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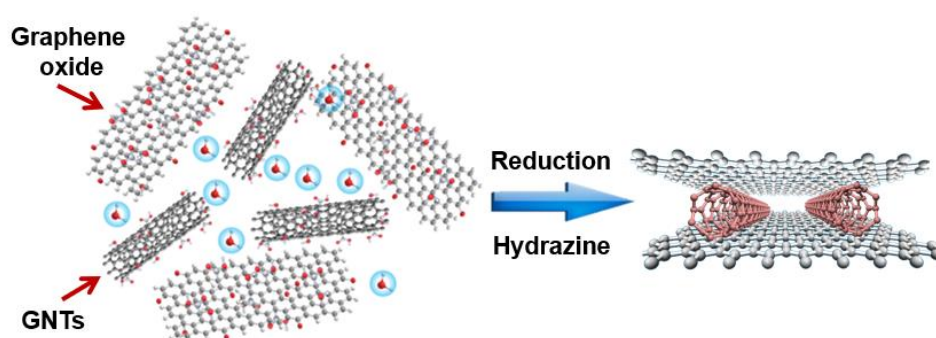
## High Energy Density Lithium Ion Capacitor Assembled with Graphene Cathode

Due to the increasing demand for rapid development of portable electronics, electric vehicles and other renewable energy products, energy-storage devices are required to provide both high energy density, power density and a longer cycle life [1]. Significant amounts of research have been devoted to environmentally friendly energy-storage devices in recent years. Lithium-ion batteries (LIBs) and electric double-layer capacitors (EDLCs) have drawn enhanced attentions because of this. However, LIBs and EDLCs are unable to satisfy the demands in terms of high energy density and power density, due to its own energy storage mechanisms. The reason for these effects are Faradaic reactions occur on the electrode in LIBs, which leads to a much higher capacity and energy density ( $100\text{-}200\text{ Wh kg}^{-1}$ ) [2]. However, due to slow kinetics processes while charging and discharging, LIBs have low power density and a short cycle life. On the other hand, EDLCs can offer high power density (as high as  $10\text{ kW kg}^{-1}$ ) and a long cycle life. Since EDLCs store energy by either reversible ion adsorption/desorption or fast surface redox reactions, though their relatively low energy density (less than  $10\text{ Wh kg}^{-1}$ ) has limited their applications [3]. Thus, it is strongly important to develop an energy storage device which can bridge the gap between LIBs and EDLCs. Lithium ion capacitors (LICs) have been proposed as a solution which combines the advantages of LIBs and EDLCs. LICs are constructed from a LIBs-type electrode (a negative electrode of LICs) and EDLCs-type electrode (a positive electrode of LICs) with a lithium salt electrolyte [4-6]. With this configuration, LICs store energy by simultaneously utilizing reversible Faradaic reactions as well as adsorption/desorption reactions in the anode and cathode respectively. Generally, the capacitance of an LICs device will be influenced by EDLCs-type cathode [7]. It is important to seek new cathode materials with lower cost and better properties in the research field of LICs so that the capacitance and energy density can be improved further. Graphene and its composite have become a focus in the research field of LICs, due to its extraordinary electrical conductivity, high specific surface area, chemical and thermal stability, and porous structure which makes it a competitive candidate as cathode material for LICs [8]. Based on above considerations, in this study we construct 3-dimensional (3D) graphene/carbon nanotubes (CNTs) structures by employing CNTs as spacers between graphene nanosheets. The schematics is shown in Figure 1. The long and tortuous CNTs can effectively inhibit graphene from aggregating in the composite, which will significantly increase the accessible surface area between the electrolyte and the active materials. Furthermore, the 3D network structure can also provide fast and efficient conductive pathways for ion adsorption/desorption. Herein, we employ graphene/CNTs nanocomposite as the EDLCs-type cathode assembled with pre-doped graphite as the LIBs-typed anode to construct a hybrid LICs as seen in Figure 2. We also studied the detailed electrochemical behaviors of graphene/CNTs LICs. The 3D network structure and porous structure of graphene/CNTs can offer better diffusive paths for ions. Through this special designed structure, graphene/CNTs LIC can achieve a high specific capacitance (about  $107\text{ F g}^{-1}$ ), a high energy density (about  $205\text{ Wh kg}^{-1}$ ), which is improved about 6 times than that of EDLC, and an improvement of 40% over AC LIC.

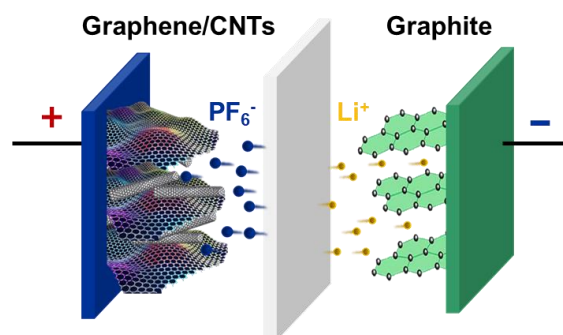
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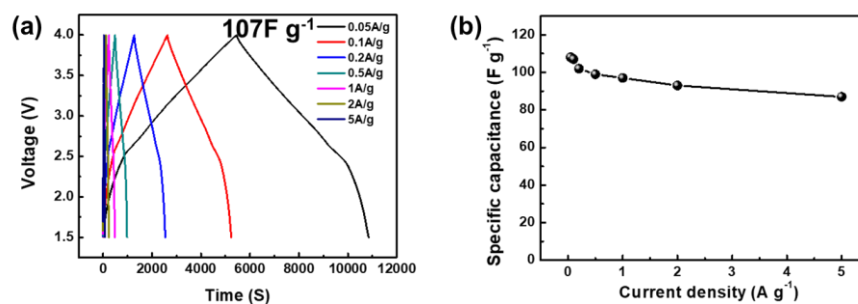
## Figures



**Figure 1:** Schematic illustration of the prepared graphene/CNTs nanocomposites



**Figure 2:** A schematic diagram of LICs in which graphene/CNTs and pre-lithiated graphite are used as the cathode and anode respectively



**Figure 3:** (a) Galvanostatic charge-discharge curves at increasing current densities. (b) Rate performance of the graphene/CNTs LIC full cell (1.5–4 V)