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Facile and Green Synthesis of Melamine-Formaldehyde@rGO Foam with Enhanced Superhydrophobicity for Oil Removal Application

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Abstract

This research presented a more effective method to synthesize superhydrophobic melamine-formaldehyde (MF) foam coated with reduced graphene oxide (MF@rGO) by a conventional heating process. This is a facile and green method based on the use of graphene oxide, GO, as a precursor, and vitamin C as a reductant. The effect of recoating GO onto MF@rGO to form MF@rGO-recoat foam on superhydrophobicity was also investigated. Then, pristine GO, rGO, MF and as-synthesized MF@rGO foams were analyzed and their structures confirmed by several techniques including FTIR, Raman, SEM, TEM, and water contact angle (WCA). The results indicated that the WCA values of MF@rGO and MF@rGO-recoat foams were increased from 144.1° to 156.9°, respectively. Furthermore, the adsorption capacity (Q_e), oil removal performance, and recyclability of MF@rGO-recoat foam were investigated. The adsorption capacity of MF@rGO-recoat foam was higher than 103.8 $g.g^{-1}$ for all the oils tested (palm oil, gasoline, diesel and lubricant oil), and the highest value was 115.9 $g.g^{-1}$ for lubricant oil. Besides, MF@rGO-recoat foam can be easily recycled up to 10 times for removal of all oils tested. Thus, this work provides a new alternative eco-friendly way to synthesize MF foam coated with rGO sheets and the synthesized MF@rGO foam can be applied and reused for the removal of oil spillages from water or wastewater.

Keywords: superhydrophobicity; reduced graphene oxide (rGO); melamine-formaldehyde (MF) foam; adsorbent; vitamin C; oil removal; water pollution
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1. Introduction

With the rapid development and growth of industry, the spillage of oils, petroleum, and organic solvents into water system has caused serious environmental and ecological damages [1, 2]. This has led to incentives to research and develop new techniques in the challenging area of oil-water

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separation. For this reason, new varieties of adsorbents or adsorber materials that can more effectively adsorb, remove, and transfer spilled oils or toxic organic solvent contaminants from water are urgently desired to help preserve and treat the water pollution [3, 4].

In the past few years, several commercially available polymer sponges or foams, for example, polyurethane (PU), polyethylene (PE), polypropylene (PP), melamine-formaldehyde (MF), and melamine, have been widely used to separate and cleanup oil spills from water because of their low operational costs, lightweight, good flexibility, easy to use and ready availability [2, 4, 5]. However, certain properties of these polymer foams, including their low oil adsorption capacity, poor selectivity, and low chemical stability in organic solvents have limited their applicability [5, 6]. In order to overcome these problems, the modifications of the commercial polymer foams are needed [7, 8].

As a single-atom sheet of two-dimensional (2D) carbon nanomaterial, graphene derivative namely reduced graphene oxide (rGO) has attracted much attention owing to its unique excellent hydrophobicity, thermal and chemical stability, very large surface area, and good flexibility [9, 10]. Recently, rGO sponges and its coating onto polymer foams have been used commonly for removing spilled oils and organic solvents from water. Usually, rGO can be produced from graphene oxide (GO) precursor by both chemical and thermal reduction processes. Unfortunately, the former process has some drawbacks in that it frequently involves the use of several toxic reductants, for instance, hydrazine (N_2H_4), hydrogen sulphide (H_2S), sodium borohydride ($NaBH_4$) and boron hydride (B_2H_6) [11, 12].

Recently, we have reported a simple, green, and effective method for the preparation of MF foam coated with rGO to form MF/rGO foams by using different types of GO as precursors and vitamin C as a reductant [13]. In this research, vitamin C was used as a reducing agent as it is natural reducing agent, environmentally friendly, non-toxicity, inexpensive and readily available. However, all the obtained MF/rGO foams had low oil adsorption ability because of their hydrophobic properties, and had water contact angle below 150°. Although the effect of high content of GO for the preparation of MF/GO was studied, a decrease in the water contact angle was observed [13].

Therefore, to further improve the superhydrophobic property of MF foam coated with rGO, this work aimed to investigate effect of GO precursors coated and recoated onto MF foam to form MF@rGO and MF@rGO-recoat foams using vitamin C as a reducing agent. To the best of our knowledge, moreover, none of the published research had reported on MF foam recoated with rGO by a facile, green, and effective method involving a direct mixing of MF@rGO foam, GO precursor, and vitamin C reductant. In this study, we found that vitamin C not only functioned as a reductant, but it also acted as a surface modifier of MF foam, enhancing the strong interaction between GO sheet precursor and the skeleton surface of MF foam. The results indicated that the as-synthesized MF@rGO-recoat foam in this present work shows a superhydrophobic property which is higher than water contact angle (as 156.9°) of 150°. Besides, the adsorption capacity (Q_e), oil removal performance, and recyclability of MF@rGO-recoat foam were also studied and reported.

2. Materials and Methods

2.1 Chemicals and materials

Melamine-formaldehyde (MF) foam was supplied from Scotch Brite, 3M Co., Ltd, Thailand. Graphite powder (99% purity), hydrogen peroxide (H_2O_2, 30%) and vitamin C, (vit C, $C_6H_8O_6$, 99%) were purchased from Sigma-Aldrich (USA). Potassium permanganate ($KMnO_4$) and sulfuric acid (H_2SO_4, 98%) were obtained from Merck (Germany). Ethanol (C_2H_5OH, 95%) was obtained from Baker (USA). All chemicals were used as received without any further purification.

2.2 Synthesis of melamine-formaldehyde/rGO (MF@rGO) foams

2.2.1 Synthesis of GO precursor and rGO samples

First, graphite oxide was synthesized through a modified Hummers method [14]. Then, the synthesized graphite oxide powder was added in DI water under sonication for 90 min, followed by centrifuging. Finally, the aqueous suspension of GO precursor (at the concentration of 5 mg/ml) was obtained. Meanwhile, rGO sample was easily synthesized by a conventional heating method at 90°C for 30 min using GO suspension and vitamin C solution, as precursor and reductant, respectively [15].

2.2.2 Synthesis of MF@rGO foams

İn this work, a commercial MF foam sheet was used, and then cut into (2.0 cm × 2.0 cm × 1.0 cm) cubes. Afterward, the MF foam pieces were cleaned and treated with distilled water, ethanol, and then dried at 60°C in an oven for 30 min. The MF foam coated with rGO (MF@rGO) was synthesized by a method based on Jirasuttisarn and Sriwong [13], using a conventional heating process. In brief, the MF foam was firstly immersed into 50 ml of distilled water and followed by adding a few drops of vitamin C solution to adjust the pH of distilled water to 3. Then, 10 mg of GO precursor was added and stirred for 5 min, followed by heating at 90°C for 30 min under stirring. Subsequently, the MF@rGO foam product was taken out and then washed with distilled water and ethanol several times. After drying in an oven at 60°C for 3 h, the MF@rGO-10 foam was obtained. Meanwhile, the recoated MF@rGO foam was also synthesized as described above, but using MF@rGO-10 foam as a starting foam with a new GO precursor at the content of 10 mg. The obtained product was named as MF@rGO-recoat foam.

2.3 Characterizations

The functional groups of GO, rGO, MF and all MF@rGO foam samples were analyzed via Fourier-transformed infrared spectroscopy (FT-IR) (Nicolet iS50, Thermo Fisher Scientific Inc., USA). The chemical structures of the samples were also well-confirmed by Raman spectroscopy technique (DXR Smart, Thermo Fisher Scientific Inc., USA). The microstructures of the samples were investigated using scanning electron microscopy (SEM), (JEOL-JSM5800LV, Japan) and transmission electron microscopy (TEM), (JEJOL-JM-2010, Japan). Water contact angle (WCA) measurements of all MF@rGO foams were performed through a contact angle analyzer using a water droplet of volume 5 ul.

2.4 Measurements of oils adsorption capacity

The MF@rGO-recoat foam was dipped into various oils, such as, gasoline, diesel, palm oil, and lubricant oil for 3 min. The adsorption capacity, Q_e (g/g) was calculated by Equation (1),

$$Q_e = (m_t - m_i)/m_i \qquad (1)$$

where m_i and m_t are the weights (g) of the foam sample before and after adsorption, respectively [10, 16]. The average adsorption capacity of MF@rGO-recoat foam for rmoval of various oils was obtained by using 3 pieces of each foam sample per test.

2.5 Recyclability

The recyclability of the MF@rGO-recoat foam was measured by repeated adsorption-squeezing methods without any washing and drying of the foam sample. This method was carried out by immersing pieces of MF@rGO-recoat foam into palm oil, gasoline, diesel or lubricant oil until the foam became saturated, and then manually squeezing the foam to seperate out adsorbed oils, a process that was repeated 10 times in order to test the recyclable performance of the MF@rGO-recoat foam.

3. Results and Discussion

3.1 Synthesis of MF@rGO foams

The photographs of pristine MF, MF@rGO-10 and MF@rGO-recoat foam samples are shown in Figure 1. It can be clearly seen that the surface of the pristine MF foam was white in color [as can be seen in Figure 1(a)], whereas both MF@rGO foams were of black color [Figures 1(b) and (c)]. This result confirmed that the surfaces of both MF@rGO foams were coated or well-covered with rGO sheet particles [10]. As a result, vitamin C not only functioned as a reducing agent, but it also acted as a surface modifier of the MF foam with the positive charges of H^+ ions on the surface of the MF foam. Therefore, the negative charges on the skeleton of GO sheets could easily adhere to the skeleton of MF surface through strong electrostatic interaction [17]. Moreover, vitamin C molecules can also react with GO molecules via good hydrogen bonding and π-π stacking, resulting in better interaction of GO precursor and vitamin C reductant, which enhances the reduction of GO to rGO. After the reduction, rGO nanosheets were easily produced from GO precursor, and still maintained cover and impregnation on the surfaces of all as-synthesized MF@rGO foams.

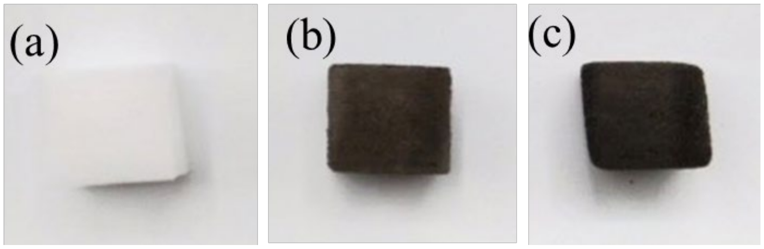

Figure 1. Photographs of (a) pristine MF, (b) MF@rGO-10 and (c) MF@rGO-recoat foams

3.2 Analysis of GO precursor and rGO

To confirm the characteristics of GO precursor and as-synthesized rGO samples, Raman, FTIR, SEM and TEM techniques were carried out, all the results of which are displayed in Figure 2. It can be observed that the Raman peaks of GO precursor and rGO are exhibited as two broad peaks at around 1336 cm^{-1} and 1586 cm^{-1} [see in Figure 2(a)], which are related to disordered carbon (D band) and graphitic sp^2 carbon (G band), respectively [6, 18]. Compared to the GO, the I_D/I_G intensity ratio of rGO was increased, which can be attributed to the transformation of GO to rGO after the reduction method using vitamin C [8, 12]. This is confirmed by the color change of GO (brown) and rGO (black) suspensions, as shown in inset of Figure 2(a) [10, 12]. Meanwhile, the FTIR spectra of GO precursor and rGO samples are also shown in Figure 2(b). After the reaction of

GO precursor, the absorption bands of some oxygen-functional groups, for instance, hydroxyl (-O-H), carbonyl (-C=O) and epoxy (C-O-C) were mostly removed [6, 19]. This result indicated that the GO precursor was transformed into rGO by the chemical reduction method using vitamin C as reductant. Figure 2(c) displays the SEM image of the rGO sample. It can be seen that the morphology of rGO particles was flake-like structure of very small sizes and high aggregation. This result agrees with the TEM image in Figure 2(d), in which it can be seen that rGO particles had formed very thin sheets [13, 17].

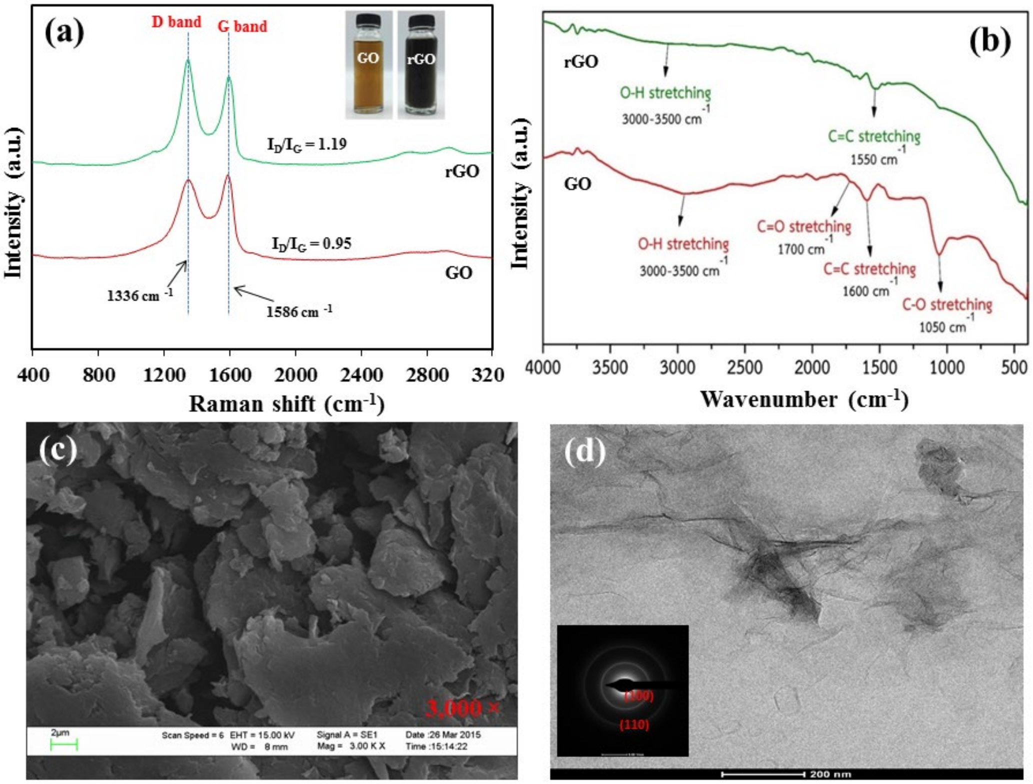

Figure 2. (a) Raman, (b) FTIR spectra of GO precursor and synthesized rGO samples; and (c) SEM and (d) TEM images of rGO sheets

3.3 SEM characterization of MF@rGO foams

Figure 3 displays the surface morphology of the pristine MF, as-synthesized MF@rGO-10 and MF@rGO-recoat foams. It can be obviously seen that the skeleton template of pristine MF foam had a very smooth surface (see in Figure 3(a), whereas both the MF@rGO foams showed rough surfaces being coated and covered with rGO sheet particles on the skeletons of the MF@rGO foams [see in Figure 3(b) – 3(c)] [13]. Compared to the MF@rGO-10 foam, the surface roughness of the MF@rGO-recoat foam has more covering of rGO nanosheets on the skeleton of the MF surface. This result indicates the recoating of MF@rGO foam can enhance the impregnation and cover of rGO particles on the foam surface.

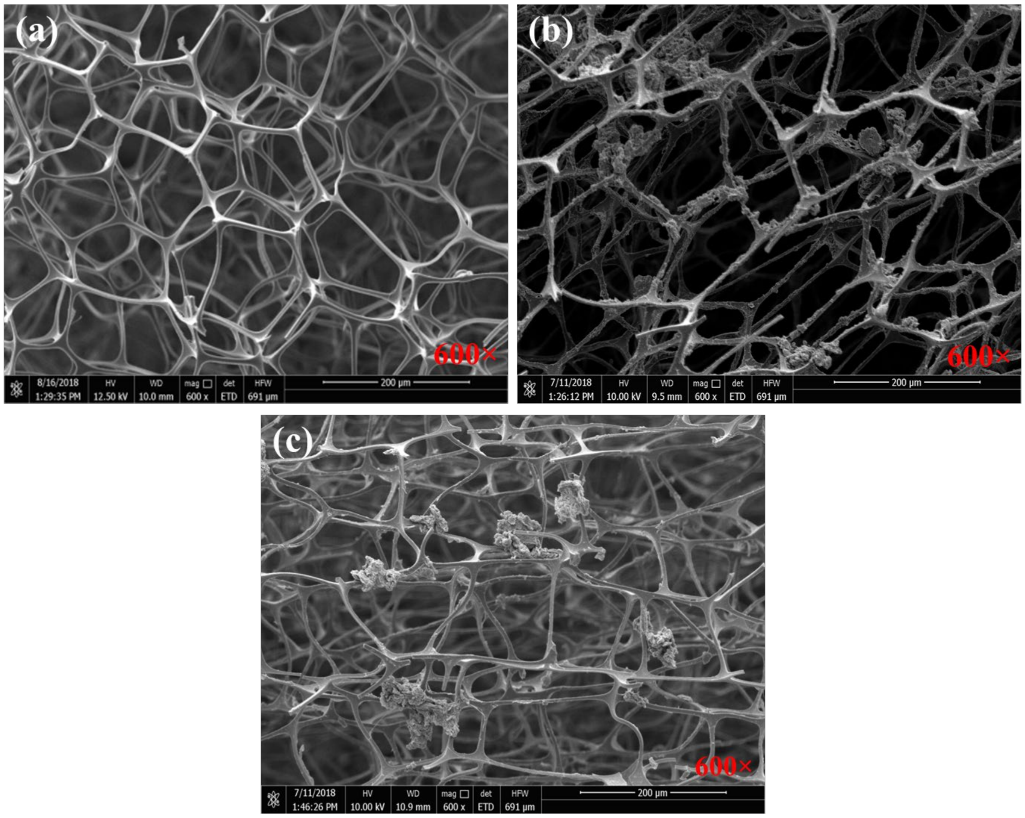

Figure 3. SEM images of (a) pristine MF, (b) MF@rGO-10 and (c) MF@rGO-recoat foams

3.4 FTIR analysis of pristine MF and MF@rGO foams

Figure 4 illustrates the FTIR spectra of pure MF and all MF/rGO foams. For the pristine MF foam, the typical absorption bands that appeared at around 3340 cm^{-1}, 2910-2848 cm^{-1}, 1556 cm^{-1}, 1450 cm^{-1}, 1320 cm^{-1}, 1018 cm^{-1} and 800 cm^{-1} were assigned to the functional groups of MF foam structure, such as, -N-H or -O-H stretching, -C-H stretching of -CH_2, the aromatic ring stretching of -C-N and -C=N groups, and triazine ring bending vibrations of MF [5, 20]. Compared to the MF foam, all the absorption bands of the MF@rGO foams can be observed but it needs to be remarked that the intensities of all characteristic bands of MF in MF@rGO foams were slightly decreased and broadened upon its coating and recoating. This result well confirmed the increase of rGO nanosheet particles that had interacted with, covered, and impregnated the surface of MF@rGO foams [13, 18, 21].

3.5 Raman analysis of MF@rGO foams

Figure 5 displays the Raman spectra of as-synthesized MF@rGO-10 and MF@rGO-recoat foam samples. In the Figure 5, two obvious peaks at around 1336 cm^{-1} (D band) and 1588 cm^{-1} (G band) of rGO can be seen with all the MF@rGO foams. The characteristic absorption peaks of rGO were more clearly observed and slightly increased along with the recoating of the foam sample. However,

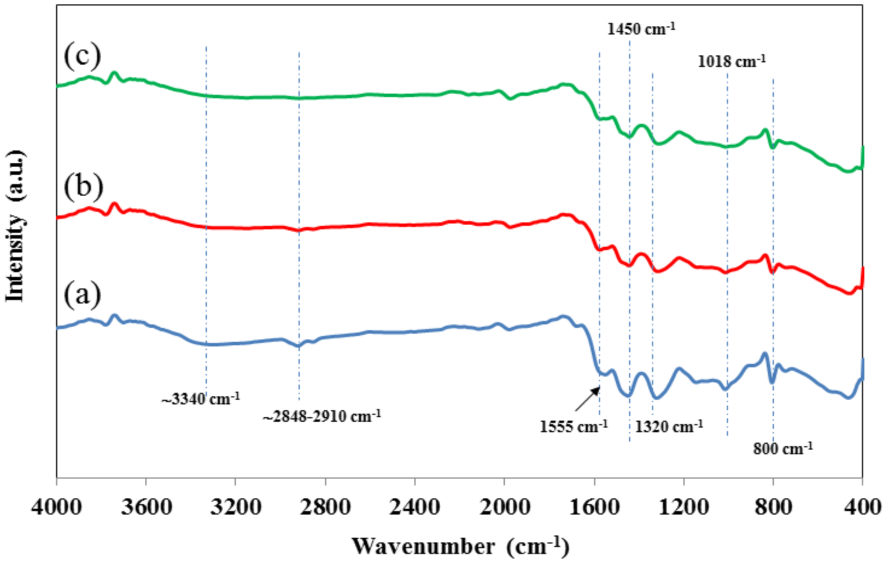

Figure 4. FTIR spectra of (a) pristine MF, (b) MF@rGO-10 and (c) MF@rGO-recoat foams

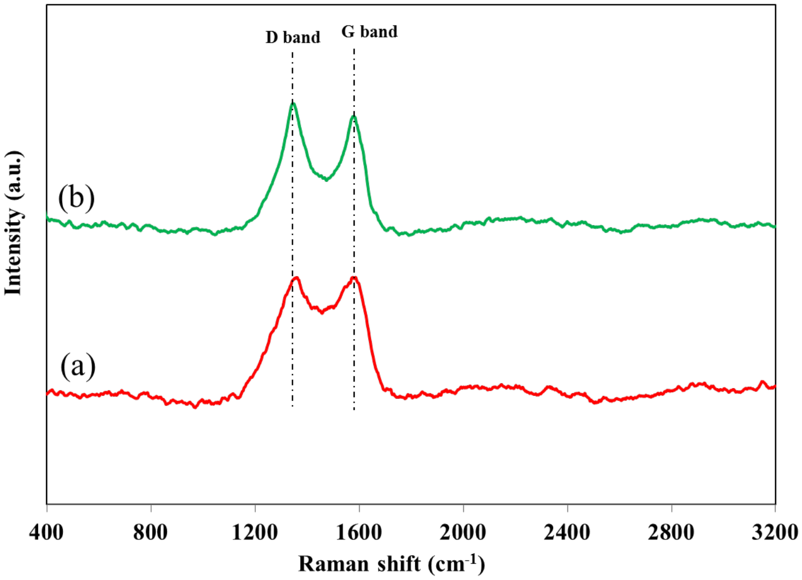

Figure 5. Raman spectra of (a) MF@rGO-10 and (b) MF@rGO-recoat foams

the typical peaks, such as a triazine ring breathing mode (at around 980-990 cm^{-1}) and C–H stretching (at around 2960 cm^{-1}) vibrations of MF structure [22] were not observed in both MF@rGO foams. This result further confirmed the rGO sheet particles had become well impregnated into and better covered the surface of the MF@rGO-recoat foam.

3.6 Water contact angle of MF@rGO foams

To understand the hydrophobic properties of all foam samples, the water contact angle (WCA) measurements were investigated, and the results are presented in Figure 6. It was found that the WCA values of the MF@rGO-10 and MF@rGO-recoat foams had increased from 144.1° to 156.9°, respectively. The WCA of MF@rGO-recoat foam had a WCA value of 156.9° which was higher than 150°, indicating that this foam was superhydrophobic as well as having excellent ability for oil adsorption. These results were directly related to the higher content of rGO sheets which existed, covered, and were coated on the surface of the MF@rGO foams. These results can be related to the data from SEM and Raman techniques. As a result, it can be concluded that the higher water contact angle of the MF@rGO-recoat foam coincided with its superhydrophobic behavior, resulting in its superior oil adsorption ability [23].

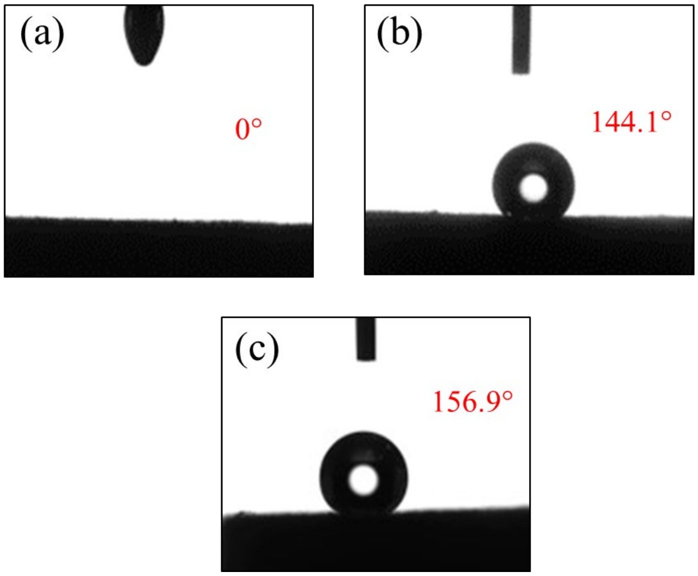

Figure 6. Water contact angle (WCA) of (a) pristine MF, (b) MF@rGO-10 and (c) MF@rGO-recoat foams

3.7 Oil removal ability of MF/rGO-recoat foam

To understand the oil removal ability, oil-water separation from water was tested using the as-synthesized MF@rGO-recoat foam. The hydrophobicity of the foam was measured by placing palm oil and water droplets on the MF@rGO-recoat foam surface, as shown in Figure 7(a). It was found that water droplets of spherical shape were still maintained on the surface of the synthesized MF@rGO foam; in contrast, the oil droplets were immediately adsorbed into the foam and disappeared. Furthermore, the removal performance of the MF@rGO-recoat for palm oil from water was also studied. As can be seen in Figures 7(b) and 7(c), when the MF@rGO-recoat foam contacted with the oil and chloroform in the water, both the palm oil and chloroform were quickly adsorbed into the foam sample within a few seconds [10, 12, 24], and moreover the adsorbed oil could be easily released by the squeezing process. These results pointed out that the synthesized MF@rGO-recoat foam had excellent ability to adsorb oil or non-polar solvents from water.

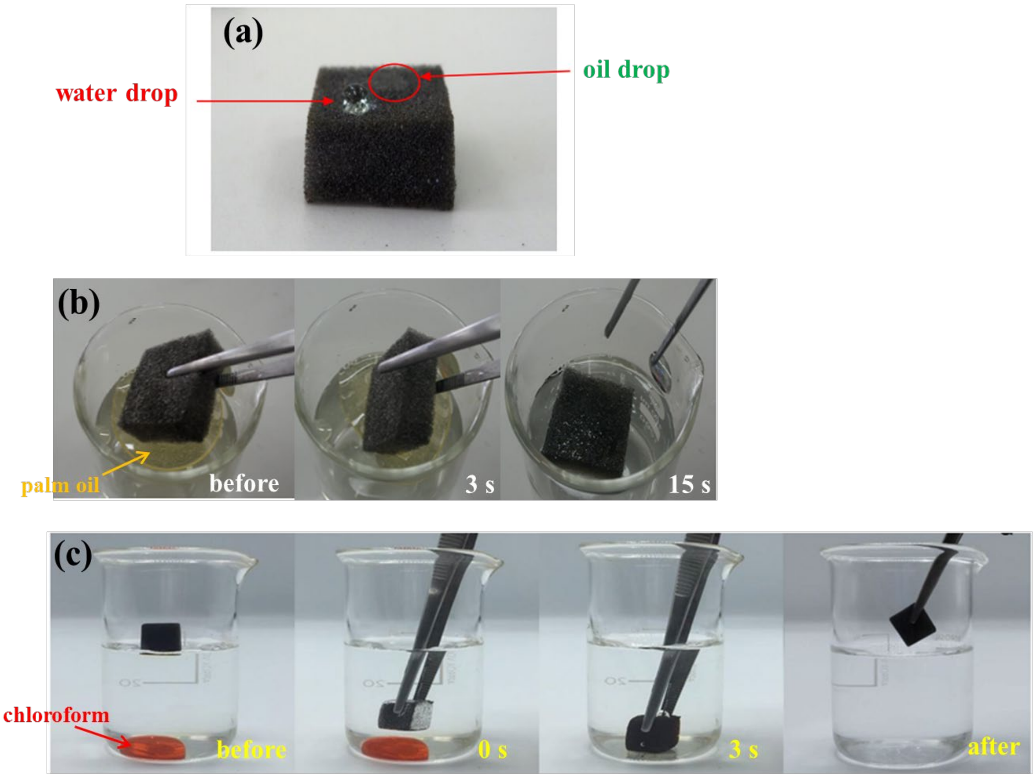

Figure 7. Photographs of (a) oil and water droplets on the surface of MF@rGO-recoat foam; (b) the removal of palm oil and (c) chloroform from water using MF@rGO-recoat foam

3.8 Recyclability of MF@rGO-recoat foam

The reusability of adsorbents is very important for practical applications. In this study, the MF/rGO-recoat foam was tested by repeated adsorption-squeezing processes. Figure 8 shows the adsorption capacity and recyclability (up to 10 times) of as-synthesized MF@rGO-recoat foam for absorbing palm oil, gasoline, diesel, and lubricant oil. From the first use, the MF@rGO-recoat foam exhibited the highest Q_e values were about 103.8 $g.g^{-1}$, 108.6 $g.g^{-1}$, 112.7 $g.g^{-1}$ and 115.9 $g.g^{-1}$ for gasoline, diesel, palm oil and lubricant oil, respectively. The lubricant oil showed the highest Q_e value compared to the other oils. Normally, excellent adsorption performance of a foam depends on the polarity and density of various oils [19, 23]. Thus, the highest Q_e value of lubricant oil with MF@rGO-recoat foam is due to the high density as well as the low polarity of this oil. After 10 cycles of all oil removal tests, the adsorption capacity values of the MF@rGO-recoat had slightly decreased but were stilled higher than 100 $g.g^{-1}$. This indicates that the MF@rGO-recoat foam retained a high and stable adsorption capacity to remove oil from water.

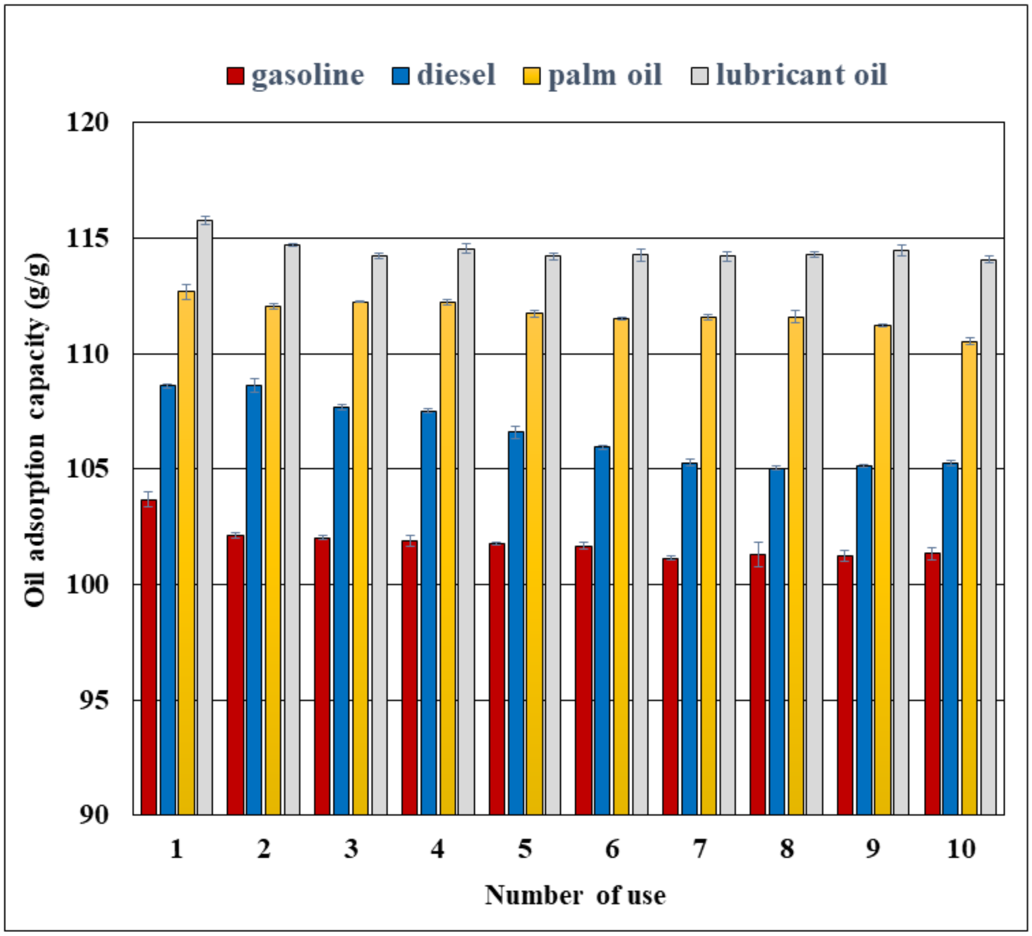

Figure 8. Oil adsorption capacity and recyclability of as-synthesized MF@rGO-recoat foam for adsorbing gasoline, diesel, palm oil, and lubricant oil, respectively.

4. Conclusions

In summary, the MF@rGO-recoat foam with excellent oil-water separation ability was successfully synthesized by a green and conventional heating process using GO as precursor, and vitamin C as reductant. The characteristics of the samples were well-confirmed and analyzed by FTIR, Raman, SEM, and TEM techniques. Depending on the appropriate content of GO and recoating cycle, the as-synthesized MF@rGO-recoat foam exhibited the highest water contact angle (156.9°) as well as excellent superhydrophobicity. The adsorption capacity (Q_e) of the MF@rGO-recoat foam was higher than 103.8 g.g^{-1} in all the oil removal tests, and the highest value seen was about 115.9 g.g^{-1} for lubricant oil. Besides, the synthesized MF@rGO foam can be used easily and recycled up to 10 times for all the oils tested in the oil removal from water.

5. Acknowledgement

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