

Analysis of the Effect of Temperature and Time for Ultrasonication on Graphite Structure

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Abstract— Graphite is a carbon allotrope with special bonding characteristics. These bonds can be altered given ultrasonic waves to be another form of bond or loosened. Changes in bonds will cause changes in bond characteristics to other forms of carbon allotropes. By varying ultrasonication process (time and temperature), the change in bonding or functional groups could be analyzed by changes in the structure of carbon. The ultrasonication process resulted in the bonding of oxygen atoms, produced bonding of C atoms with O, either as single or double bond. The new bond formed was linearly proportional to change in ultrasonication time and temperature. C bond with O caused carbon to change into allotrope of graphite oxide carbon, this was supported by the results of Raman spectroscopy characterization, where the defect of carbon was increased compared to carbon without ultrasonication. The intensity of the defect increased from 60 a.u to 265 a.u.

Keywords— Graphite, Ultrasonication, Allotrope

I. INTRODUCTION

The existence of graphite in nature is very abundant, in addition to its ease of synthesis from waste material (biomass). As carbon allotrope, graphite has hexagonal and rhombohedral structures. Graphite with hexagonal structure is more stable than rhombohedral [1]. Graphite structure can be transformed into another carbon allotrope by changing carbon bonds through several methods. The ultrasonication process can be used to change bonds in carbon, through exfoliation due to ultrasonic waves applied to graphite. Graphite with regular hexagonal structure can be turned into irregular structure, with identification of defects that occur through Raman spectroscopic characterization. The presence of defects can be identified by the changes in D-band and G-band of Raman characterization results, while for the functional groups are identified by Fourier Transform Infrared (FTIR).

A changing structure does not mean that carbon is damaged or cannot be used, but a defect in the structure will cause a change in the type of carbon to another carbon allotrope with different characteristics. Carbon with different bonding structures can be used in various purposes according to the characteristics of the carbon allotrope formed. One of the carbon allotropes obtained from ultrasonication is graphene. Graphene produced through the ultrasonication process is derived from graphite ultrasonicated with

a frequency of 40 kHz, for 2 hours [2]. Graphite structure consists of a series of overlapping parallel plane layers, the circle showing the position of the carbon atom does not represent the size of the atom. However, the atom is actually connected with other atoms. The fields arranged with hexagonal structure in graphite can be peeled off per layer or broken bonds so that the regular structure of graphite can change.

Structural irregularities in graphite allotropes are used in various purposes, including as sensor materials (gas, solution, air and others). The existence of "defects" in graphite, making graphite easier to bind or interact with other materials. The formation of graphite into other carbon allotropes can be done by various methods, including the Hummers method, the electrolysis method, the chemical and physical reaction method, the Ultrasonication method and others. Ultrasonic method, a method that uses longitudinal mechanical waves that cannot be heard by the human ear because of the high frequency they have, is capable of propagating in solid, liquid, and gas mediums [3]. Ultrasonic is an acoustic wave that has a frequency greater than 16-20 kHz [4]. The development of extraction process through ultrasonic methods continues to be done in order to get a shorter time and better results [5]. Graphite processing performed using ultrasonication process results in the formation of GO (Graphite Oxide) which then turns into Graphene Sheets depending on the process and composition given. Graphite Oxide (GO) is a single layer atomic material consisting of carbon, hydrogen, and oxygen molecules by oxidation of graphite crystals. In a previous study, GO was found to be an excellent membrane material, as it did not pass anything but water vapor and block ethanol vapors and other alcohol molecules [6]. Sheets of GO could also be used for gas separation applications [7]. GO itself is derived from the oxidation of graphite, then graphene oxide will form from GO sheets which, when in water will be exfoliated, will then leave the graphene layer after the oxygen concentration in graphene oxide can be reduced until it runs out [8].

II. MATERIALS AND METHODS

A. Materials

The basic material of carbon/graphite used was graphite with purity of 99.7% and particle size of 325 mesh (44 micrometers).

B. Methods

1. Preparation

As much as 5 g graphite was dissolved in 200 ml of distilled water, then stirred with magnetic stirrer for 30 minutes. The resulting solution was then ultrasonicated with variations in temperature and time.

2. Ultrasonication Process

Ultrasonication was performed with time and temperature variation. In this study, variation of time used was 1-3 hours [9], while temperature was 40°C, 50°C, and 60°C [10]. After the ultrasonication process was completed, the next process was to separate the solvent and the solute from the material overnight to separate the impurity, where the impurities would float above and the graphite powder settled below. Filtering was then performed and heating process was carried out for 21 hours or until material was completely dry.

3. Characterization and analysis

Dried graphite powder was crushed so as not to clot before being characterized using FTIR test (Thermo-Scientific FT-IR spectrometer) at wave number of 400-4000 cm⁻¹, with the aim to determine bonding and functional groups. Raman characterization was performed to identify the level of defect in graphite.

III. RESULTS AND DISCUSSION

Characterization using FTIR in the form of peaks used to identify functional groups and bonds contained in the graphite. The characterization was carried out on the basis of graphite and ultrasonicated graphite with time variation of 1-3 hours and temperature of 40°, 50° and 60°C. Graphite was characterized by O-H functional groups located at wave numbers 3670-3629 cm⁻¹ with stretching vibration bonding types. For the wave number 3425 cm⁻¹ there was a O-H stretching vibration, where the wave number was very close to the peak of 3428 cm⁻¹, in where an absorption zone appeared from OH anti-symmetry and stretch in free air [11].

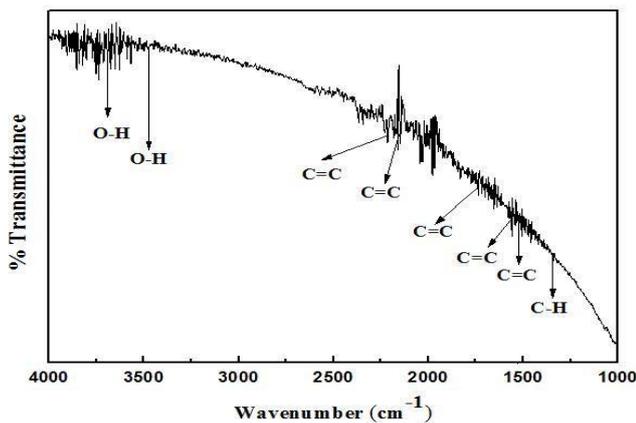


Figure 1. Results of FTIR characterization from Graphite

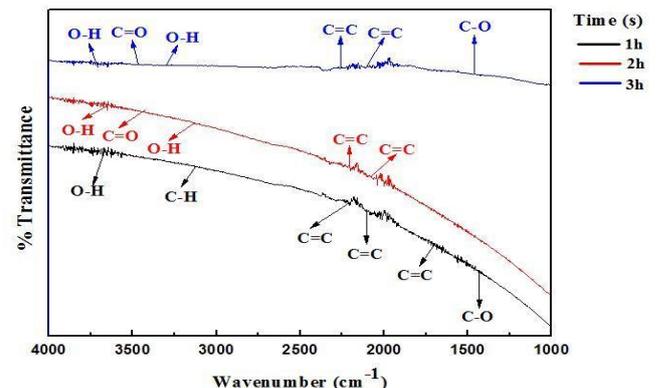
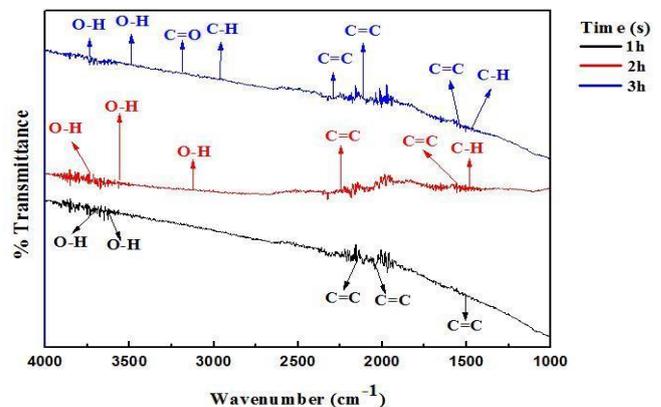
Figure 1 also shows a stretching vibration C = C in wave number of 2249-2180 cm⁻¹, where a peak detected got stretched and caused change of waveform. At wave number 2137-2106 cm⁻¹, a stretching vibration C=C was found, where a peak was detected and gave weak waveform. Wave number 1624 - 1541 cm⁻¹ had aromatic vibration stretching C = C where the peak detected tended to be medium and formed due to sp² hybridization, which was stretching vibration in a hexagonal graphite carbon ring [11]. Wave number 1557-1534 cm⁻¹ had aromatic stretching vibration C = C where the peak detected was quite strong. In the wave number 1522-1498 cm⁻¹, there was an

aromatic vibration stretching C = C, in which peak detected had irregular stretch affecting waveform to change. For wave number, 1420 cm⁻¹, there was C-H rocking vibration where the peak detected tended to be weak.

TABLE I. IDENTIFICATION WAVE NUMBER AND FUNCTIONAL GROUPS OF GRAPHITE

Wave Number (cm-1)	Functional Groups
3670–3629	vibration stretching O – H
3425	vibration stretching O – H
2249 – 2180	vibration stretching C = C
2137 – 2106	vibration stretching C = C
1624 – 1541	vibration stretching C = C
1577 – 1534	vibration aromatic stretching C = C
1522 – 1498	vibration aromatic stretching C = C
1420	vibration rocking C – H

When compared to graphite without ultrasonication, functional groups arose due to ultrasonication process were in the form of C-O and C = O, which identified the vibration stretching oxide of graphite, both single and double bonds. Functional groups in the form of oxides could be expressed as structural changes in graphite or the appearance of defects due to ultrasonication of graphite. Identification of defects in graphite bonds could also be identified from the characterization of Raman spectroscopy, where defects were expressed from the D (I_D) intensity of the Raman results. The higher the intensity of the defect, it could mean that the number of defects from graphite increased. Changes in intensity of defects were quite significant, from 60 a.u to 265 a.u. But when I_D was compared to I_G (the intensity of graphite), there had not been a change in the overall structure of graphite itself.



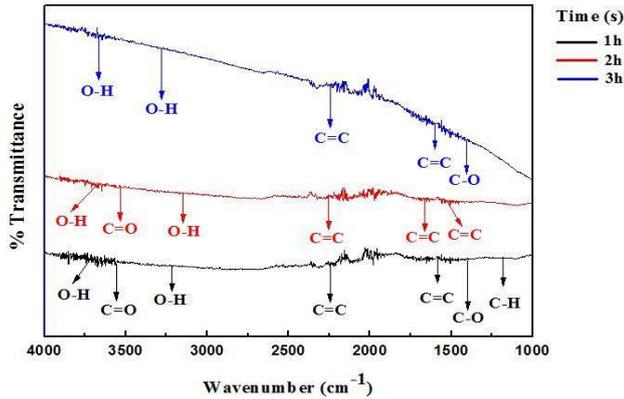


Figure 2. Comparison of FTIR results on graphite by ultrasonication at temperatures of (a) 40° (b) 50° and (c) 60°C in 1, 2 and 3 hours

The results of Raman spectroscopy could explain that ultrasonication of graphite increased the defect intensity of graphite. Increased defects due to graphite vibrations due to waves given during the ultrasonication process [12], while the peak G (IG), arose due to the vibration stretching process on bonds or graphite atomic structures previously arranged [2].

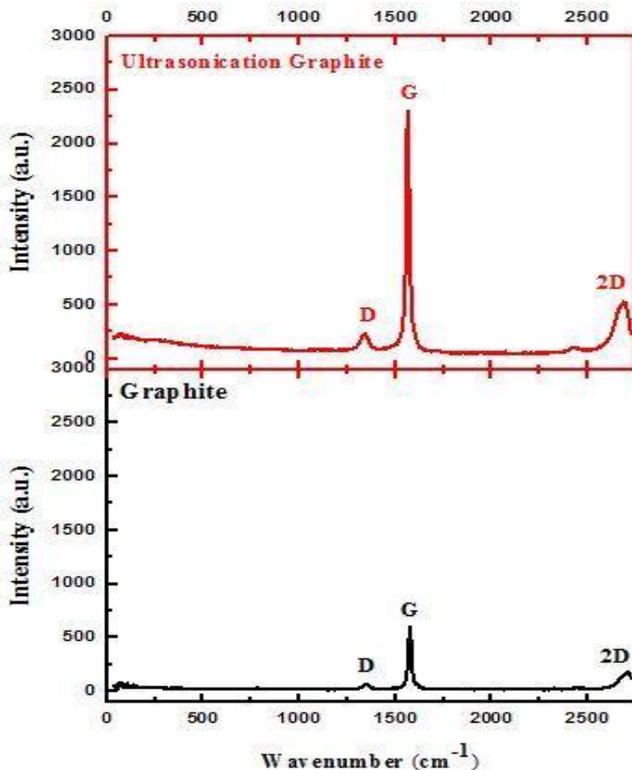


Figure 3. Result of Raman spectroscopy from both graphite and ultrasonicated graphite (60°C, 3 hours).

IV. CONCLUSION

The ultrasonication process could cause changes in functional groups in graphite and increase defects, but had not changed the structure of graphite into other carbon allotropes, only resulting in the emergence of new groups of graphite functions; C-O and C=O, when performed in higher temperature could turn into graphite oxide.

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