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# Preliminary Study on Synthesis of Composite rGO/Ni by Microwave Assisted Method

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**Abstract**. Composite reduced graphene oxide-nickel (rGO/Ni) was successfully synthesized by wet chemical process via microwave assisted method. The nickel fraction was varied 0%, 5% and 20%. Crystallinity, morphology, chemical composition, chemical bonding, and conductivity were investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), elemental dispersive X-ray spectroscopy (EDS), Fourier transform infrared spectroscopy (FTIR), 4 point probes and electrochemical impedance spectroscopy (EIS). Based on the XRD patterns, the crystal sizes were 1.62 nm for 5% rGO/Ni and 1.64 nm for 20% rGO/Ni respectively. Furthermore, vibration of C=C aromatic stretching was detected in the FTIR spectra, which corresponds to rGO fingerprint. These results give a new perspective on synthesis of composite rGO/Ni.

## 1. Introduction

The lithium ion battery, which used as energy storage device, consists of three principle parts, i.e. anode, cathode and electrolyte [1]. Lithium iron phosphate ($LiFePO_4$) is a common material for the cathode. For anode, graphite is a general material for this purpose [2]. Several research groups have reported the improvement of lithium ion batteries [3-4]. Some of them replaced the graphite with graphene as an anode material [5]. Graphene is a carbon allotrope that has a two-dimensional structure [6]. Graphene has a high electrical and a good thermal conductivity, a wide surface and a high electrical mobility [7]. However, several researchers have reported synthesis of graphene by processes such as chemical solution, chemical vapor deposition, epitaxial growing, and micromechanical exfoliation [8-9]. From these methods, chemical solution method is an easier method with a high throughput. On the other side, transition metal is a type of inorganic material that has a good electrochemical behavior. Nickel is a transition metal that is abundant in Indonesia. To improve the anode performance, some researchers have composited graphene with a transition metal by microwave assisted method, hydrothermal method, or chemical vapor deposition method. Microwave assisted method have a rapid thermal process that easier to conduct than the other methods. In this study, graphene was composited with Ni (rGO/Ni) by microwave assisted method in order to improve the anode performance. Fourier Transform Infrared (FTIR), Scanning Electron Microscopy (SEM), Elemental Dispersive X -Ray Spectroscopy (EDS), and X-Ray Diffraction (XRD) were used to investigate the structural properties of the prepared sample. Further, 4 point probe measurement and

electrochemical impedance spectroscopy (EIS) was used to study the electronic property of the sample. This result may give a new horizon in the improvement of anode material.

## 2. Experiment

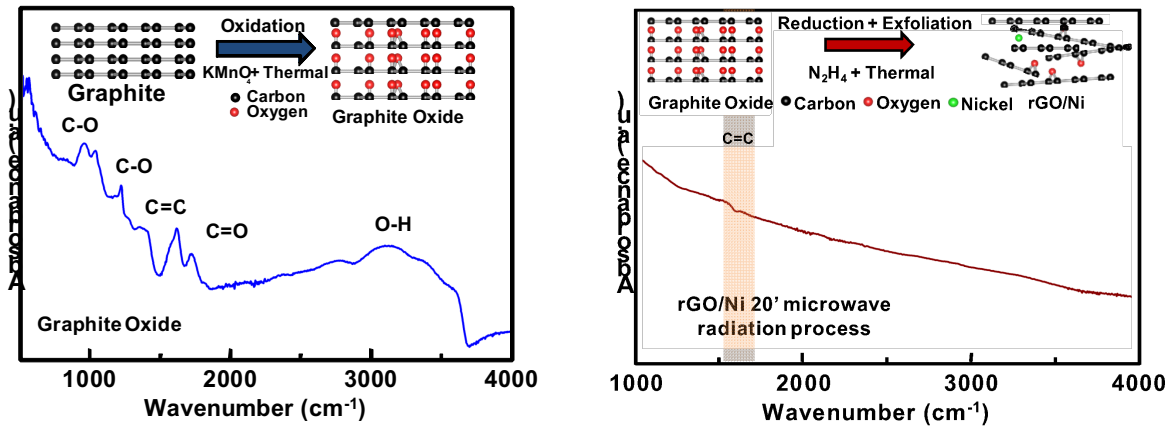
All chemical materials were used without further purification. Graphite oxide and nickel nitrate hexahydrate were used as precursors of rGO/Ni. The graphite oxide was synthesized from graphite powder (Aldrich). Sulfuric acid (Merck) and phosphoric acid (Brataco) as concentrated acids and potassium permanganate (Merck) as oxidizer agent were used to oxidize the graphite. The volume of concentrated acid was 22.5 ml for sulfuric acid and 2.5 ml for phosphoric acid, respectively, for 1 gram graphite. During this process, the temperature was kept below 10 °C. After three gram of potassium permanganate was added, the compound was stirred at 50 °C for 40 minutes. Then distilled water was added to the solution. After that 30% of hydrogen peroxide was added to stop the oxidizing process. The solvent and solute were then separated and the solute was washed using 5% hydrochloric acid, alcohol, and distilled water. Finally, the washed solute was heated for 12 hours at 60 °C in an oven.

In the composite process, the graphite oxide was added with ethylene glycol and nickel nitrate as nickel source. In the sonication process, the compound of GO, nickel nitrate and ethylene glycol was sonicated using a sonicator for 2 hours. Then hydrazine hydrate as reduction agent was dropped into the compound. The compound was reduced using a microwave assisted method for 20 minutes. The solvent and solute were separated and then washed again using alcohol and distilled water. After that the solute was heated for 12 hours at 60 °C in an oven.

Samples were characterized by XRD, FTIR, SEM, EDS and 4 point probes. X ray diffraction was used to investigate the crystal structure of the samples by a Raigaku D/max-RA X ray diffractometer with Cu K radiation. The morphology of the samples was studied using scanning electron microscopy (Jeol JCM-6000 Bench top SEM). FTIR measurement from Alpha FTIR Spectrometer was used to study the chemical bonding of the sample. Furthermore, the electrical conductivity was characterized using the 4-point probes method using an Adventest R6240A DC Voltage Current Source/Monitor as the current source. Further, the EIS measurement was done using Gamry Instruments Reference 3000$^{TM}$.

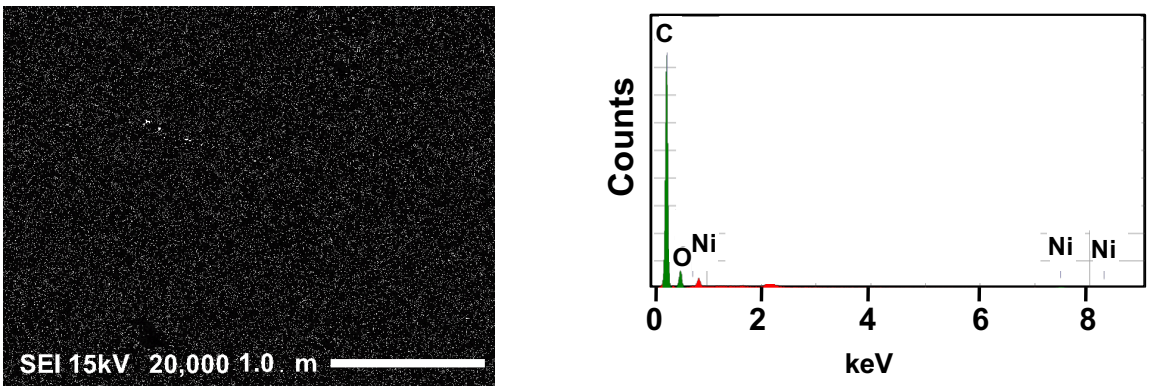
## 3. Result and Discussion

Figure 1 shows the FTIR spectra of the GO sample and the 5% rGO/Ni sample. The GO spectra show some peaks which relates to O-C bond at 1037 and 1220 $cm^{-1}$(epoxide group), O=C bind at 1718 $cm^{-1}$ (carbonyl group), C=C bond at 1613 $cm^{-1}$ and O-H bond at 3123 $cm^{-1}$ (hydroxyl group). These fingerprints indicate that GO was successfully synthesized by chemical process. Furthermore, Figure 1b shows the FTIR spectra of 5% rGO/Ni. Vibration of C=C aromatic bonding was found at ~1600 $cm^{-1}$, which corresponds to rGO. Vibration peaks of epoxide, carbonyl, and hydroxyl groups were not found in the spectra that indicate the sample (GO) was already reduced. Nickel bonding (< 500 $cm^{-1}$) was not found due to limitations of the instrument.




(a) (b)

**Figure 1.** Fourier transform infrared spectra of a) GO and b) Ni/rGO.




(a) (b)

**Figure 2.** a) SEM images of 5% rGO/Ni with 20.000 magnification and b) EDX characterization result of 5% rGO/Ni.

Figure 2a shows a SEM image of 5% rGO/Ni with 20,000 times magnification. The image shows the morphology of the sample as thin sheets. The atomic percentages from the EDX characterization were 86.61% for carbon, 11.91% for oxygen and 1.48% for nickel, respectively.

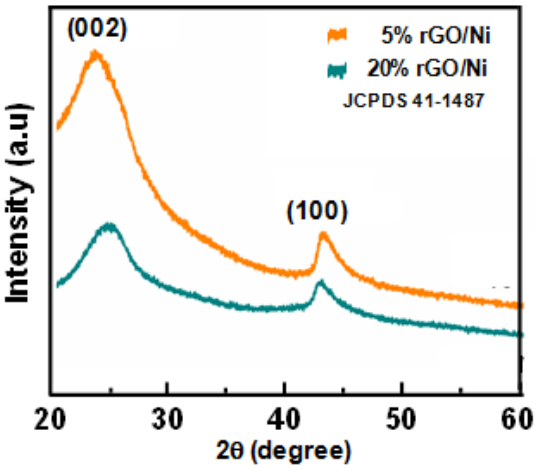



**Figure 3.** XRD pattern of 5% and 20% rGO/Ni.

The XRD patterns of the 5% and 20% rGO/Ni samples are shown in Figure 3, which correspond to graphite (JCPDS 41-1487) with diffraction peaks at 2 = 24.5° for (002) and at 2 = ~43° for (100) respectively. The graphite diffraction peak was widened at 20% sample due to stretching of the carbon layer distance. However, the nickel diffraction peaks could not be observed because the nickel was too small or its content in the sample was very slight compare to carbon content.

The electrical conductivity of the samples was measured using the 4-point probes method as shown in Figure 4a. Here, sample with 20% nickel had lower conductivity. It might be explained that bigger amount of Ni in the sample attracts the oxygen in the sample and rearrange into nickel oxide. However, this conductivity value represents the intrinsic property of the sample as single material. Hence, the EIS measurement was done to evaluate the interaction of the rGO/Ni composite as anode material with its environment. The EIS result of the samples was shown in Figure 4b with charge transfer resistance (Rct) value 3Ω, 0.86Ω, and 0.37Ω and surface resistance (Rsf) value 21.6 Ω, 9.36 Ω, and 3 Ω for 5%, 8% and 20% rGO/Ni composite, respectively. On contrary with conductivity result, the EIS result shows lower Rct and Rsf value for 20% nickel in the composite. This result shows that rGO/Ni composite can give a high charge transfer between the anode and the electrolyte and possibly to diminish the solid electrolyte interface (SEI) formation in the anode. Hence, rGO/Ni composite is potentially applied as an anode material.

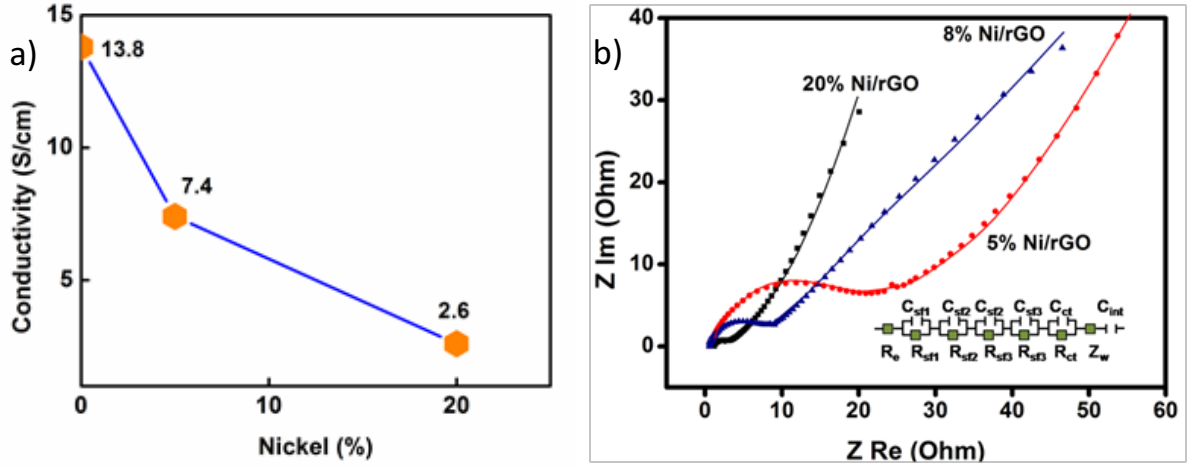



**Figure 4.** Four point probes result (a) and Nyquist plot of rGO/Ni composites.

## 4. Conclusion

Composite rGO/Ni was successfully synthesized by microwave assisted method with varying nickel content in the sample. FTIR result for rGO sample shows C=C aromatic stretching which confirms the formation of rGO sample. SEM image shows the sample in thin sheets formations which contain 86.61% carbon. Based on EIS results, rGO/Ni composite can give a high charge transfer between the anode and the electrolyte and has possibility to diminish the solid electrolyte interface (SEI) formation in the anode. Therefore, as a conclusion, rGO/Ni composite can give potential application for an anode material with microwave assisted synthesis method.

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