

Performance of N-Doped Graphene Nano Sheets on Electrode of Primary Battery Cells

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Abstract

Research on performance of N-Doped Graphene Nano Sheets (N-G) from coconut fruits as well as raw materials was carried out. The aims of this research are to produce gram scale N-G and evaluate its performance act as an electrode of primary battery cells. In this research, Graphene Nano Sheets (GNS) was produced from coconut fruits by using pyrolysis method. Then, N-G was generated with N-doped method where ammonia solution and room temperature were used as N atoms source and temperature doping, respectively. Finally, N-G was characterized with XRD, SEM-EDX, TEM, XPS, Raman and conductometer. XRD data shows that C (002) peak of N-G shifts to higher 2θ number comparing to GNS that is N-G ($2\theta = 24,72^\circ$) and GNS ($2\theta = 23.86^\circ$), indicating there is interaction between carbon and nitrogen on N-G. This data is consistent with XPS data namely the type of N-G is N-pyridin (BE = 402.0 eV) and C-N (BE = 286.8 eV). Interestingly, the TEM images show clearly N-G has the flat surface and the graphene layers distance are little bit expanding (0.36 nm) compare to graphene layers (0.34 nm). SEM images and EDX data also may prove the morphology structure like the honey comb lattices and containing N atoms dominant. Raman data succeed to clarify D-band and G-band from N-G. All of data show that N-G is well produced and proved. In addition, the good findings N-G has electrical conductivity and it may be used as an electrode on primary battery cells.

Introduction

Rising energy demands drive the development of large capacity energy storage systems. A supercapacitor is an energy storage device that is currently being explored. It has a substantially bigger energy storage capacity and a more durable discharge than regular capacitors, as well as a high energy density, a huge power density, and a rapid charging time, making supercapacitors more powerful than batteries [1–2]. Carbon-based nanomaterials are appealing for supercapacitor electrode fabrication due to their chemical stability, strong electrical conductivity, and large surface area [3–5]. Graphene, a carbon nanomaterial, has been employed as supercapacitor electrodes. Graphene has a number of intriguing physical features [6], huge surface areas ($2600 \text{ m}^2/\text{g}$) and great chemical stability, all of which might be used for prospective applications such as graphene nanoribbon field effect transistors [7–8], Supercapacitors made of graphene sheets [9] with lithium secondary batteries [10]. Graphene may be generated in a simple and low-cost manner using renewable natural waste, like discarded coconut shells [11], It comprises cellulose, hemicellulose, and lignin, all of which are composed mostly of 57.5% carbon and 42.5% oxygen [12]. Nitrogen (N-doped carbon) could be used to modify graphene for use as a non-metallic main battery electrode [13]. N-dopant synthesis Graphene may be produced in two ways: I in-situ doped and ii) after treatment doped. Dopant is created in-situ through direct integration of N-C with a high-temperature combustion process [14]. Nitrogen doping in graphene will modify its characteristics, as will the presence of ammonia as a nitrogen supply, in order to boost graphene's electrical activity [15]. Liu et al. created substitutional N-doped multiplayer graphene sheets by injecting NH_3 gas during graphene chemical vapor deposition (CVD) development [16]. Arc discharge of carbon electrodes in the presence of H_2 /pyridine or H_2 /ammonia also yielded N-doped graphene [17]. Currently, comprehensive studies of

graphene doping are required. The characterisation of N-Doped Graphene from Coconut Shell by Temperature Variation was investigated in this study. The goal of synthesizing N-Doped Graphene is to investigate its electrical conductivity.

Experimental Section

Producing of charcoal from coconut shell

The sun is used to dry the coconut shell. 1 kilogram of coconut shell is pyrolyzed within the furnace in an oxygen-free condition for 5 hours at 600°C to produce charcoal with no smoke. A mortar is used to chill and crush charcoal.

Producing of Graphene

Coconut shell charcoal was made into chips, and 15 g was combined with powder activated carbon. For one hour, the mixture was heated to 600°C. To separate the coconut shell and activated carbon, a 150 mesh screen was used. To clean the charcoal, it was rinsed with distilled water and dried at 70°C [18].

Producing of N-Doped Graphene

One gram graphene was swirled for one hour in 100 mL of distilled water. The mixture was then treated with 10 mL ammonia 10 M and stirred for 72 hours. The mixture was filtered and dried for 24 hours at 80°C. The samples were then analyzed using XRD, SEM-EDX, TEM, XPS, Raman and Electrical Conductivity [19].

Results And Discussion

In this study, graphene was used as the basic material for making primary battery electrodes synthesized from coconut shell raw materials through the pyrolysis method. This stage of graphene synthesis begins with a coconut shell dried in the sun, pyrolysis in a furnace with minimal oxygen with a temperature of 600 °C for 5 hours to produce chip-shaped charcoal. The last stage is charcoal in the form of chips and then mixed using activated carbon and re-pyrolyzed for 1 hour, then mashed with mortar and sifted with a size of 150 mesh, resulting in thin sheets of Graphene. Then, the graphene is ammonia-modified to generate N-doped graphene. The inclusion of N atoms embellishing the gbn structure is projected to change the nature of Graphene, allowing for higher Graphene activity on N-doped Graphene.

XRD Analysis

The XRD examination was carried out using a beam size of 10 mm 10 mm with Graphene monochromator Cu/K α radiation ($\alpha = 1.5406$) at 40 kV and 100 mA. The range of 2θ is 10–80° in 2.0° increments. Powder X-ray diffraction patterns were detected using a Rigaku Corporation (Japan) SWXD

diffractometer running at 18 kW and a two-dimensional diffraction pattern from the sample. The results of the analysis of Graphene and N-doped graphene using X-Ray Diffraction (XRD) are shown in Fig. 1.

The occurrence of two peaks in Graphene, namely C (002) ($2\theta = 23.86^\circ$) and C (200) ($2\theta = 44.5^\circ$) [20], shows the formation of Graphene. According to Nugraheni [21], graphene possesses weak, dilated peaks indicating a nano-sized layer of graphene placed on the graphene interlayer. Furthermore, as compared to Graphene, the peak C (002) ($2\theta = 24.72^\circ$) of N-doped Graphene is somewhat moved to the right (Fig. 1). That is, the N atom of ammonia interacts with the C atom of GBN. According to studies by Praveen [22] and Liu [23], N-doped Graphene has maxima at $2\theta = 24.5^\circ$ and 26.3° . The findings of our investigation support these findings. The nitrogen atom dopant in GBN causes a shift in its value of 2, with N-doped Graphene having an emergent peak at $2\theta = 24.72^\circ$ and Graphene having $2\theta = 23.86^\circ$.

SEM-EDX of N-doped Graphene

Surface morphology of N-doped graphene at magnification of 10000 times, the surface wrinkled and irregular. Based on the mapping findings in Fig. 2a and b, the Graphene structure has suffered defects as a consequence of the dopant of Nitrogen (N) atoms into the Graphene structure to form N-doped Graphene [24]. The abundance of elements from N-doped Graphene can be observed in Table 1.

Table 1

Abundance Element of N-doped Graphene

Element Number	Element Symbol	Element Name	Weight Conc. (%)
6	C	Carbon	93.5
8	O	Oxygen	6.0
14	Si	Silicon	0.1
17	Cl	Chlorine	0.3
19	K	Potassium	0.1

TEM of N-doped Graphene

More work has already been done to characterize the functionality, sustainability, and kinematics of various organic molecules on the graphene sheets, such as carbon and other light adatoms [25]. Indeed, TEM of the dynamics of single - molecule adsorbent surface on graphene has been researched extensively from the beginning of graphene research, which is not surprising given that examining molecules does not require as much high resolution as studying single atoms [26]. The N-doped Graphene images may be seen in Fig. 3.

After the nitrogen doping treatment (Fig. 3a), wrinkled degree of the graphene surface aggravated obviously because of the more defect formation and the N atoms induced into the graphene (Fig. 3b). This N-doped graphene defect is believed to improve its electrochemical performance [27]. The N-doped

technique and mechanism are comparable to nitrogen-doped carbon nanotubes' bamboo-shaped characteristic [28]

Xps Of N-doped Graphene

The XPS N1s spectra are divided into four groups: I pyridine-N (397.6–398.1 eV); (ii) pyrrolic-N (399.0–399.7 eV); (iii) grafitic (400.0–401.7 eV); and (iv) pyridine-N oxidation (402.7–404.6 eV) [29–30]. In Fig. 4a is showed N aoms has been in graphene. Its because oxidized pyridinic nitrogen and pyridone nitrogen are both oxidized, whereas pyridinic nitrogen is reducing [31]. Yamada et al. [32] presented computed values for peak shifts and associated FWHMs. They discovered that the average shift in BE values of N1s reported by other authors is pretty closely associated with their estimates for various functional groups, including pyridinic, pyrrolic, and quaternary nitrogen. They also discovered that functional groups emanating from edges had a lower FWHM than functional groups originating from basal planes. Zhang et al. [33] observed a change in the N1s peak location when the growth temperature of N-doped graphene was increased, which was explained by a changing local environment of N atoms due to defect formation.

In data X1s spectra of N-doped Graphene showed the three peaks with binding energy 284.7; 286.8 and 287.8 eV. The primary component at 284.6 eV relates to the existence of a C atom inside graphene's sp^2 C = C bond [34–35]. While compared to the peak location of sp^2 C = C in graphene, the peak of the major component of N-doped Graphene would shift to a higher binding energy because the N atom (3.10) is more electronegative than the C atom (2.55) (Fig. 5b). The presence of a C - N (pyridine) bond in N-doped Graphene is shown by binary peaks centered at 284.7 and 287.8 eV [36–37]. The component of N-doped Graphene is showed in Table 2.

Table 2
Element Components of N-doped Graphene

Component N-GBN	Binding Energy (eV)	Mass Concentration (%)
N-Pyridin	402,0	1,98
Carbon sp^2 (C = C)	284,7	12,49
C – N	286,8	33,68
C – O	287,8	36,42

Raman of N-doped Graphene

The raman spectra of N-dopen graphene is shown in Fig. 5.

Raman data were acquired after being shot with a laser, demonstrating the link between raman intensity and wavelength displayed in graphene form. The D band indicates disorder, which can be caused by

flaws like as voids or grain boundaries [38–39] and amorphous carbon species. The G band is slightly shifted from the position of that of Graphene [40]. Figure 5 shows all spectra two characteristic peaks of graphene carbon materials as G band near 1580 cm^{-1} and D band near 1350 cm^{-1} , representing the degree crystallization and the degree of disorder or defect in the sample respectively. The former is created by in-plane stretching vibration in the E_{2g} mode, whereas the latter is induced by phonon scattering at the disordered hexagonal Brillouin zone's border [41]. N-doped graphene samples were created using nitrogen doping and reduction treatment, with a little drop in ID/IG value (from 0.84 to 0.85). This is the consequence of numerous variables acting in concert: (1) less oxygen-containing groups resulted in a drop in ID/IG value; and (2) more structural flaws in graphene generated by N doping resulted in an increase in ID/IG value. The ID/IG values of Grafena-N were comparable, indicating that the N-doped samples had a similar defect density but a distinct defect type and distribution, as validated by XPS data [42].

Electrical conductivity

In order to know the electrical properties of N-doped Graphene, we carried out the electrical conductivity measurement (Fig. 6).

Figure 6 shows that the electrical conductivity of N-doped Graphene (Fig. 6b) is higher compared to graphene (Fig. 6a). This is because the N atom in N-doped Graphene is able to reduce electrons captured from the anode that oxidize electrons, resulting in a higher electrical conductivity.

Conclusions

Producing the large scale and sustainable N-doped Graphene is possible produced with coconut fruits as raw material and doped with ammonia in room temperature. The N-doped Graphene shows the outstanding properties namely C-sp², flat surface, and N-Pyridin type. The existence of N atoms may improve the electrical conductivity of N-doped Graphene. All of data prove that the N atoms doped on Graphene surface may be expected to modulate performances of Graphene and that is good news, we may use it as a potentially electrode material for battery.

Declarations

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Author Contributions

Rikson Siburian: Conceptualization, Writing – review & editing, Supervision, Funding acquisition, Project administration. **Yatimah Alias:** Investigation, Validation, Writing – review. **Elsa Pasaribu:** Investigation,

Methodology, Visualization, Writing – original draft. **Minto Supeno**: Investigation, Validation. **Ronn Goei**: Methodology, Validation, Resources, Investigation. **Alfred ling Yoong Tok**: Methodology, Resources. Validation. **Mehran Sookhakian**: Validation, Writing – review.

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Ethics Approval and Consent to Participate

Not applicable.

Consent for Publication

Not applicable.

Competing interests

The authors declare that they have no competing interests.

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Figures

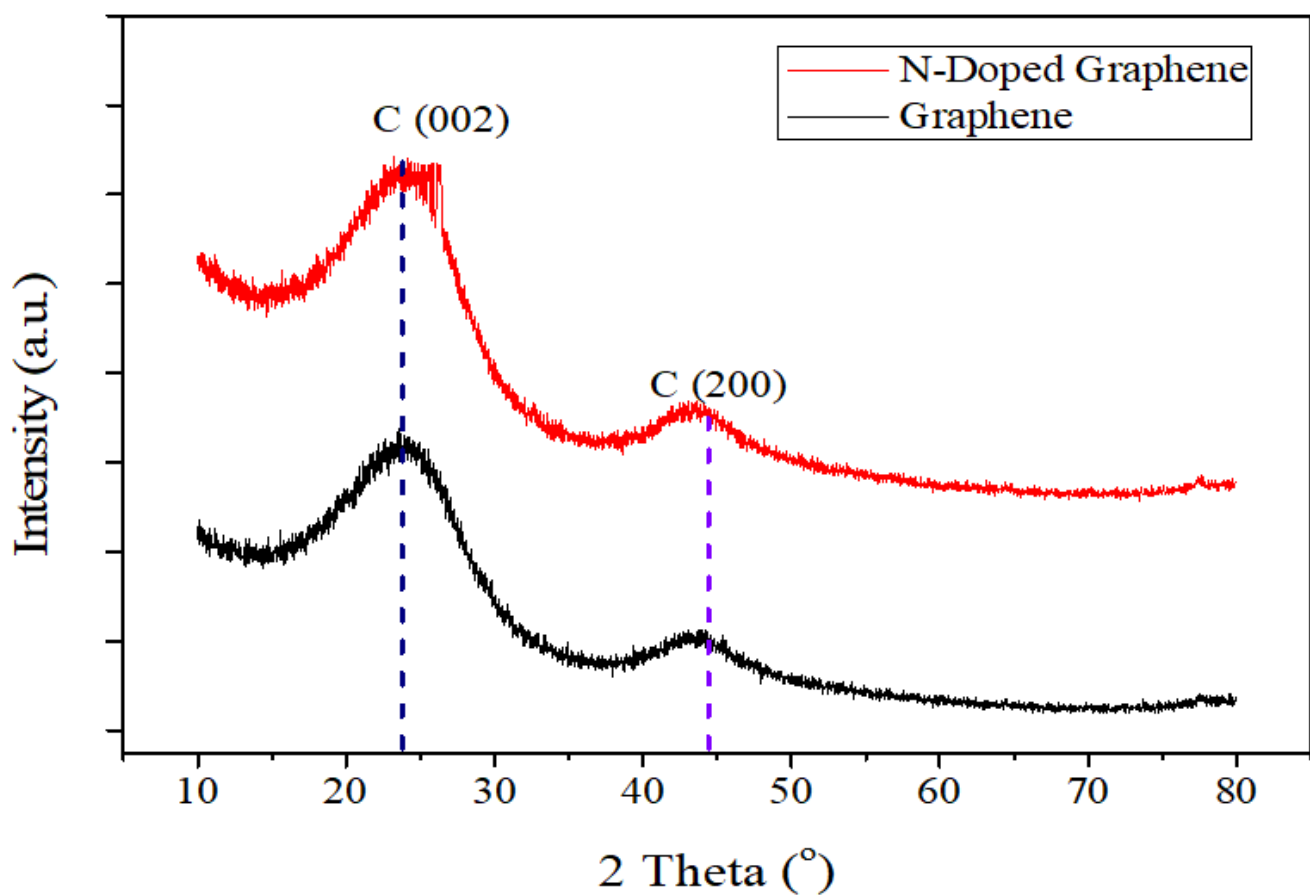


Figure 1

XRD Spectra of Graphene and N-Doped Graphene.

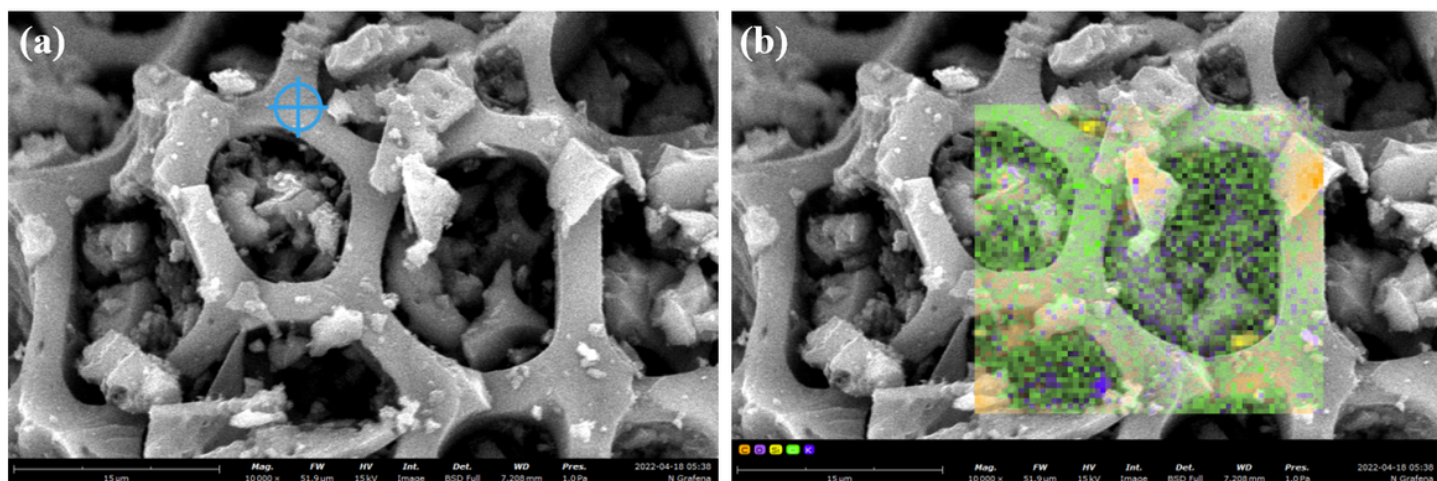


Figure 2

Surface morphology (a) and mapping of N-doped Graphene (b).

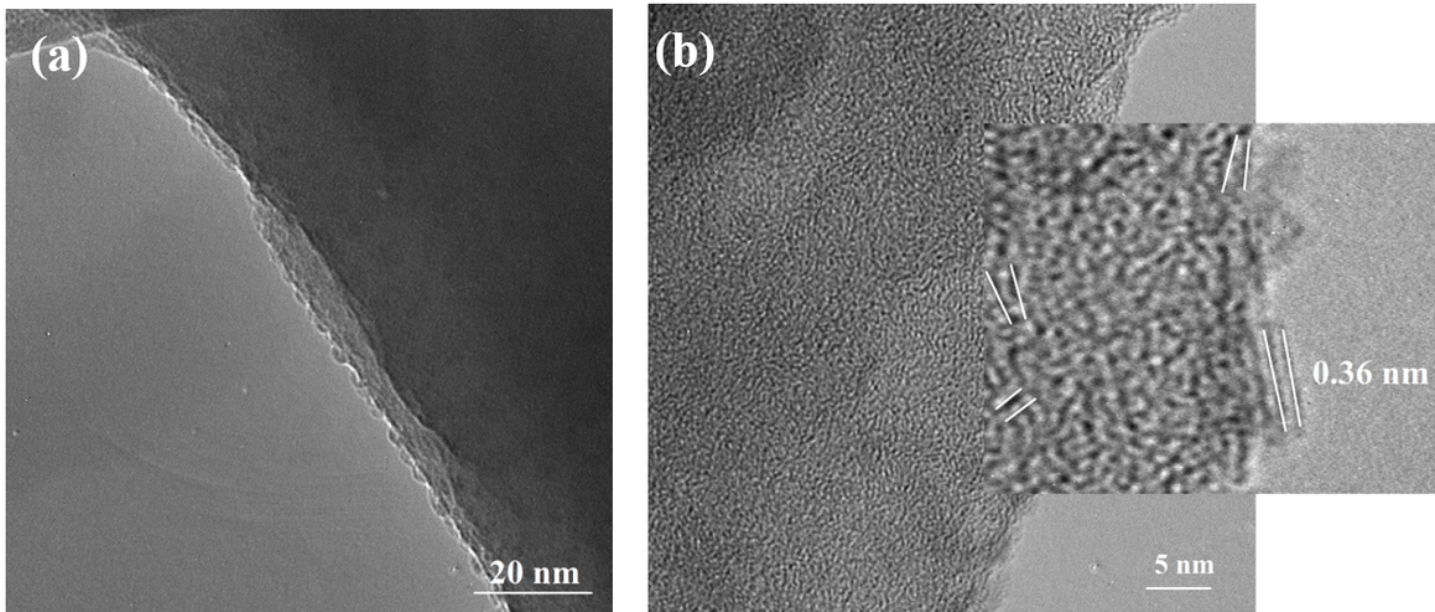


Figure 3

TEM images of (a) N-doped Graphene and (b) of N-doped Graphene distance layers

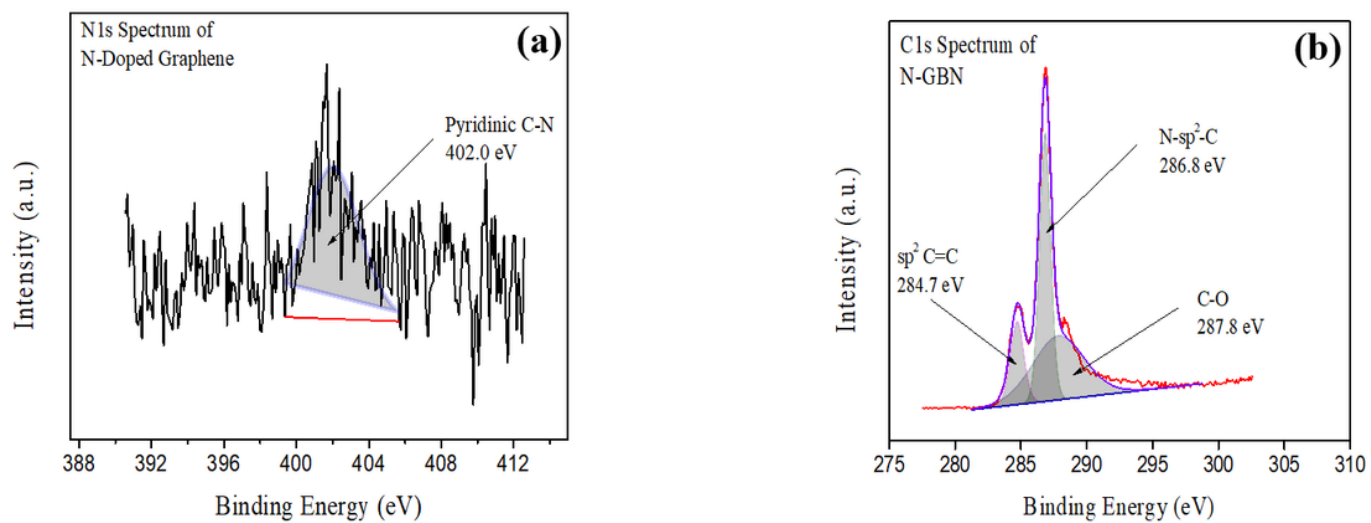


Figure 4

XPS Spectra N-doped Graphene (a) N1s and (b) C1s

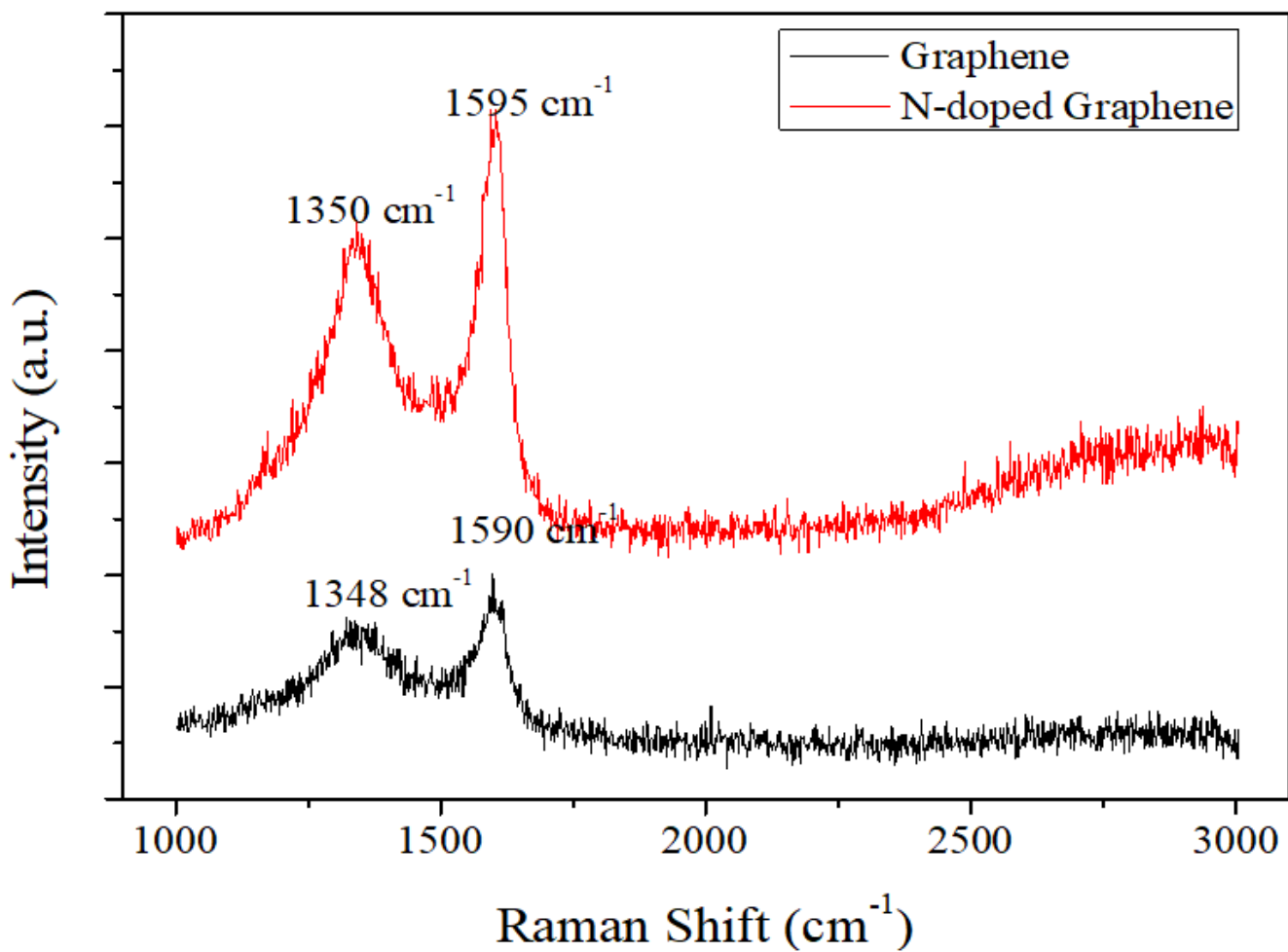


Figure 5

Raman spectra of Graphene and N-doped Graphene

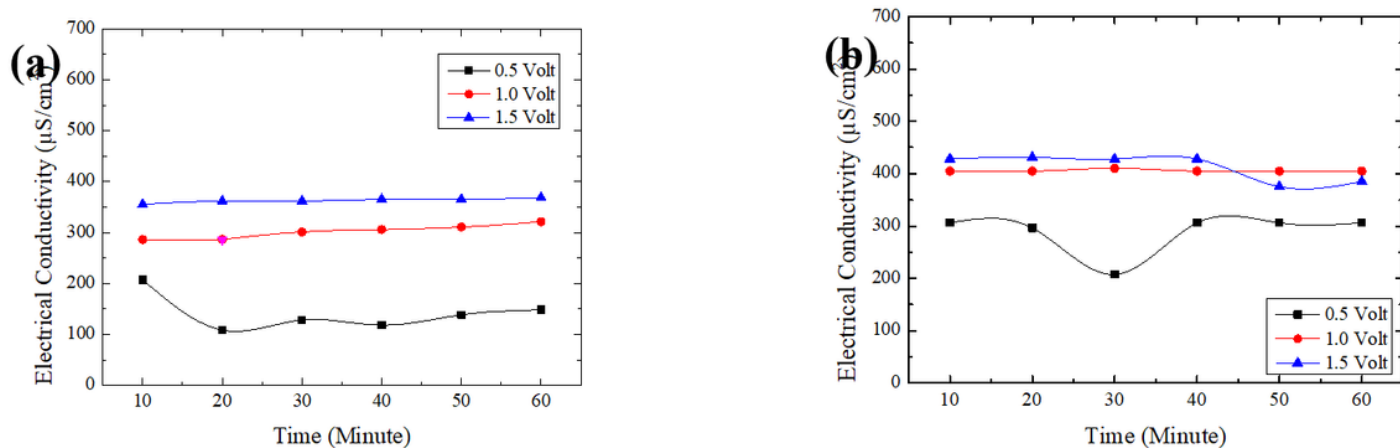


Figure 6

Electrical Cnductivity of (a) Graphene and (b) N-doped Graphene