STUDY ON MORPHOLOGY AND ELECTRICAL PROPERTIES OF GRAPHENE OXIDE DECORATED WITH NICKEL OXIDE FOR ELECTROCHEMICAL SENSOR

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ABSTRACT

In this work, graphene oxide (GO) and graphene oxide decorated with nickel oxide (GO-NiO) were synthesized by using modified Hummer's method and hydrothermal method respectively. The synthesized nanoparticles were subjected to morphology and electrical testing. The EDS data shows oxygen, carbon and nickel elements present in the synthesized product. The FESEM images of GO shows the nanoparticles is in flakes form whereby the FESEM images for graphene oxide decorated with nickel oxide in oval-flakes form. Impedance Spectroscopy (IS) were used to study an electrical properties of synthesized product. Later, the result were discussed using Nyquist graph to confirm the conductivity of the GO and GO-NiO with presence of semicircle and 45° straight line. Since GO-NiO consists elements of GO and NiO, it has the ability to conduct an electricity. As a conclusion, it was observed that there is significant different in the reading of conductivity with an addition of NiO into GO.

Keywords: Graphene oxide, graphene oxide-nickel oxide, Hummer's method, hydrothermal method.

KAJIAN MORPHOLOGI DAN SIFAT ELEKTRIK GRAPHENE DIHIASI DENGAN NIKEL OKSIDA UNTUK SENSOR ELEKTRIK

ABSTRAK

Dalam kerja ini, graphene oksida (GO) dan graphene oksida yang dihiasi dengan nikel oksida (GO-NiO) telah disintesis dengan menggunakan kaedah Hummer yang telah diubahsuai dan kaedah hidrotermal. Nanopartikel yang disintesis telah tertakluk kepada morfologi dan ujian elektrik. Data EDS menunjukkan unsur oksigen, karbon dan nikel yang terdapat dalam produk yang disintesis. Imej FESEM untuk GO telah menunjukkan bahawa nanopartikel GO berada dalam bentuk serpihan dan bagi imej FESEM untuk graphene oxide dihiasi dengan nikel oksida dalam bentuk bujur-serpih. Spectroscopi Impedans (IS) digunakan untuk mengkaji sifat elektrik produk sintetik. Kemudian, hasilnya dibincangkan menggunakan grafik Nyquist untuk mengesahkan kekonduksian GO dan GO-NiO dengan kehadiran separuh bulatan dan garis lurus 45 °. Oleh kerana GO-NiO terdiri daripada unsur-unsur GO dan NiO, ia mempunyai keupayaan untuk menjalankan elektrik. Kesimpulannya, di dalam kajian ini didapati terdapat perbezaan yang signifikan dalam pembacaan kekonduksian dengan penambahan NiO ke dalam GO.

Keywords: Graphene oksida, graphene oksida-nickel oksida, kaedah Hummer, kaedah hidrotermal.

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A-GO	:	Acetone graphene oxide
C=O	:	Carbon monoxide double bond
cm	:	Centimeter
-COOH	:	Carboxyl group
CVD	:	Chemical Vapour Deposition
EDS	:	Energy Dispersive Spectroscopy
E-GO	:	Ethanol graphene oxide
FESEM	:	Field Emission Scanning Electron Microscopy
FTIR	:	Fourier-transform infrared spectroscopy
GO	:	Graphene Oxide
H_2O_2	:	Hydrogen peroxide
H_2SO_4	:	Sulphuric Acid
IS	:	Impedance Spectroscopy
KMnO4	:	Potassium permanganate
kV	:	kilovolt
mg	:	Milligram
mL	:	Millimeter
NaOH	:	Sodium Oxide
NF	:	Nickel Foam
Ni	:	Nickel
$Ni(NO_3))_2$	:	Nickel nitrate
Ni-AC	:	Nickel Acetate
NiO	:	Nickel Oxide
NiO	:	Nickel Oxide

nm : Nanometer

-OH : Alcohol group

PEO : Polyethylene oxide

rpm : Revolutions per minute

SEM : Scanning electron microscopy

TEM : Transmission electron microscopy

W : Watt

XRD : X-ray Diffraction

CHAPTER 1: INTRODCUTION

1.1 Overview

Nanotechnology is a combination of science, engineering, and technology sector that produce many devices applications in nanoscale which ranging from 1 until 100 nanometers. For the last decades, the nanotechnology industry has grown rapidly. Due to that, many researchers around the world trying to found a new alternative element. Graphene oxide (GO) has received much attention as it is believed as a promising element in the nanotechnology industry.

There is the various synthesis method has been developed all around the world via a physical and chemical method to synthesis GO. GO has become a demand in industry to create new electronic devices with a vast range of application (Vincenza Modafferi, 2019).

Among various method, chemical oxidation from graphite to graphene oxide and followed by the reduction in the production of graphene oxide is the most appealing method been used in many research and study. Both of this method is considered as fewer pollutant manners in the production of graphene oxide

There are raised an issue regarding concerns in the pollution of environmental impact from the nanotechnology industry. This is because the production of electronic devices is believed to have a negative impact on the environment. This is because the production of nanotechnology produces a massive mass of toxic chemicals. Therefore, more study needs to conduct to overcome this issue. The environmental pollution issue has become a big debate among manufacturers and activist.

Graphene consists of a functional group of oxygen which are hydroxyl and epoxy group that present at the plane of carbon element. With the presence of oxygen groups, GO can act as potential anchoring sites for adsorbing diverse metals oxides of nanocomposites.

Other than that, at the edge of the structure of graphene oxide are consist of carbonyl and carboxylic groups. The presence of carbon functional group makes graphene oxide excellent with hydrophilic character and chemical reactivity. Due to that, GO is a remarkable electrocatalytic, electrochemical sensing and electrochemical energy conversion properties, which make it a promising material for a wide of application.

With the recent exploration of production graphene oxide in large scale, the number of publications related to graphene oxide and graphene-metal oxide-based materials has increased exponentially.

Four methods have been widely used for the preparation of graphene oxide. The most known method is based on the introduction of sulfuric and nitric acids into the solution by Schafhaeutl's method. Then, it is followed by the introduction of potassium chlorate by Brodies', one-pot method by Staudenmaier's and Hummer and Offeman method (Lacina, 2018). It is well known that back in 1859, Brodie was the first person who synthesized by added potassium chlorate to the graphite oxide in the condition of fuming nitric acid. And after 40 years in 1958, Hummer's and Offeman has developed a new method to synthesized graphite oxide by dissolved $NaNO_3$ and $KMnO_4$ in concentrated of H_2SO_4. This method is faster than Brodie's method as Hummer's method, graphite oxide can be synthesized within a few hours. Hummer's method has widely adopted by many researchers in the whole world.

However, in the past 20 years, various studies have been conducted to modify the original Hummer's method to avoid toxic gas generation and residual of nitrate by removing $NaNO_3$. There are much research has been reported that GO composites decorated with metal oxide have a uniform distribution of metal oxide on GO sheets which can eliminate the restacking of sheets during synthesis and stabilized the volume changes in metal oxide during charge-discharge cycling. GO-NiO composite getting more attention lately as it can improve the electrochemical performances of nanocomposites.

Nickel oxide has high theoretical specific capacitance and low cost. It is also reported that nickel oxide has poor electrical conductivity. However, this problem can be overcome by an added conductive material such as graphene and other conductive polymers to improve the electrochemical performance (Bai, 2017).

After a lot of reading, this study is focused on the interest of producing a GO/rGO by modified Hummer's method. Then GO/rGO is decorated with NiO via hydrothermal method.

1.2 Problem Statement

Because of the presence of conduction and valence band, GO/rGO have a gap and act as a semiconductor in photocatalytic transformations. The presence of oxygen functional groups in GO/rGO can also act as nucleation centers or anchoring sites for the landing of nanoparticles, limiting the growth of the nanoparticles and improving their stability and dispersion on the rGO surface. At the same time, these nanoparticles can help to enlarge the interplanar spacing of the rGO sheets, limiting their stacking, and thus maintain the excellent properties of individual rGO nanosheets.

1.3 Focus and Objective of the Research

The main focus of this study is to synthesis graphene oxide (GO) by using modified Hummer’s method.

The hydrothermal method is believed will be the best method to use to decorate nickel oxide on graphene oxide. This due to several advantages that are very simple setup and could minimize defects due to its closed system of relatively high temperature and internal pressure.

Next, the intended of this study is to investigate the morphology and electrical properties of graphene oxide decorated with nickel oxide. For this work, the morphology and electrical properties rGO-NiO will be study using field emission scanning electron microscopy (FESEM), X-ray diffraction (XRD) and Impedance Spectroscopy (IS) machine.

Below is the objectives on this study:

1. To synthesis graphene oxide (GO) using modified Hummer's method.

2. To decorate nickel oxide on graphene oxide using hydrothermal method.

3. To investigate morphology and electrical properties of graphene oxide decorated with nickel oxide by using FESEM, XRD and IS machine.

1.4 Thesis Outline

This report is divided into five main chapters including 1. Introduction, 2. Literature review, 3. Methodology of experiment, 4. Result and discussion and 5. Conclusion. The overview about graphene oxide and nickel oxide of this research are introduced in the first chapter.

Next, a brief discussion and review about methodology of Hummer's method in production of graphene oxide and hydrothermal methods to produced graphene oxide decorated with nickel oxide can be find in Chapter 2. Literature review focusses on the morphology and structural of GO and GO decorated with metal oxide. Other than that, in this chapter also focuses on the electrical properties.

In Chapter 3, Methodology of experiment were discussed including the procedure in preparation of sample of graphene oxide using modified Hummer's method and graphene oxide decorated with nickel oxide using hydrothermal method.

Chapter 4, Results and Discussion are divided into two parts. In the first part, the results of experimental reduced graphene oxide decorated with nickel oxide were discussed. The second part of this chapter contained result of morphology and electrical properties of GO and GO-NiO.

The conclusion on the research findings are presented in Chapter 5. Besides, several suggestion regarding the improvement method and also ideas for future work are included in the last chapter.

CHAPTER 2: LITERATURE REVIEW

This chapter covers a brief introduction of graphene oxide and nickel oxide. It also discussed several journals on the preparation method of the sample using the modified Hummer's method and hydrothermal method. Besides, a material characterization of GO by FESEM, XRD and IS machine is being discussed as well.

2.1 Graphene Oxide

Since the discovery of graphene in 2014, it has attracted remarkable attention because of its properties of a single atomic thick layer of graphite with honeycomb framework that packed closely with conjugated and hexagonal connected carbon atoms. Graphene comprises of two equivalent sub-lattices of carbon atoms linked to each other with sigma bond of 0.142 nm long. Graphene mainly consist of pure carbon at the edge and oxygen group of an element at the base framework of carbon that presents different electrochemical characteristics.

Graphene has advantages such as a large specific surface area with a theoretical surface area of $2630 m^2 g^{-1}$. Besides, GO has reported having high electron mobility, good mechanical strength has high electric and thermal conductivity, good optical transparency and tunable band gap. For all these adaptable properties and obvious advantages, the rapid development of graphene-based materials has been witnessed in the fields especially in nanotechnology (Xin Jiat Lee, 2019) (Halder, 2016).

Graphene oxide (GO) contains a cluster of reactive carbon-oxygen functional groups such as C-O, C=O and -OH, which make it a stronger nominee for use in many applications through chemical functionalization.

GO is constructed from graphite and graphite rich with exogenous functional groups, carbon, hydrogen, and oxygen.

There has high interest been concentrated on assimilating GO with other materials by hybridizing it with good electrical properties materials (Norhayati Hashim, 2016).

The non-stoichiometric structure and composition are highly dependent on the production step since there are the different method used for synthesis GO. The transformation of GO to reduced graphene oxide (rGO) can be achieved through chemical or physical reduction. Through the reduction process, the oxygenous functional groups of in the GO are eliminated to form GO with carbon to oxygen (Xin Jiat Lee, 2019).

Graphene oxide is graphite that has been oxidized to intermingle the carbon layers with oxygen molecules and then reduced to separate the carbon layers into individual or few-layers of graphene. Due to the disruption bonding networks of *sp2*, graphene oxide is often described as an electrical insulator. The ability for graphene oxide to conduct electrons depends on the amount of oxidization in the compound as well as the method of synthesis.

The oxidization in the solution will disturb the electrical conductivity, so while highly oxidized graphene oxide would be in a very poor conductor of electricity state. However, some methods can be used to counteract this situation. There are a few procedures that can modify the substance to develop its properties to suit a specific application (Arthi G & Bd, 2015).

There are various ways and different substrate s can be used to synthesize GO. By exfoliated graphite mechanically to produce GO, it has caused high interest as this technique has a simple and low budget technique (Avouris & Dimitrakopoulos, 2012).

GO can be used as a supercapacitor because of its unique properties, such as high surface area, excellent flexibility, chemical inertness, and good electrical conductivity. The practical use of the entire surface area of a GO sample is difficult. So it is often used in combination with an active metal oxide for application as electrode material in supercapacitors. GO has a layered structure, but the plane of carbon atoms is heavily decorated by oxygen-containing groups which expand the interlayer distance and make graphite oxide hydrophilic (Salimian et al., 2015).

Combining a transition of metal oxides with highly conductive materials is one of the techniques to stop accumulation and increase the conductivity. GO grows epitaxial on most metals, on polycrystalline metal substrates it has a polycrystalline structure in 2D. GO is one of the commonly used material as substrate for composite. This is due to the atomic structure of GO where it has thick two dimensional structure.

2.2 Addition of Nickel Oxide into Graphene Oxide

Nickel is an alloy metal that widely used in various sectors such as aerospace, electronics, power tools, etc. Nickel also is known for its strength and improve the properties of the surface of another substrate. It has widely used to improve the conductivity of another substrate (Meng Li, 2017). Nickel is believed is a good alternative as it is cheaper to be used to improve electrical properties.

The interfaces of between graphene oxides with different transition metals have been studied thoroughly in recent years. Metal oxide nanostructure such as nickel oxide, zinc oxide, iron oxide, etc. has attracted much attention because of their extraordinary properties in different fields of optics, optoelectronics, catalyst, solar cell materials, and sensors (S. Fahad, 2017). The interfaces between GO and metal oxide differ in two main aspects which are lattice mismatch and metal-graphene interaction. Strong interacting metals the graphene-metal separation is around 0.21 nm and for weak interaction is around 0.33 nm (Dahal, 2014).

There is study has been conducted to investigate the interaction between graphene oxide and metal oxide. In the study found that interface of GO and metal oxide can be divided into two groups which are physisorption interface and chemisorption interface. The interaction between carbon presented in graphene oxide with metal oxide which is in this study is Nickel atom has destroyed the perfect lattice structure of GO. This has caused lower interface resistivity due to the opening of π-d band gap to take place in the graphene oxide. It also has been reported that NiO can be applied for fabricate high performance of sensor.

There is various method to synthesis graphene oxide decorated with nickel oxide. In this study, our work focused more on preparation of graphene oxide with modified Hummer's method and decorated the nickel oxide via hydrothermal method.

2.2.1 Preparation of graphene oxide with Hummers Method and hydrothermal methods

The bottom-up and top-down are two different routes of methods that can be used to synthesis graphene oxide. The bottom-up route is a method that uses a source of the carbonaceous gas to break graphene. The bottom-up route consists of mechanical and chemical exfoliation and also chemical synthesis.

While the top-down route is by break the layer of graphene sheets by exfoliating or separate graphite derivatives. It can be done by using pyrolysis, epitaxial growth, chemical vapor deposition (CVD) and others.

Hummer's methods are one of the chemical exfoliation method and one of the best appropriate method for synthesizing graphene oxide. In this method, the chemical produces colloidal suspension which modifies the GO from graphite and graphite intercalation compound. There are many different types of study has been conducted for the preparation of GO using Hummer’s method (Md. Sajibul Alam Bhuyan, 2016).

When considering the synthesis of graphene oxide and its derivatives, the desired structure and properties are largely dependent upon the size, shape and functional groups attached to the surface of the material. The ideal structure is single-layer graphene oxide consisting of a single-atom, thin hybridized carbon structure with minimal defects (N.I. Zaaba, 2017).

According to study conducted in Shanghai, the reduced graphene oxide-Ni nanostructure was been prepared using the hydrothermal method. The synthesis of GO was from natural graphite powder using modified Hummer’s method. For a sample of Ni-rGo was carried out by a microwave involved heating approach in a specialized microwave system. The procedure is the same as the hydrothermal method where the mixture of 100mg GO and 1 mol of nickel acetate was dispersed in 100 mL of ethylene glycol and been in ultrasonification for 30 minutes. In this study, the pH value was turned to 12 by adding the ethylene glycol containing 2mol/L of NaOH. In this hydrothermal method, the microwave system was used to dry out the sample at 800W for 150s after addition of 2mL hydrazine hydrate. After that, the sample was washed a few times with excessive water and ethanol before the sample been dried out again. However, in this study nickel decorated with reduced graphene oxide was carried out using microwave heating approach. The GO ultrasonically dispersed in 100 mL of ethylene glycol. Then the 1M of $Ni(AC)_2$ was added into the homogeneous dispersion before ultrasonicated for 30 minutes. Because of the increase of pH value to 12, the mixture was microwave for 150 s before washed adequately with water and ethanol. Then the products were freeze-dried to obtain Ni-rGO composites (Qiu, 2017).

Also, study conducted by in 2016, the GO was synthesized chemically using a modified Hummer's method. In this study, 5g of graphite powder is treated with 5% of HCI before added with concentrated of sulphuric acid, H_2SO_4. Later the mixture was added with $KMnO_4$ while being kept in an ice bath while being stirring for 2 hours. Then the mixture was washed with HCI for several times before being dried out at 50°C. GO was dispersed in 400mL distilled water and being ultrasonicated for 40 minutes before added with hydrazine hydrate. Then the nickel acetate tetrahydrate was dissolved in the separate flask with a minimum amount of water. Later, the mixture of GO and NiO were mixed. In this study, there are five (5) different amount if nickel acetate tetrahydrate used which are 100, 200, 400 and 600 mg utilized to study the influence of metal loading. The end product of GO-NiO was dried under vacuum after washed several times by using distilled water (Nasser A.M. Barakat, 2016).

Before that, there is study conducted in 2015, modified Hummer's method was used to synthesis graphene oxide to study the influence of different solvent on the structural and electrical properties of graphene oxide. This study is focusing on the production of acetone graphene oxide (A-GO) and ethanol graphene oxide (E-GO). The solution of GO was dissolved in the acetone/ethanol solution. As typical Hummer's method procedure, graphene oxide is extracted from graphite powder. However, in this study, the procedure is a bit different with the previous study where there is an addition of 3mL phosphoric acid (H_3PO_4) was added in the solution of 27 ml of sulphuric acid with ration 9:1 and stirred for several minutes. 1.32 g of potassium permanganate $KMnO_4$ was added into the mixture solution and stirred for 6 hours until the color of the solution turns to dark green. In this procedure, hydrogen peroxide H_2O_2 was added slowly into the mixture solution to remove the excess of potassium permanganate (Zhang, 2015).

Graphene oxide was prepared by using a modified Hummer's method. In this study, the same method was used where the graphene oxide synthesized from graphite powder. However, in this study, hydrogen peroxide, H_2O_2 was added the formed the residual of potassium permanganate and manganese oxide MnO_2. Because the formation of the residual in the solution, the color of the solution changes from brown to light yellow. However, the brown-colored slurry of graphene oxide was obtained. The removal of the impurities was done by centrifuged the dispersion at 5000 rpm for 30 minutes (Anu Saini, 2016).

In other study conducted, rGO was prepared using chemical oxidation method, Hummer's method at different temperature which are 150°, 180°, 210°and 240°C. The obtained product was dried at 80°C for 10 hours after washed with water and ethanol. At the same time, nickel foam (NF) was immersed in the aqueous solution of 25 mg of GO, 1 mmol of cobalt salt and 2 mmol of thiourea (Shen et al., 2011).

The graphene oxide was prepared by using the modified Hummer's method. The resultant suspension was extensively washed with distilled water by filtration and freeze-dried. The GO dispersed in the deionized water before mixed with a solution of nickel nitrate $(Ni(NO_3))_2$. After the sonification, the hydrazine hydrate was added into the solution. The final nanocomposites were washed with deionized water. The sample morphology was observed using XRD, various types of electron microscopy.

2.3 Material Characterization Technique for GO-NiO Nanocomposite

Characterization has been one of the foundational pillars of material science to gain knowledge and understand the properties, structures, and mechanism of the samples. The material characterization can be done using field emission scanning electron microscopy (FESEM) for morphology and x-ray diffraction (XRD) for structural information.

2.3.1 Field Emission Scanning Electron Microscopy (FESEM)

Normally in all study has been conducted, the surface of the sample is study by a high resolution field emission scanning electron microscope. According to research study by N.I. Zaaba, is used SEM model Jeol JSM-6010LV to observe the morphologies of the sample. The SEM was used at the magnification of 500x and 10,000 x with 20 kV of accelerating voltage. Based on the SEM, it is shown that graphene oxide that dissolved in acetone shows comprehensive extensive spread on the silicone surface while for ethanol-GO looks clustered on the entire surface. This is can be caused by the GO was not fully dissolved in ethanol solvent.

Study conducted by Qiu (2017), the sample were characterized by using FESEM with accelerating voltage of 20 kV. The value of voltage used are same with previous study. However, the brand model used in study is JEOL S-4800. The morphology of GO was found stacked and crumpled which is due to unsettled exfoliation and restacking process. For Ni-rGO morphology found that the nickel nanoparticles decorated homogenously onto rGO sheets. It also found that the active sites in GO laminates facilitate the formation and uniform landing of nickel nanoparticles.

The rGO surface were observed using SEM. NF nanoflakes was observed as very thin flakes and its insert rGO vertically which are leading to porous three-dimensional structures. This is give an advantages for an access to electrolyte and transportation of an electron during electrochemical reactions (Shen, 2011).

2.3.2 X-ray Diffraction (XRD)

XRD was used to determine the crystal structure and interlayer spacing of A-GO and E-GO. In the tabulated data shown that the interlayer spacing of both acetone-GO and ethanol-Go was slightly different. The increasing interlayer in acetone-GO is because of the intercalated functional group of oxygen and water molecule into the carbon layer structure. The interlayer spacing of graphene oxide was controlled on the degree of oxidation of graphite and amount of water molecules intercalated into interlayer spacing. Based on the literature study of this study stated that when the graphite was oxidized and become graphene oxide, the XRD peak is shifted from 26°C to 11°C. However, based on their study found there is no sharp peak appeared in their XRD result. However, the peak shows a broad range from 10°C to 25°C. In this study also studied the interaction bonding of the samples. They concluded that graphite powder was successfully oxidized with the concentration of potassium permanganate (N.I. Zaaba, 2017).

In this study, the reduction of graphene oxide was done by one-step hydrothermal approached with additional of 2 ml of Hydrazine hydrate in the dispersion. In this study, XRD was used to analyze the crystallinity of the reduced graphene oxide. Based on their findings, the strong diffraction peaks corresponded to the crystalline plane were observed at $2\Theta= 12.5°$. Here, the study concluded where the oxygen was introduced intro graphite layer without annealing process by oxidizing the agents and increase its interplanar distance with 0.61 nm. For annealed reduced graphene oxide, the peak showed at $2\Theta=26.56°$ with an interplanar distance of 0.335 nm. The distance becomes shorter is because the distance disappeared after the exfoliation. The increase of the interplanar distance is because of the formation of the oxygen-containing functional groups. While scanning electron microscope (SEM) was used to study the physiochemical changes occurred in the graphite. The SEM was prepared at different magnification, 1K, 5K, 2K, and 3K. Based on their finding, graphene oxide powder are showed layered structure, folded and irregular in shape. But for GO nanosheets, the images are more crinkled and more bubble structure was observed which resulted from the deformation upon the exfoliation and restacking processes (Anu Saini, 2016).

Also, the sample was observed using XRD to investigate the composition of the specimens with ranging from 10° to 80°. The peak diffraction is shown slightly different where for acetone is showed at 11.7° while ethanol-GO at 123°. Both samples also showed the difference in interlayer spacing. However, in this study, the sharp peak does not appear in an XRD result. Also, it’s studying about its electrical characterization using Pico ammeter (N.I. Zaaba, 2017).

2.3.3 Impedance Spectroscopy (IS)

Electrochemical Impedance Spectroscopy (IS) is a measurement technique to measure the electrical properties of a variety different medium which can be either ionic, semiconductor or insulator. The mechanism of the machine is based on the interaction between an external field and the electric dipole moment of the sample. It is also known as permittivity. This technique measures the impedance of a system over a range of frequencies that are dependent on the impedance of the electrochemical system. The result of the permittivity of the sample is expressed in bode plot graph or cole-cole/Nyquist plot. This is because of ion transport through the layers of medium.

The dielectric properties of graphene oxide containing polymer nanocomposites are significantly influenced by the aspect ratio of graphene oxide and the interfacial adhesion with the polymer matrix. During the application of current flow through the polymer nanocomposites, the charges are accumulated at the interfaces between two dielectric materials with dissimilar relation times.

CHAPTER 3: METHODOLOGY

The purpose of this chapter is to present the research strategy including the methods of synthesizing and characterizing the samples. The content of this chapter is partitioned into two sections with the first section covers the experimental procedures for preparing the samples started from producing graphene oxide until decorating the nickel oxide nanocomposite. The second section is to describe the characterization techniques that been used in this study.

Production of low friction, long-lasting and highly conductive coatings is highly desired for many electrochemical applications. However, the production is a great challenge for a long time. GO with excellent mechanical and electrical properties make it very attractive for a wide range of electrochemical applications. There is a various method used in the production of graphene oxide. However, in this study only focused in modified Hummer's method. (M. Ghorbani, 2015)

3.1 Materials

All of the material reagents used were analytical grade and were utilized without further purification in this experiment. The materials used in this work are listed in Table 3.1 below.

Table 3.1 List of chemicals used in the experiment

No.	Types	Molar mass (g/mol)	Brand
1.	Graphite powder	12.00	Aldrich Chemistry
2.	Potassium Permanganate - Powder ($KMnO_4$)	158.03	R & M Chemicals
3.	Nickel Acetate Tetrahydrate (Gotoh et al.)		Aldrich Chemistry
4.	Polyethylene oxide	900.00	ACROS Organics
5.	Sulphuric acid – Liquid (H_2SO_4, 98%)	98.00	AJAX Chemicals
6.	Absolute Ethanol – ethanol	46.07	AJAX Chemicals

3.2 Preparation of Graphene Oxide using modified Hummer's Method

Graphene oxide (GO) was prepared from natural flakes graphite as the precursor. GO was synthesized via the modified Hummer's method by oxidation of graphite flakes in the acidic solution with the presence of $KMnO_4$.

Synthesis of GO started with weighing 1.049 gram of graphite powder and 3.0615 gram of potassium permanganate powder in ratio 1:3. Both powders were dissolved in 80 ml of sulphuric acid and cause an increase in temperature. The solution was subjected to magnetic stirred using for 3 hours and the temperature was maintained at 50°C. Then the solution was cooling at the room temperature for a while. The color of the solution has change its color to become a dark-brownish color as shown in Figure 3.1 below from dark black color.

Figure 3.1 The mixture solution of graphite and potassium permanganate.

50 mL of deionized water was added into the solution to stop the reaction and stirred for 15 minutes. By added deionized water into the dispersion, a large amount of heat was released. Then, the beaker that contained a mixture of the solution was moved from magnetic stirrer to big beaker contained ice. Quenched is need in the procedure is to remove heat from the mixture. The mixture quenched for 15 minutes in the ice bath.

Next, the procedure proceeds with the washing step where the GO dispersion moved into 4 separate test tube as shown Figure 3.2 below.

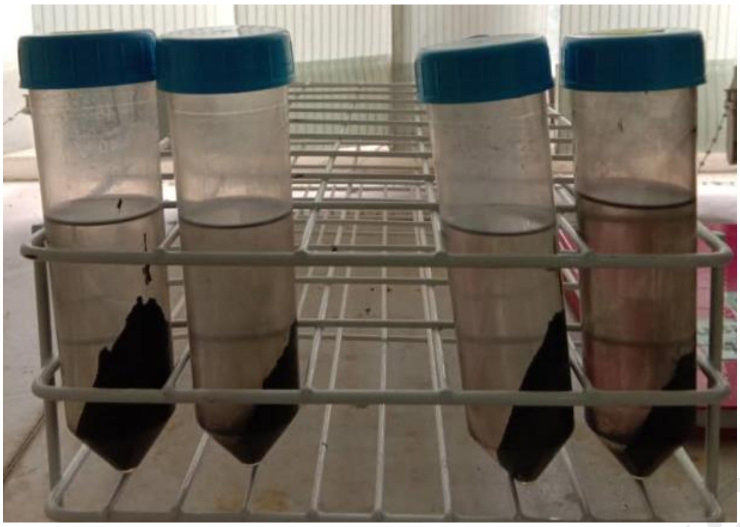

Figure 3.2 The mixture of GO separate into 4 test tubes for washing.

The GO dispersion was centrifuged to separate the supernatant with 6000 rpm for 5 minutes with 20mL of distilled water to remove residual metallic ions. The washing start with centrifuged the dispersion in distilled water for 5 minutes, then shake well the tubes by hands and centrifuged again for 5 minutes. Figure 3.3 below shows the mixture of GO is shake well to wash thoroughly.

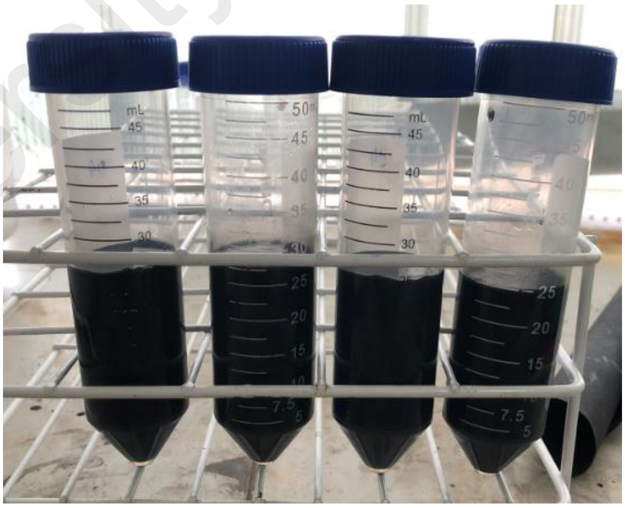

Figure 3.3 GO mixture shake by hand to washing after centrifuged for 5 minutes.

This is one full cycle of washing steps. This is to ensure the dispersion washed equivalent. Then the procedure was repeated with 20 mL of ethanol and hydrochloric acid. An adequate amount of washing is needed to have a good quality of GO with fewer impurities. The pH of the dispersion was calculated after every cycle of washing. In this study, the GO dispersion was achieved until pH 5.1.

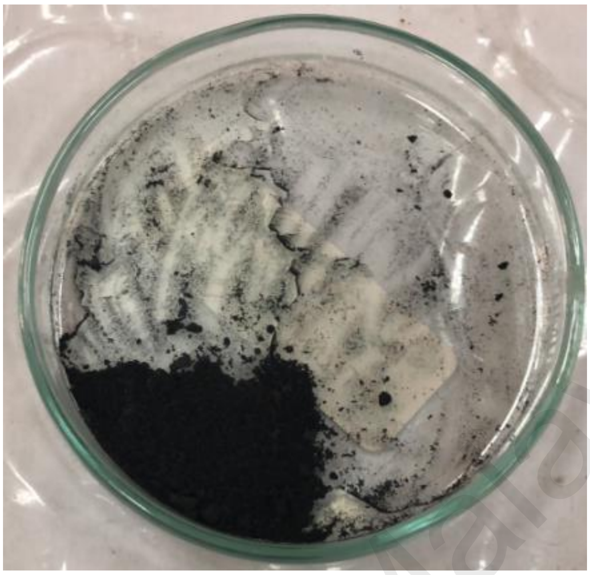

Figure 3.4 GO powder after drying in the oven for 24 hours.

Subsequently, the dispersion was transferred into a petri dish and drying the dispersion in the oven with temperature 110°C for 24 hours. The final mixture obtained is reduced powder graphene oxide as show in Figure 3.4 above.

3.3 Preparation of Graphene Oxide decorated with Nickel Oxide using Hydrothermal Method.

Figure 3.5 Flakes of GO-NiO after drying in the oven for 18 hours.

The second stage was done to prepare mixture graphene oxide (GO) and nickel oxide (NiO) via the hydrothermal method. 1.0146-gram nickel acetate tetrahydrate ($Ni(NO_3)_2.6H_2O$) as prepared by dissolving in 20 mL of ethanol. Then, 0.1079 gram of powder of graphene oxide were added into the dispersion and stirred for 60 minutes by using magnetic stirrer. The dispersion was transferred into a petri dish and then baked in the oven for 18 hours in 110°. The powder of GO-NiO obtained and shows in Figure 3.5 above.

3.4 Preparation of polymer nanocomposites.

Solution casting was used in preparation of GO-polymer based. The polymer used in this study is polyethylene oxide (PEO). Figure 3.6 below show the process weighing PEO.

Figure 3.6 weighing process of PEO powder

In this study, the nanocomposites were prepared in three different solutions which are pure polymer, GO polymer and GO-NiO polymer. 1.0213 gram of PEO was dissolved in 40mL of distilled water. The mixture was stirred for 4 hours by using magnetic stirrer at a speed of 220 rpm until a homogenous solution was obtained. Figure 3.7 below showed the process of pure PEO dissolved in distilled water.

Figure 3.7 PEO dissolved in distilled water and stirred for 4 hours to obtained homogenous solution.

After the PEO was completely dissolved in distilled water, the dispersion moved into a petri dish and left in the oven for 5 hours with 30°C. The thin film of pure PEO was obtained.

Then, the procedure was repeated three times by added PEO powder into dissolved solution of GO and GO-NiO. Firstly, the GO powder was prepared with 5wt% which is 0.0520 gram of powder dissolved in 40 mL of distilled water. The powder stirred for another 15 minutes until all powder of GO and GO-NiO dissolved completely. Then 1.011 gram of PEO powder was added slowly into the dispersion. The dispersion of GO and PEO was stirred for 4 hours at 220 rpm. Figure 3.8 below show how the GO-NiO powder dissolved in distilled water before PEO powder were added into the solution.

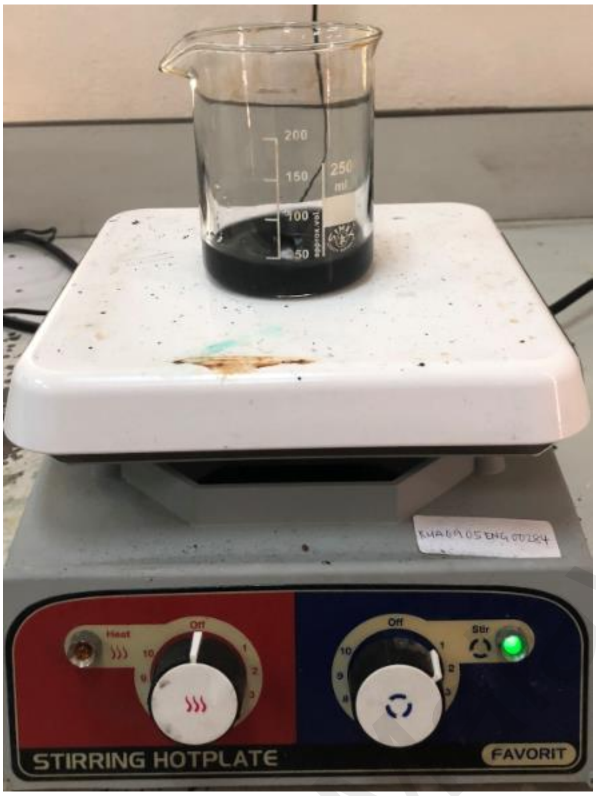

Figure 3.8 GO-NiO powder dissolved in the distilled water and PEO was added into solution.

After that, the dispersion was transferred into a petri dish and left dried at the oven for 5 hours in 30°C.

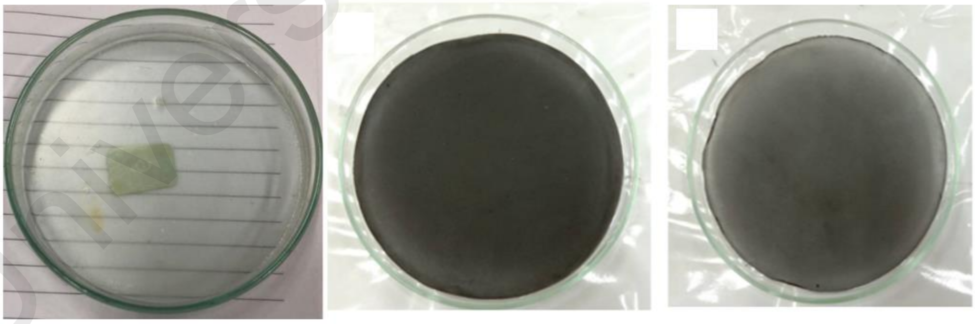

Figure 3.9 From left is a) Pure PEO-polymer, b) GO-polymer nanocomposite and c) GO-NiO-polymer nanocomposite.

The thin film of polymer nanocomposites was obtained. The step was repeated with GO-NiO powder. Figure 3.9 above shows the final product of thin film obtained.

3.5 Characterization Technique

Characterization has been one of the foundational pillars of material science to gain knowledge and understand the properties, structures, and mechanism of the samples. The characterization techniques used in this study to characterize the samples shown in Figure 3.10 below.

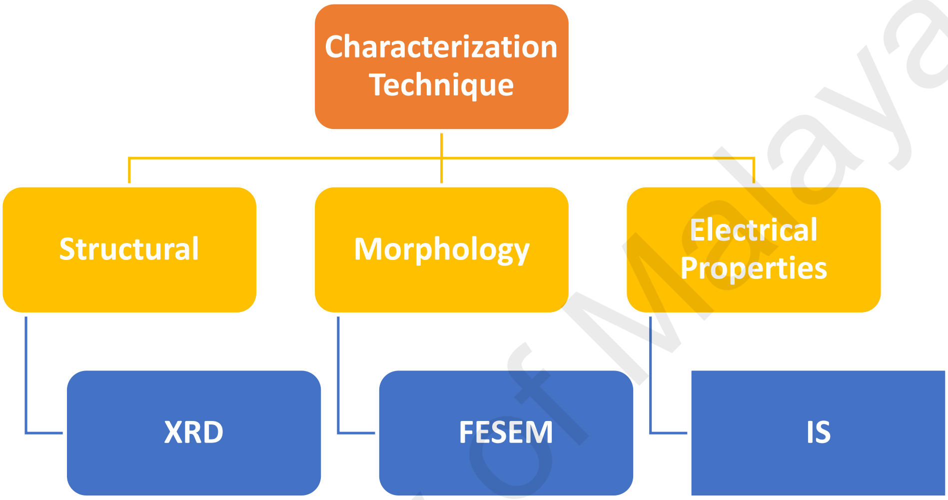

Figure 3.10 Flow chart of characterization technique used in this study.

The field emission scanning electron microscopy (FESEM) was used to study morphology for all 5 samples included thin film and powder which are a thin film of pure PEO, thin-film GO-PEO, thin-film GO-NiO-PEO, GO powder and GO-NiO powder. Energy Dispersive Spectroscopy (EDS) was used to check the element present in the samples. X-ray diffraction (XRD) was carried out to determine the elemental compound of GO and GO-NiO powder. Then Impedance Spectroscopy (IS) was used for studying the electrochemical system.

3.5.1 Determination of Morphology by FESEM and Element by EDS

FESEM is used to study the morphology of the sample. In this study, the images of the samples were taken using the Carls Zeiss Auriga machine as shows in Figure 3.11 below. This machine also included with EDS. FESEM can provide the topographical and elemental information at various magnification ranging from 10x until 300,000x. A scanning electron microscope is a tool to visualize the images of the order nanometer to micrometer.

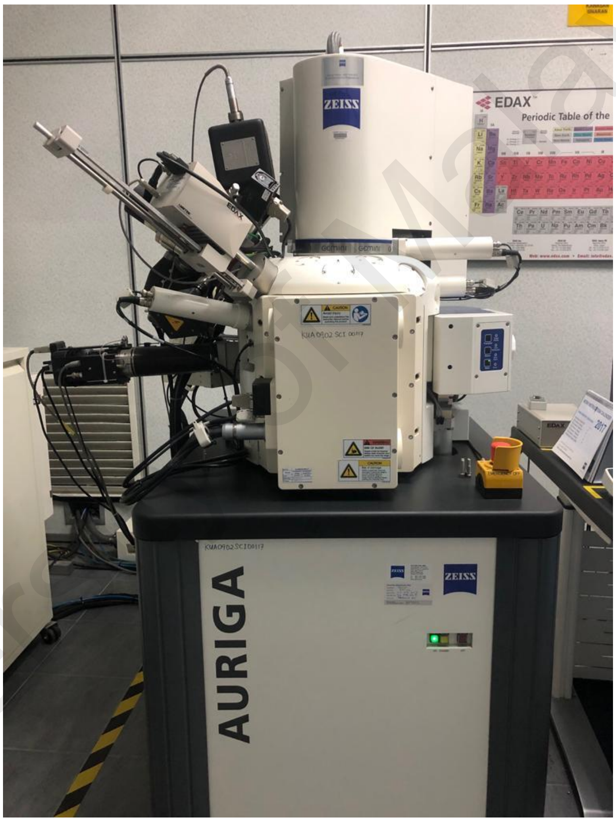

Figure 3.11 AURIGA FESEM and EDS machine.

The thin film of pure polymer, GO-polymer, and GO-NiO was cut into small pieces and the sample attached to sample holder. While a small amount of powder of GO and Go-NiO also attached to the sample holder. The adhesive tape was taped on top on sample holder to hold the sample and hand blower was used to clean the loss debris particle. After that, all five samples entering the vacuum chamber.

After the images of all sample taken, the procedure proceeds with EDS technique to analyze the element present in each sample. The analysis of EDS shows the detected element and showed in terms of the peak.

3.5.2 Determination of the Structural by XRD

X-ray diffraction (XRD) is one of the important technique to study the structural and phase compositions of materials. In this study, the XRD pattern of GO and GO-NiO powder was analyzed with 45keV. Figure 3.12 below shows the model of XRD machine used and the process of structural analysis for GO and GO-NiO powder sample.

Figure 3.12 Model of PANalytical EMPYREAN XRD were used to analyze GO and GO-NiO powder sample.

The incident beam was used ranging from 10° to 90°. Each of the sample was analyzed with 30 minutes. The X-ray intensity is plotted against the angle of 2ϴ with step size of 0.02°/min and sharp peak causes by the diffraction appears. X-ray diffraction (XRD) is a characterization tool to analyze the crystal configuration and orientation of the nanomaterials. A diffraction pattern is formed when X-rays come in contact with the crystalline phase of the film. It can provide information about different orientation and interlayer spacing of the atomic layers. The XRD is mainly dependent on the shape of the peak to determine the compound of the sample. It can measure *d-spacing* to determine the crystallization of the element.

3.5.3 Determination of the Electrical Properties by IS

In this study, the electrochemical properties of the sample are employed by using Impedance Spectroscopy as shown in Figure 3-13. HIOKO 3532-50 LCR Hi-Tester with a frequency range from 50 Hz to 5 MHz. The sample of thin-film pure polymer, GO polymer and GO-NiO polymer was placed on the sample holder. The results were analyzed using the HIOKI software to measure the conductivity of the electrolyte for each sample. This technique is using the application of an alternating voltage inappropriate frequency range to measure the conductivity of the electrolyte.

Figure 3.13 LCH Hitester for testing electrical properties of sample.

The difference in the oxidation process can cause the presence of defects, impurities, structural disorder, wrinkle, crack, fragmentation and other structural attributes are possible and this can affect the electronic, optical and adsorption properties of GO (Xin Jiat Lee, 2019).

CHAPTER 4: RESULT AND DISCUSSION

In this chapter, the result and discussion regarding the steps of experimental processes are presented. The results are presented in three main parts: 1) synthesis of graphene oxide, 2) decorated graphene oxide with nickel and 3) morphology and electrical study of rGo-NiO nanoparticles. Each of these parts come with the discussion of the characterization techniques such as FESEM, XRD and IS.

4.1 Synthesis of GO and GO-NiO Powder by using XRD, FESEM and EDS

For morphology and electrical properties testing, the sample of the thin film for each sample was cut to become 20 cm x 20 cm width and length respectively. Figure 4.1 below shows the cut of 20 cm x 20 cm size of sample of pure PEO-polymer, GO-polymer and GO-NiO-polymer nanocomposites respectively.

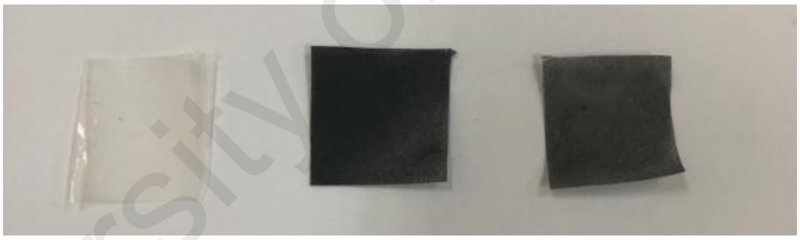

Figure 4.1 20 cm x 20 cm of a) Pure PEO-polymer nanocomposite, b) GO-polymer nanocomposite and c) GO-NiO-polymer nanocomposite.

4.1.1 Graphene Oxide (GO)

An X-ray diffraction graph is important in order to obtain detailed structure on the GO. The Figure 4.2 below show the X-ray diffraction peak when 2ϴ = 26.3648° with d-spacing of 0.338054 nm at the polycrystalline structure [002] (Hai Tan, 2018). High diffraction peak present in the XRD pattern of GO powder below corresponds to the well-arranged layer polycrystalline structure of GO.

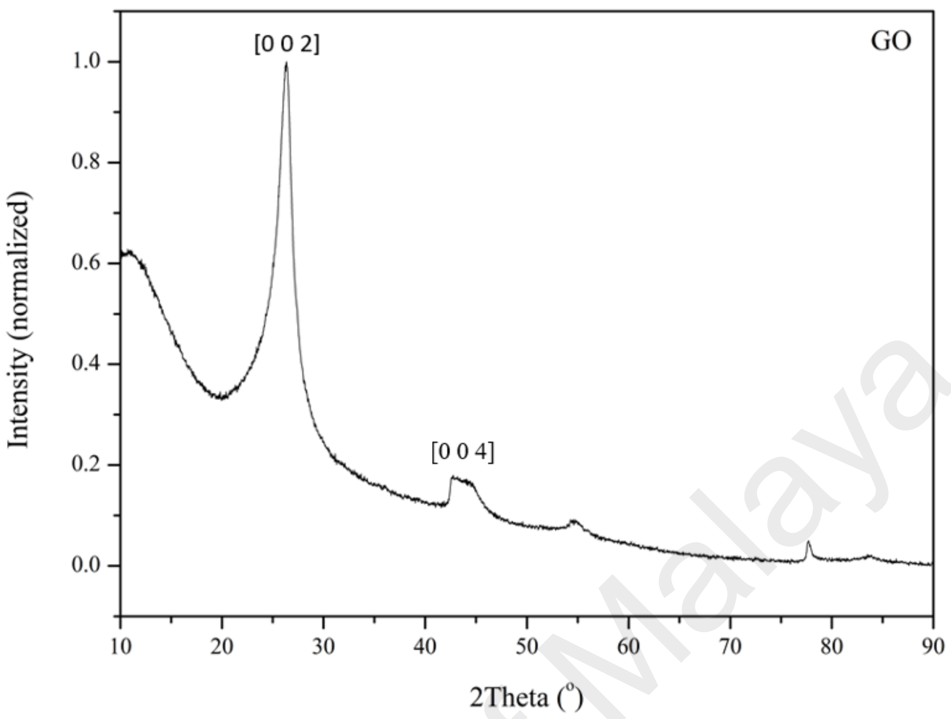

Figure 4.2 XRD graph of GO powder.

Based on FESEM images Figure 4.3 below shows for images for GO powder where all the morphology of the powder is in flakes and wrinkled sheet structure found at the edge of the powder for all images. However, the captured images is estimated shape of graphene oxide structure only.

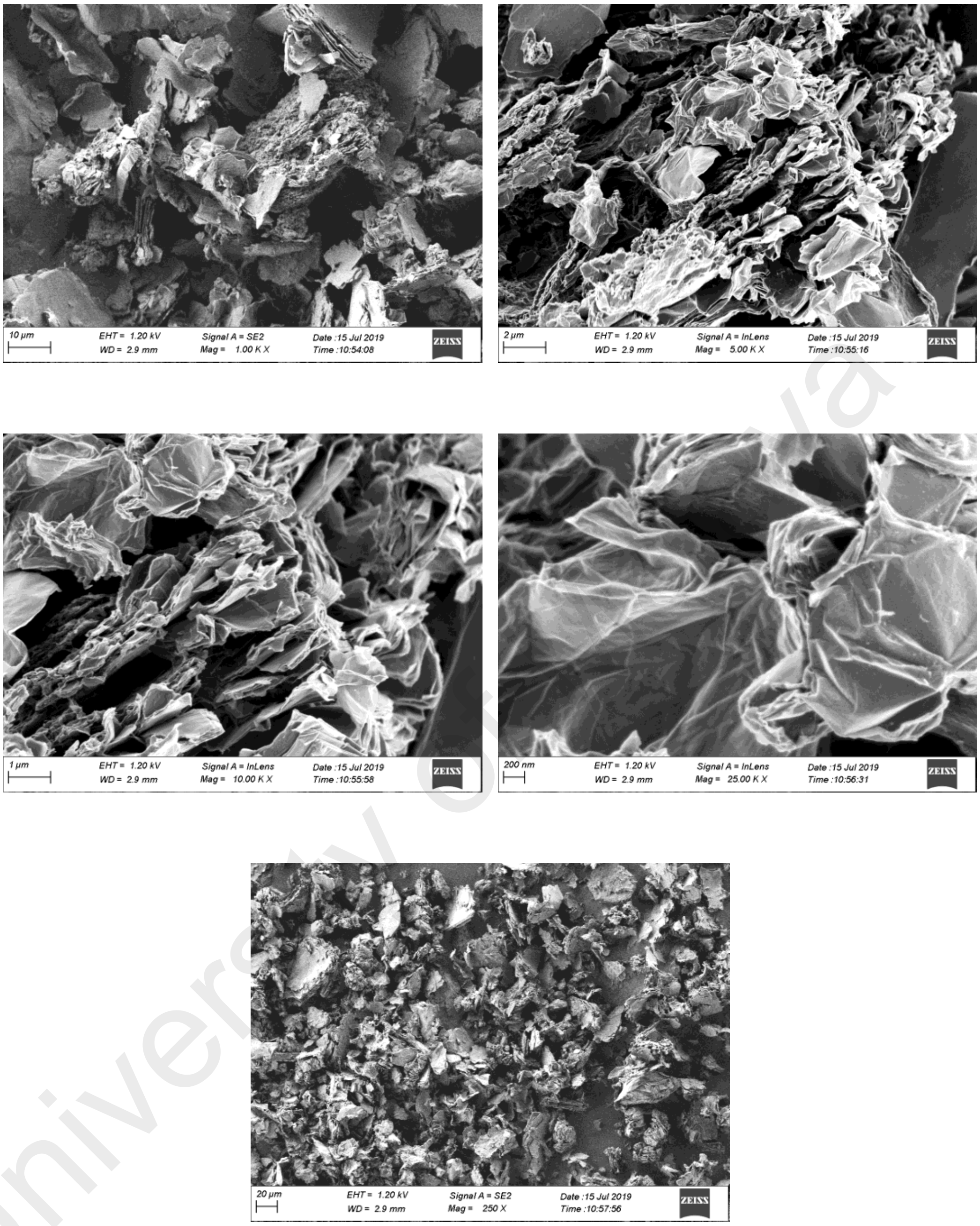

Figure 4.3 The FESEM images of GO powder with magnification of (a) 250x, (b) 1,000x, (c) 5,000x (d) 10,000x and (e) 25, 000x

$KMnO_4$ powder that added into the solution were acted as an oxidation agent. A $KMnO_4$ were believed to have activated species of Mn_2O_7.The equation is believed interaction between potassium permanganate and sulphuric acid:

$$KMnO_4 + 3H_2SO_4 \rightarrow K^+ + MnO_3+ + H_3O^+ + 3HSO_4^-$$

$$MnO_3^+ + MnO_4^- \rightarrow Mn_2O_7$$

It is believed the edge of graphite was oxidized and intercalated with the aid of the oxidizing agent. The hydroxyl group –OH was formed during this process. Figure 4.2 above shows SEM morphologies of GO which is dried 24 hours at 110° (Cao & Zhang, 2015).

The removal of –OH and the other functional groups from GO during rapid exfoliation of GO results in flakes and wrinkled structure. The high magnification result also confirms the wrinkled and flakes structure presence in GO sample. It clearly can be seen at magnification 25,000x. This is supported with journal where GO was reduced with thermal treatment in the oven for 24 hours (M. Ghorbani, 2015).

The presence of GO in the sample were confirmed by using EDS graph shows in Figure 4.4 below. Based on the EDS, C and O elements are found with C 72.02 wt% and O, 27.97 wt% respectively shows in Table 4.1 below.

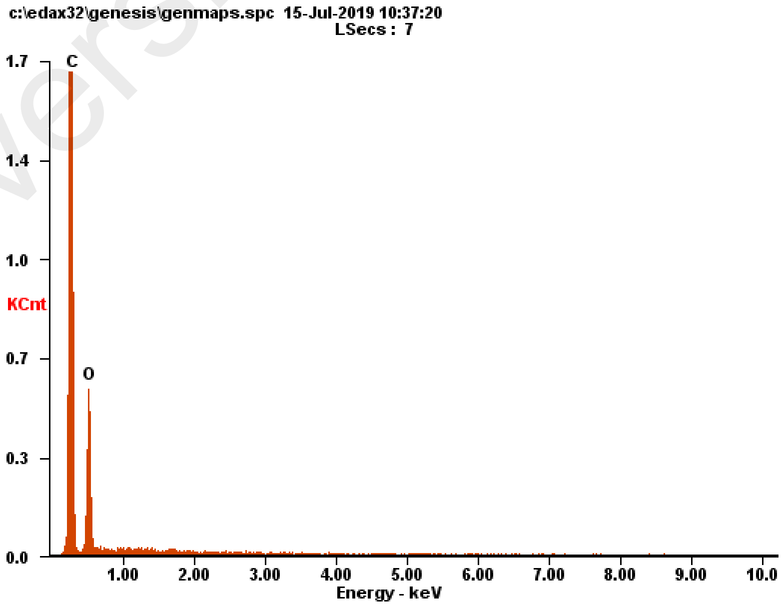

Figure 4.4 EDS Graph for GO Powder

There are no other element can be found during the analysis and the GO powder is free from any impurities. The presence of the carbon peaks in the GO powder shows GO also contains small quantity of carboxyl and hydroxyl groups (Muhammad Irfan Raza, 2017).

Table 4.1 Element present in GO powder

Element	Wt%	At%
CK	72.03	77.43
OK	27.97	22.57

4.1.2 Graphene Oxide-Nickel Oxide (GO-NiO)

The characteristic peak of GO-NiO is obtained 2ϴ = 26.4237° shows in Figure 4.5 below with *d-spacing* of 0.337314 nm, this study confirmed there are present of NiO nanoparticles in the GO composites. This is supported based on conducted study done by Muhammad Irfan Raza (2017), where the reflection peak of Ni-GO found when 2ϴ = 37.4°. This is due to the indication formation of graphene by the reduction process of graphene oxide. In addition, the presence of Ni in the compound were confirmed by EDS at Graph 4.7 below.

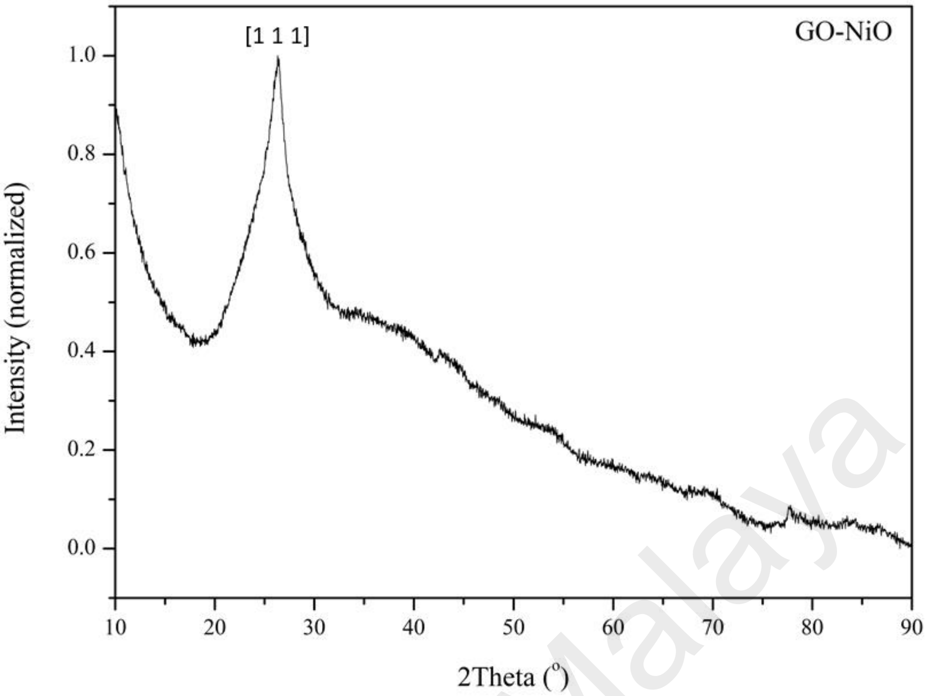

Figure 4.5 XRD graph of GO-NiO powder.

Based on study conducted by Gopiram (2019), the XRD result shows the presence wide peak for Ni-GO was observed at 2Ɵ = ~25° and the result can supports our finding. This is because the metallic nickel nanoparticles show a strong diffraction peaks at 2Ɵ = 44.45°, 51.73° and 76.84°. The broadness of the peaks of GO-NiO has revealed it has small size nanocrystalline of nickel oxide particles. Besides, widen of the peak also can be causes composition of nickel is small compare to carbon element in the sample. (Fakhri, Husnah, Aimon, & Ferry, 2017)

The presence of NiO nanoparticles on the surface of GO was confirmed using XRD analysis to measure the distance between two layers. The structural characterization of the sample carried out by powder sample with angle 2Ɵ recorded from 10° to 90°. The structural of GO-NiO is a polycrystalline structure. The interaction of the incident rays with the sample produces constructive interference and a diffracted ray when the conditions satisfied with Bragg's Law. Bragg's law relates the wavelength of electromagnetic radiation to the diffraction angle and the lattice spacing (*d-spacing*) in a crystalline sample. Bragg's law is calculated using below equation:

$$n\lambda = 2d \sin \theta$$

d – distance between atomic layers in a crystal

λ – wavelength of the incident x-ray beam

n – an integer

Due to the random orientation of the powdered sample, scanning the sample trough a range of 2ϴ angles, the possible diffraction of the lattice was attained. The *d-spacing* of the sample has its own unique distance.

Figure 4.6 below is believed shows magnification images of NiO nanoparticles are attached to GO sheets with the linkage between Ni_2^- and –COOH groups at the edge of GO sheets, induced by the electrostatic interactions.

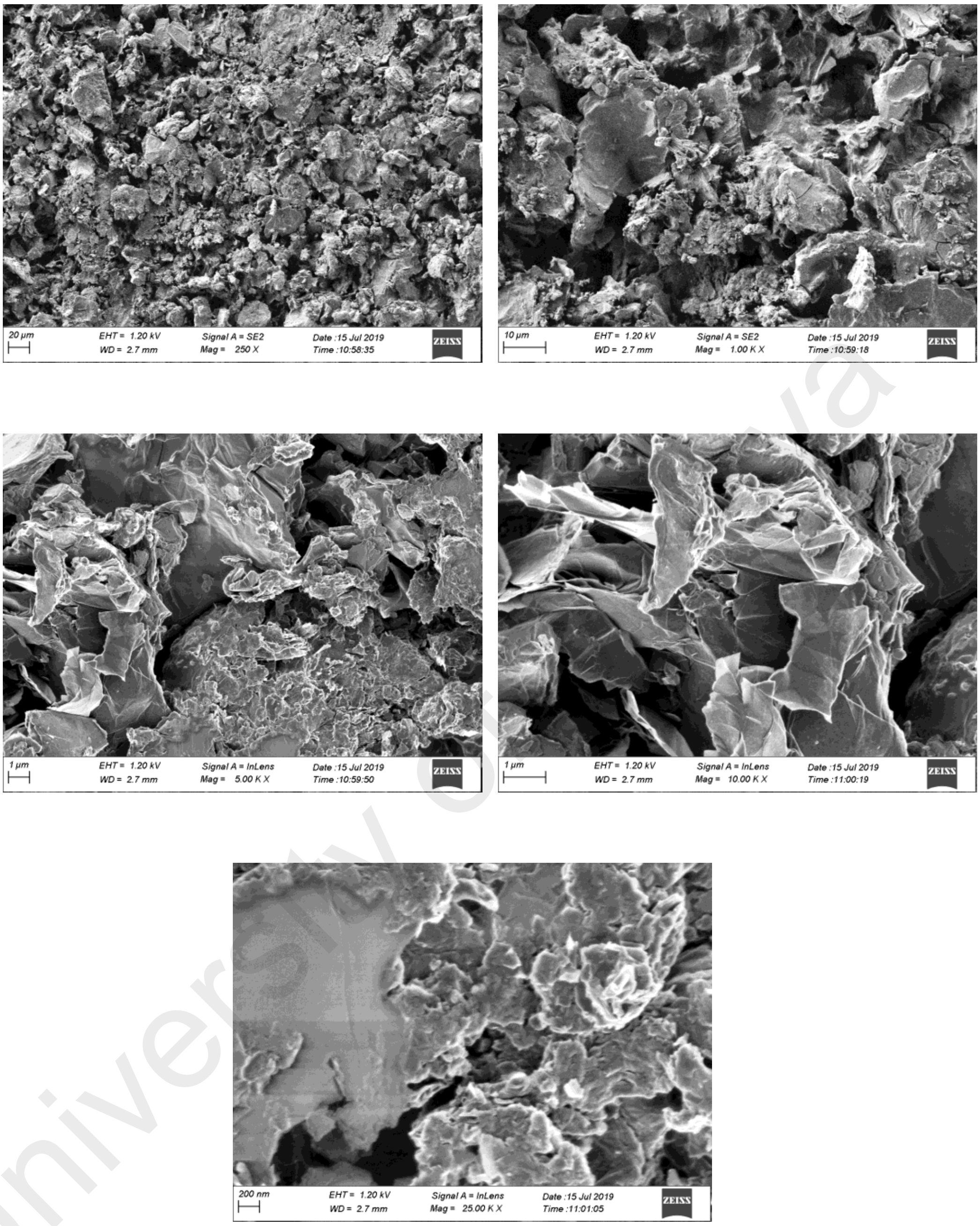

Figure 4.6 The FESEM images of GO-NiO powder with magnification of (a) 250x, (b) 1,000x, (c) 5,000x (d) 10,000x and (e) 25, 000x

The less concentration of –OH groups on the surface of GO may result in the weak attachment of NiO nanoparticles to the sheets. The mixture of nickel into GO leads to structural changes. The oval-shaped structural elements typical for dimpled structure transformed into polygonal with clearly defined angles and edges. However, the clear oval-shaped structural of NiO cannot be seen using FESEM. To have clear images needs to use TEM machine. As it seen from Figure 4.6 FESEM images above, the typical structure elements of pure nickel surface are spherical agglomerates with characteristic linear dimensions (V.G. Konakov, 2017)

However, in our study find, the structure of GO were widely can be seen in GO-NiO powder. This is due to the weight percent of Ni presence in the sample only 21.22% after the oxidation and reduction hydrothermal process occurred. Graph 4.7 below shows the EDS graph peak of element presence in the sample. The EDS graph shows result of analysis of GO-NiO powder.

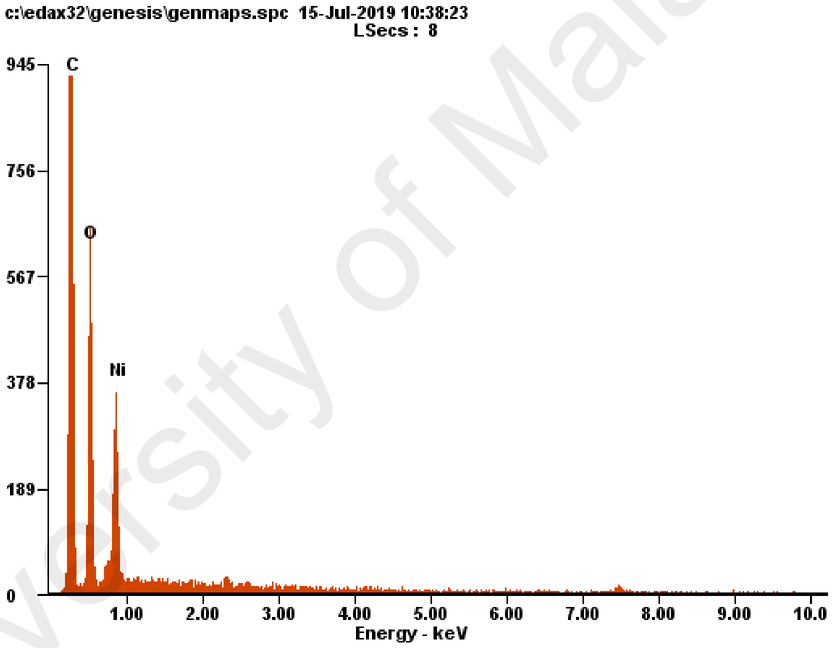

Figure 4.7 EDS graph of GO-NiO Powder

The EDS analysis spectrum were composed of C, O and Ni element with weight percentage is tabulated in Table 4.2 below.

Table 4.2 Element present in GO-NiO Powder

Element	**Wt%**	**At%**
CK	55.14	71.39
OK	23.65	22.99
NiL	21.22	05.62

The value properties for both GO and GO-NiO obtained during XRD analysis including *d-spacing* and peak position, 2Ɵ is summarized in Table 4.3 below.

Table 4.3 Properties of GO and GO-NiO obtained by XRD analysis.

Material	Peak Position, 2Ɵ (°)	*d-spacing* [Åm]
GO	26.3648	3.38054
	42.5602	2.12422
	54.4030	1.68651
	77.5937	1.22941
GO-NiO	26.4237	3.37314
	77.6119	1.23018

As we make comparison in Figure 4.8 below between XRD peak of GO and GO-NiO, there are not much differences in the analysis. As tabulated in Table 4.3 above, sharp peak of GO and GO-NiO can be seen at 2ϴ=26.3648° with *d-spacing* of 0.338054 nm and 2ϴ=26.4237° with *d-spacing* of 0.337314 nm respectively.

Most of conducted study for GO will have high peak when angle around 8° to 10° due to oxidation of graphite (Arthi G & Bd, 2015). The sharp peak present at around the range of GO graph is due to presence of oxygen functional groups in the layers (Cao & Zhang, 2015) (Arshadi Rastabi et al., 2019). However, in this study found wide peak appeared at 26.3648° and it may be causes by the poor arrangement of the graphene along the stacking direction. The weak diffraction peak indicates the structure of GO is in amorphous structure. The broadening of the peak can be causes by the poor order of GO sheets along the stacking direction. It were revealing that there are many nanosheets presence in the sample

Apparently, the characteristic sharp peak of GO did not appeared in our study and only widen peak obtained. The product was completely oxidized after the chemical and exfoliation procedure (Seyyedeh et.al, 2017) (Arthi G & Bd, 2015).

Also when make comparison, the diffraction peak intensity of both GO and GO-NiO found in this study shows in Figure 4.8 below, it shows both of the diffraction peak is around 26°. There are no obvious different diffraction of peaks. This is can be cause by its amorphous structure and low content in the composite. The different in the peak of intensity can be cause by the crystalize size of NiO is bigger than GO (Arifin, et.al, 2016).

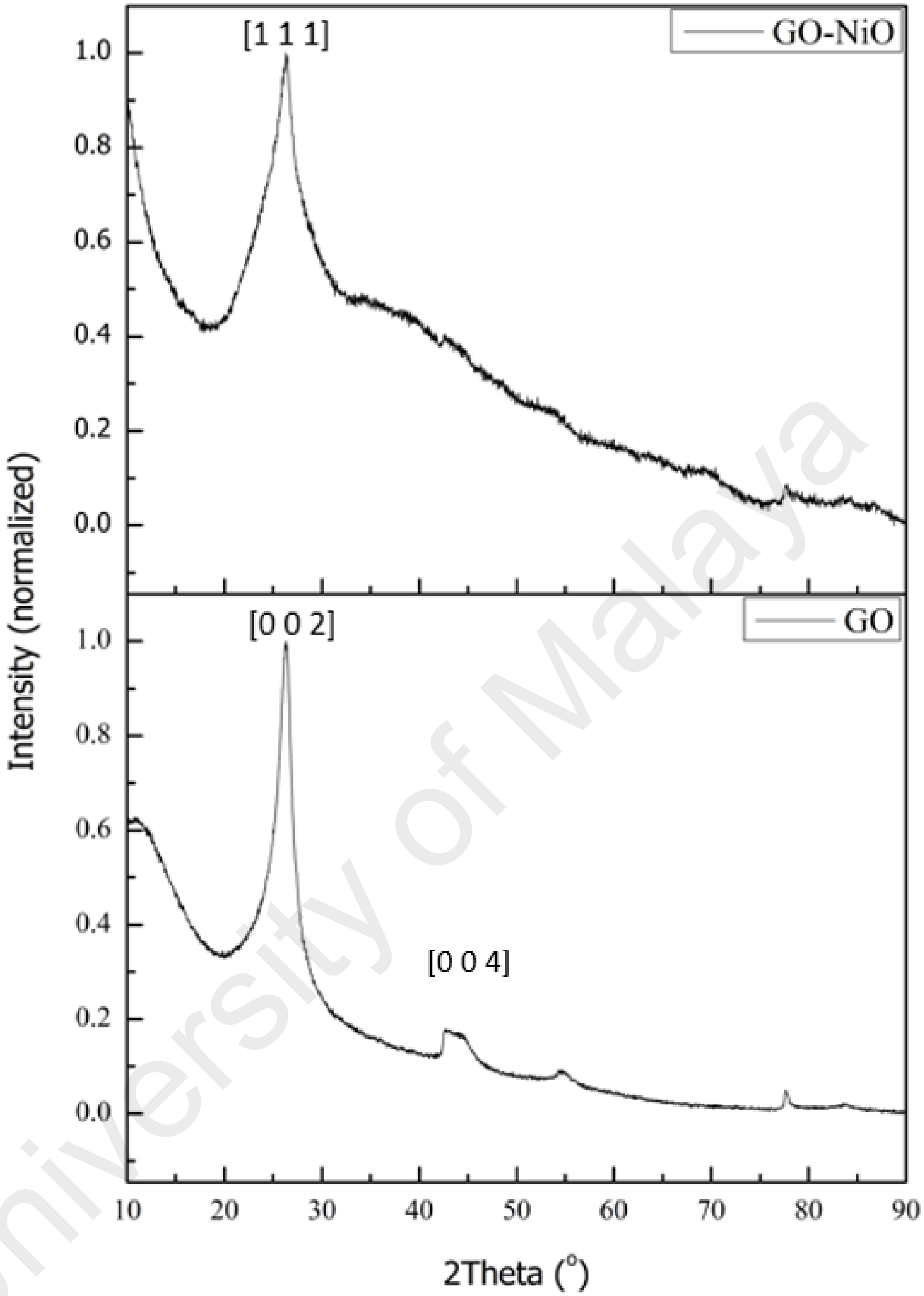

Figure 4.8 XRD graph for comparison between GO and GO-NiO powder.

There is conducted study stated that the normal peak diffraction of GO were found around 8° to 15°, however different value of peak find in our study. This can be lead from various factor whose affected the peak diffraction. One of the factor is can be causes by the crystallization of the sample. Both GO and GO-NiO powder has polycrystalline structure. It is believe, GO-NiO has high crystallinity. Other than that, because of our sample did not undergo enough thermal reduction and the value of 2Ө is similar with flake graphite peak of diffraction of 26.42° (Syed Nasimul Alam, 2017). Other than that, GO-NiO should have high ability as a conductor of electricity. Crystallites of the powder sample for both GO and GO-NiO are assumed to have random oriented structure.

4.2 Synthesis of PEO, GO-PEO and GO-NiO-PEO using FESEM and EDS

Polyethylene oxide (PEO) is added into the sample and thin film were produced. Three sample being analyzed by using FESEM and EDS.

Surface morphology and elemental composition of GO and GO-NiO nanocomposites were examined by field-emission scanning electron microscope (FESEM). The FESEM images is shows the morphologies 5 samples of nanocomposites that have been taken at different magnification. The images of samples was captured with voltage of 1.0 kV at magnification of 250x, 1,000x, 5,000x, 10,000x and 25,000x. The lower accelerating voltage were used to get more through images as it produces clearer images.

In FESEM, the signal to noise ratio will determine the clearer of images. However, for thin film, the procedure cannot stay longer at one place as it will break the line structural. In this study, the images of five sample with different magnification taken at a different place.

Energy dispersive spectroscopy (EDS) was performed to ensure the element of components of the synthesized of Ni element has been successfully decorated into prepared GO.

4.2.1 Pure Polyethylene Oxide (PEO) Polymer Nanocomposites.

Figure 4.9 below show PEO analyzed by using different magnification and at images of taken at different places. The pure PEO samples show uniformly distributed particles on the thin film surface with 250x magnification. However, as increase the magnification of microscopy, it can be seen that there is crumple. It can be caused by PEO are not fully dissolved. On the other hand, the roughness of the sample surface has dependence on the thickness of polymerization layer of sample.

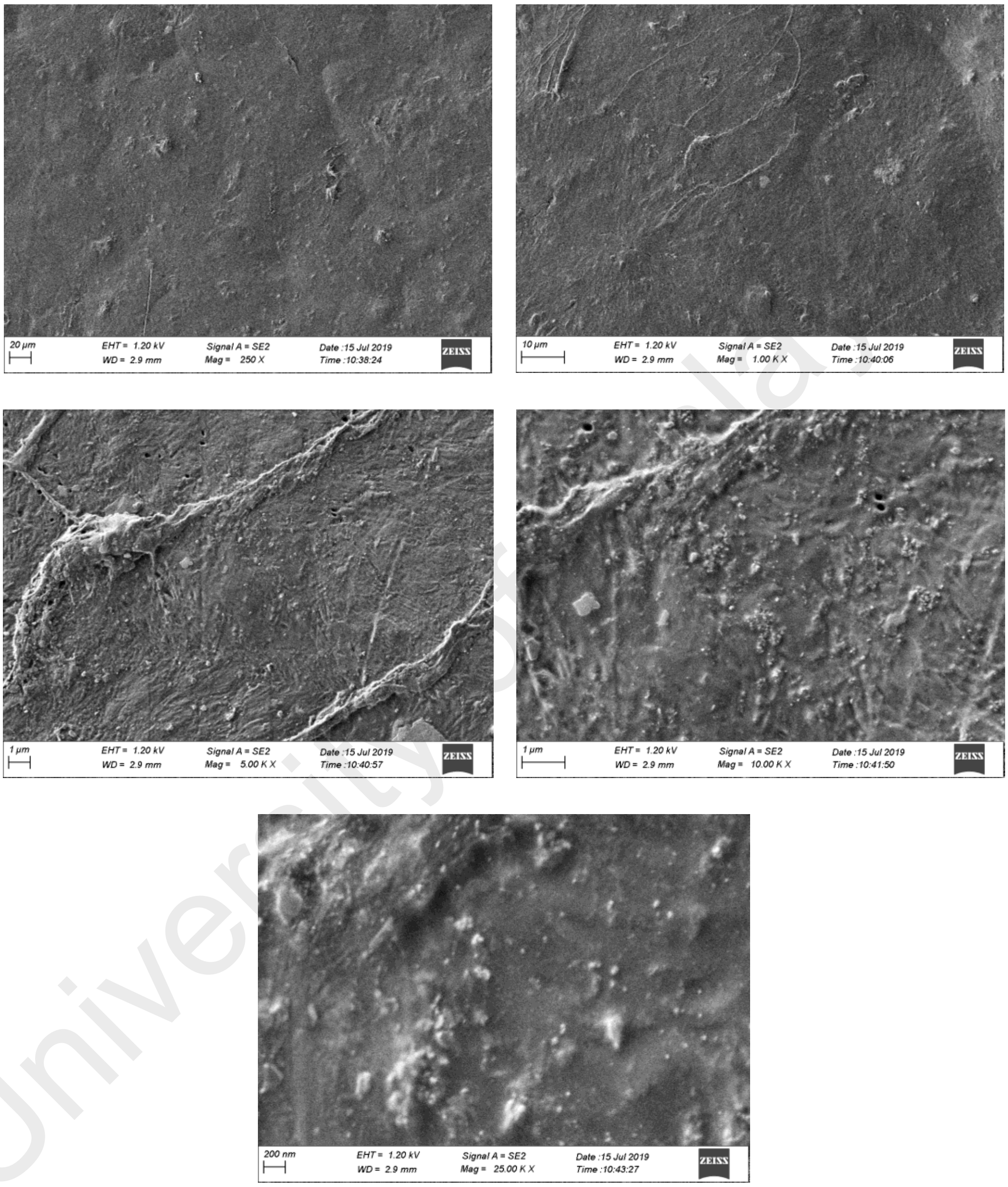

Figure 4.9 The FESEM images of pure polymer nanocomposite with magnification of (a) 250x, (b) 1,000x, (c) 5,000x (d) 10,000x and (e) 25, 000x

As shown in Figure 4.10 below, pure polymer only contained element of carbon with 57.03%, oxygen with 41.66% and silicone with 1.31% by showed the respective peaks.

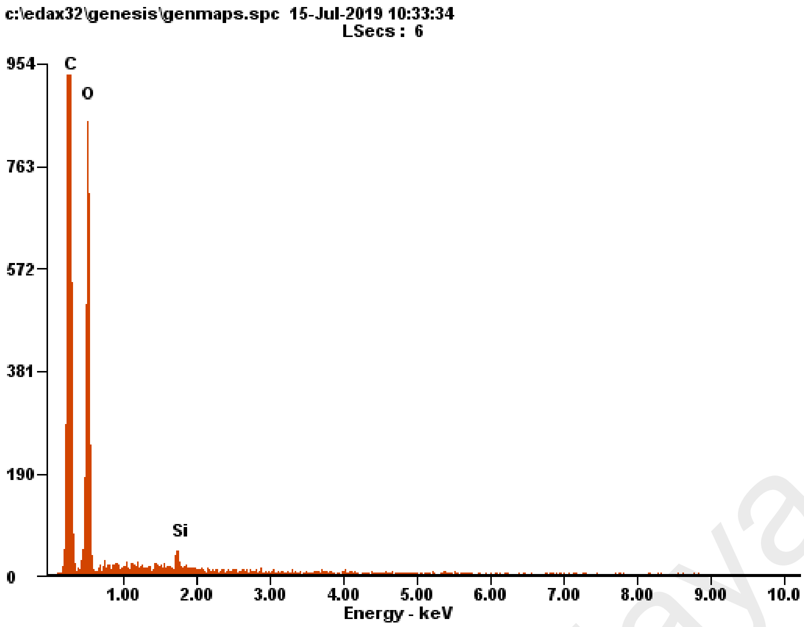

Figure 4.10 Pure polymer (PEO) thin film nanocomposite.

The value were of each element present in the sample tabulated in Table 4.4 below.

Table 4.4 Element present in PEO thin film nanocomposite

Element	Wt%	At%
CK	57.03	64.17
OK	41.66	35.19
SiK	01.31	00.63

4.2.2 Graphene Oxide (GO)-Polymer Nanocomposites.

From Figure 4.11 below shows the FESEM images analysis morphology of thin film of GO-polymer nanocomposites. With the addition of GO into the polymer matrix, it can be seen there are changes in the morphology of the sample.

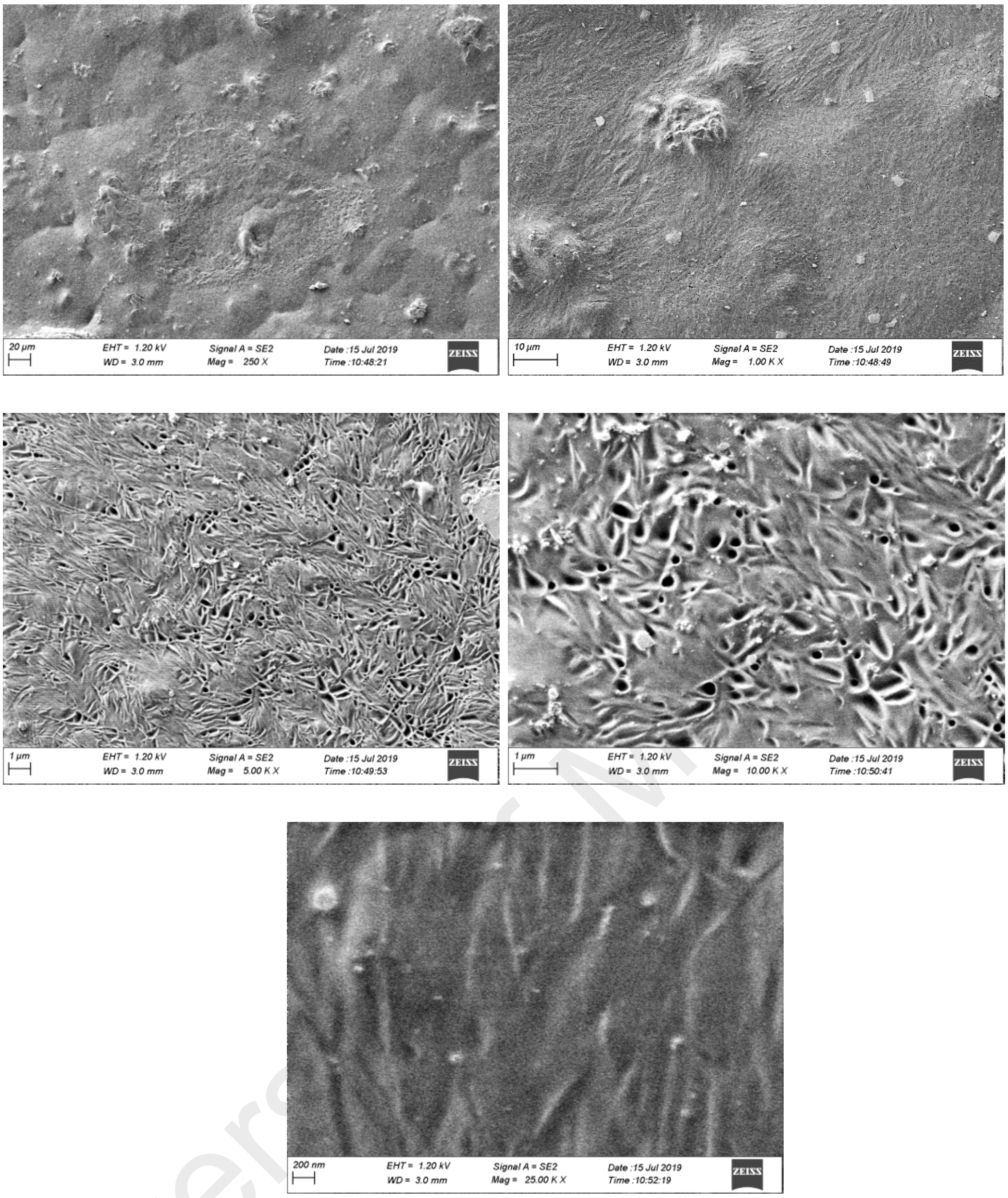

Figure 4.11 The FESEM images of GO-polymer nanocomposites with magnification of (a) 250x, (b) 1,000x, (c) 5,000x (d) 10,000x and (e) 25, 000x

This study find, normal configuration for single-layer of graphene has many wrinkles and the height fluctuate on its surface within range 0.5 nm (Feng Dai, 2015). The random morphology of graphene oxide can lead to unpredictable electronic properties.

Figure 4.12 below shows the EDS graph to shows the element presence in the GO-polymer nanocomposites.

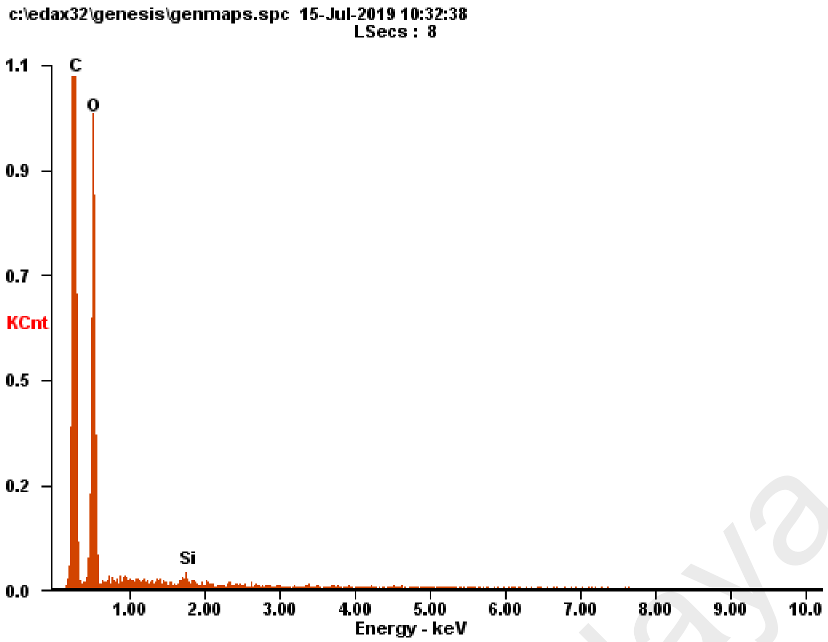

Figure 4.12 EDS graph of GO-polymer nanocomposite.

The weight percent of GO element were tabulated in Table 4.5 below.

Table 4.5 Element present in thin film of GO-polymer nanocomposite.

Element	Wt%	At%
CK	56.98	63.94
OK	42.52	35.82
SiK	00.50	00.24

4.2.3 Graphene Oxide-Nickel Oxide (GO-NiO)-Polymer Nanocomposites.

Figure 4.13 below shows the FESEM images of GO-NiO-polymer nanocomposites with different magnification and at different places. The graphene oxide-coated with NiO surface is visualized below.

GO found to be embedded in the Ni matrix. It was understood that once the GO deposited on the cathode and the Ni ions from the solution were attracted to the cathode and deposited on it quicker than on Ni. This is due to the higher current density of graphene oxide and resulted in GO dispersed in the Ni matrix. As can be seen from the FESEM images below, the surface morphology of GO-NiO is increased in roughness as an increase in the number of layers (Meng Li, 2017).

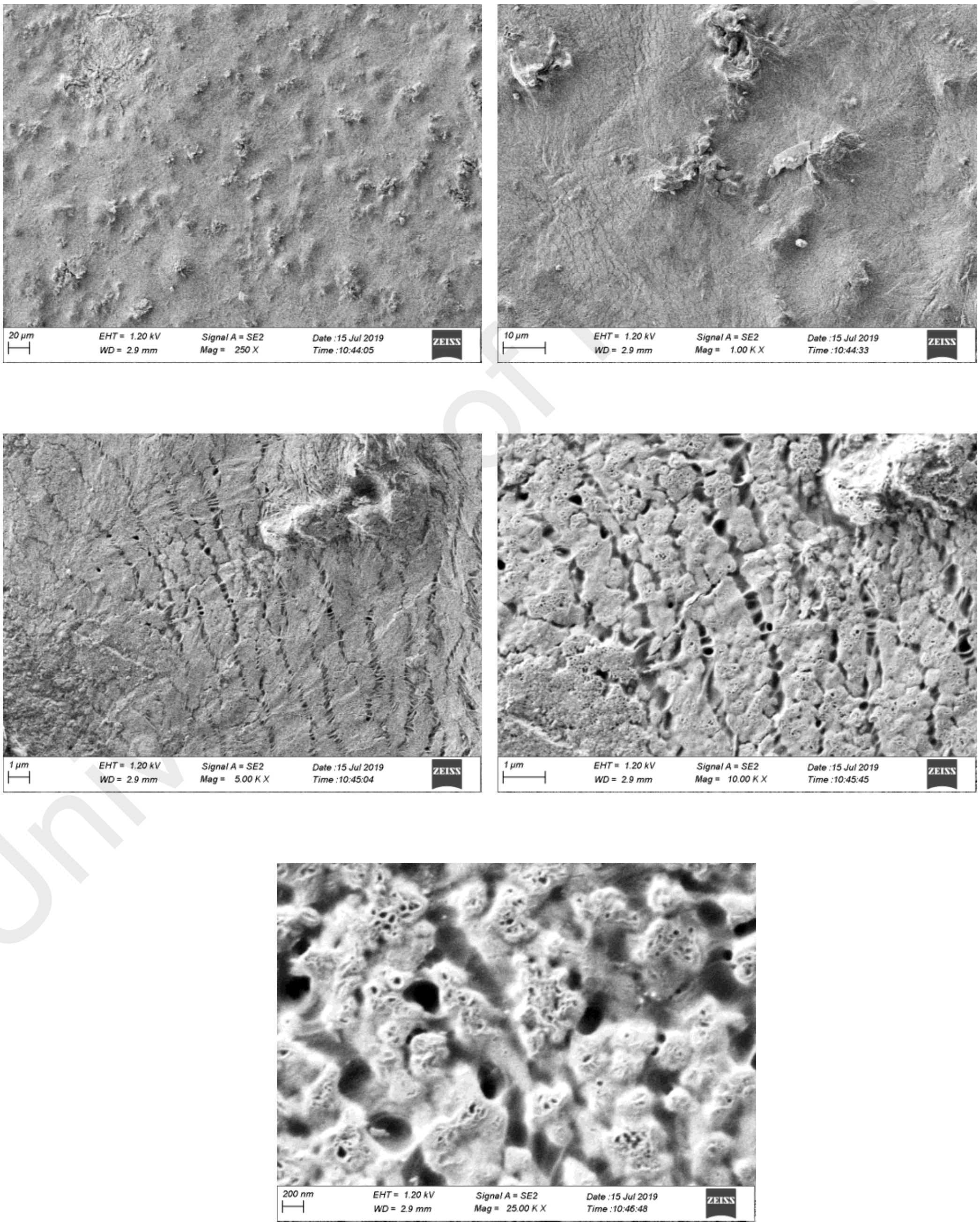

Figure 4.13 The FESEM images of GO-NiO-polymer nanocomposites with magnification of (a) 250x, (b) 1,000x, (c) 5,000x (d) 10,000x and (e) 25, 000x

GO-NiO-polymer nanocomposites is characterized by a high degree of surface development and incompletely build-up flakes of graphene can be seen. The formation of such an extensive structure on the surface of the coatings is the result of the tendency of the GO flakes to form conglomerates and of the good conductivity of the GO. As a result of an electric current transfer, the nickel deposits directly on the flakes leading to the development of the surface of the produced composite GO-NiO coatings (Grzegorz Cieślak, 2018).

Based on EDSD graph shows in Figure 4.14 below, the graph has showed peak for carbon, oxygen and nickel. Here can be conclude that nickel element has been successfully decorated in GO nanostructures.

From the Figure 4.14 below, clearly can be seen that the presence of well-defined peaks of Ni with 8.21% element confirmed the purity of GO-NiO powders. The presence of GO powder is confirmed by the presence of carbon element with 52.19% and oxygen element 39.23% in the EDS spectrum. The result has shown that the prepared nanocomposites are free from impurities. The EDS spectra confirmed there is the presence of C and Ni in the contaminants of surface roughness. With increase in the number of layers, it can caused the surface become rougher (Meng Li, 2017).

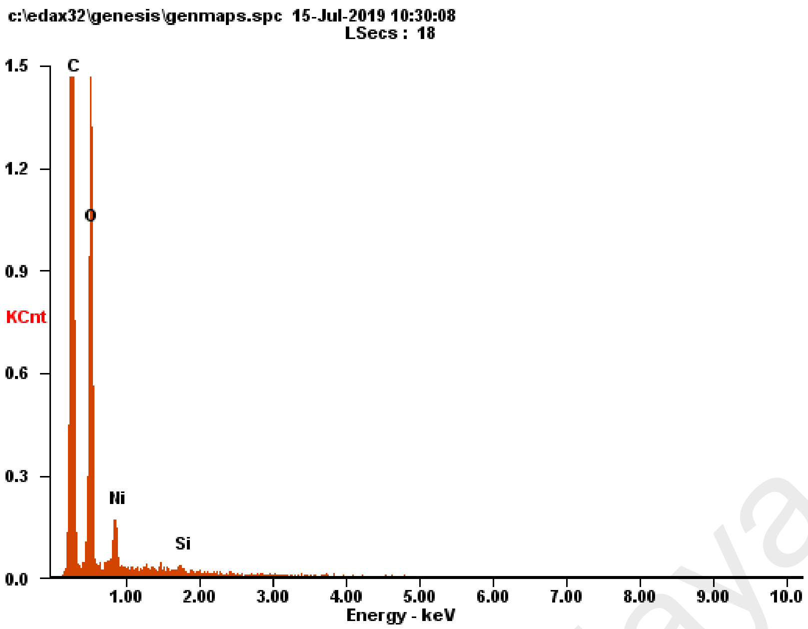

Figure 4.14 EDS graph for thin film of GO-NiO-polymer nanocomposites.

Table 4.6 below shows tabulated weight percent of all elements present in the GO-NiO-polymer nanocomposites sample.

Table 4.6 Element present in thin film of GO-NiO-polymer nanocomposites.

Element	**Wt%**	**At%**
CK	52.19	62.52
OK	39.23	35.28
NiL	08.21	02.01
SiK	00.38	00.19

As increase the thickness of the sample, the surface roughness will be increase. This can causes the affect to GO and GO-NiO. The physical properties of the graphene oxide is dependent on the thickness of the sample. The rough of the surface of the sample can because by the sample are not properly dried. The EDS spectra confirmed that only C, O and Ni element presence in the sample and no other contaminants caused the roughness.

4.3 Impedance Spectroscopy (IS)

Impedance Spectroscopy (IS) is a dielectric spectroscopy that measures the dielectric properties of the sample as a function of frequency. It is based on the interaction between an external field and the electric dipole moment of the sample. In this study, IS were used to study the conductivity ability of the sample.

4.3.1 Electrical Properties Analysis using Impedance Spectroscopy

Dielectric spectroscopy data can be analyzed in different terms of a) dielectric permittivity, b) conductivity, c) electric modulus and d) complex impedance. In this study, we are focusing on the conductivity. Dielectric permittivity is measured in medium of the function of frequency where it measures the interaction of an external field with the electric dipole sample.

In our study, we are focusing on the conductivity ability of the sample. This stud focus on the ability of the sample to allow the passage of an electron. The conductivity of a material were measured using below equation:

$$\sigma = \frac{l}{RA}$$

Where

l = electrolyte thickness between two electrodes

A = surface area of the electrolyte

R = total bulk electrolyte of resistance

Graphene oxide has extraordinary of property with high electrical conductivity. This is because the dispersion of GO in a polymer matrix enables the transition of neat polymer from electrical insulator to electrical conductor. (Menchaca-Campos et al., 2018)

To measure the conductivity of the sample was measured using Impedance Spectroscopy machine. As each sample was analyzed using HIOKO software to measure the impedance of the conductivity of the electrolyte, the Nyquist graph was obtained. Figure 4.19 below shows the Nyquist plot for pure polymer sample. The Nyquist graph shows below represent for the plotting of imaginary versus real part of the complex impedance.

The average thickness of sample was measured and tabulated in Table 4.7 below. The thickness of the sample thin film is not uniform due to the solution did not equally poured at constant rate.

Table 4.7 Average thickness of thin film polymer samples.

Sample thin film of polymer	**The average thickness of sample thin film (mm)**
Pure PEO	0.12
PEO with 5 wt% of GO	0.11
PEO with 5wt% of GO-NiO	0.10

4.3.2 Pure Polyethylene Oxide (PEO)-Polymer Nanocomposites.

The semicircle of the Figure 4.15 below is obtained where it represented the information of bulk electrolyte behavior. The curves to the right of the semicircle characterize the properties of the electrode. Since pure polymer did not contain any metal element that can conduct electricity (Poi Sim Khiew, 2013).

In the impedance plots of Figure 4.15 below, the value of bulk resistance is estimated around 8.56 x 10^6 Ω. The conductivity of PEO-polymer calculates based on the bulk resistance is 2.85 x 10^{-10} S cm^{-1}.

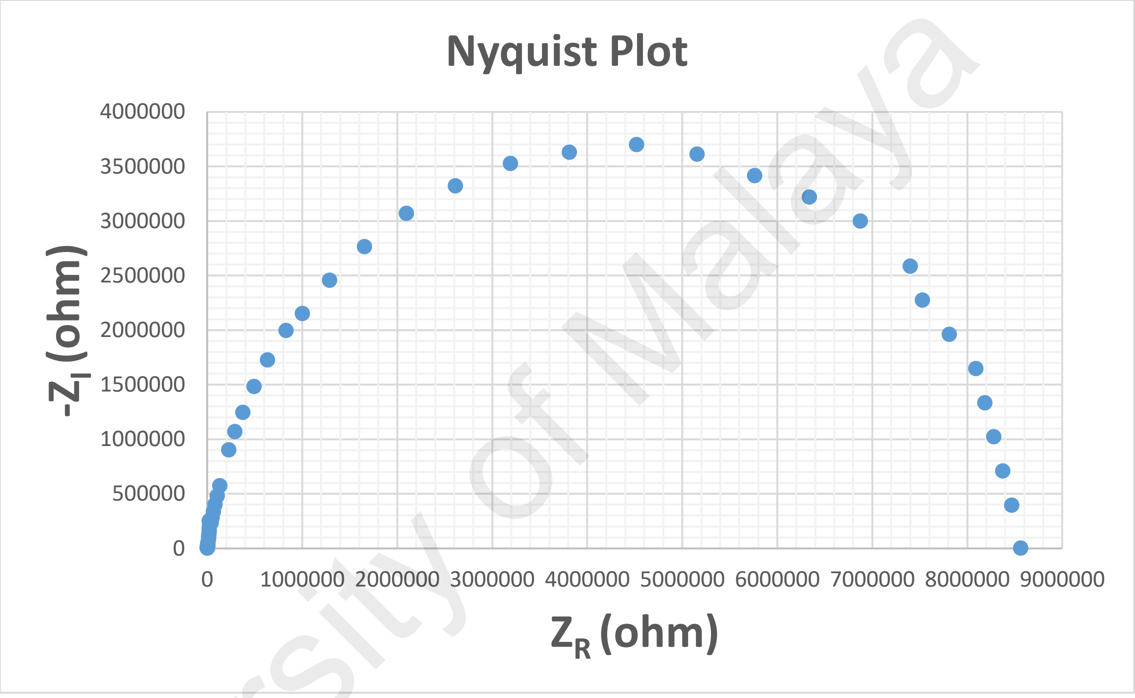

Figure 4.15 Nyquist Graph for thin film of pure PEO-polymer nanocomposites.

The Figure 4.15 above shows it have large diameter. This can be due to presence of high resistance cause by non-conducting behavior of pure PEO-polymer nanocomposites. Other than that, it was observed the semicircle of the plot is lessen when increase the value of resistor.

4.3.3 Graphene Oxide-Polymer Nanocomposites.

The semicircle Figure 4.16 below is shown the characteristic of a single time constant. The equivalent of the Nyquist plots are based on the normal circuit with a resistor and capacitor. The bend of the semicircle of the plotted graph is indicated there is charge transfer resistance in the sample. Based on Figure 4.16 below, this study estimated the value of bulk resistance is 7.1 x 10^6 Ω. Based on calculated electrical impedance, the value of conductivity is 3.156 x 10^{-10} S cm^{-1}.

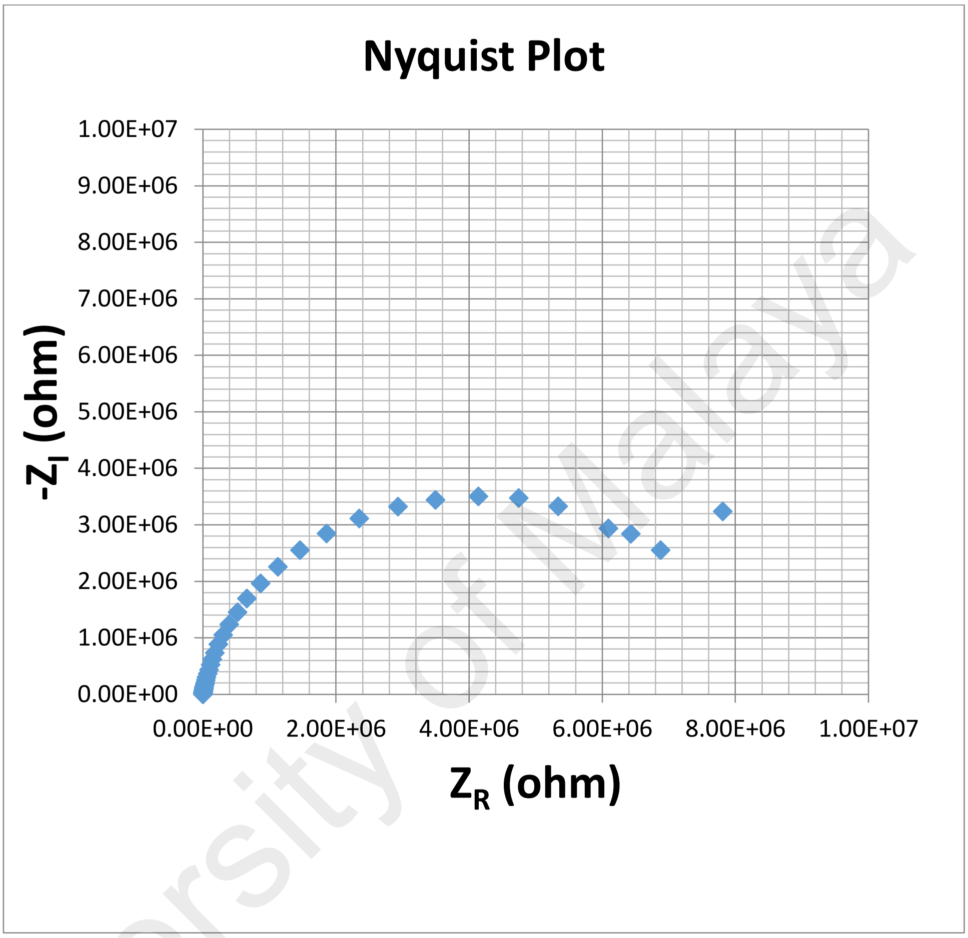

Figure 4.16 Nyquist graph for thin film of GO-polymer nanocomposites.

The high resistance of GO is due to the existence of oxygen-containing groups. The real portion in the low frequency range strongly related to the resistor value measured on the Nyquist graph, imaginary portion.

The general criteria influence the dielectric features of graphene oxide containing PEO nanocomposites can be the state of dispersion, the significant reinforcement of graphene oxide to the PEO matrix, the sample thickness also the amount of filler loading.

4.3.4 Graphene Oxide-Nickel Oxide (GO-NiO)

The semicircle part of the Figure 4.17 below shows the ohmic resistance (R_s) where it is related to the electron transfer limited process and revealed the process of electron transfer and its conductivity. While the linear part of the graph is related to the diffusion process. The smaller the electron transfer resistance is better in the electrode. This proves the synergistic effect of GO and GO-NiO nanoparticles at the electrode (Menchaca-Campos et al., 2018).

Based on study conducted, it is found that GO-NiO were shows the same resistor behavior as GO. According to EDS result tabulated in Table 4.6 above, the value of Ni presence in the sample only 8%. It doesn't affect much the electrical conductivity even Ni were believed to have high electrical conductivity due to its metallic nature. Therefore, the value of resistance of Nyquist graph were estimated with 1.26 x 10^6 Ω and the value of conductivity of GO-NiO-polymer nanocomposites is estimated around 1.61 x 10^{-9} S cm^{-1} (Olena Okhaya, 2015).

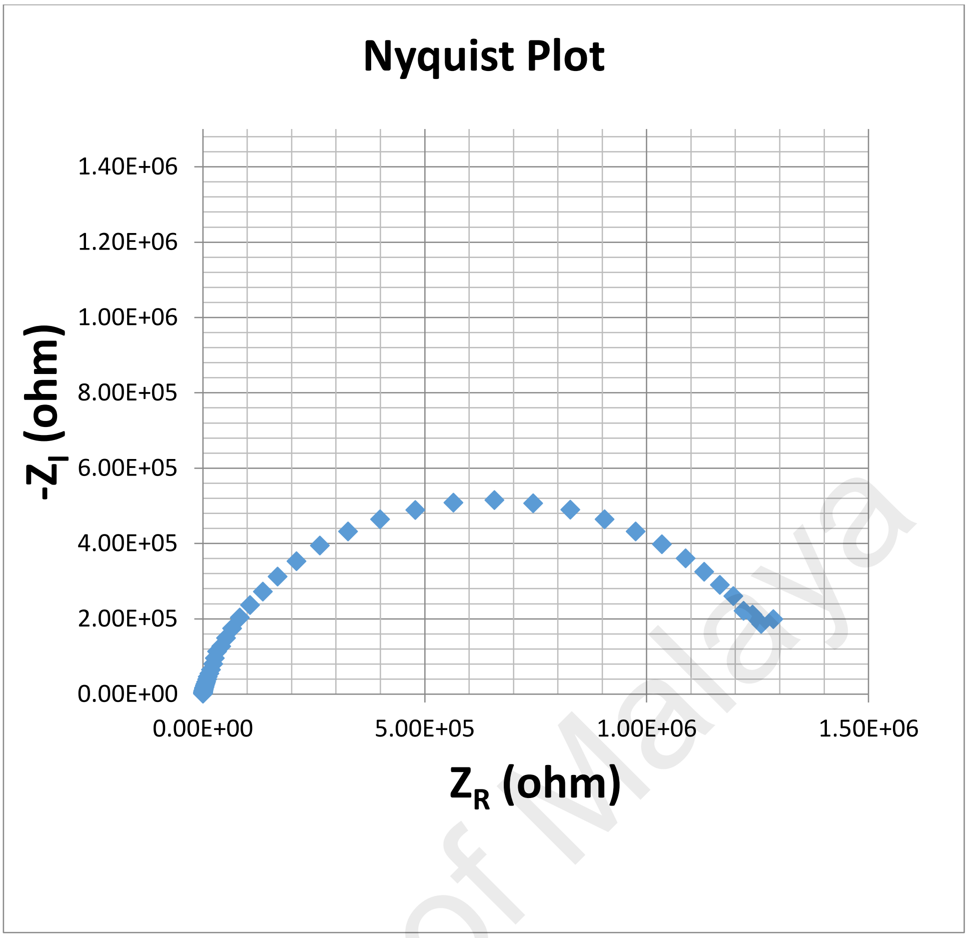

Figure 4.17 Nyquist plot for thin film of GO-NiO-polymer nanocomposite.

Based on this study found, there are no significant different between all two samples of PEO-polymer nanocomposites and GO-polymer nanocomposites cause all it has same magnitude. Nonetheless, the conductivity of GO and GO-NiO thin film have not much different in the value. The conductivity value for all three samples were tabulated in Table 4.8 below.

GO-NiO-polymer nanocomposites has the higher ability to conduct an electron. This is because GO-NiO-polymer nanocomposites contains metal elements, nickel. PEO-polymer and GO-polymer not have significant different as both sample has same magnitude. However, GO-polymer has more ability as a conductor compared to PEO-polymer. The radius of semicircle is indicative for conductivity and the charge transfer resistance of the sample.

Table 4.8 below is the summary conductivity value for all three samples of polymer nanocomposites.

Table 4.8 Value of Conductivity for three samples.

Sample thin film	**Conductivity (S cm^{-1})**
PEO-polymer nanocomposites	2.85×10^{-10}
GO-polymer nanocomposites	3.156×10^{-10}
GO-NiO-polymer nanocomposites	1.61×10^{-9}

In this study found for GO-NiO-polymer nanocomposites dielectric properties showed by the presence of semi-circle in the Nyquist graph, Figure 4.17 above. The probability values of conductivity were similar is due to long time exposure of the nanocomposites at room temperature. This is because, when the sample exposed to room temperature, the adsorbed oxygen molecule can capture free electron of NiO and formation of charge depletion layer which induces the resistance.

In this study, it found Nyquist plot for GO-polymer nanocomposites and GO-NiO-polymer nanocomposites, Figure 4.16 and Figure 4.17 respectively were contains two parts which are semicircle portion and straight-line portion. At the high middle frequency region, the semicircle portion reflects the change transfer process around the electrolyte interface. While for lower frequency range it is the impedance process. The smaller diameter of the semicircle represents the reduced charge transfer resistance. Enhanced electron transfer with fewer resistance properties was observed using highly conductive reduce graphene oxide nanocomposite. The polarization rate was depressed due to the reduced charge transfer resistance. The line is caused by mass transfer due to ion diffusion.

Nickel has advantages for the synthesis of complex graphene oxide-based materials. For instance, the high catalytic activity of Ni allows the growth of graphene oxide on graphene oxide-related sp2 carbon structures at fairly low temperatures on prepatterned Ni –films to form patterned graphene oxide structure on substrates. High-temperature evaporation of the nickel catalyst may be used to just leave the graphene oxide behind on dielectric support without the presence of metal. However, by addition of nickel in graphene oxide, is a good choice for graphene oxide to become efficient electrical material. Nickel has efficient electric contact with graphene oxide. The strong chemical interaction between graphene oxide and nickel may improve the contact. It can lower the contact resistance. A single graphene oxide layer on Ni can protect the Ni-surface from corrosion. This is a useful property, especially for material like Ni that easily oxidizes when exposed to atmospheric condition. A graphene oxide layer on a Ni-film may protect it from oxidation during the growth of the oxide dielectric and this may result in a sharper metal or oxide interface (Dahal & Batzill, 2014).

CHAPTER 5: CONCLUSION

5.1 Conclusion

In summary, graphene oxide successfully synthesized by using modified Hummer's method. By analysis the element present using EDS, it has been confirmed that for GO, only carbon and oxygen elements are existed in the synthesized GO. Other than that, FESEM images shows the morphology of the GO in the flakes condition.

Next, GO were successfully decorated with Nickel oxide. The GO-NiO has been synthesized using hydrothermal method. The presence of Ni in the element were confirmed by EDS with peak shows 21.22%.

Electrical properties of GO and GO-NiO were discussed by Nyquist graph. Since GO-NiO consists elements of GO and NiO, the nanocomposite has the ability to conduct an electricity. This can be confirmed by the presence of the semicircle and straight 45° line in the Nyquist graph and the value of conductivity of GO-NiO tabulated in Table 4.8 above. This confirmed there is conductivity and also there is diffusion where related to movement of an electron to indicate there is electrical conductivity activity presence in the sample. In this study conclude, there are a slight different in reading of conductivity between sample of GO and GO-NiO. This is due to presence of nickel in the element. It is estimated 28% improvement of electrical conductivity were recorded for GO-NiO was compared to NiO.

5.2 Suggestion for future work

In this research, there were some obstacles encountered during the synthesis process. As the washing process takes a long time, the comprehensive modified Hummer's method can be studied, future. The study should focus on the best solution which is suitable to wash the graphene dispersion in less amount of time.

As graphene oxide has no band gap, further studies can be conducted to introduce artificial band gap. Furthermore, there can also be additional studies on the commercialization of the graphene with low cost but has the best properties, to be manipulated for future advanced technology.

There are various studies that have been carried out to investigate the parameters and quality of GO. GO produced under high temperature parameters can cause more defects to the structure it is applied to. This could lead to a low-quality GO-based structure. However, the quality can be increased by introducing some annealing time to the GO, under extremely high temperature (Hai Tan, 2018).

Other than that, GO decorated with metal oxides, especially Ni, has excellent properties and can be a promising candidate for several other applications. Therefore, more in-deep studies need to be conducted to know the effect of Ni addition to the electrical properties of GO made components. Besides, more study of GO decorated with other metal oxides also can be conducted to study the effect of the conductivity properties.

More analysis on graphene oxide decorated with nickel oxide by using TEM or FTIR can be done. The morphology and the surface roughness of GO can also be further studied, as the effect of GO substrate interaction remains elusive.

REFERENCES

Anu Saini, A. K., Vijay Kumar Anand, Suresh Chander Sood. (2016). Synthesis of Graphene Oxide using Modified Hummer's Method and its Reduction using Hydrazine Hydrate. *International Journal of Engineering Trends and Technology (IJETT), 40*(Number 2), 67-71.

Arifin, M., Iskandar, F., Aimon, A. H., Munir, M. M., & Nuryadin, B. W. (2016). Synthesis of LiFePO4/Li2SiO3/reduced Graphene Oxide (rGO) Composite via Hydrothermal Method. *Journal of Physics: Conference Series, 739*. doi:10.1088/1742-6596/739/1/012087

Arshadi Rastabi, S., Sarraf Mamoory, R., Dabir, F., Blomquist, N., Phadatare, M., & Olin, H. (2019). Synthesis of NiMoO4/3D-rGO Nanocomposite in Alkaline Environments for Supercapacitor Electrodes. *Crystals, 9*(1). doi:10.3390/cryst9010031

Arthi G, P. B., & Bd, L. (2015). A Simple Approach to Stepwise Synthesis of Graphene Oxide Nanomaterial. *Journal of Nanomedicine & Nanotechnology, 06*(01). doi:10.4172/2157-7439.1000253

Avouris, P., & Dimitrakopoulos, C. (2012). Graphene: synthesis and applications. *Materials Today, 15*(3), 86-97. doi:10.1016/s1369-7021(12)70044-5

Bai, Y. (2017). Reduced Graphene Oxide/Nickel Oxide/Polyaniline: Preparation and Properties Investigation as Supercapacitor Electrode Material. *International Journal of Electrochemical Science*, 652-662. doi:10.20964/2017.01.20

Cao, N., & Zhang, Y. (2015). Study of Reduced Graphene Oxide Preparation by Hummers' Method and Related Characterization. *Journal of Nanomaterials, 2015*, 1-5. doi:10.1155/2015/168125

Dahal, A., & Batzill, M. (2014). Graphene-nickel interfaces: a review. *Nanoscale, 6*(5), 2548-2562. doi:10.1039/c3nr05279f

Fakhri, H. A., Husnah, M., Aimon, A. H., & Ferry, I. (2017). Preliminary Study on Synthesis of Composite rGO/Ni by Microwave Assisted Method. *Journal of Physics: Conference Series, 877*. doi:10.1088/1742-6596/877/1/012073

Gopiraman, M., Saravanamoorthy, S., Deng, D., Ilangovan, A., Kim, I. S., & Chung, I. M. (2019). Facile Mechanochemical Synthesis of Nickel/Graphene Oxide Nanocomposites with Unique and Tunable Morphology: Applications in Heterogeneous Catalysis and Supercapacitors. *Catalysts, 9*(5). doi:10.3390/catal9050486

Gotoh, K., Kinumoto, T., Fujii, E., Yamamoto, A., Hashimoto, H., Ohkubo, T., . . . Ishida, H. (2011). Exfoliated graphene sheets decorated with metal/metal oxide nanoparticles: Simple preparation from cation exchanged graphite oxide. *Carbon, 49*(4), 1118-1125. doi:10.1016/j.carbon.2010.11.017

Grzegorz Cieślak, M. T. (2018). Preparation and properties of nanocrystalline Nigraphene composite coatings deposited by electrochemical method. *Journal of Checmical Technology, 20*(1), 29 - 34. doi:10.2478/pjct-2018-0005

Hai Tan, D. W. a. Y. G. (2018). Thermal Growth of Graphene- A Review. *MDPI, 40*(8). doi:10.3390/coatings8010040

Halder, A., Zhang, M., & Chi, Q. (2016). Electrocatalytic Applications of Graphene–Metal Oxide Nanohybrid Materials. In Advanced Catalytic Materials - Photocatalysis and Other Current Trends.

Hussain, N., Gogoi, P., Khare, P., & Das, M. R. (2015). Nickel nanoparticles supported on reduced graphene oxide sheets: a phosphine free, magnetically recoverable and cost effective catalyst for Sonogashira cross-coupling reactions. *RSC Advances, 5*(125), 103105-103115. doi:10.1039/c5ra22601e

Lacina, K., Kubesa, O., Horáčková, V., Moravec, Z., Kuta, J., Vanýsek, P., & Skládal, P. (2018). Graphene Oxide from Improved Hummers' Method: Is This Material Suitable for Reproducible Electrochemical (Bio)Sensing? *ECS Journal of Solid State Science and Technology, 7*(10), M166-M171. doi:10.1149/2.0171810jss

M. Ghorbani, H. A., M. R. Golobostanfard. (2015). Graphene nanosheets by low-temperature thermal reduction of graphene oxide using RF-CVD. *Procedia Materials Science*, 326-330. doi:10.1080/17458080.2012.676679

Md. Sajibul Alam Bhuyan, M. M. I., Md. Nizam Uddin & Ferdaushi Alam Bipasha. (2016). Synthesis of graphene. *Int Nano Lett, 6*, 65–83. doi:10.1007/s40089-015-0176-1

Menchaca-Campos, C., Pereyra-Laguna, E., García-Pérez, C., Flores-Domínguez, M., García-Sánchez, M. A., & Uruchurtu-Chavarín, J. (2018). Synthesis and Characterization of Reduced Graphene Oxide/ Polyaniline/Au Nanoparticles Hybrid Material for Energy Applications. In *Graphene Oxide - Applications and Opportunities*.

Meng Li, X. Z., Yaqing Tian, Jie Ren, Kyle Jiang, Jian Liu, Sai Priya Munagala. (2017). *Surface properties of graphene platelets-nickel composite coatings*. Paper presented at the IEEE International Conference on Manipulation, Manufacturing on the Nanoscale, Shanghai.

Muhammad Irfan Raza, E. S., Naseem Iqbal and Safeer Ahmed. (2017). *Synthesis of Graphene Supported Nickel and Cobalt Nanoparticles and Their Applications for Methanol Oxidation in Alkaline Medium*. Paper presented at the May 22-24, 2017 Kuala Lumpur (Malaysia) ICLTET-2017, ACBES-2017.

N.I. Zaaba, K. L. F., U. Hashima, S.J.Tan, Wei-Wen Liu, C.H. Voon. (2017). Synthesis of GO using modified hummers method-solvent influence. *Procedia Engineering*, 469-477. doi:10.1016/j.proeng.2017.04.118

Nasser A.M. Barakat, M. M., Zafar Khan Ghouri, Ahmed S. Yasin, Mohamed H. El Newehy & Salem S. Al-Deyab. (2016). Nickel nanoparticles-decorated graphene as highly effective and stable electrocatalyst or urea electrooxidation. *Journal of Molecular Catalysis A: Chemical*, 83-91. doi:10.1016/j.molcata.2016.05.011

Norhayati Hashim, Z. M., Mohd Zobir Hussein, Illyas Md Isa, Azmi Mohamed, Azlan Kamari, Suriani Abu Bakar, Mazidah Mamat, Adila Mohamad Jaafar. (2016). A brief review on recent graphene oxide-based material nanocomposites: Synthesis and applications. *Journal of Materials and Environmetal Science*(9), 3225-3243.

Olena Okhaya, L. M. G., João Ventura, Alexander Tkach, Mathias Kläui, Rahul Krishna and Elby Titus. (2015). *Electrical and Magnetic Properties of Graphene Films with Ni Nanoparticles.* Paper presented at the 5th International conference on Advanced Nano Materials.

Poi Sim Khiew, M. Y. H., Thian Khoon Tan, Wee Siong Chiu, Roslinda Shamsudin, Muhammad Azmi Abd-Hamid, and ChinHua Chia. (2013). Synthesis-and-Electrochemical-Characterization-of-Iron-Oxide--Activated-Carbon-Composite-Electrode-for-Symmetrical-Supercapacitor. *World Academy of Science, Engineering and Technology International Journal of Materials and Metallurgical Engineering, 7*(No 8).

Qiu, H., Qiu, F., Han, X., Li, J., & Yang, J. (2017). Microwave-irradiated preparation of reduced graphene oxide-Ni nanostructures and their enhanced performance for catalytic reduction of 4-nitrophenol. *Applied Surface Science, 407*, 509-517. doi:10.1016/j.apsusc.2017.02.159

S. Fahad, M. A., S. Ahmed, S. Khan, S. Alam, S. Akhtar. (2017). <91 Effect of Metal Contact and Rapid Thermal Annealing on Electrical Characteristics of Graphene Matrix.pdf>. *34*, 106801-106801 - 106801-106805. doi:10.1088/0256-307X/34/10/106801

Salimian, M., Ivanov, M., Deepak, F. L., Petrovykh, D. Y., Bdikin, I., Ferro, M., . . . Goncalves, G. (2015). Synthesis and characterization of reduced graphene oxide/spiky nickel nanocomposite for nanoelectronic applications. *Journal of Materials Chemistry C, 3*(43), 11516-11523. doi:10.1039/c5tc02619a

Seyyedeh Saadat Shojaeenezhad, M. F., Iraj Kazeminezhad. (2017). Effects of initial graphite particle size and shape on oxidation time in graphene oxide prepared by Hummers' method. *Journal of Science: Advanced Material and Devices, 2*, 470-475. doi:10.1016/j.jsamd.2017.09.003

Shen, J., Yan, B., Shi, M., Ma, H., Li, N., & Ye, M. (2011). One step hydrothermal synthesis of TiO2-reduced graphene oxide sheets. *Journal of Materials Chemistry, 21*(10). doi:10.1039/c0jm03542d

Syed Nasimul Alam, N. S., Lailesh Kumar. (2017). Synthesis of Graphene Oxide (GO) by Modified Hummers Method and Its Thermal Reduction to Obtain REduced Graphene Oxide (rGO). *Graphene, 6*, 1-18. doi:10.4236/graphene.2017.61001

V.G. Konakov, O. Y. K., I.V. Lomakin, N.N. Novik1, S.N. Sergeev, E.N. Solovyeva, A.P. Zhilyaev, I.Yu. Archakov and I.A. Ovid'ko. (2017). Graphene-Modified Bulk Nickel Composite - Investigation Of Structure And Mechanical Characteristics. *50*.

Vincenza Modafferi, S. S., Michele Fiore, Enza Fazio, Claudia Triolo, Salvatore Patanè,Riccardo Ruffo and Maria G. Musolino. (2019). Transition Metal Oxides on Reduced Graphene Oxide Nanocomposites Evaluation of Physicochemical Properties. *Journal of Nanomaterials, 2019*. doi:10.1155/2019/1703218

Xin Jiat Lee, B. Y. Z. H., Kar Chiew Lai, Lai Yee Lee, Suyin Gan, Suchithra Thangalazhy-Gopakumar, Sean Rigby. (2019). Review on graphene and its derivatives- Synthesis methods and potential industrial implementation. *Taiwan Institute of Chemical Engineers, 98*, 163-180. doi:10.1016/j.jtice.2018.10.028