The Effect of Sonication Duration and Citric Acid Concentration on The Yield of Reduced Graphene Oxide (r-GO) Isolated from East Kalimantan Bituminous Coal

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ABSTRACT: Reduced graphene oxide (r-GO) is a derivative of graphene which has good electrical conductivity, so it can be used as a composite in the industrial sector. The manufacture of graphene oxide (GO) as the basic material for r-GO is carried out using green chemistry one of them is using the sonication technique and citric acid reductant. This research aims to isolated r-GO from East Kalimantan Bituminous coal which has 70,24% carbon content. The citric acid was varied on 10%, 30%, and 50% and the applies sonication duration was 10, 30, and 50 minutes. In order to obtain the optimal r-GO which was evaluated according to the lowest yield. The optimal condition took place at 10% citric acid under 50 minutes sonication. The r-GO was characterized using FTIR and it showed that the peaks of C-OH, C=C, and C-O-C exhibiting the high %T. The morphology of selected r-GO was closed to the references based on SEM analysis. There was an increase about 50,20% of carbon content in r-GO (84,412%) compared to the GO (56,199%). On the other hand, the percentage of oxygen decreased by 64,27% for GO (23,785%) to selected r-GO (8,498%). There was a change of C:O ratio, from 2,409 (GO) to 9,935 (r-GO). Its density and pH for r-GO were 1,4 g/cm^3 and 2.

KEYWORDS: r-GO, GO, Citric acid, Bituminous, Sonication

1. INTRODUCTION

Coal is a sedimentary rock derived from dead plant residues with the main constituent composition in the form of organic compounds that undergo a coalification process. In the process of coalification, cellulose will be converted into various categories of coal, namely lignite, sub-bituminous, bituminous, and anthracite [1]. The percentage of different type of coal in East Kalimantan are: low quality coal in the form of lignite with a distribution of 58.6%. Medium to high quality coal is subbituminous with 26.6%, bituminous with 14.4%, and anthracite with 0.4% [2]. One of the largest coal producers in Indonesia is East Kalimantan with 47.58 billion tons [3]. In coal, there are external impurities in the form of soil attached to the coal surface and internal impurities in the form of SiO_2, Al_2O_3, $CaSO_4$, $Al_2Si_2O_5(OH)_4$, $FeCO_3$, Fe_2O_3, FeS_2 and other metal oxides. In addition, coal generally has the highest silica content followed by alumina, sulphur dioxide, and other oxides. However, in Loa Janan Coal, iron/Fe (Fe oxide) content has the highest composition followed by silica. The iron content in Loa Janan Coal reaches twice the silica content and bituminous coal which has 70,24% carbon content [4]. Coal is generally used as a fuel or energy source in an industry. Coal is one of the natural sources of graphite with a degree of graphitization that can reach more than 80% [5]. Thus, coal can be used as a precursor or basic material for the manufacture of r-GO (reduced graphene oxide).

Graphene is a semiconducting carbon material that has a two-dimensional hexagonal structure consisting of thin layers of sp^2 carbon atoms connected through Van der Waals forces [6]. Graphene that has a carbon atom bonded with three carbon atoms will create a honeycomb lattice [7]. Graphene has strong electrical, thermal and mechanical conductivity so that it can be an innovation for the development of the electronics industry, aircraft, ultrasensitive sensors, thin-film transistors, and anti-corrosion, batteries, supercapacitors, and so on [8]. The manufacture of graphene can be done by various methods using graphite as a base material. Graphite has a multi-layered structure, while graphene has a structure consisting of one layer and is very different from the graphite structure. Graphite will be oxidized to graphite oxide which will then be converted to graphene oxide (GO) and reduced to graphene which has high selling value and wider utilization [5]. Graphene oxide has a sp^3 hybridized structure with a carbon layer formed from oxygen functional groups. Graphene oxide will dissolve in a number of organic solvents, so it can be used as a combination of polymer and ceramic materials because it has a high surface area [9]. Graphene has a derivative in the form of r-GO (reduced graphene oxide). r-GO is the result of deoxygenation of graphene oxide which has reduced oxide groups and has a conductivity of 8000 S/m [10].

There are several efforts that have been made in the isolation of graphene oxide from graphite in coal, namely methods such as the Brodie method, Hoffmann method, Staudenmaier method, and Hummers method which uses hazardous compounds [11]. The Brodie method is a method of making GO with graphite added with $KClO_3$ and reacted in fuming HNO_3 for 4 days at 60ºC. The Staudenmaier method is a modification of the Brodie method by replacing fuming with concentrated H_2SO_4 and adding $KClO_3$ for 4 days. The Hoffmann method is a method of making GO using graphite with the addition of HNO_3, H_2SO_4, and $KClO_3$. The Hummers method is a method of making GO by adding concentrated H_2SO_4 to graphite and adding $KMnO_4$ and $NaNO_3$. However, these methods have disadvantages such as requiring a long time process, using hazardous materials, and producing toxic gases such as NO_2 and N_2O_4. In addition, these methods produce residual Na^+ and NO^{3-} ions that are quite difficult to remove from wastewater [12]. However, the latest method that has been discovered and carried out by Mutrofin *et al.* graphene oxide can be produced by adjusting pH 4 on coal leached using 0.1 M HCl assisted by a mechanical method, namely sonication for 3 hours. This method has the advantage of being able to speed up the isolation time of graphene oxide as a precursor to graphene isolation, cost-effective, and simple [4]. Isolation of r-GO from GO can be produced from the reduction of oxide groups using organic acids such as citric acid [13]. Citric acid in the isolation of r-GO from coal serves to remove minerals and metal ions present in coal. The organic structure will change into a heterogeneous aromatic structure with the help of citric acid as a reductant. The increase in aromatic structure will result in better coal quality than before [14]. The presence of a high energy source in the form of sound waves and high frequency can break crystals into smaller sizes and have more active sides to react with the reductant to obtain isolated graphene. The sonication method and the use of citric acid reductant also use the principles of green chemistry with the aim of reducing hazardous waste generated by the use of solvents in the common r-GO manufacturing process.

This research aims to isolate r-GO from the isolated GO of East Kalimantan Bituminous coal using variations of citric acid concentration as a reductant and variations in sonication approach duration. The isolation of r-GO is expected to produce optimal yields with proper citric acid concentration and sonication duration, so that the resulting r-GO can be a potential candidate as a composite in the industrial world to increase the thermal conductivity value of a material. The resulting r-GO obtaining under the best citric acid concentration and sonication duration will be analyzed using Fourier Transform Infrared (FTIR) Spectrophotometer and Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray (EDX) to see changes in the oxide group content in the compound.

2. METHOD

2.1 Material and Tools

The materials used were bituminous coal from Anugerah Bara Kalimantan company, HCl p.a, NaOH p.a. Citric acid, n-hexane, toluene, and aquadest (Hydrobatt) which have analytical grade. 11 µm filter paper (Hawach Scientific).

Tools and instruments used were drop pipette (Onemed), 1 mL measuring pipette (Iwaki Pyrex), suction ball (Sicherheits-Pipettierball), glass stirrer (Pyrex), mortar stamper pestle (Onemed), 150 and 200 mesh sieve (Test Sieve), watch glass (Pyrex), digital balance, oven, glass funnel (Pyrex), desiccator (Duran), 250 mL and 500 mL beaker (Pyrex), 250 mL and 500 mL erlenmeyer (Pyrex), 100 mL measuring cup (Pyrex), 100 mL and 10 mL volumetric flask (Pyrex), stative and clamp (Trivi), porcelain cup (Pyrex), magnetic stirrer (Thermo Scientific Cimarec), hot plate (Thermo Scientific Cimarec), aluminum foil, plastic wrap, 1 mL pycnometer (Pyrex), pH indicator (MQuant), FTIR spectrophotometer Variation fts 1000, ultrasonic cleaning bath DELTA D68H, and SEM-EDX thermo scientific Phenom ProX.

2.2. Coal Preparation

The coal chunks were ground and pulverized using a mortar and pestle. Then, the coal that has been refined was sieved with 150 mesh and 200 mesh sievers. Coal with a size of 74.000 nm that has passed to 150 mesh siever and was retained on the 200 mesh siever was heated for 10 hours at 40ºC. The dried coal was then washed using aquadest with the ratio of coal and aquadest of 1:5. Stirring was carried out for 3 hours at room temperature at a speed of 1500 rpm. After that, filtering was carried out with 11 µm filter paper. The filtrate from the filtration was collected in an erlenmeyer and the filtered coal was re-heated for another 10 hours at 40ºC. The dried coal was cooled in a desiccator until cold and was ready to be used.

2.3. Isolation of Graphene Oxide (GO)

A total of 5,0 g of prepared coal was added to 250,0 mL demineralized water and 0.1 M HCl or 0.1 M NaOH was added until the solution reached pH 4. Next, the solution was sonicated for 3 hours. Then, the solution was filtered using 11 μm filter paper. The filtrate from the filtration was collected in an erlenmeyer and the filtration results were oven for 10 hours at 40°C. The dried coal was cooled in a desiccator until cold. The resulting graphene oxide (GO) was identified and characterized using FTIR spectrophotometer FTS 1000 variation, SEM-EDX, density, pH, and solubility in n-hexane, toluene, and aquadest solvents.

2.4. Isolation of Reduced Graphene Oxide (r-GO)

A total of 1,0 g of isolated GO was added to 50,0 mL of aquadest into an erlenmeyer and covered with plastic wrap. Next, the solution was sonicated for 2 hours. After sonication, the solution was added with various concentrations of citric acid, that is: 10%, 30%, and 50% in 10 mL of aquadest. The solution was sonicated again for 30 minutes and the solution was filtered using 11 μm filter paper. The filtrate from the filtration was collected in an erlenmeyer and the filtration results were put in the oven for 24 hours at 50°C. The dried r-GO was cooled in a desiccator until cold and the weight and yield calculations were carried out. The concentration variation was repeated twice for each citric acid concentration. The result of adding citric acid concentration that was the lowest yield would be used in the variation of sonication duration after the addition of citric acid. The lowest yield was chosen because citric acid would reduce impurities in GO, so that the molecular weight would decrease. Reduced graphene oxide with the lowest yield would also be characterized using an FTIR spectrophotometer of fts 1000 variation.

The isolated GO was weighed about 1,0 g and added 50,0 mL of aquadest into an erlenmeyer. Next, the solution was covered with plastic wrap and sonicated for 2 hours. After sonication, the solution was added with citric acid solution with optimal concentration. Then, the solution was sonicated again with variations in sonication duration of 10 minutes, 30 minutes, and 50 minutes. Afterwards, the solution was filtered using 11 μm filter paper. The filtrate from the filtration was collected in an erlenmeyer and the filtration results were baked for 24 hours at 50°C. The dried r-GO was cooled in a desiccator until cold and weighed and calculated its yield. The sonication duration variation was repeated twice for each sonication duration variation. The results of the sonication duration that was the lowest yield would be identified and characterized using FTIR spectrophotometer FTS 1000 variation, SEM-EDX, density, pH, and its solubility in n-hexane, toluene, and aquadest. The selection of the lowest yield was because citric acid would reduce impurities in GO with an optimal sonication time, so that the molecular weight would be reduced as well.

2.5. Characterization and Identification of GO and r-GO

The results of the study were characterized using FTIR spectrophotometer FTS 1000 variation, SEM-EDX, density, pH, and solubility in n-hexane, toluene, and aquadest. Fourier Transform Infrared (FTIR) spectrophotometer was used to identify the functional groups of GO and r-GO that could be analyzed such as carboxyl, epoxide, hydroxyl, and carbonyl groups [15,16]. Scanning electron microscopy (SEM)-energy dispersive X-ray (EDX) was a combination of two types of instruments, namely SEM and EDX. Scanning electron microscopy (SEM) is an instrument to determine the morphology or topology of the materials. The Energy dispersive X-ray (EDX) is an instrument to determine the elemental components or chemical composition of material [17]. Thus, SEM-EDX could be used in calculating the C:O ratio from GO to r-GO. In addition, for supporting data of r-GO isolation, characterization is carried out using pH indicator paper on GO and r-GO filtrates, density measurements on GO and r-GO, and solubility with n-hexane, toluene, and aquadest.

3. RESULTS AND DISCUSSION
3.1. Coal Preparation

Coal that has been pulverized with a size of 74.000 nm aims to expand the surface or active side in the coal. The more active sides number of coal, the more GO will be produced. After washing and stirring the coal, the resulting filtrate was black which indicated that there was coal passed the filtration. This indicated that stirring for 3 hours using aquadest produced a coal size that was smaller than the pore of filter paper (11 μm). The coal retained on the filter paper was heated for 10 hours at 40°C. The use of low temperature aimed to reduce organic volatile impurities in the coal without damaging the main carbon structure of the coal. Thus, in coal preparation, more active side of coal would be obtained with concerning to minimize impurity content. Coal preparation would result in an increased carbon content value and a decrease in the content of oxygen and other impurity minerals [4].

3.2. Isolation of Graphene Oxide (GO)

Graphen oxide isolation was carried out by adding aquadest to the prepared coal and adjusted using 0,1 M HCl to obtain pH of 4. Chloric acid solution was an oxidizing agent that would bind metal oxides. Graphene oxide isolation was obtained without going through the stages of making graphite from coal. This was in accordance Mutrofin *et al.* who have successfully isolated GO from coal using a mechanical method, namely sonication for 3 hours [4]. Thus, this method had the advantages of being cost-effective, simple, and can speed up the isolation time of GO as an r-GO precursor. After sonication, the resulting filtrate was colorless and had a pH value of 5. This indicated that the impurities or oxide groups present in the coal after washing had been successfully reduced by the sonication method which could facilitate the release of hydrogen bonds into H_2O and carbocations. The GO retained on the filter paper was oven for 10 hours at 40°C. The used of this temperature aimed to reducing the organic volatile impurities in the coal and the water content in the coal from the washing process would disappear slowly without damaging the carbon owned by the coal. Thus, in coal preparation, more active side of coal would be obtained with reduced impurities.

3.3. Isolation of Reduced Graphene Oxide (r-GO)

The isolation of r-GO was done by adding aquadest into GO and sonicated for 2 hours. The sonication method was done in order to break/cut the Van der Waals forces between layers so as to produced r-GO sheets that can improve electronic, mechanical, thermal, and chemical properties [18]. This method can also break metal ions in a molecule, so that carbon bonds with oxide groups could be released and form complex bonds with citric acid [19]. After sonication, the solution was added with varying concentrations of citric acid, 10%, 30%, and 50%. Then, the solution was sonicated again for 30 minutes and the solution was filtered using 11 µm filter paper. The resulting filtrate was yellow and had a pH value of 2. This proves that citric acid had successfully reduced the oxide groups found on GO. Citric acid would oxidize and form oxidation compounds so that it would produce a yellow color in the filtrate. After filtering, r-GO would be dried in the oven for 24 hours at 50°C and the yield calculation was carried out. The selection of the lowest yield was because citric acid would reduce impurities in GO, so that the molecular weight would decrease. The yield percentage of the influence of citric acid concentrations and sonication duration are in **Table 1** and **Table 2**, respectively.

Table 1. Yield of Citric Acid Concentration Variation

Concentration Variation			
Repetition	**10%**	**30%**	**50%**
1	89,66	102,38	117,35
2	89,64	103,69	117,75
Average	89,65	103,035	117,55

Table 2. Yield of Sonication Duration Variations

Variation of Sonication Time			
Repetition	**10%**	**30%**	**50%**
1	87,89	89,66	90,38
2	88,11	89,64	90,18
Average	88	89,65	90,28

The lowest yield produced in citric acid variations was at a concentration of 10%, with an average of 89.65%. The yield values that exceed 100% were at 30% and 50% citric acid concentrations. This could be caused due to citric acid concentrations that were too saturated, thus making the oxide groups on GO will re-bond and make citric acid trapped in the GO layer so that the final GO was greater than the initial GO weight. In addition, a t-test was conducted to see whether there was a difference between the three concentration variations. From the calculation results, it could be concluded that the t-count was smaller than the t-table, so there was no real difference between the three variations. Thus, a concentration of 10% citric acid was chosen to be used in the preparation of r-GO with varying sonication duration.

Isolation of r-GO with variations in sonication duration was carried out by adding aquadest into GO and sonication for 2 hours. After that, citric acid was added with the lowest yield, 10% citric acid concentration and continued with sonication again with variations in sonication duration of 10 minutes, 30 minutes, and 50 minutes. The lowest yield of r-GO produced in the sonication duration variation was 10 minutes with an average of 88%. The sonication duration of 30 minutes and 50 minutes had a higher yield value. In addition, a t-test was conducted to see whether there was a difference between the three variations of sonication duration.

From the calculation, it can be concluded that the t-count was smaller than the t-table, so there is no real difference between the three variations. Thus, the r-GO isolated under a sonication duration of 10 minutes and citric acid concentration of 10 % was chosen for further characterization.

3.4. Characterization and Identification
3.4.1. Density Indentification

Density is a measurement of mass per unit volume with standard units of measurement such as kg/m^3 or g/cc. The higher the density or density of a compound, the greater the mass per volume [20,21]. Density functions in determining a compound. Each compound would has a different density as its finger print. The difference in the density value of the compound could be caused by the constituent components and the empty gaps between the constituent compounds [22]. Density measurements of GO and r-GO isolation results were carried out using a 1 mL pycnometer according to ASTM standards D70-03. From the density measurement results, it was found that the density of GO isolation was 1.1 g/cm^3 and r-GO had a density of 1.4 g/cm^3. This has similarities with Torrisi et al. which states that GO has a density of 0.981 g/mL and r-GO has a density of 1.5-1.9 g/cm^3 [23]. The formula for measuring the density of solids is as follows:

$$\rho = \frac{W_2 - W_1}{(W_4 - W_1) - (W_3 - W_2)}$$

Description:

W1: Empty pycnometer

W2: Pycnometer + solids

W3: Pycnometer + solids + water

W4: Pycnometer + water

3.4.2. Characterization Using Solubility

Solubility analysis on GO and r-GO isolation results was carried out using toluene, n-hexane, and aquadest. In aquadest GO showed solubility which indicated the presence of carbonyl and carboxyl groups, thus causing GO to be hydrophilic, which was easily soluble in water, producing a brown solution. In r-GO with the addition of aquadest showed insoluble results because r-GO had hydrophobic properties that was hard to miscible in water. Furthermore, in toluene and n-hexane, GO was insoluble and r-GO was partially soluble [24]. The isolated GO that was difficult to dissolve in organic solvents such as n-hexane and toluene was because GO had strong hydrogen bonds between GO layers. All data related to GO and r-GO solubility are in **Figure 1**.

Solvents	GO	r-GO
Toluene	Insoluble	Partially Soluble
N-Hexane	Insoluble	Partially Soluble
Aquademineralization	Soluble	Insoluble

Figure 1. Solubility Spot Test of GO and r-GO

3.4.3. Characterization Using FTIR

Fourier transform infrared (FTIR) spectrophotometer analysis aimed to predict the type of bonding functional groups contained in a compound. The functional groups of GO and r-GO that could be analyzed are carboxyl, epoxide, hydroxyl, and carbonyl groups. On GO, the FTIR isolation results could interfere with oxygen-containing functional groups in sp^2 hybridization. The spectra of GO and selected r-GO is in **Figure 2**. The infrared spectra of GO isolation resulted in peaks at 1607 cm^{-1} and 1212 cm^{-1} due to the presence of a large number of aromatic C = C and epoxide groups with %T of 55.98% and 54.45%. Peak At 3696 cm^{-1} showed the O-H stretch in water with a transmittance of 96.13%. Peak at 3264 cm^{-1} showed the O-H group of alcohol with a transmittance of 75.14%. Peaks at 2919 cm^{-1} and 1434 cm^{-1} showed the presence of C-H alkyl group and C-OH vibrations with %T of 77.18% and 62.6%. In addition, at 469 cm^{-1} showed mineral bonds with oxygen (Me-O) with a %T of 10%.

In the FTIR spectra of r-GO isolation resulted from isolations in sonication duration of 10 minutes, 30 minutes, and 50 minutes, the spectrum at 3600-3200 cm^{-1} increased in %T because the O-H groups in water and alcohol decreased due to deoxygenation with citric acid and sonication. Peaks at 2950-2850 cm^{-1} and 1600 cm^{-1} also increased which confirmed the presence of C-H alkyl and C=C aromatic bonds that remained before and after oxygen reduction using citric acid. Peaks at 1400 cm^{-1} and 1200 cm^{-1} showed an increase in %T indicating a decrease in C-OH vibrations and epoxy groups indicating successful deoxygenation with citric acid. In addition, at 460 cm^{-1} there is also an increase in %T which indicates that the oxygen reduction process using citric acid and sonication can reduce the mineral content in GO by forming mineral complexes with citric acid such as Si-O-Si. Thus, the highest %T of FTIR results is owned by a sonication time of 50 minutes with a concentration of 10% citric acid which indicates that this sonication time was optimal in producing r-GO which had the least oxide groups. The predicted reaction mechanism is in **Figure 3.**
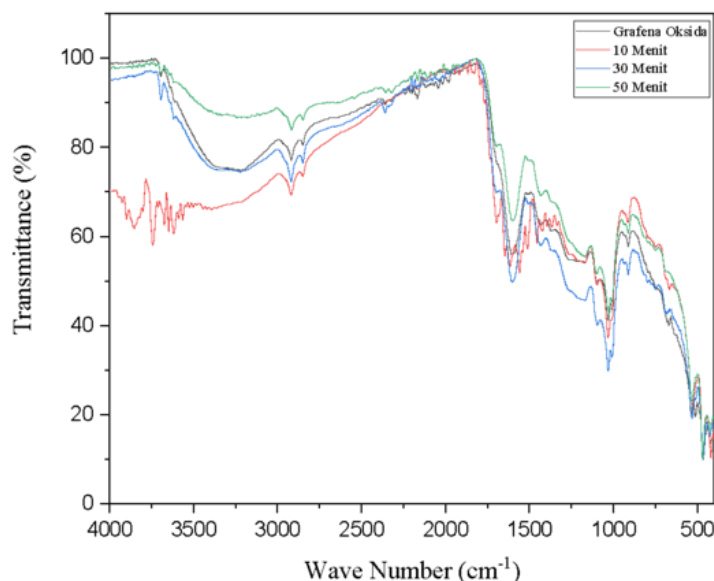

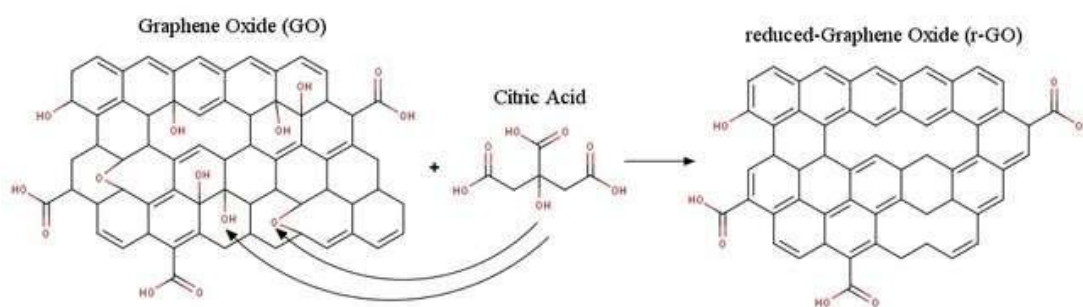**Figure 2.** Infrared Results of GO and r-GO Isolation under Sonication Duration

Figure 3. Predicted Reaction Mechanism between GO and Citric Acid

3.4.4. Characterization Using SEM-EDX

Analysis using scanning electron microscopy (SEM) with energy dispersive X-ray (EDX) is to determine the morphology or topology of GO/r-GO and the composition or chemical components. Scanning electron microscopy analysis conducted on GO shows a thick and wrinkled surface with elemental composition near the surface showing a high percentage of carbon (C) and oxygen (O). This indicated the presence of functional groups and the presence of weak Van der Wals bonds visible under the microscope with added wrinkles [25]. The image of SEM in **Figure 4** (a-b) showed that the morphology of GO was in the form of indentation or fold lines. This indicated that the GO surface was very thin and folded due to the thermodynamic instability of the 2D material which forms folds to reduce it was energy (Fauzi et al., 2021). The indentations were also seen in the morphology of r-GO but the surface of r-GO showed more wrinkled indentations. This indicated that the r-GO sheet was thinner than GO so that the r-GO surface was easier to roll. The wrinkles and indentations were caused by the reduction of oxide groups on the GO surface [26]. In addition, the morphological differences between GO and r-GO could be seen in the pores formed. There was no pore formation in GO, while in r-GO pores with a diameter of 659.52-1060 nm were formed. The used of citric acid with sonication had succeeded in removing oxygen groups on the GO surface, resulting in the formation of layers and a decrease in the distance between GO layers that would be seen in EDX analysis.
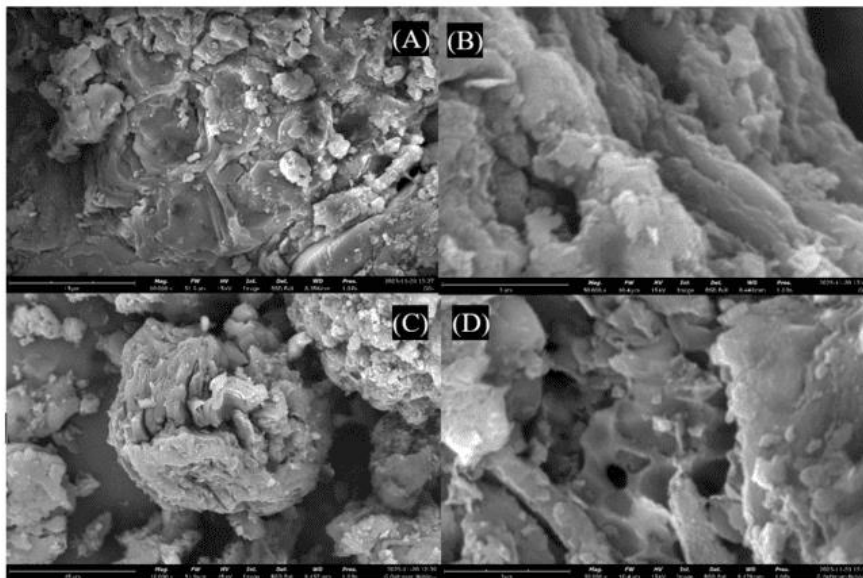

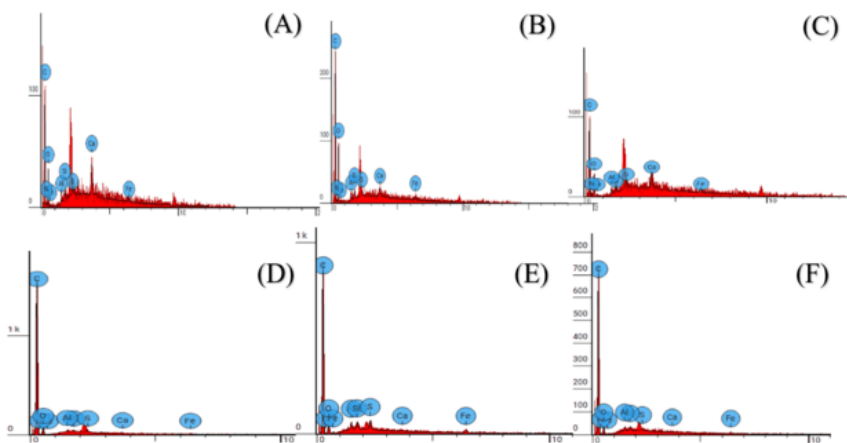**Figure 4.** SEM results for GO (A-B) and r-GO (C-D)

Figure 5. EDX spectra for GO (A-C) and r-GO (D-F)

The difference between the EDX results of GO and r-GO lies in the oxygen element which was less than GO. This indicated that GO had been successfully reduced to r-GO by reducing the oxygen-containing functional groups. The chemical composition of EDX analysis can be used to determine the ratio of carbon and oxygen (C:O) in GO and r-GO. From the calculation, the C:O ratio analyzed is 2.409 and r-GO had increased by 9.935. In addition, there was an increase in carbon content of about 50.20% in r-GO (84.412%) while GO (56.199%). On the other hand, oxygen decreased by 64.27% for GO (23.785%) to r-GO (8.498%). This indicated that the used of citric acid with sonication succeeded in reducing oxide groups (-OH, -O-, -C=O, and -COOH) and increasing the carbon number of GO. The results were comparable to those reported by Tran *at al*. in an effort to reduce GO using ascorbic acid which states an increase in GO (1.60) to r-GO (4.03) [27]. Thus, the used of citric acid reduction using sonication could effectively remove oxide groups on the GO surface significantly. All data extracted from EDX analysis is in **Table 3** and **Table 4**.

Table 3. Carbon-Oxygen Atomic Concentration Ratio (C:O) of GO

GO				
Element	Point 1 (%)	Point 2 (%)	Point 3 (%)	Average C:O
C	53,552	56,806	58,241	
O	23,318	27,911	20,125	
C:O	2,297	2,035	2,894	2,409

Table 4. Carbon-Oxygen Atomic Concentration Ratio (C:O) of r-GO

r-GO				
Element	Point 1 (%)	Point 2 (%)	Point 3 (%)	Average C:O
C	85,603	84,856	82,777	
O	8,631	8,605	8,257	
C:O	9,918	9,861	10,025	9,935

4. CONCLUSION

It can be concluded that the isolation of r-GO from GO bituminous coal of East Kalimantan was successfully carried out the GO and r-GO are 1.1 g/cm^3 and 1.4 g/cm^3. The filtrate in the manufacture of GO produces a pH of 5 and r-GO of 2. GO isolation shows solubility in aquadest and is insoluble in toluene and n-hexane. Meanwhile, the isolated r-GO is partially soluble in toluene and n-hexane and is insoluble in aquadest. The results of the FTIR spectra of GO and r-GO experienced an increase in %T which indicates the success of the deoxygenation process in the O-H water, O-H alcohol, C-H alkyl, C=C aromatic, C-OH, C-O-C and Me-O functional groups. In addition, the analysis using SEM shows that there are changes between GO and r-GO which the r-GO has larger pores and more layers than the GO one. The EDX analysis reveals that the C:O ratio of r-GO is higher than the C:O ratio of GO, that is 9.935 (r-GO) and 2.409 (GO). Thus, the isolation of r-GO from GO coal has been successfully carried out using harmless materials and can potentially become a potential candidate as a composite in the industrial world to increase the thermal conductivity value of a material.

REFERENCES

1. Putri, R. I., Situntun, Y. B., Rindawati, P. I., & Anjarwati, R. (2022). Geologi dan Pemodelan Geometri Ketebalan SertaSebaran Batubara Pada Daerah Purwajaya Kecamatan Loa Janan Kalimantan Timur. *Jurnal Teknologi Mineral FT UNMUL*, *10*(2), 38–49. https://doi.org/http://dx.doi.org/10.30872/jtm.v10i2.9724
2. Yustanti, E. (2012). Pencampuran Batubara Coking dengan Batubara Lignite Hasil Karbonisasi sebagai Bahan Pembuatan Kokas. *Jurnal Teknologi Pengolahan Limbah*, *15*(1).

3. Haryadi, H., & Suciyanti, M. (2018). Analisis Perkiraan Kebutuhan Batubara untuk Industri Domestik Tahun 2020-2035 dalam Mendukung Kebijakan Domestic Market Obligation dan Kebijakan Energi Nasional. *Jurnal Teknologi Mineral dan Batubara*, *14*(1), 59. https://doi.org/10.30556/jtmb.vol14.no1.2018.192

4. Mutrofin, S., Wardhani, S., & Khunur, M. M. (2021). *Preparasi Nanopartikel Batubara Loa Janan Kalimantan Timur Hasil Pencucian dengan Teknik Sonikasi*. Malang.

5. Hou, D., Li, K., Ma, R., & Liu, Q. (2020). Influence of order degree of coaly graphite on its structure change during preparation of graphene oxide. *Journal of Materiomics*, *6*(3), 628–641. https://doi.org/10.1016/j.jmat.2020.04.009

6. Khan, M., Tahir, M. N., Adil, S. F., Khan, H. U., Siddiqui, M. R. H., Al-Warthan, A. A., & Tremel, W. (2015). Graphene Based Metal and Metal Oxide Nanocomposites: Synthesis, Properties and Their Applications. *Journal of Materials Chemistry A*, *3*(37), 18753–18808. https://doi.org/10.1039/c5ta02240a

7. Jiříčková, A., Jankovský, O., Sofer, Z., & Sedmidubský, D. (2022). Synthesis and Applications of Graphene Oxide. *Materials*, *15*(3), 920. https://doi.org/10.3390/ma15030920

8. Tiwari, S. K., Sahoo, S., Wang, N., & Huczko, A. (2020). Graphene Research and Their Outputs: Status and Prospect. *Journal of Science: Advanced Materials and Devices*, *5*(1), 10–29. https://doi.org/10.1016/j.jsamd.2020.01.006

9. Kigozi, M., Koech, R. K., Kingsley, O., Ojeaga, I., Tebandeke, E., Kasozi, G. N., & Onwualu, A. P. (2020). Synthesis and Characterization of Graphene Oxide from Locally Mined Graphite Flakes and Its Supercapacitor Applications. *Results in Materials*, *7*. https://doi.org/10.1016/j.rinma.2020.100113

10. Kusumattaqiin, F., Ramli, R., Kurnyawaty, N., Halik, A., & Hira, T. (2020). ANALISA STRUKTUR OKSIDA GRAFENA TEREDUKSI. *Dalton : Jurnal Pendidikan Kimia dan Ilmu Kimia*, *3*(1). https://doi.org/10.31602/dl.v3i1.3107

11. Olabi, A. G., Abdelkareem, M. A., Wilberforce, T., & Sayed, E. T. (2021, January 1). Application Of Graphene In Energy Storage Device. *Renewable and Sustainable Energy Reviews*. Elsevier Ltd. https://doi.org/10.1016/j.rser.2020.110026

12. Chen, J., Yao, B., Li, C., & Shi, G. (2013). An improved Hummers method for eco-friendly synthesis of graphene oxide. *Carbon*, *64*, 225–229. https://doi.org/10.1016/j.carbon.2013.07.055

13. Andrijanto, E., Shoelarta, S., Subiyanto, G., & Rifki, S. (2016). Facile synthesis of graphene from graphite using ascorbic acid as reducing agent (p. 020003). https://doi.org/10.1063/1.4945457

14. Manoj, B., & Narayanan, P. (2013). Study of Changes to the Organic Functional Groups of a High Volatile Bituminous Coal during Organic Acid Treatment Process by FTIR Spectroscopy. *Journal of Minerals and Materials Characterization and Engineering*, *01*(02), 39–43. https://doi.org/10.4236/jmmce.2013.12008

15. Rasyida, K., Kuswandi, B., & Kristiningrum, N. (2014). Deteksi Kemurnian Air Zamzam Menggunakan Metode Spektrofotometri Fourier Transform Infrared (FTIR) dan Kemometrik (Detection of Zamzam Water Purity Using Fourier Transform Infrared (FTIR) Spectroscopy Technique and Chemometrics). *E-Jurna Pustaka Kesehatan*, *2*(2), 320–326.

16. Sanjiwani, N. M. S., Paramitha, D. A. I., Wibawa, A. A. C., Ariawan, I. M. D., Megawati, F., Dewi, N. W. T., … Sudiarsa, I. W. (2020). Pembuatan Hair Tonic Berbahan Dasar Lidah Buaya dan Analisis dengan Fourier Transform Infrared. *Journal IKIP PGRI Bali*, *21*(1), 249–262. https://doi.org/10.5281/zenodo.3756902

17. Saeedkia, Daryoosh. (2016). *Materials Characterization Using Nondestructive Evaluation (NDE) Methods*. Elsevier. https://doi.org/10.1016/C2014-0-00661-2

18. Noroozi, M., Zakaria, A., Radiman, S., & Abdul Wahab, Z. (2016). Environmental Synthesis of Few Layers Graphene Sheets Using Ultrasonic Exfoliation with Enhanced Electrical and Thermal Properties. *PLOS ONE*, *11*(4), e0152699. https://doi.org/10.1371/journal.pone.0152699

19. Firnando, H. G., & Astuti. (2015). Pengaruh Suhu Pada Proses Sonikasi Terhadap Morfologi Partikel dan Kristalinitas Nanopartikel Fe3O4. *Jurnal Fisika Unand*, *4*(1). https://doi.org/10.25077/jfu.4.1.%25p.2015

20. Abdurrojaq, N., Devitasari, R. D., Aisyah, L., Nur, Faturrahman, Bahtiar, S., … Maymuchar. (2021). Perbandingan Uji Densitas Menggunakan Metode ASTM D1298 dengan ASTM D4052 pada Biodiesel Berbasis Kelapa Sawit. *Lembaran Publikasi Minyak dan Gas Bumi*, *55*(1), 47–55. Retrieved from http://www.journal.lemigas.esdm.go.id

21. Sitindaon, D., & Harahap, M. H. (2014). Pengaruh Penambahan Styrofoam Pada Pembuatan Beton Ringan Menggunakan Pasir Merah Labuhan Batu Selatan. *Jurnal Einstein*, *2*(3), 14–19. Retrieved from http://jurnal.unimed.ac.id/2012/index.php/einstein

22. Fatimura, M., Daryanti, & Santi. (2016). Pembuatan Biodiesel dari Minyak Jelantah Bekas Rumah Makan dengan Variasi Penambahan Katalis KOH pada Proses Transesterifikasi. *Jurnal Redoks*, *1*(2), 35–44.

23. Torrisi, L., Cutroneo, M., Torrisi, A., & Silipigni, L. (2022). Measurements on Five Characterizing Properties of Graphene Oxide and Reduced Graphene Oxide Foils. *physica status solidi (a)*, *219*(6). https://doi.org/10.1002/pssa.202100628

24. Konios, D., Stylianakis, M. M., Stratakis, E., & Kymakis, E. (2014). Dispersion behaviour of graphene oxide and reduced graphene oxide. *Journal of Colloid and Interface Science*, *430*, 108–112. https://doi.org/10.1016/j.jcis.2014.05.033

25. Devi, S. C., & Khan, R. A. (2020). Effect of Graphene Oxide on Mechanical and Durability Performance of Concrete. *Journal of Building Engineering*, *27*. https://doi.org/10.1016/j.jobe.2019.101007

26. Fauzi, F., Sunu, D. W., & Dwandaru, B. (2021). Analisis Karakteristik Graphene Oxide dan Reduksinya melalui Gelombang Mikro. *Jurnal Fisika*, *11*(1), 9–18. https://doi.org/https://doi.org/10.15294/jf.v11i1.28136

27. Tran, D. T., & Nguyen, V. N. (2020). rGO/persulfate metal-free catalytic system for the degradation of tetracycline: effect of reaction parameters. *Materials Research Express*, *7*(7), 075501. https://doi.org/10.1088/2053-1591/ab9e47