Performance of electrodes Mg/Graphene nanosheet (GNS) and Mg/N-Graphene nanosheet (N-GNS) as anode of battery

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Abstract. The aims of this research are to know the electrode preparation method of Mg/GNS and Mg/N-GNS, and to evaluate performance Mg/GNS and Mg/N-GNS as anode of battery. This research is an experimental laboratory research. GNS and N-GNS were synthesized by using modified Hummer and N dopants method, respectively. The electrode of Mg/GNS and Mg/N-GNS were synthesized with impregnation method. GNS, N-GNS, Mg/GNS and Mg/N-GNS characterized by using SEM-EDX and Conductivity. The research data shows that GNS and N-GNS are well formed (SEM data) and indicates that Mg metals were deposit well on GNS and N-GNS confirmed with EDX data. The smallest Mg particles size are found on Mg 1.84%/GNS and Mg 2.26%/N-GNS, those are 0.337 and 0.353 µm. Meanwhile, Mg 1.82%/GNS and Mg 2.15%/N-GNS have the largest Mg particles, those are 0.481 and 0.495 µm (SEM data). Conductivity data show, the highest conductivity numbers are 69.9301 µS/cm for Mg 1.86%/GNS and Mg 2.43%/N-GNS (96.1538 µS/cm), comparing among Graphite (79.15 µS/cm) and a node of commercial primary battery (10 µS/cm). All of data conclude that the character and performance of Mg/GNS and Mg/N-GNS may be modified with supporting materials GNS and N-GNS, respectively.

1. Introduction

Batteries are electrochemical devices that convert chemical energy into electrical energy during the process of exchanging electrons between the anode and cathode [1]. The main components of the primary battery are carbon on the anode and carbon metal alloy at the cathode [2]. That is, the quality and cost of producing primary batteries is determined by their constituent materials (cathodes, anodes, and electrolyte pastes) [3]. Therefore, we need an alternative new material that can be used to produce primary batteries in large quantities [4]. Graphene has become an interesting material to study specifically for electrochemical energy storage devices [5]. Graphene is a carbon monolayer layered with 2D lattice [6]. Features of graphene: carbon 2D [7], sp2 [8], the surface area of 2600 m2g-1 [9], very high strength [10], thermal conductivity (4840–5300 WmK-1) [11,12] and electrical conductivity (1250 Scm-1) [13] and graphene can be considered to be an energy storage material for Li and Na ions in battery ions, for example Li/Graphene has a specific capacity (744 m Ah g-1) and Li/Graphite (372 m Ah g-1) [14].
Electrons and therefore the ability of the battery cathode to deliver will be better. The main problem with development battery is that the price of lithium is very expensive and becomes waste after usage, the density and energy capacity of primary batteries is still low, weak interactions between electrodes and transport of electrons and loss of electrical contact at long usage times. The solution to overcoming this problem is to use graphene.

2. Methodology
2.1. General
This research used an experimental method which was carried out in the Laboratory of Analytical Chemistry, Chemistry Department, Universitas Sumatera Utara for preparation of materials. Characterization of samples of SEM-EDX instrument was carried out in the Laboratory of Materials, National Taiwan University Science and Technology. Electrical conductivity test was carried out in the Laboratory of Basic Physics Sciences, Universitas Sumatera Utara.

2.2. Materials preparation
There are some materials that used in this research like graphite commercial powder (carbon 98 wt%, ash wt 15%), sulfuric acid (H₂SO₄, 98 wt%), potassium permanganate (KMnO₄, 99.5 wt%), hydrogen peroxide (H₂O₂, 30 wt%), and ammonia (NH₃, 25 wt%) were purchased from Sigma-Aldrich (Singapore). Sodium nitrate (NaNO₃, 99 wt%) and Magnesium chloride (MgCl₂, 99 wt%) were purchased from Merck (Singapore). All the chemical reagents as approved without any further purification.

2.3. Synthesis of graphene oxide
Graphene oxide was synthesized base on graphite powder by using a modification of Hummers and Offenman’s method [15]. Briefly, 0.2 g of graphite was added with 0.2 g NaNO₃ and 15 ml H₂SO₄, were stirred together for 1 hour in an ice bath condition. The end of 1 hour, 1.0 gram of KMnO₄ was slowly added into the solution and stirred for 1 day to generate graphite solution. Finally, 20 mL of H₂SO₄ 5% and 1 mL H₂O₂ 30%, stirred for 1 hour, respectively. The solution was centrifuged at 6.500 rpm for 20 minutes and ultrasonicated for 6 hours, cropping a solution of graphene oxide.

2.4. Synthesis of graphene
Graphene oxide solution was added with a 5 mL of NH₃ 10 M and stirred for 3 days, filtered and dried at 80°C to produce graphene powder.

2.5. Preparation of Mg/GNS
As much as 1.0 g of graphene is put into the beaker glass which contains 1.0 ppm standard magnesium series solution. Then, put the magnetic bar into the beaker glass and stirrer them for 2 hours. Then filtered using Whatman filter paper no. 42, will be become filtrate and sediment. The filtrate was not given any treatment. While the sediment was weighed and tested using SEM-EDX. The same treatment will be done for the 2.0 standard magnesium series solution; 3.0; 4.0; 5.0 and 10 ppm, then Mg/GNS was characterized by SEM-EDX.

2.6. Preparation of Mg/N-GNS
As much as 1.0 g of N-graphene was put into the glass beaker which already contained 1.0 ppm Magnesium standard solution. Insert the magnetic bar into the beaker glass and cook for 1 hour. Then filtered using Whatman filter paper no. 42. So as to obtain filtrate and sediment. The filtrate was not given any treatment. While the precipitate obtained was weighed and characterized using SEM-EDX. The same is done for the standard Magnesium 2.0 solution; 3.0; 4.0; 5.0 and 10 ppm.
2.7. Measurement of electrical conductivity
Put 0.25 g of graphite powder into the fuse, compacted, then covered with a fuse cover. The crocodile clamp cable is connected to the negative and positive poles of the Digital multimeter and Regulated DC Power Supply. Measured electrical conductivity with variations in voltage 5, 10, 15, 20, 25, and 30 volts and recorded the current. The same treatment will be done for the graphene nanosheet powder; commercial primary battery anode, Mg 1.0; 2.0; 3.0; 4.0; 5.0; 10 ppm/graphene nanosheet powder and Mg/N-graphene.

3. Results and Discussions
3.1. SEM-EDX Analysis
3.1.1 Graphite, Graphene, and N-Graphene
Scanning Electron Microscope-Energy Dispersive X-ray (SEM-EDX) Images of graphite, graphene, N-graphene, and anode. SEM-EDX analysis used type of EM 30 COXEM with a voltage of 20,000 kV with enlargement 100-4000 times.

Figure 1a shows the surface of graphite which is piled up, solid, in the form of flakes and dense hexagonal structure piles. The surface of graphene is very different from graphite (Figure 1b). The structure of the graphene looks like a sheet and does not appear hexagonal structure, meaning that the graphene was successfully synthesized. The N-graphene surface shows that the N-graphene surface looks very different from graphene, where the surface looks more encased than graphene (Figure 1c). That is, the N atoms interact with the structure of graphene [16].
3.1.2. Mg/Graphene

Figure 2. SEM images of (a) 1.78 wt% Mg/GNS, (b) 1.80 wt% Mg/GNS, (c) 1.82 wt% Mg/GNS, (d) 1.84 wt% Mg/GNS, (e) 1.86 wt% Mg/GNS, (f) 1.90 wt% Mg/GNS with 5000x magnifications.

Based on (Figure 2), it is clearly seen that Mg metal is deposited in a diffused manner characterized by the presence of distributed white spots on the graphene surface at each Mg weight percent (1.78; 1.80; 1.82; 1.84; 1.86, and 1.90 %wt Mg/GNS). Weight concentration element variations wt% Mg/GNS in Table 1.

| Sample          | Weight Concentration element (%) | SUM |
|-----------------|----------------------------------|-----|
|                 | C    | O    | Mg   | SUM |
| 1.78 wt% /GNS   | 62.02 | 37.37 | 0.61 | 100 |
| 1.80 wt% /GNS   | 51.48 | 47.07 | 1.45 | 100 |
| 1.82 wt% /GNS   | 46.68 | 51.39 | 1.39 | 100 |
| 1.84 wt% /GNS   | 44.77 | 54.03 | 1.20 | 100 |
| 1.86 wt% /GNS   | 45.47 | 52.86 | 1.67 | 100 |
| 1.90 wt% /GNS   | 62.02 | 37.37 | 0.61 | 100 |

Based on the table above it can be seen that a lot of magnesium deposited in GNS varies at each weight percent of concentration Mg. To make it easier to see the relationship between particle size and variations in Mg concentration can be seen in Figure 3.
Figure 3. Mg particle size with variation Mg concentrations in graph

The graph above shows that at a concentration of Mg 1.78-1.80% / GBN Mg particle size decreased, then rose significantly at Mg 1.82% / GBN, and dropped significantly at Mg 1.84% / GBN and rose slowly until Mg 1.90% / GBN. Shows that Mg 1.82% / GBN has the highest particle size that is 0.481 µm and Mg 1.84% / GBN has the smallest particle size that is 0.337 µm. This is due to the number of Magnesium atoms in GBN, each of which is different so that the particle size also varies.

3.1.3. Mg/N-Graphene

Figure 4. SEM images of (a) 2.05 wt% Mg/N-GNS, (b) 2.13 wt% Mg/N-GNS, (c) 2.15 wt% Mg/N-GNS, (d) 2.17 wt% Mg/N-GNS, (e) 2.26 wt% Mg/N-GNS, (f) 2.43 wt% Mg/N-GNS with 5000x magnifications.
Based on (Figure 4) the surface of N-graphene shows more irregularity than graphene but it can be clearly seen the spots attached to the spread N-graphene sheet also indicate that Mg is deposited in N-graphene surface at each Mg weight percent (2.05; 2.13; 2.15; 2.17; 2.26, and 2.43 wt % Mg/N-GNS).

Table 2. Weight Concentration element variation wt % Mg/N-GNS with used EDX data.

| Sample            | C   | O   | Mg  | SUM |
|-------------------|-----|-----|-----|-----|
| 2.05 wt % /GNS    | 44.67 | 54.30 | 1.03 | 100 |
| 2.13 wt % /GNS    | 55.32 | 44.21 | 0.47 | 100 |
| 2.15 wt % /GNS    | 65.31 | 34.22 | 0.47 | 100 |
| 2.17 wt % /GNS    | 53.86 | 45.49 | 0.65 | 100 |
| 2.26 wt % /GNS    | 52.12 | 47.34 | 0.54 | 100 |
| 2.43 wt % /GNS    | 55.71 | 43.36 | 0.93 | 100 |

Based on the table above it can be seen that a lot of magnesium deposited in N-GNS varies at each weight percent of concentration Mg. To make it easier to see the relationship between particle size and variations in Mg concentration can be seen in Figure 5.

Based on (Figure 5) shows that at a concentration of Mg 2.05-2.15% / N-GBN Mg particle size rises slowly, then decreases at Mg 2.13-2.17% N-GBN then rises slowly at Mg 2.43% / N-GBN. So from (Figure 5) Mg 2.15% has the highest particle size which is 0.495 µm and 2.26% Mg has the smallest particle size which is 0.353 µm. This is due to the number of Magnesium atoms in the N-GBN with the addition of NaCl electrolytes, each of which varies by weight percent variation of Mg deposited into the N-GBN so that the particle size also varies.
3.2. Electrical Conductivity

3.2.1. Current Measurement of 1.86 wt% Mg/GNS and 2.43 wt% Mg/N-GNS with time variations at a voltage of 30 volts.

To know the stability of strong current of 1.86 wt% Mg/GNS and 2.43 wt% Mg/N-GNS, the measurement of current strength at a voltage of 30 volts at time variation of 10-60 minutes.

![Figure 6. Currents strength of 1.78 wt% Mg/graphene and 2.43 wt% Mg/N-graphene](image-url)

The picture above shows the current of 1.78 wt% Mg/graphene and 1.43 wt% Mg/N-graphene is relatively stable decreases with increasing time, the graph that 1.78 wt% Mg/graphene and 1.43 wt% Mg/N-graphene loses electrons each time with relatively little increase so that it can control electron mobility better. This is because Mg has the ability to store electrons and release electrons slowly. The stability of the conductivity of graphene and n-graphene made of metal conducts electric current and the lifetime of graphene and n-graphene-based batteries is affected by the stability of the conductivity modification of metal concentrations [17, 18].

4. Conclusions

Preparation of electrodes on battery anodes by conducting graphite synthesis into graphene by the Hummer's method using magnesium reducing agent. And continued with the impregnation method to deposit metal in graphite, graphene and N-graphene. Modification of the battery anode gets good results. Mg/graphene nanosheet and Mg/N-graphene nanosheet can be used as an alternative battery anode because of its higher conductivity (63.6945 μS/cm) compared to the commercial battery anode (26 μS/cm).

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