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## Preparation and characterization of zinc–aluminum layered doubled hydroxide/ graphene nanosheets composite for supercapacitor electrode

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Keywords: Supercapacitor ZnAl-LDHs Graphene Hybrid system EIS	Graphene nanosheets were synthesized using the liquid phase exfoliation technique while zinc-aluminum layered doubled hydroxides (ZnAl-LDHs) were fabricated using the chemical bath deposition method. A composite of graphene/Zn–Al LDHs was synthesized and used as an electrode for the supercapacitor. ZnAl-LDHs present a well-standing nanosheets-like structure that covered the entire area of the substrate. The thickness of these nanosheets was found to be between 60 and 80 nm. The SEM images of the graphene/Zn–Al LDHs composite display graphene nanosheets next to the LDHs nanosheets. The optical band gap of this composite was found to be about 4 eV. The obtained electrode exhibited excellent supercapacitive performances with the highest specific capacitance of ~ $140 F/g$ along with the maximum energy, and power densities of $15.359 kW Kg^{-1}$ and $2.016 Wh Kg^{-1}$ , respectively. The as-prepared composite showed long-term stability and it retains 97% of the initial capacitance after 2000 charge-discharge cycles. The electrochemical impedance spectroscopy (EIS) results confirm the contribution of both ZnAl-LDHs and graphene in the supercapacitor system. The fabricated electrode using the present simple strategy in this report can be further extended to manufacture other electrodes using other composites from the same family for the upcoming energy storage systems.

## 1. Introduction

Supercapacitors (SCs) are a type of energy storage system that providing high specific capacitance, high energy and power densities, as well as rapid charge-discharge comparing to other conventional devices like batteries and capacitors [1–6]. SCs, also known as an electrochemical capacitor, is considered as one of the most reliable energy storage system [7–12]. This system has become the bridge between the conventional battery and the well-known capacitor due to its higher energy density over capacitor and higher power density compared to the battery [13]. Although, SCs have improved drastically in the last few years, their energy density is still lag behind comparing to the conferential battery. Therefore, researchers have worked to improve the overall performance of SCs using more suitable electrodes in order to replace batteries in future electrochemical energy storage systems [14–17].

SCs can be classified based on the way of energy storage in their electrodes into (1) electric double-layer capacitors (DLSCs), (2) Pseudocapacitance, and (3) hybrid capacitors. The first one is DLSCs, which stores energy electrostatically by absorption/desorption of electrolyte

ions at the electrode-electrolyte interfaces. The DLSCs system is mostly carbon-based materials such as carbon nanotube (CNTs), graphene (Gr), carbon nanofibers (CNFs), and active carbons (ACs). This system provides a rapid and reversible non-faradic electrostatic adsorption process on the surface of electrodes along with an excellent life cycle and highpower density. However, this system has the limitation of offering high energy density. The second type is called Pseudocapacitors which takes a faradaic response on the electrodes-electrolyte interfaces. Pseudocapacitors stores energy by the quick and reversible oxidation-reduction reaction. This type of SC exhibits higher energy density and lower stability comparing to the DLSCs case. The electrodes are made from metal oxide materials such as ZnO [18], V2O5 [19], and RuO2 [20] or conducting polymers [21]. Pseudocapacitors system provides higher energy density compared to DLSCs, but it suffers from lower energy density and short life cycle in comparison with DLSCs. Therefore, to reach an optimum condition, the combination of DLSCs and Pseudocapacitors (also known as (3) hybrid capacitors) have been developed to become the smart option and also the substitution of both DLSCs and Pseudocapacitors. Hybrid capacitors system stores energy by the combination of Faradaic and non-Faradic pathways in order to achieve both high energy

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