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Graphene/Polymer Composites for Supercapacitor

Graphene has attracted interest as an electrode material for supercapacitors because of its large specific surface area (~2630 $m^2 \cdot g^{-1}$) and low in-plane electrical resistivity (1 μΩ·cm).[1-3] It is reported that the orientation of graphene sheets in the electrode could affect the performance of supercapacitors.[4] If graphene sheets are oriented in a parallel stacked geometry, this structure often restricts direct access of electrolyte ions to the charge-storage surfaces and hinders them from penetrating deep inside the stacked graphene planes, This results in the incomplete utilization of the electrochemical surface area of the graphene sheet.[5,6] Furthermore, in the formation of electric double layers at the electrode−electrolyte interfaces, the high electrical resistivity (3×10^5 μΩ·cm)[7] between stacked graphene sheets also hinders the electron transfer process, which contributes to additional electronic resistance in the electrochemical process. In order to avoid this inherent flaw in parallelly-stacked graphene-based supercapacitors, vertically-aligned graphene electrodes have been proposed.[4,8] When compared to parallelly-stacked graphene, generally the vertically-aligned graphene electrode has two main advantages in supercapacitors. On one hand, graphene sheets have direct contact with the current collector, which leads to fast electron transferring and minimizes electronic resistance because of the high in-plane conductivity of graphene. On the other hand, the vertically-aligned graphene electrode allows electrolyte ions to have more open and direct access to the charge-storage surfaces. This results in more rapid electrochemical kinetics and enhancing the rate capability of the supercapacitor. In this study, a vertically-aligned graphene-PEDOT:PSS (poly(3,4-ethylenedioxythiophene) polystyrene sulfonate) composite film was fabricated by volatile solvent evaporation in a strong magnetic field (12 Tesla). PEDOT:PSS was applied as the conductive polymer binder in electrode film fabrication. Figure 1 shows the schematics of graphene alignment under an external magnetic field and graphene without alignment when there is no external magnetic field. With the extra magnetic field, graphene reaches alignment. Without the extra magnetic field, graphene sheets become randomly oriented. The structure and electrochemical rate capability of the film were also investigated. Electrochemical properties of two specimens were fabricated by the solvent evaporation method, with one specimen processed with a vertical 12 Tesla magnetic field, and another without the magnetic field (0 Tesla). Electrochemical tests were carried out by a two-electrode capacitor system with a 1M H_2SO_4 electrolyte. The technique of 2D small-angle X-ray scattering is used to characterize the alignment of the fabricated sample both with and without the magnetic field. The alignment of graphene/polymer composites under the magnetic field is enhanced. With the external magnetic field, the cyclic voltammetry plot of the vertical 12 Tesla sample kept its rectangular shape well at increased scan rates, while the 0 Tesla sample showed significant shape distortion. This work provides a facile and effective method to fabricate ultra-high rate graphene supercapacitor.

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Figures

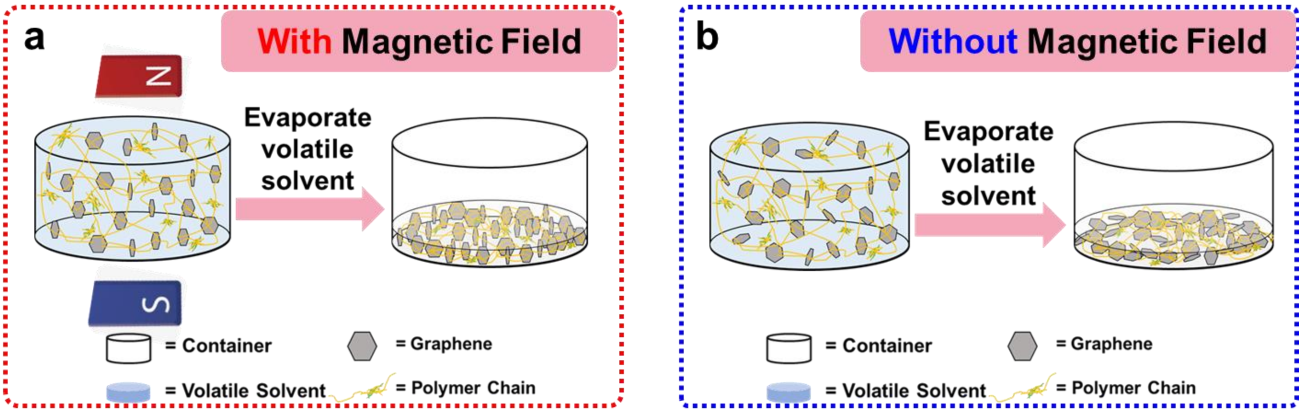

Figure 1: (a) The schematics of graphene alignment under external magnetic field; (b) The schematics of graphene without alignment when there is no external magnetic field.