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SYNTHESIS AND CHARACTERIZATION OF GRAPHITE OXIDE, GRAPHENE OXIDE, AND REDUCED GRAPHENE OXIDE FROM GRAPHITE WASTE USING MODIFIED HUMMERS' METHOD AND ZINC AS REDUCING AGENT

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ABSTRACT

In this paper, synthesis of graphite oxide, graphene oxide (GO), and reduced graphene oxide (rGO) from the spent pot lining (SPL) of aluminum industrial waste using modified Hummers' method and zinc as reducing agent is examined. The effects of ultrasonication time from 1 to 2 h and Zn mass ranging from 8 to 24 g as reducing agent were observed in detail for reduction reaction of GO into rGO. The chemical structures and morphology of the samples were confirmed through Fourier Transform Infrared Spectroscopy (FTIR), Particle Size Distribution (PSD), Scanning Electronic Microscopy-Energy Dispersive X (SEM-EDS), and X-ray Diffraction (XRD) characterizations. The FTIR analysis confirmed the formation of GO. Although some restacking/unexfoliated graphite structures showed a diffraction peak at 2θ of 26.54°, the XRD analysis clearly exhibited a peak at 2θ of 20.04°, assigned to rGO after reduction of the GO. The smallest particle size of rGO was observed in the range of 1 to 10 μm when under ultrasonication time of 1 h and Zn mass of 8 g. The FTIR spectrum of GO showed that there was a functional group C=C, which is an indication of rGO formation due to the covalent bonding of the graphene structure. SEM image of the rGO showed that the morphology seemed thick and layer stacking. The quality of rGO produced in this study needs to be improved further to meet requirements for applications.

Keywords: Graphene oxide; Modified Hummers' method; Reduced graphene oxide; Ultrasonication time; Zinc as reducing agent

1. INTRODUCTION

The aluminum smelting process using electrolysis produces a huge amount of waste of graphite electrodes (Reny et al., 2016). In this electrolysis process, graphite is used as cathode, since it is inert and capable of conducting heat and electric current efficiently. Some of the graphite

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used in the electrolysis processes will be recycled, while the remaining will be recycled, while the remaining will be dumped as waste. To present, the reutilization of electrode graphite waste has only been as filler in the production of steel. To improve the use and value of this graphite waste, investigations for synthesis and characterization of graphite waste as valuable products, such as adsorbents (Kusrini et al., 2017; Kusrini et al., 2018) have been reported.

The aluminum industry continues to grow rapidly year by year with rate increase of 5% per year, and the aluminum production was predicted to reach 68 million tons in 2020 (Pei & Cheng, 2012). Increasing aluminum production prompts an increase in the use of graphite electrodes in the electrolysis of Al_2O_3. The electrolytic cell lining should be replaced every 3–5 years, and spent pot lining (SPL) becomes solid waste. The main content of SPL is carbon in the form of graphite, which can actually be utilized as raw material for the production of graphene-related materials. As we know, graphite is the most common feedstock for the synthesis of graphene using a top-down method. The grade of graphite depends on flake size and carbon content; thus, the price ranges from \$430 to \$20,000 per MT (Lee et al., 2019). Increasing the carbon content in graphite makes it more valuable.

Graphene is commonly a monoatomic two-dimensional sheet-like material with sp2 hybridized carbon atoms configured in a hexagonal or honeycomb-like structure, and its thickness is similar to an atom diameter (Novoselov et al., 2004; Lee et al., 2019). Graphene is the world's thinnest material—a single layer of carbon atoms that has a hexagonal structure (Zhi-gang et al., 2012). The excellent electrical properties of graphene can make it play a large role in energy storage, material composites, sensors, and other fields (Dikin et al., 2007). Graphene is recognized as an advanced material due to its advantages and unique properties. With a thickness of about one carbon atom, graphene has optical transparency of up to 97.7% (Nair et al., 2008). The structure of graphene, consisting of layers, makes graphene highly conductive with a carrying mobility of up to 200,000 $cm^2V^{-1}s^{-1}$ and thermal conductivity of up to 5,300 $Wm^{-1}K^{-1}$ (Bolotin et al., 2008; Balandin et al., 2008). Graphene oxide (GO) and its reduced graphene oxide (rGO) are classified as graphene family materials and have many applications, such as optical, in biomedical water treatment, and as adsorbents (Raghavan et al., 2017; Ahmad et al., 2019).

Graphene can be synthesized using two methods, namely bottom-up and top-down (Supriadi et al., 2017). In this study, a top-down method including the exfoliation of graphite and the chemical reduction of graphite oxide and graphene oxide (Marcano et al., 2010) was used. Graphite oxide can be synthesized by oxidizing graphite. The Hummers method is commonly used to synthesize graphite oxide because the final product has a higher oxidation rate than the final product of the Staudenmaier method (Hummers & Offeman, 1958). The materials used in the Hummers method are also easier to obtain and are less dangerous than the Staudenmaier method. In the Hummers method, graphite oxide is reacted with potassium permanganate ($KMnO_4$) and sodium nitrate ($NaNO_3$) in a sulfuric acid solution (H_2SO_4). Several reducing agents, such as sodium borohydrate, hydrazine, and ascorbic acid, have been used for conversion of GO to rGO. On the other hand, chemically reducing GO into rGO using Fe or Zn in an acidic medium under ambient conditions has also been reported (Jassby et al., 2012; Wang et al., 2009).

Herein, the synthesis of rGO from SPL aluminum industrial waste using modified Hummers' method and Zn as a reduction agent is reported. The effects of ultrasonication time from 1 to 2 h and Zn mass ranging from 8 to 24 g as reducing agent were observed in detail for reduction reaction of GO into rGO.

2. MATERIALS AND METHODS

2.1. Materials

SPL which was graphite waste from an aluminum industry (Inalum Co., Indonesia) was used as the raw material. Before being used as a graphite source, the electrode powder was smoothed using a ball mill, then the crushed powder was sieved using a 200 mesh (75 μm) sieve to uniform particle size. The synthesis process was conducted using modified Hummers' method, with graphite waste from the SPL, $KMnO_4$, $NaNO_3$, and H_2SO_4 as base materials.

2.2. Synthesis of Graphite Oxide from Graphite Waste (SPL)

The synthesis of graphite oxide was begun by dissolving graphite waste (2 g) in H_2SO_4 (98 mL, 98%) for 1 h. Then, 4 g $NaNO_3$ was added and stirred for 1 h. After that, 8 g $KMnO_4$ was added gradually then stirred for a further 2 h. The reaction was then moved to an ice bath to keep the temperature under 20°C. In the process of this addition, the solution changed color from dark black to greenish black. To homogenize the reaction, further stirring was done at a temperature of 35°C for 24 h. When the sediment was too thick, 100 mL of distilled water was added. After 24 h, stirring was switched off and H_2O_2 added to the solution, and the solution had turned to a light yellow. After stirring for 30 min, the solution was centrifuged to separate the deposition of graphite oxide, residual liquid, and impurities. After centrifugation, HCl was added to the graphite oxide to remove the metal impurities. Finally, the sample of graphite oxide was washed with deionized water until the pH was 7. Graphene oxide (GO) was thus synthesized by the modified Hummers method.

2.3. Reduction of GO into rGO using Zn as Reducing Agent

GO (0.4 g) was dissolved in 400 mL of distilled water under ultrasonication for 1 and 2 h, respectively, at 54,000 Hz. The solution was then added with various masses of Zn powder (8, 16, and 24 g) in 100 mL 35% HCl and under stirring for 1 h. After the reduction reaction finished, 100 mL 35% HCl was added to remove the excess Zn. The suspension of rGO was filtered and washed with distilled water until the pH was neutral and to remove all the impurities. The result of the black suspension of rGO was obtained and kept for further characterization.

2.4. Characterization

FTIR was recorded on a Perkin Elmer system 2000 FTIR spectrophotometer in the range of 4000–400 cm^{-1} using the conventional KBr pellet method. The surface morphology was measured using a scanning electron microscopy (JEOL JSM-6360LA). The size distribution of particles was characterized by a particle size analyzer (PSA). All characterizations were carried out at room temperature.

3. RESULTS AND DISCUSSION

3.1. Synthesis of Graphite Oxide

The preparation of rGO was started with the oxidation processing of graphite into graphite oxide. The graphite oxide was exfoliated in aquadest until the GO was formed by using ultrasonication. A reduction reaction then followed to reduce the oxygen functional groups in the GO, using Zn powder as reducing agent. The preparation of graphene from graphite waste is shown in Figure 1.

Here, the oxidizing agent used was $KMnO_4$, which is a strong oxidizing agent with a reduction potential (E°red) of 1.679. Potassium permanganate is an important oxidizer that is widely used in various organic and inorganic reactions because of its capability to oxidize various kinds of functional groups. This process can only occur in strong acid conditions (H_2SO_4). In addition to acting as a solvent of graphite, functions in other oxidation processes (Sorokina et al., 2005; Marcano et al., 2010).

The oxidation reaction occurred in a strong acid condition, resulting in a dark brown mixture. The change in color from dark green to dark brown is due to the reaction between graphite, H_2SO_4, and $KMnO_4$ and indicates that the oxidation process of graphite has started. During this oxidation process, some functional groups, such as phenol groups, epoxy groups, ketones, carboxyl groups, and carbonyl groups, are formed on the surface of carbon (Shao et al., 2012). These functional groups cause the material to be highly hydrophilic and to exfoliate easily into graphite oxide. The addition of $NaNO_3$ during the process decreases the number of layers and slightly increases the distance between layers, whereas the addition of H_2O_2 stops the oxidation process.

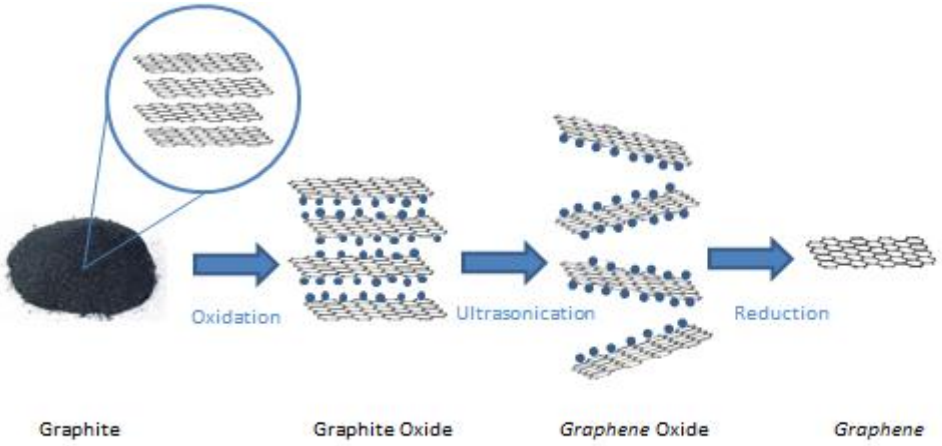

Figure 1 Schematic illustration of synthesizing graphene from graphite waste using modified Hummers' method and zinc as reducing agent

3.2. Synthesis of GO

The synthesis of graphene began with the manufacture of graphene oxide (GO) as precursor. GO was obtained by dispersing graphite oxide in water using an ultrasonication process. In ultrasonication, ultrasonic waves function to process exfoliating graphite oxide into GO. Through the ultrasonication, graphite oxide was exfoliated in water, and a homogenous water-based graphene oxide suspension was produced. The interaction between graphite oxide and ultrasonic waves caused the shear forces and cavitation processes experienced by the medium in the form of water. The cavitation process occurs due to differences in pressure during the ultrasonication process, resulting in the initiation of the graphite oxide exfoliating process that causes the formation of GO (Marcano et al., 2010). The spacing of layers in graphite oxide is greater due to the addition of water and oxygen molecules, thus facilitating the exfoliation process, which causes the formation of GO (Shen et al., 2010). This exfoliation process was done using a mechanical technique because an ultrasonic wave is a mechanical wave with a frequency range of more than 20,000 Hz. The ultrasonication used in this study had a frequency of 54,000 Hz. In order to obtain the structure of graphene or rGO in a single or small layer, a longer ultrasonication process varying from 1 to 2 h was used.

3.3. Reducing GO into rGO using Zn as Reduction Agent

The reduction reaction of GO into rGO was carried out in an acidic condition with Zn as a reducing agent. The reduction of GO occurred because Zn was added to the solution and reacted with H_2O, as shown in Equation 1, forming the H^+ ion that initiates the reduction process (Zhu et al., 2010).

$$Zn + 2H_2O \rightarrow Zn(OH)_2 + 2H^+ + 2e^-$$

$$Zn(OH)_2 \rightarrow ZnO + H_2O$$

$$GO + 2H^+ + 2e^- \rightarrow rGO \qquad (1)$$

HCl is used to form an acidic condition. The addition of Zn powder was carried into a solution of GO that has an acidic condition. Zn powder reacted with GO to produce gas bubbles due to the reduction reaction of oxygen functional groups. When the bubbles have ceased to react, HCl is again added to remove ZnO, which is an impurity. Zn powder made the rGO morphology thinner. In this study, to observe the effects of Zn powder as a reducing agent in the reduction reaction, the mass of Zn was varied from 8 to 24 g.

3.4. FTIR Studies

The FTIR spectra of graphite waste, graphite oxide, and GO were measured between 4000–400 cm^{-1}. For the SPL, the functional groups of C-F (1384 cm^{-1}), C-Cl (700 cm^{-1}), and C-I (586 cm^{-1}) were observed. These are impurities in SPL waste. Figure 2 shows that graphite oxide and GO have a similar peak, but the transmittance intensity is different. The functional groups in graphite oxide and GO are similar: there are carbonyl C=O (1754 cm^{-1}), aromatic C=C (1573 cm^{-1}), carboxyl C-O (1238 cm^{-1}), alcohol C-O (1065 cm^{-1}) and C-F (1399 cm^{-1}). In the FTIR spectrum of graphite oxide, however, a C-OH absorption peak was not observed. It can be seen that the intensities of graphite waste (SPL) peaks increase after treatment by the oxidizing process and ultrasonication into graphite oxide and GO, indicating that the oxidizing and the synthesis of GO were successful. An absorption band at 1573 cm^{-1} is assigned for the C=C aromatic functional group on GO. Based on FTIR spectral analysis, it can be concluded that ultrasonication with Zn as a reducing agent has potential as a method for the production of GO and rGO. As we know, graphene is made from pure carbon, where each carbon atom is connected by covalent bonding in the same planar within a monolayer graphene sheet, linked by the van der Waals interaction (Lee et al., 2019).

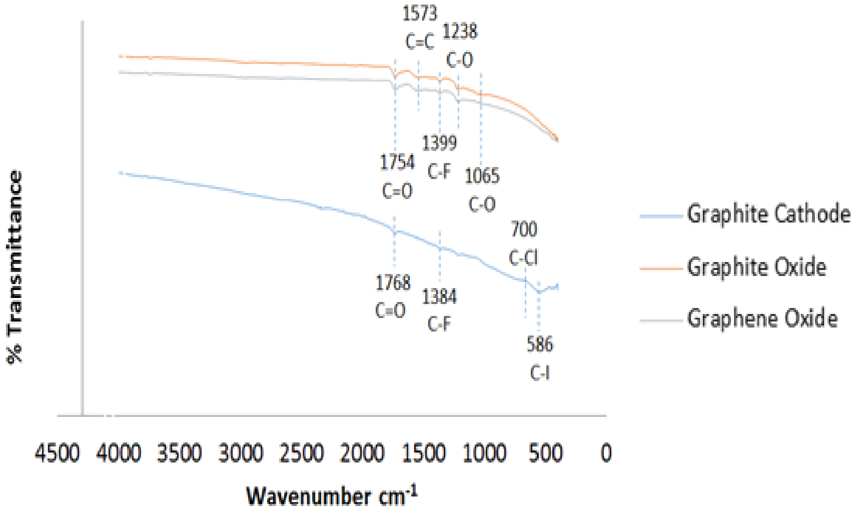

Figure 2 FTIR spectra of graphite waste, graphite oxide, and GO

3.5. XRD Studies

The phase structures of graphite waste and GO were investigated by XRD. XRD patterns of the graphite waste and graphite oxide are shown in Figure 3. It can be seen that there is the same peak in both samples. In the graphite waste, a peak at 2θ of 26.54° with d-spacing of 3.36

Å and a very high intensity of 469,590.8 a.u was observed. This is similar to previously reported graphite waste with a diffraction peak at 26.5° (Huang et al., 2011; Shao et al., 2012; Naebe et al., 2014). In the XRD spectra, the graphite waste has high crystallinity with a sharp peak. However, upon comparing with database, impurity elements such as sodium, fluorine, aluminum, magnesium, and ferrum, forming NaF compounds at $2\theta = 38.85°$, $Na_3(AlF_6)$ at $2\theta = 46.71°$, CaF_2 at $2\theta = 54.63°$, and valleriite at $2\theta = 7.82°$ were also observed. In the graphite oxide, a peak at 2θ of 26.64° with d-spacing of 3.35 Å and a much lower intensity (15,144.49 a.u.) was observed. Naebe et al. (2014) reported the diffraction peak of graphite oxide at 2θ of 10.5°. In this study, the graphite oxide showed that the oxidation reaction was incomplete, because there was no diffraction peak observed at 2θ of 10.5°, the peak of graphite oxide. We assumed that the graphite waste contained many impurities because no purification treatment was conducted before we used it as a precursor for synthesis of rGO, and we assumed that this would influence the oxidation reaction in the formation of GO and rGO.

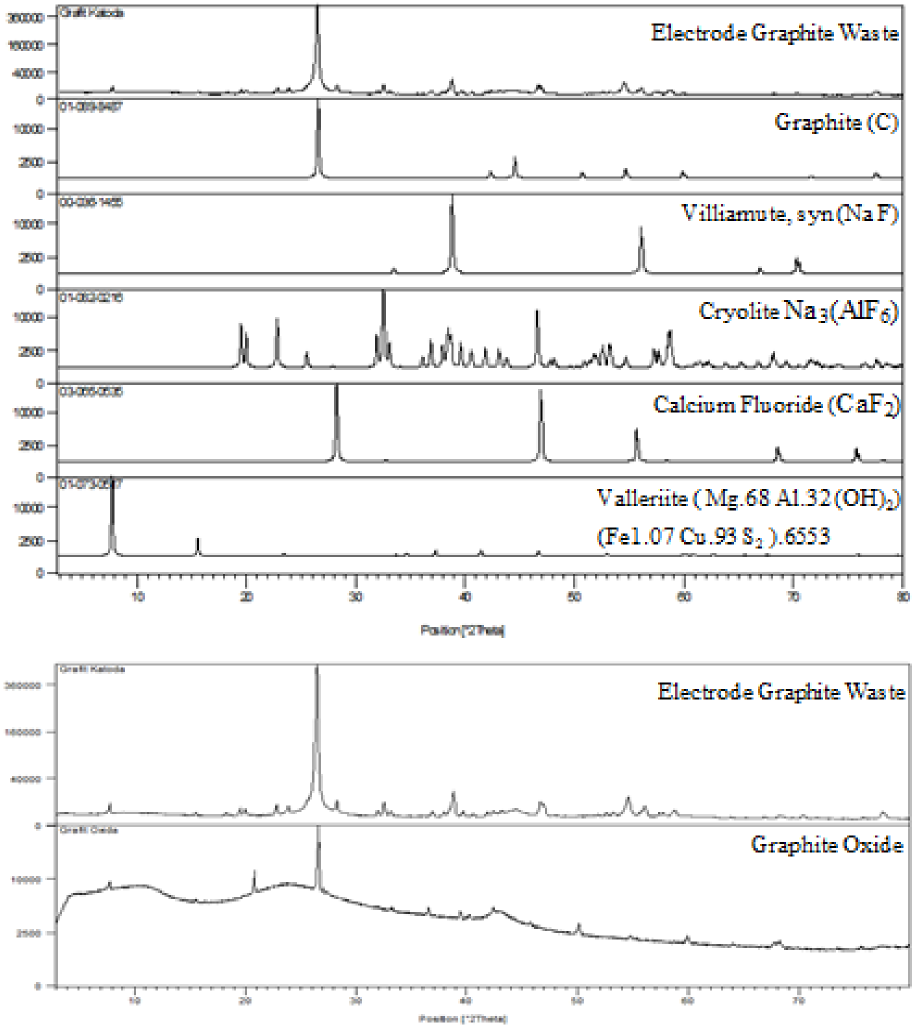

Figure 3 XRD patterns of graphite waste and graphite oxide

3.5. Particle Size Distribution Studies

The PSD data of the graphite oxide are shown in Figure 4. The particle distribution size of 10% was 22.33 µm, 50% was 148.25 µm, and 90% was 231.79 µm. These results indicated that the particle size of graphite oxide was larger than the graphite waste (75 µm), which indicates that agglomeration occurred in the graphite oxide, so that the particle size became larger.

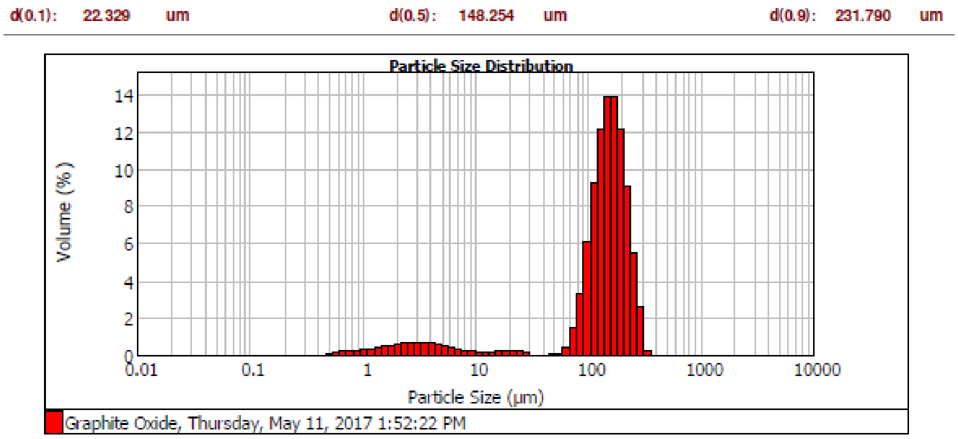

Figure 4 PSD Characterization of graphite oxide

The PSD characterization of the GO is shown in Figures 5a and 5b. It can be seen that the GO with 2 h of ultrasonic time had a larger particle size compared to the GO with 1 h of ultrasonic time. However, when compared with graphite oxide, the size of the GO particles was much smaller. This shows that the graphite oxide was dispersed due to ultrasonic waves, so that the particle size became smaller.

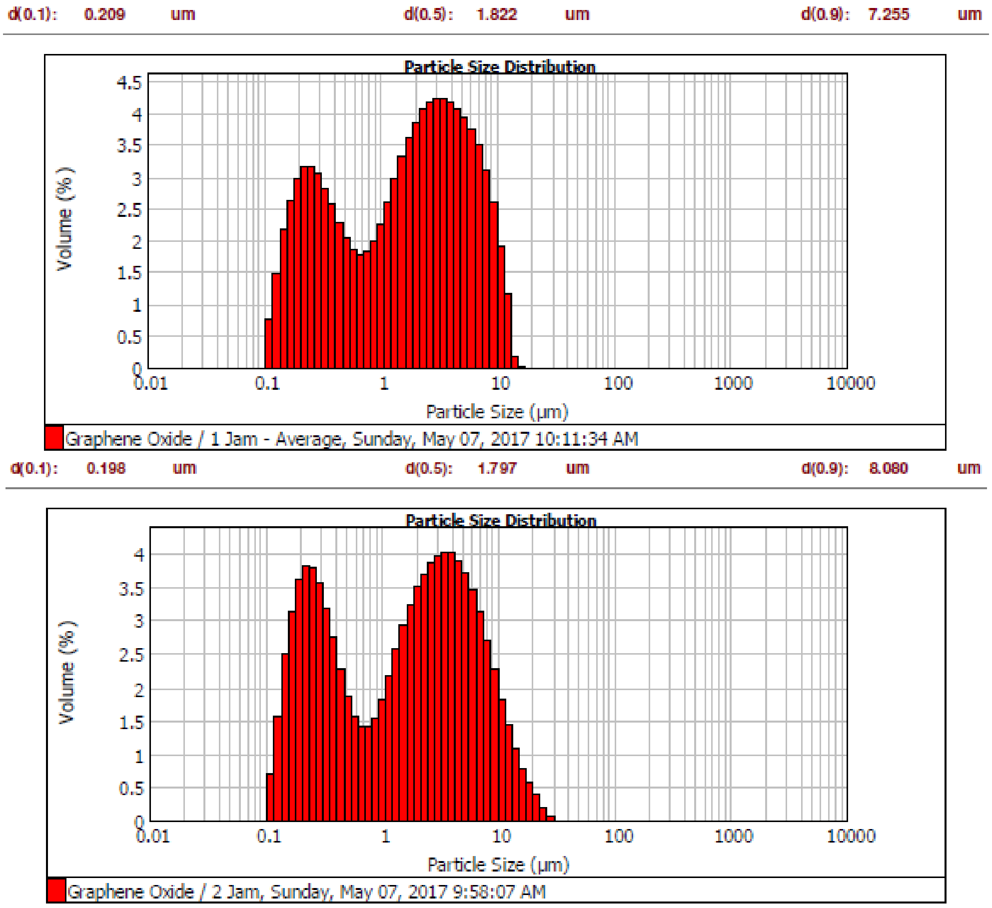

Figure 5 PSD patterns of GO synthesized for 1 h (a) and 2 h (b)

The PSD characterization of rGO with ultrasonication times of 1 h and 2 h and Zn mass of 8 g, 16 g, and 24 g are shown in Figures 6a–6c. It can be seen that rGO synthesized with 1 h of ultrasonic time and 8 g of zinc had the smallest particle size. It is notable that by increasing the ultrasonication time and the mass of zinc as a reducing agent, the particle size distribution of rGO also increased (see Table 1).

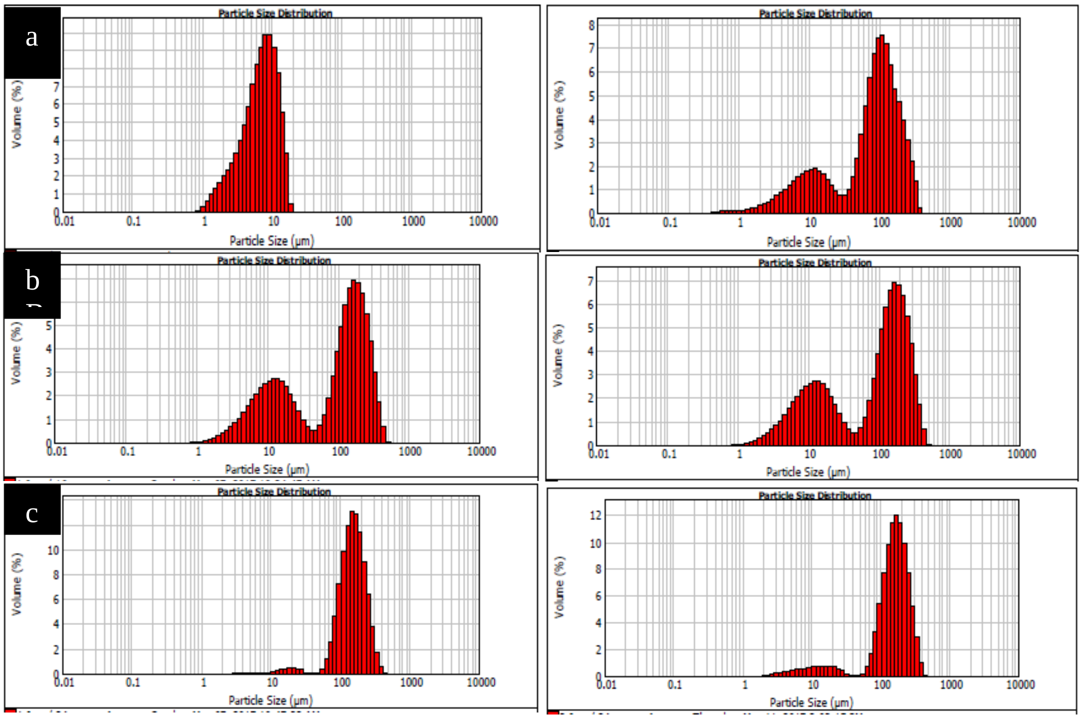

Figure 6 PSD characterization of rGO with ultrasonication times of 1 h (left), 2 h (right), 8 g (a), 16 g (b), 24 g (c)

Table 1 Particle size distribution characterization of rGO with variation of ultrasonication time and mass of Zn as reducing agent

Ultrasonication time (h)	Mass of Zn (g)	Particle size (µm)		
		10%	50%	90%
1	8	2.64	7.12	12.95
1	16	7.09	113.49	273.92
1	24	86.05	151.60	251.41
2	8	7.77	89.92	214.16
2	16	6.48	50.16	243.96
2	24	65.70	159.17	271.03

3.6. Morphology and Composition Studies

The morphological and surface chemistry of graphite waste, graphite oxide, and rGO with variation of ultrasonication times and mass of Zn as reducing agent are shown in Figures 7a–7e. As previously reported, SEM is the most important technique for characterization of morphology of rGO and its derivatives because of the highly energetic electrons used to produce the SEM images of the samples (Lee et al., 2019). In addition, it can be used to observe the topography, crystallographic structure, shape, size, and composition of materials (Ahmad et al., 2019). Indeed, the thickness and number of layers of graphene and its derivatives can be calculated based on the color depth (Dong & Chen, 2010).

SEM tests were conducted only for the three samples with the smallest particle size based on the PSD characterization, namely: (1) rGO with 1 h ultrasonic time and zinc as reducing agent of 8; (2) 16 g; and (3) rGO with 2 h ultrasonic time and zinc of 8 g. The SEM images of the rGO are shown in Figures 7c, 7d, and 7e.

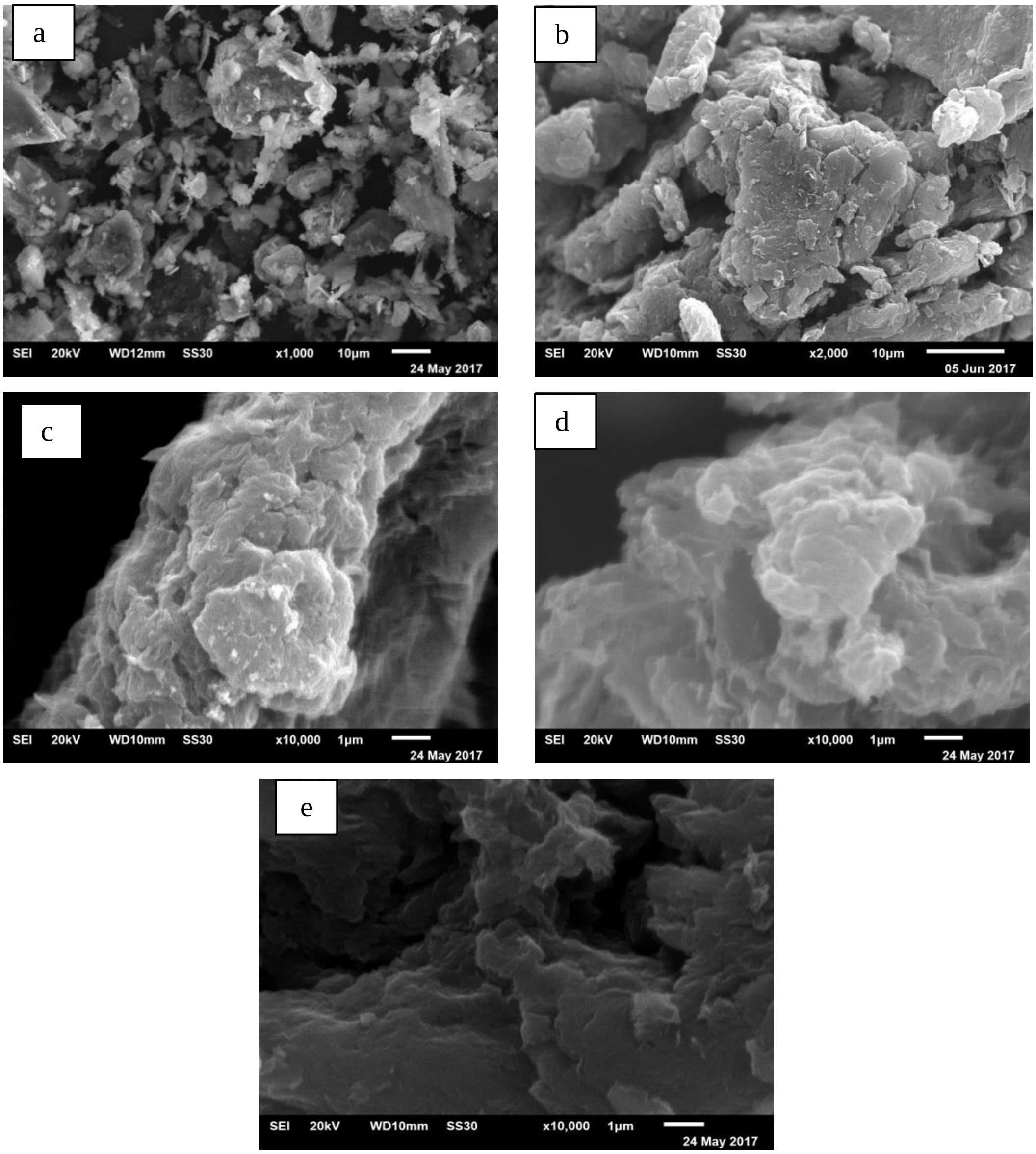

Figure 7 SEM images of graphite waste (a), graphite oxide (b), rGO 1 h with Zn of 8 g (c), rGO 2 h with Zn of 8 g (d), rGO 1 h with Zn of 16 g (e), where graphite waste with 1,000× magnification, graphite oxide with 2,000× magnification and rGO with 10,000× magnification

Single-layer graphite (graphene) was not formed in this study; however, the structure of the resulting rGO seems thick and layer stacking, which may be caused by the preceding graphite oxide that was not reacting completely; thus, it influenced the rGO produced. As was noted, imperfections of oxidation and reduction reactions are caused by impurity elements present in the graphite waste without pre-purification treatment, so that the purity of the graphite waste as a precursor was low. Previous research reported that graphite non-modified (GNM) has a carbon content of 80.09% (Kusrini et al., 2017). According to Lee et al. (2019), the graphite waste in this study included amorphous powder with a carbon content of 80–85% and estimated price of about $430–$480/MT.

The SEM-EDS data was collected to determine the carbon and oxygen (C/O) ratio of the resulting graphite oxide. The EDS characterization showed that the C/O ratio of the resulting graphite oxide was 6.22 (see Table 2). This ratio is higher than the effectiveness of the oxidation process with the C/O ratio between 2.1 and 2.9 (Hummers & Offeman, 1958). We concluded that the oxidation method in this study is not effective in forming graphite oxide, mainly because the reactive metal elements will react first with reagents used for oxidation and graphite reduction, causing the oxidation and reduction reactions not to complete. It can be concluded that the purity of the graphite waste used is very important in increasing the oxidation and reduction reactions so that these can be done in perfectly, thus producing rGO.

Table 2 EDS data of graphite oxide

Element	Mass (%)
C	77.89
O	12.53
Au	9.57

4. CONCLUSION

In this study, graphite oxide, graphene oxide (GO), and reduced graphene oxide (rGO) were successfully synthesized from graphite waste. The chemical structures and morphology of graphite oxide, GO, and rGO were confirmed through FTIR, PSD, SEM-EDS, and XRD characterizations. The FTIR analysis confirmed the formation of GO. Although some restacking/unexfoliated graphite structures showed a diffraction peak at 2θ of 26.54°, the XRD analysis clearly exhibits a peak at 2θ of 20.04°, assigned to rGO after reduction of the GO. The XRD patterns of the graphite oxide show that the oxidation reaction was not perfect, because there was no diffraction peak at 2θ of 10.5°, which is peak of graphite oxide. Graphite oxide has a C/O ratio of 6.22 and an average particle size of 148.25 μm. The FTIR spectrum of the GO showed that there was a functional group C=C, which is an indication of rGO formation due to the principal bonding of the graphene structure. The SEM images of the rGO showed that the morphology seemed thick and layer stacking. Further research, with pre-purification treatment of graphite waste using acid and base leaching, is important to obtaining high purity graphite as a starting material to produce graphene.

5. ACKNOWLEDGEMENT

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