Effect of Electrolysis Voltage on Graphene Structure Synthesized by One-step Electrolytic Exfoliation in PEDOT/PSS Solution

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Graphene has recently attracted great interest in a wide range of technological applications due to its extraordinary electronic, physical and chemical properties. Nevertheless, its local structures and characteristics are considerably dependant on synthesis method and process conditions. Several graphene production methods including micromechanical cleavage, epitaxial growth via ultra-high vacuum graphitation, chemical synthesis through oxidation of graphite, chemical vapor deposition (CVD), solvothermal synthesis and electrolytic exfoliation have been proposed [1-3]. Among these, electrolytic exfoliation is a relatively new organic solution-based route that is highly attractive due to its high-quality graphene production, low cost, low temperature processing and ease of large-scale production. In this work, we present a new electrolytic exfoliation of graphene in Poly (3,4ethylenedioxythiophene)/poly-styrene-sulfonic acid (PEDOT/PSS) conducting polymer and study the effect of electrolysis voltage on graphene formation.

Electrolytic exfoliation was conducted in an electrolysis cell filled with a commercial PEDOT: PSS solution and a constant voltage ranging from 3 to 12 V was applied between two graphite electrodes. The anode was corroding and a black precipitate was gradually formed in the reactor. The electrolysis time was varied from 5 to 40 hours to obtain stable graphene-PEDOT/PSS dispersion with different graphene concentrations. The dispersed product was centrifuged at 12000 rpm to separate large agglomerates and supernatant portion of the dispersion was decanted. The graphene concentration was estimated from weight loss of the graphite electrode.

The concentration of graphene in PEDOT/PSS solution estimated from weight loss of graphite electrode as a function of electrolytic voltage for various synthesis times are shown in Fig. 1. It can be seen that the concentration of graphene slowly increases as electrolytic voltage increases from 3 to 8 V and then rises more rapidly when the voltage increases further. For the effect of time, the concentration monotonically increases with time as expected. The detailed structure of graphene-PEDOT/PSS composites prepared at various electrolytic voltages for 40 hours are identified and characterized by TEM and selected area electron diffraction (SAED) as illustrated in Fig. 2. It can be seen that structures prepared at different voltages are polygon sheets with different shapes and morphologies. In addition, the size and thickness of graphene structure are increased as the electrolytic voltage increases and crystallinity is improved with the synthesized voltage of 8 V being a critical voltage that produces highly crystalline graphene structure.

Fig. 3 demonstrates Raman spectra from extracted graphene powder synthesized at various electrolytic voltages for 40 hours. It is evident that 2D/D peak ratio increases as the electrolytic voltage increases. This implies that the quality of graphene is improved as the synthesized voltage increases. In addition, 2D bands of all spectra are relatively broad compared to G band suggesting that all synthesized graphenes have multiple layers.

The functional groups of PEDOT/PSS, graphene-PEDOT/PSS and washed graphene powders are characterized by FTIR as illustrated in Fig. 4. It can be seen that the functional groups of graphene-PEDOT/PSS are almost exactly the same those of PEDOT/PSS and the FTIR spectrum of washed graphene confirms that the synthesized structures are graphene not graphene oxide, which typically

exhibits C-O and C=O peaks between 1600-1800 cm^{-1}. From the characterization results, 8 V is thus seen as an optimum electrolysis voltage that produces high-quality single crystal graphene structures in PEDOT/PSS solution.

References

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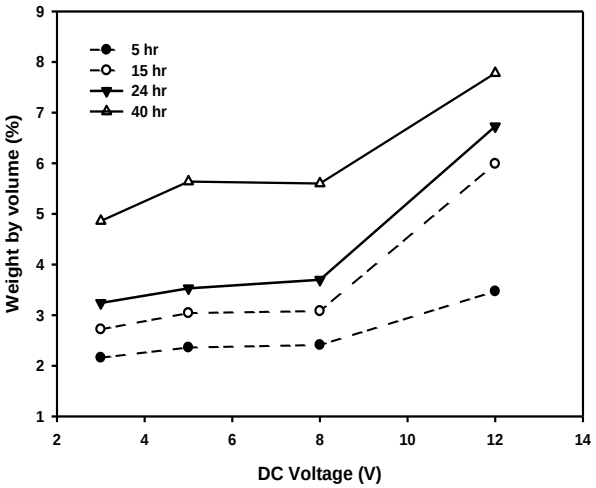

Figure 1. The concentration of graphene in PEDOT/PSS solution estimated from weight loss of graphite electrode as a function of electrolytic voltage for various synthesized times.

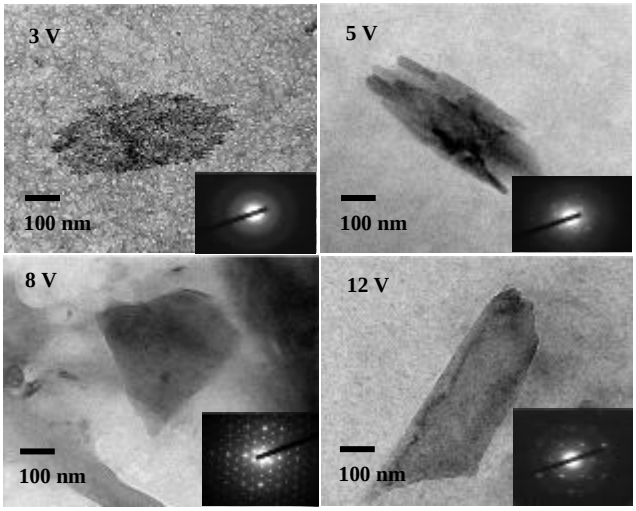

Figure 2. Bright field TEM images with SAED patterns of graphene-PEDOT/PSS composites synthesized at various electrolytic voltages for 40 hours.

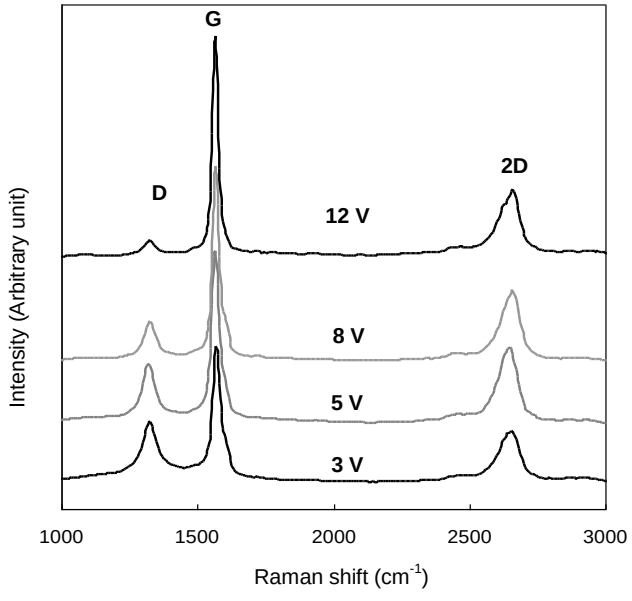

Figure 3. Raman spectra from graphene powders extracted from graphene-PEDOT/PSS composite synthesized at various electrolytic voltages for 40 hours.

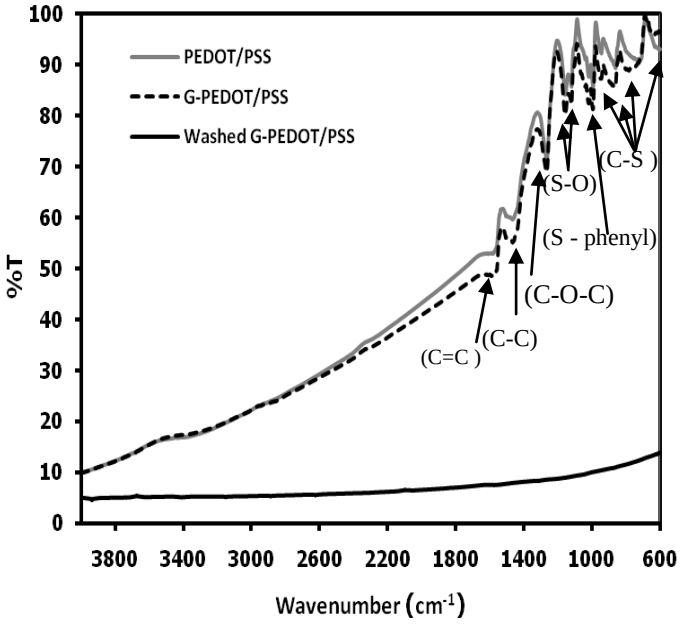

Figure 4. Typical FTIR spectra of PEDOT/PSS, graphene-PEDOT/PSS and washed graphene-PEDOT/PSS. Synthesized voltage and time are 8V and 40 hours, respectively.