

*Article*

## Synthesis and Adsorption Performance of Graphene Oxide-Polyurethane Sponge for Oil-Water Separation

Eny Kusrini<sup>1\*</sup>, Indy Ramadhani<sup>1</sup>, Muhammad Idrus Alhamid<sup>2</sup>, Nyuk Yoong Voo<sup>3</sup>,  
and Anwar Usman<sup>4</sup>

<sup>1</sup> Department of Chemical Engineering, Faculty of Engineering, Universitas Indonesia, Kampus Baru UI-Depok 16424, Indonesia

<sup>2</sup> Department of Mechanical Engineering, Faculty of Engineering, Universitas Indonesia, Kampus Baru UI-Depok 16424, Indonesia

<sup>3</sup> Department of Applied Physics, Faculty of Science, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong BE1410 Negara Brunei Darussalam

<sup>4</sup> Department of Chemistry, Faculty of Science, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong BE1410 Negara Brunei Darussalam

\*E-mail: ekusrini@che.ui.ac.id (Corresponding author)

**Abstract.** Graphene oxide was prepared from pencil type 2B by modified Hummers method and used for coating of polyurethane sponge. The graphene oxide-modified polyurethane (GO/PU) sponge was applied as an adsorbent for oil-water separation, which was represented by the removal of diesel oil from water. The formation of graphene oxide and GO/PU was characterized by spectroscopic, imaging, and X-ray diffraction methods. The best adsorption performance of GO/PU was 71.2% efficiency within 10 s of contact time. It was demonstrated that the incorporation of graphene oxide enhanced the adsorption performance of the PU sponge which showed 54.9% efficiency diesel removal from water under the same experimental condition. The result also suggested that graphene oxide (GO) has a synergistic effect of superhydrophobicity and good mechanical performance in the GO/PU sponge.

**Keywords:** Adsorbent, graphene oxide, polyurethane sponges, separation of water-oil.

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## 1. Introduction

Owing to their hydrophobic properties, graphene and graphene oxide (GO) been extensively explored as promising adsorbents to remove a variety pollutants such as dyes, metals, and oils from water [1, 2]. The graphene sheets can be simply prepared and exfoliated from graphite contained in a pencil, and graphene oxide is generally synthesized by oxidation of graphene using the modified Hummers method [3]. In particular, graphene and graphene oxide aerogels have a large number of macropores, which provide adsorption of oil rapidly [4, 5]. Graphene oxide has been also used to modify polyurethane sponges to enhance adsorption efficiency of oil from water, and such a graphene oxide/polyurethane (GO/PU) is found to have high adsorption capability and it is recyclable [6]. In this sense, polyurethane that has the characteristic of the urethane (-NHCOO-) and isocyanate functional groups in its polymer backbone is very oleophilic to clean up oil spills quickly [7]. Graphene oxide in GO/PU not only to expands the spatial interaction between adsorbent and oil, but also plays as an active adsorbent for the oil-water separation [8-18].

It is noteworthy that graphene oxide membranes [8], aspartic acid functionalized graphene oxide mixed with matrix membranes [9], and amine-modified graphene oxide particles have been developed for the separation of oil-water separation [10]. Further development of adsorbents for the oil-water separation has prompted the use of graphene oxide to modify or to decorate metal organic framework [11], melamine sponge [12], melamine sponge with silk fibroin [13], sepiolite [14], lignin-based carbon aerogels [15], polydopamine for the oil-water separation [16, 17]. On the other hand, graphene oxide nanosheets incorporated with membrane have been explored for olive oil/water emulsion separation [18]. In general, the surface chemical property particularly surface micro- and nano-structure of the composites has been pointed out to be crucial in determining the hydrophobicity and effective oil-water separation [19-24].

The hydrophobic adsorbents for oil spill cleanup have received a lot of attention because of many cases of offshore oil spills in sea [25]. Therefore, in this study, a new material based on GO/PU was developed for oil-water separation. The objective is to develop GO-coated PU sponges, and the prepared GO/PU was applied as an adsorbent for separation of diesel oil from water, as a representative model of oil spills or oil-water separation. In particular, graphene oxide (GO) was synthesized from pencil graphite waste type 2B, which is the allotropic form of carbon, by using the modified Hummers method. The prepared graphene oxide and GO/PU were thoroughly characterized by Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and scanning electron microscope-energy dispersive X-ray spectroscopy (SEM-EDX).

## 2. Experimental

### 2.1. Materials

Graphite waste pencil type 2B was obtained from local market. Analytical reagent grade sodium hydroxide (NaOH) (99%), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) 30%, distilled water, potassium permanganate (KMnO<sub>4</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), acetone, and hydrochloric acid (HCl) were purchased from Merck (Darmstadt, Germany). Polyurethane (PU) sponge and diesel oil were purchased from a local market and a local gas station (Indonesia).

### 2.2. Purification of Graphite

Graphite from pencil type 2B was purified using the two-step leaching method. Briefly, graphite was mixed with 1 M NaOH with ratio of 1 g/6 mL. The mixture was heated at a temperature of around 90 °C and stirred for 1 h. Graphite was separated from the liquid and then rinsed with distilled water and dried. Graphite resulting from the first leaching step was mixed with HCl (5%) with the ratio of 1 g/4 mL. After heating, stirring, and rinsing similar to the first step, the purified graphite was then dried at 60°C.

### 2.3. Synthesis of Graphene Oxide

Graphene oxide was synthesized based on the modified Hummers method [26], according to the procedure reported previously by Kusrini et al. [27], as schematically illustrated in Fig. 1. Here, the purified graphite and 98% H<sub>2</sub>SO<sub>4</sub> with the ratio of 1 g: 25 mL were mixed in a beaker glass. The temperature of the mixture was set and maintained at 20 °C. Into the mixture, 3 g KMnO<sub>4</sub> was added gradually while stirring to allow oxidation reaction. A 50 mL of distilled water was slowly added by dripping to stop the reaction, followed by addition of 100 mL of distilled water. 30% H<sub>2</sub>O<sub>2</sub> was then added until a yellowish-brown mixture was obtained. The solution then washed with 5% HCl to remove unreacted KMnO<sub>4</sub>, and the precipitated solid was washed with distilled water for several times until the pH of the solution was 4. The graphene oxide was filtered and dried for further used.

### 2.4. Synthesis of GO/PU Sponge

Polyurethane (PU) sponge was sterilized according to the method reported by Kusrini et al. [28], where it was soaked in acetone for 1 h, followed by rinsing with distilled water. The sterilized PU sponge was then dried at 100 °C for 5 mins, and it was then immersed in a solution of graphene oxide. The obtained GO/PU sponges was dried at 60°C for 1 h in an oven.

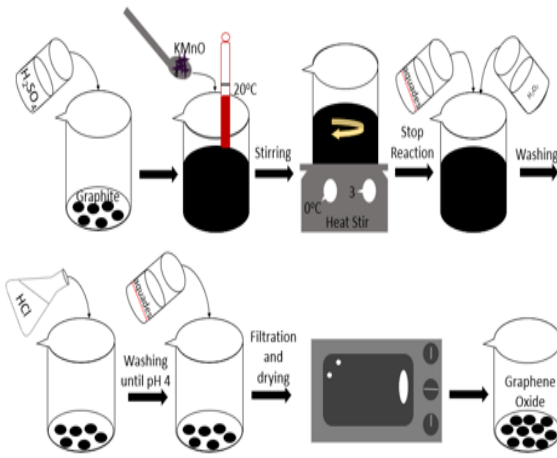


Fig. 1. Schematic illustration of synthesis graphite to graphene oxide.

## 2.5. Performance Test of GO/PU Sponge For Oil-Water Separation

A mixture of 5 g diesel oil with 50 mL water was used as a model of an oil-water mixture. The oil adsorption was performed by the simple sorption-mechanical squeezing test method. GO/PU sponge was put in the prepared oil-water mixture. The mass of the GO/PU sponge was weighted before and after the adsorption test. In this test, the contact times were set to be 1, 5, and 10 s.

Adsorption efficiency was calculated according to the following Eq. (1):

$$Q_w = \frac{m_1 - m_0}{m_0} \quad (1)$$

where  $m_0$  and  $m_1$  are the initial and final weight of GO/PU sponge, and  $Q_w$  is the saturated adsorption capacity of oil on the GO/PU sponge.

The efficiency ( $\eta$ ) was calculated to evaluate the oil rejection coefficient according to Eq. (2):

$$\eta(\%) = \left(1 - \frac{C_1}{C_0}\right) \times 100 \quad (2)$$

where  $C_0$  and  $C_1$  are the oil concentration in oil-water mixture before separation and the concentration of oil collected after separation, respectively.

## 2.6. Characterization

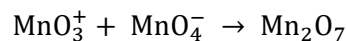
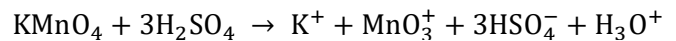
The crystalline of graphene oxide was characterized using X-ray diffraction on an XRD 700 diffractometer (Shimadzu). The vibrational spectra of graphite and graphene oxide were recorded using an FTIR 8201PC spectrophotometer (Shimadzu), and their surface morphology was analyzed by SEM-EDX (ZEISS EVO® MA-10).

## 3. Results and Discussion

### 3.1. Graphene oxide

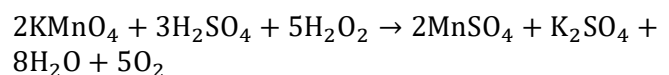
The modified Hummers method to synthesize graphene oxide involve oxidation reaction which was carried out at room temperature, so that it can be considered to be safer compared to the conventional Hummers method at high temperature. Moreover, this method did not use sodium nitrate ( $\text{NaNO}_3$ ) agent which could produce poisonous  $\text{NO}_x$  gas as it is used in the conventional Hummers method. The modified Hummers method is also technically simple and gives a high conversion from graphite to graphene oxide. The resulted graphene oxide has high quality, large surface area, and good mechanical, physical and chemical properties.

Though the oxidation reaction to produce graphene oxide was carried out at a short time and in less extreme acidic conditions, the strong oxidizing agents, such as  $\text{H}_2\text{SO}_4$  and  $\text{KMnO}_4$ , were used for exfoliation and functionalization. The use of these strong oxidizing agents is unavoidable, as graphite has high chemical stability [29]. The reaction between  $\text{H}_2\text{SO}_4$  and  $\text{KMnO}_4$  produced  $\text{Mn}_2\text{O}_7$ , according to the following reaction schemes.



$\text{Mn}_2\text{O}_7$  is a strong oxidizing agent and which can oxidize graphite. However,  $\text{Mn}_2\text{O}_7$  can be explosive when it was heated at temperatures above 50 °C or when it comes into contact with organic compounds. Therefore,  $\text{KMnO}_4$  was added slowly at temperatures below 20 °C to control the amount of  $\text{Mn}_2\text{O}_7$  in the solution and to ensure the temperature of the exothermic reactions to be below 40 °C.

The oxidation process of graphite was monitored by the color change in the solution. The solution will turn black-greenish with the addition of  $\text{KMnO}_4$ , indicating that the graphite oxidation process was started [27,28]. During the oxidation reaction, the color of the solution will change to brown then turn reddish-brown due to the formation of the  $\text{Mn}_2\text{O}_7$  accompanied by an observable increase in the viscosity of the solution. At the final stage, the oxidation reaction was stopped by adding distilled water and hydrogen peroxide (30%). The hydrogen peroxide was added slowly until the color of the solution changed to brownish-yellowish. The resulting brown-yellow color indicates a high level of oxidation [26]. The function of hydrogen peroxide is to stop the oxidation reactions and reduce the excess of  $\text{KMnO}_4$  that does not react during the oxidation process according to the following reaction.



The resulting graphite oxide was then washed with 5% HCl to remove the Mn content as a side reaction of the oxidation process in the resulting solid. Furthermore, it was washed with distilled water repeatedly until the pH of the solution was 4. During the washing process, an exfoliation process occurred, and it was recognized as the graphene oxide gel formation [26]. In this study, the oxidation reaction of 1.00 g purified graphite can produce 1.49 g graphene oxide.

### 3.2. FTIR Studies

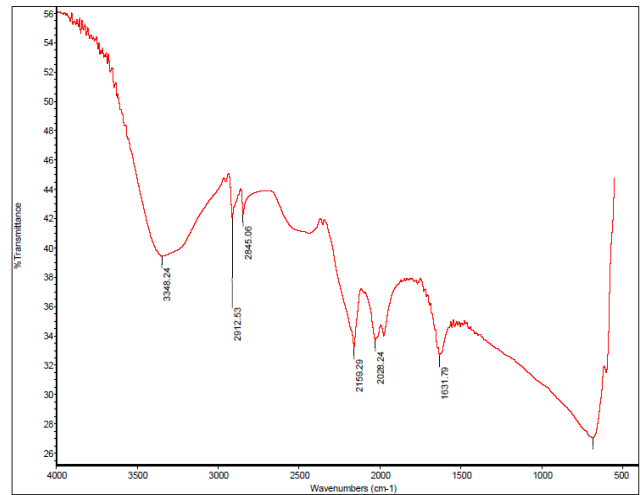
Figure 2 shows FTIR spectra of graphite and graphene oxide, which were used to identify vibrational bands of their functional groups. After the oxidation process was carried out, it was seen that there was a new peak at 3256  $\text{cm}^{-1}$  and it was assigned to the oxygen-containing functional groups. This peak suggested the presence of OH bonds that originated from hydroxyl and carboxylate groups. The peak at 1623  $\text{cm}^{-1}$  was due to C=C bonds of unoxidized carbon, while the peak at 1084  $\text{cm}^{-1}$  was originated from the C–O bond of the epoxy group that presented in the graphene oxide structure. These results are in accordance with those reported in the literature [20, 30]. The FTIR results indicate that the oxidation process has been successfully carried out at this stage.

### 3.3. XRD Characterization

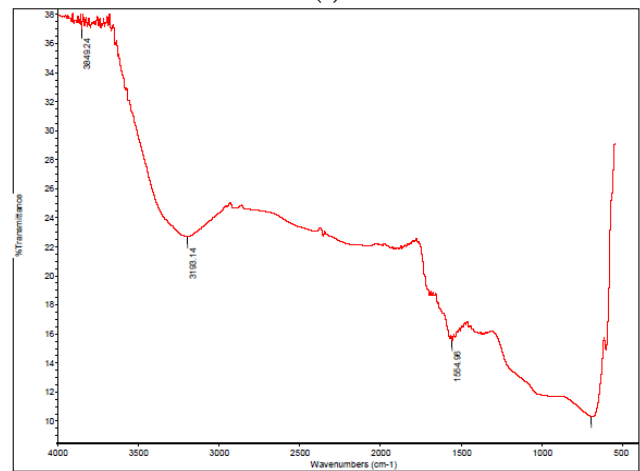
The purified graphite shows a diffraction peak with a very high intensity at a value of  $2\theta = 26.88^\circ$  with an interlayer distance of 0.33 nm, which was estimated using the Bragg's equation (see Fig. 2). It is similar with the peak for purified graphite waste that reported by Kusriani et al. [27, 28]. The highest intensity shows the crystalline nature of the graphite structure which is arranged regularly due to  $\text{sp}^3$  bonding between the graphene layers. After the oxidation process, the diffraction peak value shifts to  $10.46^\circ$  (Fig. 3) with an interlayer distance of 0.84 nm. This diffraction peak is identical to that of reported graphene oxide ( $2\theta = 10.34^\circ$ ) [26]. The interlayer distance of the synthesized graphene oxide is within the range reported in the literature (0.56–0.95 nm) [31]. The large interlayer distance of the graphene oxide in this study is due to the presence of oxygen-based functional groups such as epoxy, carboxyl, hydroxyl, and carbonyl on the carbon layer. The diffraction peaks of graphene oxide were much wider than in graphite, which indicates the occurrence of an exfoliation process in the graphite layer, so that a more irregular (amorphous) crystal structure was obtained.

The presence of functional groups containing oxygen after the oxidation process causes the distance between layers of graphene oxide to increase. Theoretically, the large interlayer distance of the graphene oxide causes weak van der Waals forces between the layers, making it easier for the exfoliation process at the washing stage when the synthesis of graphene oxide takes place. As a result, a single layer/several layers of graphene can be produced.

The XRD results reveal that the oxidation process has been successfully occurred and it causes exfoliation of the graphene layer on the graphite structure.



(a)



(b)

Fig. 2. FTIR spectra of (a) purified graphite and (b) graphene oxide.

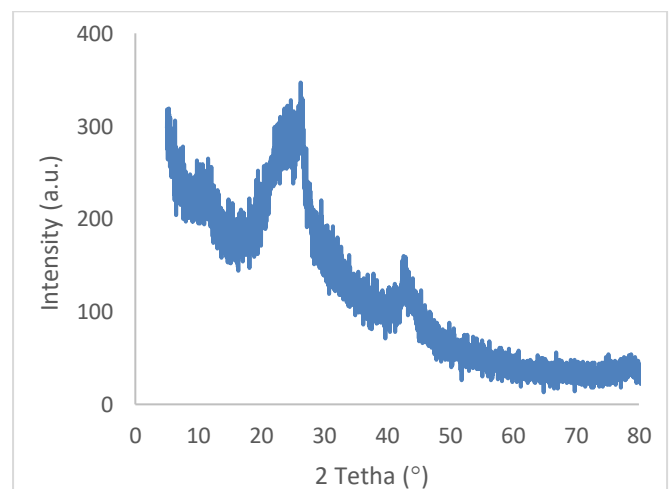


Fig. 3. XRD pattern of GO.

The XRD data can be used for determination of the interlaminar distance ( $t$ ) using the Bragg's equation and the number of layers ( $n$ ) of graphite and graphene oxide using the Debye-Scherrer equation [27]. The calculation results are summarized in Table 1. It was found that oxidation process significantly reduced the number of layers of graphite. The graphite possess 109 layers of carbon sheets, while graphene oxide has only 10 layers. This data is comparable with graphene oxide produced from spent pot lining from industrial aluminum wastes which has 9 layers of carbon sheets [27]. This result is better than graphene oxide and reduced graphene oxide derived from graphite waste using the modified Hummers's method with zinc as reducing agent [32, 33].

Table 1. Number of layers and interlaminar distances of graphite and graphene oxide.

Sample	$2\theta$ ( $^{\circ}$ )	FWHM ( $^{\circ}$ )	$d$ (nm)	$t$ (nm)	$n$
Graphite	26.88	0.34	0.33	35.99	109 [26]
Graphene oxide	10.46	1.84	0.84	8.63	10

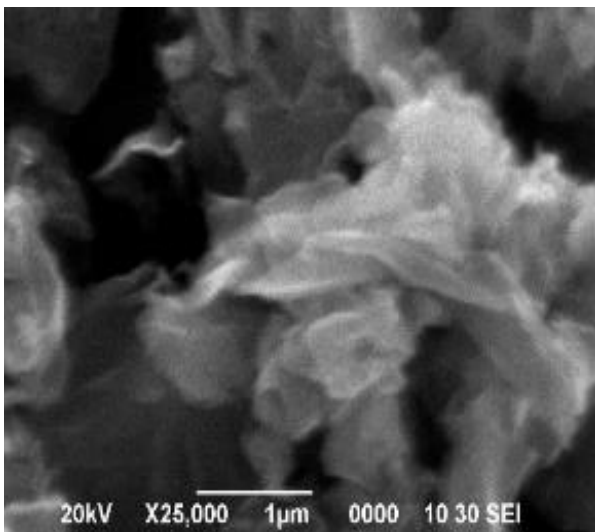


Fig. 4. SEM image of graphene oxide with a magnification of 25.000 $\times$ .

### 3.4. SEM-EDX Characterization

The synthesized graphene oxide was characterized using SEM-EDX method to assess its surface morphology and elemental contents. As shown in Fig. 4, layered sheets of the graphene oxide are thick due to the flake structure of graphite [27], and the spacing distances between the layers are clearly visible. SEM image of the graphite generally shows its layered structure build up of carbons, and it forms coarse flakes with irregular sizes. After converting to the graphene oxide, the surface becomes smoother and the layer sheets are thinner. As the GO/PU sponge was prepared by mixing the graphene oxide without any physical and chemical modification, one could

consider that the structure of graphene oxide remains intact in the GO/PU sponge [27, 34-36].

As listed in Table 2, the C and O contents of graphene oxide were found to be 54.27 and 31.14 w%. These data were in accordance with the commercially available graphene oxide with commonly contains 45-56 w% C and 30-50 w% O (see Table 3) [37]. Nevertheless, some impurities, such as Si and Al elements, were also found in the graphene oxide.

Table 2. The EDX analysis of GO.

Element	w%
	GO
C	54.27
O	31.14
Si	8.45
Al	6.14

Table 3. Specification of commercially available graphene oxide [37].

Specification	Graphene oxide
Carbon (%)	45-56
Oxygen (%)	30-50
Hydrogen (%)	$\leq 4,5$
Nitrogen (%)	$\leq 1,5$
Lateral Size ( $\mu\text{m}$ )	$\geq 7,0$
Thickness (Nm)	$< 5$
Specific Surface Area ( $\text{m}^2/\text{g}$ )	500-1000

### 3.5. Performance Test of GO/PU Sponge

To adsorption performance of diesel oil onto GO/PU sponge, as a model of separating oil spills from water, was carried out using the simple adsorption method [28, 33]. In comparison, superhydrophobic kaolinite modified graphene oxide-melamine sponge (K-GOMS) were tested in various types of organic solvents and oils such as diesel oil, motor oil, liquid paraffin, and kerosene [12].

As summarized in Table 4, the masses of oil adsorbed onto GO/PU sponge at contact times of 1, 5, and 10 s are 1.952, 3.081, 3.558 g, respectively, suggesting that the effect of contact time on the oil adsorption. Elongation of contact time without any additional force from outside increase the removal efficiency of oil from water, as shown in Fig. 5. Within 10 s of contact time, the removal efficiency of oil on the synthesized GO/PU sponge was 71.2%. It is, however, smaller compared to the GO@CuO mesh for separation of water-oil with efficiency of 99.49% [16], but it is comparable was observed adsorption oil on K-GOMS and K-MS and the adsorption was also saturation within 10 s [12]. Under similar experimental conditions, adsorption efficiency of diesel oil on PU sponge was lower compared with those of CO/PU sponge. At 10 s of contact time, the adsorption efficiency of oil on PU sponge was only 54.9%. The results showed that graphene oxide (GO) could improve the adsorption capacity of oil

on the PU sponge, suggesting that graphene oxide improved superhydrophobicity and good mechanical performance of the GO/PU sponge. However, this efficiency for oil-water separation is not so high compared to other modified graphene oxide [10-16].

In the present work, the synthesised GO/PU sponge adsorbs diesel oil less than 5 g diesel oil in 50 mL of water. Thus, the GO/PU sponge is not considered appropriate adsorbent for separation of water-oil due to its low adsorption efficiency (71.2%), less than that of reduced graphene oxide (rGO)/PU sponges which are more appropriate for separating oil from water due to superhydrophobic character of the rGO [28]. Nevertheless, graphene oxide can be modified further by introducing some functional groups such as amine [10], or it can also be blended with other natural or synthetic materials, such as semiconductor nanoparticles [38], metal organic framework [11], kaolinite [12], lignin [15], aspartic acid [9] to increase their capability as adsorbents for oil-water separation.

Table 4. Total mass of oil adsorbed on PU and GO/PU sponge.

Time (s)	Oil Mass (g)	
	PU	GO/PU
1	0.85	1.952
5	1.97	3.081
10	2.74	3.558

Table 5. Efficiency of GO/PU sponge.

Time (s)	Efficiency (%)	
	PU	GO/PU
1	17.02	39.04
5	39.42	61.62
10	54.78	71.16

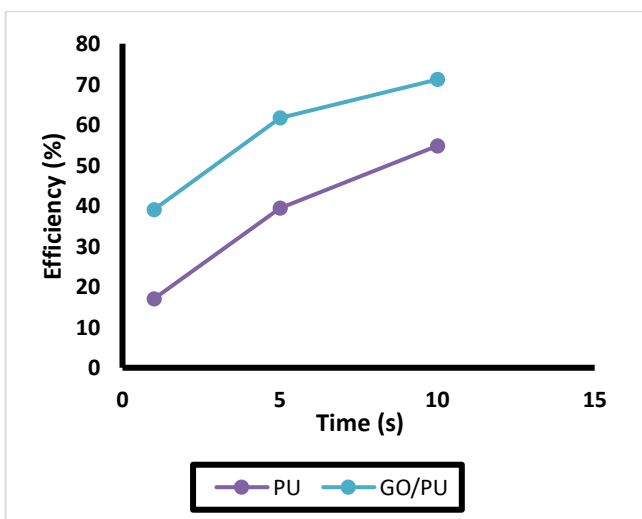


Fig. 5. Comparison the performance of PU [28] and GO/PU sponges for oil-water separation.

## 4. Conclusion

In summary, graphene oxide was prepared from graphite derived from pencil type 2B by using the modified Hummer's method. The chemical, crystalline structure, and elemental composition of the synthesized graphene oxide were characterized by FTIR, XRD and SEM-EDX. The graphene oxide was then used to functionalize polyurethane, generating GO/PU sponges which was used as adsorbent for oil-water separation by simple sorption method. The results suggested that the adsorption efficiency of oil on GO/PU sponges is 71.2% within 10 s of contact time. It is better than the adsorption efficiency of oil on pristine polyurethane (54.9%), under the same experimental conditions. The result suggested that the prepared GO/PU has a synergistic effect of superhydrophobicity and good mechanical performance in the GO/PU sponge. However, the efficiency for the removal of oil is not so high, less than that of reduced graphene oxide (rGO)/PU sponges and other chemically modified rGO. Therefore, further chemical modification of graphene oxide to enhance its superhydrophobicity with introducing new functional groups or other components such as metal framework, lignin, kaolinite or other type of acid would be of interest for future studies.

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**Eny Kusrini** received her B.Sc degree in chemistry from Gadjah Mada University, Indonesia in 2002, and the Ph.D. degree in chemistry from Universiti Sains Malaysia, Malaysia in 2007. Since 2009, she joined Department of Chemical Engineering, Universitas Indonesia. Her research interests include extraction and separation of lanthanide and versatile application of lanthanides and their derivative compounds. She also studied nanotechnology, quantum dot, synthesis of graphene oxide, reduced graphene oxide and their derivative compounds, adsorption and utilization of agro-wastes as adsorbents and utilization of natural kaolin.



**Indy Ramadhani** received her B.Sc degree in chemical engineering from Universitas Indonesia in 2020. Her research interests include in graphene and its application.



**Muhammad Idrus Alhamid** received his Bachelor engineering degree from Universitas Indonesia, Indonesia in 1978 and his PhD degree in Mechanical engineering from Katholieke Universiteit, Leuven, Belgium in 1989. Since 1980, he has been a lecturer in Department of Mechanical Engineering, Universitas Indonesia. His research interests include heat exchangers, refrigerant and energy.



**Nyuk Yoong Voo** graduated from the University of Southampton with a Ph.D. degree in Optoelectronics in 2006 and then joined Universiti Brunei Darussalam from 2006 until present. Her main research interests include: 1) synthesis and application of graphene-based materials; 2) Thermochromic materials for architectural glazing. She has published over 40 peer-reviewed journal articles with over 500 citations.



**Anwar Usman** received his PhD degree in Chemistry from Tohoku University, Japan in 2000, and extended his research in several places, including Universiti Sains Malaysia (Malaysia), Max-Born-Institut für Kurzzeitspektroskopie, Berlin (Germany), Osaka University (Japan), and École Normale Supérieure du Chimie, Paris (France), National Chiao Tung University, Hsinchu (Taiwan), and King Abdullah University of Science and Technology, Thuwal (Saudi Arabia). Since 2015, he joined Department of Chemistry, Universiti Brunei Darussalam. He may be contacted at Email: [anwar.usman@ubd.edu.bn](mailto:anwar.usman@ubd.edu.bn)