	Diesel oil	26	Zhou et al. (2016)
	n-hexane	26	
Copper film PU sponge	Decane	13	Zhu et al. (2011)
	Labricating oil	20	
TTEAT-rGO PU sponge	Pump oil	30	Tjandra et al. (2015)
	Gasoline	50	
rGO melamine sponge	Diesel oil	99	Liu et al. (2015)
Graphene coated melamine sponge	Pump oil	100	Zhao et al. (2016)
	Diesel oil	108	
Polymethylsilsesquioxane coated melamine sponge	Diesel oil	78	Zhang et al. (2017)
	Gasoline	70	
Graphene based-foam	DMF	75	Ji et al. (2017)
	Ethanol	60	
K-MS	DMF	89	Present work
	Diesel oil	76	
K-GOMS	DMF	113	Present work
	Diesel oil	96	

110

100





Fig. 9. Effect of pH values (a) and temperature (b) on the adsorption capacity of K-GOMS for DMF.

different temperatures for 30 min, then absorbed by K-GOMS. Interestingly, as shown in Fig. 9b, with the increase temperature of organic liquid, the adsorption capacity decreased, indicating that the adsorption process was exothermic in nature. Moreover, it is noteworthy that

Fig. 8. Adsorption recyclability of K-MS and K-GOMS for DMF (a) and diesel oil (b).

Cycles

the adsorption of organic liquid by K-GOMS can sustain during a wide temperatures ranging from -20 °C to 90 °C. All these results indicated that the adhesion within K-GOMS was quite strong and tight, which confirmed that the sponge could be suitable for water treatment and oil cleanup at a wide range of pH values and temperatures.

4. Conclusion

In summary, a superhydrophobic, flexible, and porous K-GOMS was fabricated through a facile and cost-effective process. The GO sheets and kaolinite were coated on the melamine sponge surface, forming a rough and superhydrophobic surface, as well as maintaining the initial flexible and hierarchical porous structure of melamine sponge. The results showed that the fabricated K-GOMS possessed many crucial properties, such as high adsorption capacity, superhydrophobic surface, outstanding recyclability, and excellent environmental tolerance. All of these remarkable performances enable K-GOMS great potential in the removal of organic solvents and oils from water and a promising candidate for practical treatment of oil spillage and chemical leakage.

Compliance with ethical standards

Conflict of interest

The authors declare that they have no conflict of interest.

Ethical approval

This article does not contain any studies with human participants or animals performed by any of the authors.

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