Layered assembly of hierarchical graphene/Ni-Al hydroxide composites for supercapacitors

Yasodinee Wimalasiri and Linda Zou

SA Water Centre for Water Management and Reuse, University of South Australia, Mawson Lakes, SA 5095, Australia.
E-mail: kanaththage.wimalasiri@mymail.unisa.edu.au

Abstract

A high performance supercapacitor electrode material was synthesized by a novel route using 2D nanosheets of graphene oxide and delaminated-layers of Ni-Al double hydroxide (LDH) in aqueous dispersion. The composite— G/Ni-Al LDH-based electrodes provided a specific capacitance of 915 F/g at a current density of 2 A/g based on the total mass of active materials in the absence of conductive additives. After 1500 cycles at 10 A/g current density, 95% of the initial capacitance was retained.

Supercapacitors (SCs), also known as electrochemical capacitors (ECs), are recognized as a class of promising electrical energy storage devices that possesses high power performance, long cycle life, short charging time and good safety[1, 2], which are complements to batteries and conventional capacitors. The capacitance of a supercapacitor is largely dependent on the electrode material and thus research advancement in developing high performance electrode materials is of great importance[3]. Graphene based electrodes have been comprehensively studied as supercapacitors due to high theoretical electrical double layer capacitance[4]. Significant improvements in capacitance can be achieved by the controlled structural design of graphene based composites. Utilizing pseudocapacitive materials such as layered double hydroxides (LDH) could further enhance capacitance in such composites[5], and the research on this specific topic is still limited. This study presents an innovative route to prepare a composite material with graphene and Ni/Al LDH and evaluates the performance of the composite as supercapacitors.

Ni-Al LDH was synthesized by urea hydrolysis under hydrothermal conditions and the wet cake was exfoliated in water by simple stirring and mild ultrasonication. A reasonably stable colloidal suspension having a zeta potential of +36.2 mV at pH 3 was formed. This Ni-Al LDH suspension was mixed with an aqueous dispersion of graphene oxide with a zeta potential of –47 mV at pH 5. Subsequently the graphene oxide was reduced to graphene by chemical reduction using hydrazine monohydrate in the presence of Ni-Al LDH. The resultant graphene/Ni–Al LDH (G/Ni-Al LDH) composite had 40% of Ni-Al LDH and revealed a hierarchical nanostructure where graphene and Ni-Al LDH sandwiched each other. The electrical resistivity of the composite was 0.67 Ω.cm. The composite material demonstrated a superior electrochemical performance due to the synergistic effect from the electrical double layer capacitance of graphene and pseudocapacitance of Ni-Al LDH. The high electrochemical performance and facile aqueous-based synthesis route demonstrated that the G/Ni-Al LDH composite can be a promising electrode material for supercapacitor applications.

References

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Figures

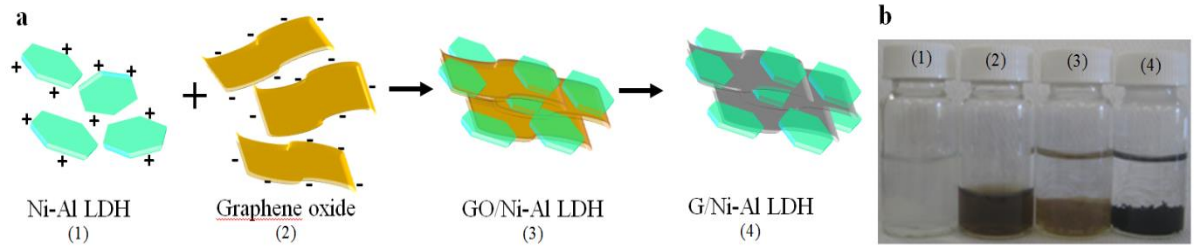

Fig. 1 Synthesis process steps of the G/Ni-Al LDH composite (a) the schematic representation and (b) the corresponding digital photographs.

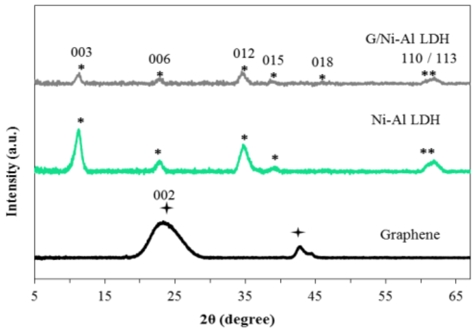

Fig. 2 The XRD patterns for graphene, Ni-Al LDH and G/Ni-Al LDH composite

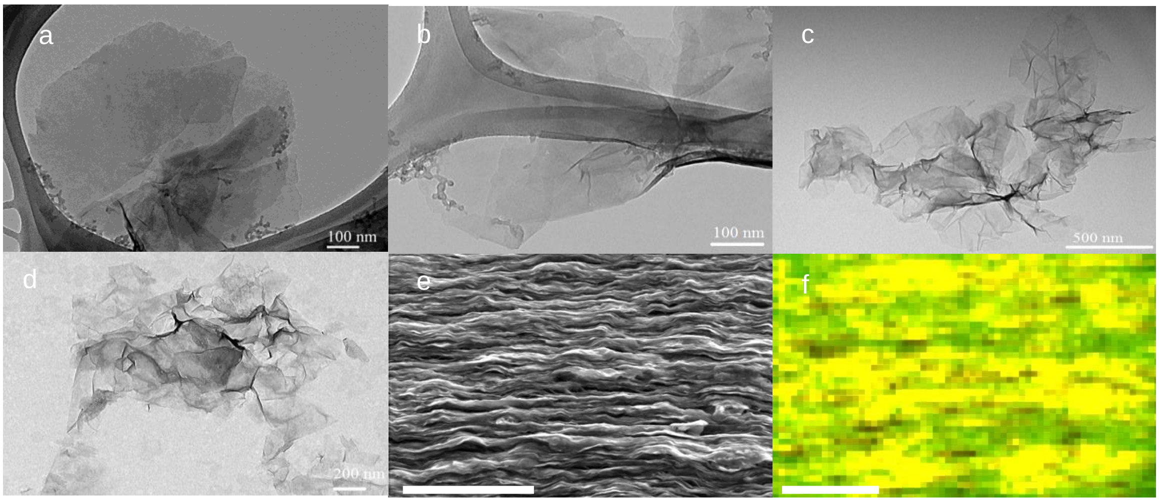

Fig. 3 TEM images of (a) dry Ni-Al LDH crystals; (b) exfoliated Ni-Al LDH nanosheets; (c) graphene; (d) G/Ni-Al LDH (e) cross sectional SEM image of G/Ni-Al- LDH, scale bar is 5 µm, and (f) the corresponding overlay of elemental distributions of C, Ni and Al by EDX, scale bar is 2 µm, green-C, yellow and brown- Ni-Al LDH

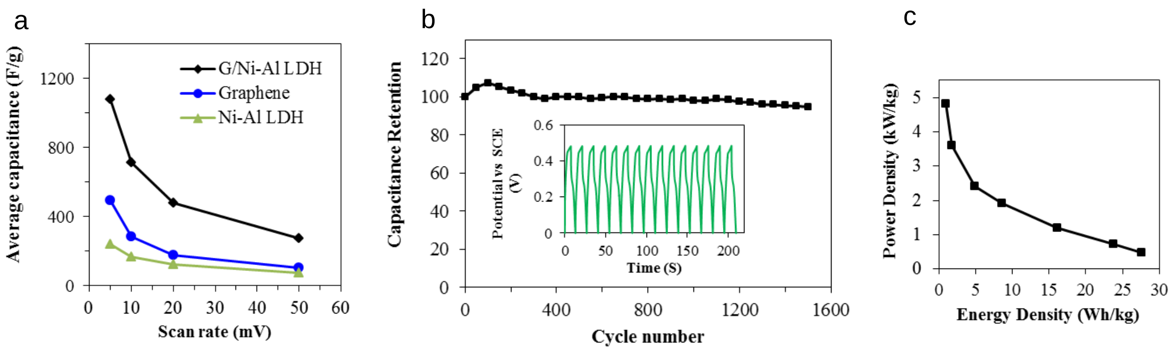

Fig. 4 electrochemical characterization (a) Variation of the average specific capacitance of G/Ni-Al LDH, Ni-Al LDH and graphene with scan rate (b) Galvanostatic discharge performance and the specific capacitance vs Cycle number at a current density of 10 A/g; (c) Ragone plot based on the mass of active material for G/Ni-Al LDH composite. The power densities and energy densities were calculated from the galvanostatic discharge curves at various current densities.