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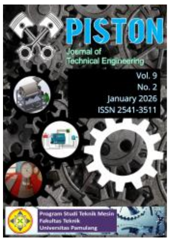

Utilization of Coconut Shells as a Source of Graphene Nanosheets Fe/N-GNS for Environmentally Friendly Primary Battery Electrodes

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Abstract: The research on the performance of carbon (C)/d-orbital metals (graphite/graphene, graphene/N-graphene, graphite/Fe-graphene, graphite/Fe-N-graphene, and graphene/Fe-N-graphene) in primary battery electrode systems was carried out using a simple technology by mixing coconut shell powder with N and Fe. The purpose of this study was to determine the preparation method of Fe/GNS and Fe/N-GNS electrodes and to evaluate the performance of the electrolyte on electron distribution in Fe/GNS and Fe/N-GNS electrodes as primary battery anodes based on electrical conductivity values. This research was conducted as a laboratory experimental study. GNS and N-GNS were synthesized using a modified Hummers method, while Fe/GNS and Fe/N-GNS electrodes were synthesized using the impregnation method. GNS, N-GNS, Fe/GNS, and Fe/N-GNS after electrolyte combination were characterized using SEM–EDX and a multimeter, respectively. The SEM–EDX results at 170 °C and 500–600 °C showed a folded and wrinkled graphene structure with dispersed Fe (5.3 wt% by EDX), dominated by C and O. The addition of Fe–NH_3 acted as a catalyst to form more regularly structured graphite. The DHL test showed the highest electrical conductivity (~51,400 at 40 V) for Fe-N-GNS samples synthesized at 170 °C and 600 °C, which were identified as the most optimal synthesis conditions.

Keywords: Graphite/Fe-graphene, Graphene/Fe-N-graphene, Pyrolysis, Electrodes, Primary Battery.

Abstrak: *Penelitian ini mengkaji kinerja material karbon (C) berbasis logam d-orbital (grafit/graphene, graphene/N-graphene, grafit/Fe-graphene, grafit/Fe-N-graphene, dan graphene/Fe-N-graphene) sebagai sistem elektroda pada baterai primer. Material disintesis menggunakan teknologi sederhana melalui pencampuran serbuk tempurung kelapa dengan nitrogen (N) dan besi (Fe). Tujuan penelitian ini adalah menentukan metode preparasi elektroda Fe/GNS dan Fe/N-GNS serta mengevaluasi pengaruh elektrolit terhadap distribusi elektron pada elektroda Fe/GNS dan Fe/N-GNS sebagai anoda baterai primer berdasarkan nilai konduktivitas listrik. Penelitian dilakukan secara eksperimental di laboratorium. GNS dan N-GNS disintesis menggunakan metode Hummers termodifikasi, sedangkan elektroda Fe/GNS dan Fe/N-GNS disintesis menggunakan metode impregnasi. Karakterisasi morfologi dan komposisi unsur GNS, N-GNS, Fe/GNS, dan Fe/N-GNS setelah penambahan elektrolit dilakukan menggunakan SEM–EDX, sedangkan pengukuran konduktivitas listrik dilakukan menggunakan multimeter. Hasil SEM–EDX pada suhu 170 °C dan 500–600 °C menunjukkan struktur graphene yang terlipat dan berkerut dengan distribusi Fe yang merata (5,3 % berat berdasarkan EDX), dengan unsur dominan C dan O. Penambahan Fe–NH_3 berperan sebagai katalis dalam pembentukan struktur grafit yang lebih teratur. Uji DHL menunjukkan nilai konduktivitas listrik tertinggi (sekitar 51.400 pada tegangan 40 V) pada sampel Fe-N-GNS yang disintesis pada suhu 170 °C dan 600 °C, sehingga kondisi tersebut diidentifikasi sebagai kondisi sintesis paling optimal.*

Kata kunci: *Grafit/Fe-graphene, Graphene/Fe-N-graphene, Pirolisis, Elektroda, Baterai Primer.*

INTRODUCTION

The research institute of the Thai Institute of Technology and Science revealed that coconut shell charcoal has a calorific value of around 4,830 kcal/kg. Arif further stated that Indonesian coconut shell charcoal

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has a higher calorific value, ranging from approximately 6,700 to 7,100 kcal/kg. This difference is attributed to Indonesia's fertile soil composition and its geographical location in a tropical climate, which are considered the main contributing factors [1]. The main components of charcoal include carbon, ash, moisture, nitrogen, and sulfur. In addition, some large pores are still covered by impurities such as tar and other organic compounds. Charcoal also contains inorganic elements, including magnesium (Mg), aluminum (Al), potassium (K), calcium (Ca), and iron (Fe), as reported by Rampe et al [2]. The results of artificial anode studies indicate that palm oil biomass can be utilized as a promising and low-cost material to improve the performance of microbial fuel cell (MFC) anodes [3]. This highlights the potential of biomass-derived carbon materials as sustainable electrode materials.

Graphene is a two-dimensional (2D) material that has attracted significant attention from researchers worldwide due to its outstanding properties, such as large specific surface area, excellent chemical stability, high electron mobility, and superior electrical conductivity [4]. Structurally, graphene has a honeycomb-like hexagonal crystal lattice and exhibits semimetallic behavior with a zero band gap [5]. Cheap and renewable coconut shell biomass waste can therefore be selected as a carbon source to prepare 2D graphite- and graphene-based electrode materials with excellent capacitive performance [6]. During the carbonization process, carbon is gradually transformed into graphite and subsequently into charcoal with a more ordered structure [7]. Furthermore, graphene or modified graphene sheets can also be produced from this biomass source [8].

Graphene nanosheets (GNS) and Fe/GNS were synthesized using a modified Hummers method combined with an impregnation technique [9]. Graphite oxide was reduced to graphene by dissolving the graphite oxide in distilled water, followed by ultrasonication for 90 minutes to obtain a homogeneous solution [10]. The resulting reduced graphene oxide (rGO) was then subjected to repeated washing and heating processes [11]. In related material processing studies, cellulose fibers were isolated from the bark of jicama tubers (*Pachyrhizus erosus*). A suspension consisting of distilled water, starch, and glycerol was mixed with various cellulose loadings (0, 2, 6, and 10 g) and gelatinized using a hot plate equipped with a magnetic stirrer. The biocomposite gel was subsequently sonicated using an ultrasonic probe (47.78 W/cm² for 4 minutes) [12]. Characterization of the synthesized materials was carried out using scanning electron microscopy (SEM), density measurements, and Vickers hardness number (VHN) analysis [13]. In addition, the development of low-cost potentiostats as electrochemistry-based analytical instruments was considered, while the roles of NH_3 and Fe as catalysts in improving the structural quality of graphene oxide (GO) reduction to rGO were evaluated. Further analyses included crystallinity and phase identification, morphology and compositional characterization, assessment of structural defects and regularity, functional group identification, and electrical conductivity measurements [14].

METHODS

The graphene material used in this study was synthesized from old coconut shells that had been dried and soaked in an aluminum kettle at temperatures of 170 °C and 220 °C for 2 hours. The coconut shells were then cleaned from coconut flesh and fibers and burned with an initial combustion at a temperature of 300 °C for 1 hour. After 1 hour, the temperature was increased to 500 °C and 600 °C for 1 hour. After becoming charcoal, the charcoal was ground using a mortar and sieved to a particle size of 200 mesh.

Coconut shell charcoal was soaked in distilled water in a test tube for 24 hours to clean the charcoal from impurities, then filtered using Whatman filter paper and dried at room temperature for 48 hours. The sample was then dried at a temperature of 90 °C for 1 hour and 30 minutes using a furnace. This process aimed to remove moisture content. The sonication process was carried out for 30 minutes, and the sample was dried at room temperature for 24 hours. Subsequently, the drying process was carried out using a 100 °C hotplate for 2.5 hours. The sample was then doped with 25% ammonia solution (NH_3) using a hotplate magnetic stirrer.

First, an Erlenmeyer was prepared and filled with 30 mL of aquadest. Then, 1.2 g of sample and 1 mL of NH_3 were added. The flask was placed on a hotplate and set to 100 °C, with stirring at 150 rpm. A magnetic stir bar was inserted into the Erlenmeyer flask, and a temperature sensor rod was placed inside the flask to detect the internal temperature. The heater and magnetic stirrer were then turned on. This process lasted for approximately 50 minutes. During this process, the moisture content in the sample evaporated due to heating. After approximately 50 minutes, the sample in the flask dried and returned to powder form, which was ready to be transferred into a sample bottle for the next step involving mixing with $FeCl_3$ and $NaBH_4$.

A total of 0.12 g of $FeCl_3$ was mixed with 100 mL of aquadest and stirred for 15 minutes using a magnetic stirrer at a speed of 300 rpm. 0.2 g of GNS was mixed with 200 mL of aquadest and then sonicated for 30 minutes. The $FeCl_3$ solution was then poured into the GNS suspension and stirred for 12 hours at 650 rpm. Subsequently, 0.12 g of $NaBH_4$ was mixed with 100 mL of aquadest and stirred for 15 minutes at 300 rpm. After completion, the $NaBH_4$ solution was added to the GNS–$FeCl_3$ mixture and stirring was continued for 1 hour. After the stirring process was complete, the mixture was filtered using Whatman filter paper No. 42, then placed in a bowl and dried using a furnace at 60 °C for 6 hours until Fe–N doped graphene powder was obtained.

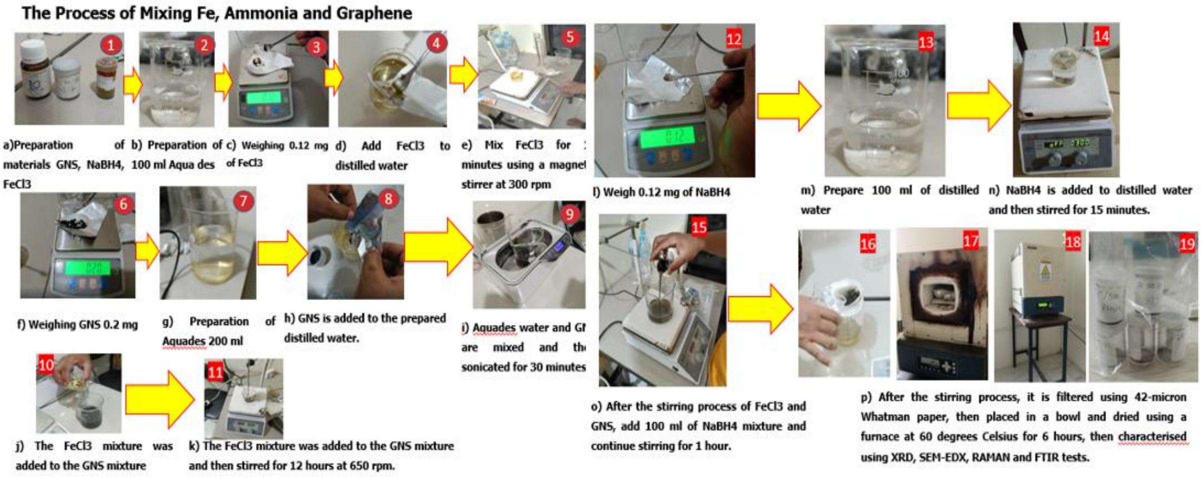


Figure 1. Synthesis process of Fe–N-doped GNS

SEM–EDX analysis was conducted at an accelerating voltage of 15.00 kV to observe the morphology of graphene (folded, wrinkled, and sheet-like structures) and to determine the elemental composition (C, O, and Fe). The electrical conductivity test was performed using a pellet compacted from approximately 1.00 g of sample and covered with a pellet holder. Crocodile clip cables were connected to the negative and positive terminals of a digital multimeter and a regulated DC power supply. Electrical conductivity was measured by applying voltage variations of 40, 44, 48, 51, and 55 V, while recording the corresponding current values. The instrument used for this conductivity test was a multitester power supply, which functions as a device to supply electric current with various voltage levels.

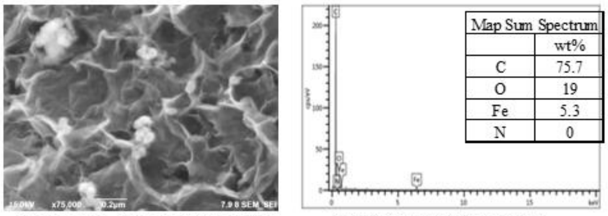
RESULT AND DISCUSSION

SEM–EDX Test Results of Pyrolysis at 170 °C, 500 °C, and 600 °C (Fe–N-Doped GNS)

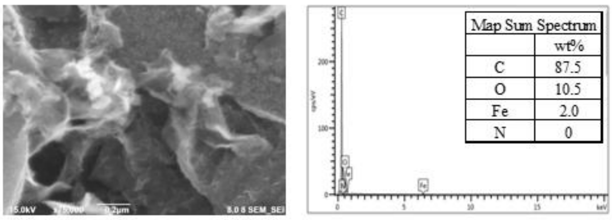
SEM examination was carried out on graphite, graphite oxide, and graphene samples to observe the morphology of graphene resulting from Fe doping using 0.12 g of $FeCl_3$ and 0.12 g of $NaBH_4$ to form Fe–N-GNS. Figure 2(a) shows morphological differences in the graphene samples, where graphene appears in the form of small flakes irregularly dispersed. After oxidation into graphite oxide, the morphology changes into thicker layered structures, which then revert to graphene after the reduction process. The particle dispersion shows the presence of white spots scattered over the surface of the GNS sheets; these particles correspond to Fe nanoparticles formed during the pyrolysis process.

The EDX results indicate that the main constituent is carbon (75.7%), corresponding to graphene nanosheets, followed by iron (5.3%), which confirms the successful doping of Fe nanoparticles, and oxygen (19.0%), originating from residual functional groups formed during the synthesis and oxidation processes. Figure 2(b) shows a wrinkled, sheet-like, and folded GNS morphology with Fe nanoparticles dispersed as white spots on the surface. However, the number of these particles appears reduced and more uniformly distributed compared to the sample pyrolyzed at 500 °C.

The EDX analysis further reveals that the carbon content increases significantly from 75.7% to 87.5%, indicating that pyrolysis at 600 °C is more effective in reducing oxygen-containing functional groups and improving the crystallinity of the GNS carbon framework. The oxygen content decreases from 19.0% to 10.5%, supporting the effectiveness of thermal reduction, while the iron content decreases from 5.3% to 2.0%, likely due to phase transformation of Fe crystals resulting in a smoother and more homogeneous distribution.



a. SEM 75.000x -170-500 ⁰C Fe-N-GNS EDX -170-500 ⁰C Fe-N-GNS



b. SEM 75.000x -170-600 ⁰C Fe-N-GNS EDX -170-600 ⁰C Fe-N-GNS

Figure 2. SEM images (75,000×) and EDX spectra of Fe–N-doped GNS: (a) pyrolysis at 500 °C and (b) pyrolysis at 600 °C after baking at 170 °C

Conductivity Test

Electrical conductivity tests were conducted at baking temperatures of 170 °C and 220 °C and pyrolysis temperatures of 500 °C (Fe–N-GNS) and 600 °C (Fe–N-doped GNS) using a compressed pellet with a mass of approximately 0.50 g under various applied voltage conditions. The electrical conductivity was determined based on the relationship between voltage and current using the equations $R = V/I$ and DHL $= 1/R$. The complete electrical conductivity data obtained from these measurements are presented in Tables 1 and 2.

Table 1. Electrical conductivity of Fe–N-doped GNS after baking at 170 °C and pyrolysis at 500 °C

Voltage (V)	Current (μA)	Resistance (Ω)	Conductivity (μS)
40	2033	0.019675356	50.825
44	2102	0.020932445	47.772
48	2198	0.021838034	45.791
51	2224	0.022931654	43.607
55	2344	0.023464163	42.618

Table 2. Electrical conductivity of Fe–N-doped GNS after baking at 170 °C and pyrolysis at 600 °C

Voltage (V)	Current (μA)	Resistance (Ω)	Conductivity (μS)
40	2056	0.019455252	51.400
44	2282	0.019281332	51.863
48	2328	0.020618556	48.500
51	2359	0.021619330	46.254
55	2421	0.022717885	44.018

Based on the conductivity graphs, current measurements were conducted at voltages of 40, 44, 48, 51, and 55 V to evaluate electrical conductivity accuracy and stability. The results indicate that graphene exhibits relatively stable electrical behavior, with a gradual decrease in conductivity corresponding to increasing voltage. This behavior suggests controlled electron mobility, attributed to graphene's ability to store charge carriers and release electrons gradually, resulting in stable electrical performance.

CONCLUSIONS

1. SEM analysis at a baking temperature of 170 °C and pyrolysis temperatures of 500 °C and 600 °C for Fe–N-doped GNS, observed at a magnification of 75,000×, reveals that the morphology of Fe–N-GNS particles consists of tightly packed structures resembling graphene sheets. The EDX results confirm the successful incorporation of iron (Fe) into the graphene-based material, with a relatively high Fe content of 5.3 wt%. Carbon is identified as the dominant element, while oxygen is present as residual functional groups originating from the synthesis process.
2. Electrical conductivity decreases with increasing applied voltage. The highest conductivity value (~51.4 μS at 40 V) was obtained for Fe–N-doped GNS synthesized under baking at 170 °C and pyrolysis at 600 °C, indicating that these conditions are the most optimal for achieving superior electrical conductivity.

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