Supporting Information

Solvothermal Hybridization of $LiMn_{1/3}Ni_{1/3}Co_{1/3}O_2$ and Reduced Graphene Oxide to Promote Lithium-ion Cathode Performance

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Experimental details

Preparation of materials

Preparation of LMNC: The layered LMNC powders were synthesized by a sol-gel method using citric acid as a chelating agent. A stoichiometric amount of lithium acetate, manganese acetate, and nickel acetate and cobalt acetate were chosen as starting materials to prepare the precursor. All the salts were dissolved in an appropriate quantity of distilled water and added dropwisely into citric acid solution under continuous stirring at 80 ºC until the clear viscous gel was formed. The gel was dried at 120 ºC for 20 h to obtain the precursor powder. After heated at 450 ºC in air for 5 h, the powder was ground and sintered at 850 ºC for 12 h.

Preparation of GO: Aqueous colloidal suspension of GO was prepared by a modified Hummer's method. Briefly, 1g flake graphite and 0.5 g $NaNO_3$ were added into 23 mL of concentrated H_2SO_4 (98%, w/w) and cooled to 0 ºC in an ice bath. 3 g $KMnO_4$ was added slowly into the mixture under vigorous stirring. The reaction temperature was maintained below 20 ºC for 4 h, then 35 ºC for 24 h. The mixture gradually became thick and appeared dusty green. Additional 3 g $KMnO_4$ was added slowly and stirred for 4 h, then 23 mL H_2SO_4 (98%, w/w) was mixed in and stirred for 24 h. The reaction mixture was cooled to the room temperature and poured onto ice (500 mL) with 10 mL H_2O_2 (30%, w/w). The color of the mixture turned into light yellow. The centrifugal product was washed with HCl (10%, w/w) and water. Dialysis was carried out to further remove the remaining metal species. The graphite oxide dispersion was sonicated under ambient conditions for 1.5 h. The resultant homogeneous brown GO dispersion was obtained, which is stable for several months.

Preparation of RGO/LMNC: The sintered LMNC powders were dispersed in GO water/ethanol or water/ethylene glycol (volume ratio of 2:1) mixture solution, and the weight proportion of GO with respect to LMNC was applied about 5%; this mixture solution was transferred into a 100 mL Teflon-line autoclave with a filling degree of 80%. The autoclave was placed into an oven to heat at 120 ºC for 3 h. After cooling to room temperature, the powdery precipitates were collected and washed thoroughly with distilled water, and then dried at 120 ºC in air. The obtained composite materials hybridized by the sintered LMNC with ethanol reduced graphene oxide and ethylene glycol reduced graphene oxide are denoted as EtRGO/LMNC and EGRGO/LMNC, respectively.

Characterization and measurements

The crystal structures and the surface morphologies of pristine pre-sintered LMNC, EtRGO/LMNC and EGRGO/LMNC nanocomposites were examined by powder X-ray diffraction (XRD) and field emission-scanning electron microscopy (FE-SEM), respectively. The microcrystal shape and local atomic arrangement of nanocomposites were observed with high resolution-transmission electron microscopy (HR-TEM). Carbon contents in EtRGO/LMNC and EGRGO/LMNC nanocomposites were estimated by thermogravimetric analysis (TGA), which was performed at a heating rate of 10 **ºC** min^{-1} from room temperature to 800 **ºC** in ambient atmosphere.

The electrochemical measurements were carried out using coin-cell setup (CR-2016), in which pure lithium foil was used as a counter electrode and the electrolyte consisted of 1M $LiPF_6$ in ethylenecarbonate (EC) / dimethylcarbonate (DMC) / diethylcarbonate (DEC) (1:1:1 in volume). The cathodes were prepared by coating the slurry of a mixture composed of 85 wt% active material, 10 wt% acetylene black, and 5 wt% poly(vinyl difluoride) (PVDF) onto an cleaned and polished aluminum foil. Subsequently, the electrodes were dried at 100 **ºC** under vacuum for 24 h. Cyclic voltammograms (CVs) were recorded in the voltage of 2.5–4.3 V at a scanning rate of 0.05 mV s^{-1}. Galvanostatic charge-discharge tests were performed at C rates of 0.1, 0.5, 1, 2, 5 in the voltage range 2.5–4.3 V. The theoretical capacity of LMNC assumed in this work is 200 mAh g^{-1}. The potential values mentioned in the text are with respect to Li/Li^{+}.

Characterization data of GO

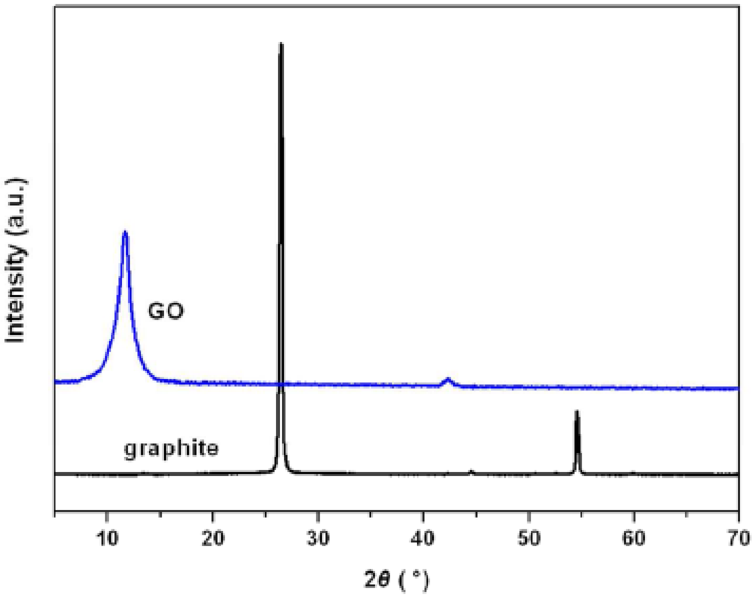

Fig.S1 XRD of the prepared GO and the used pristine graphite.

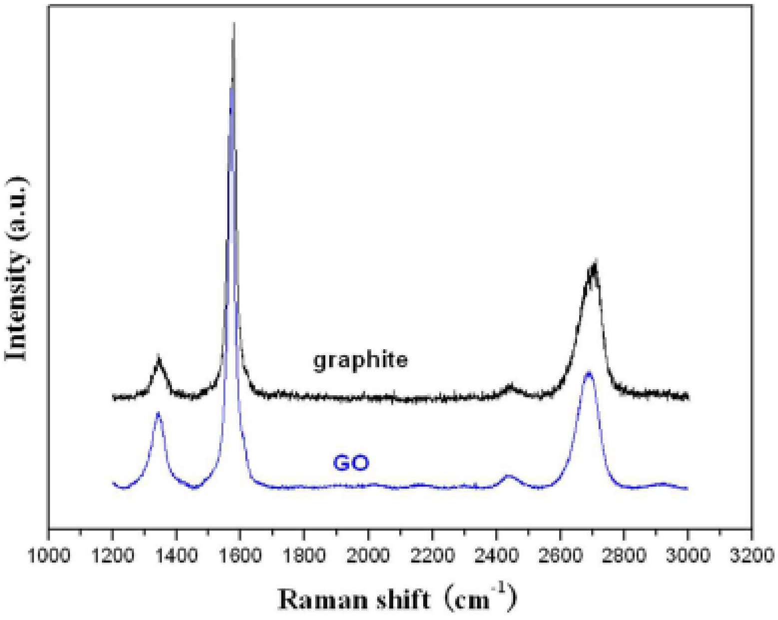

Fig.S2 Raman spectra of the prepared GO and the pristine graphite

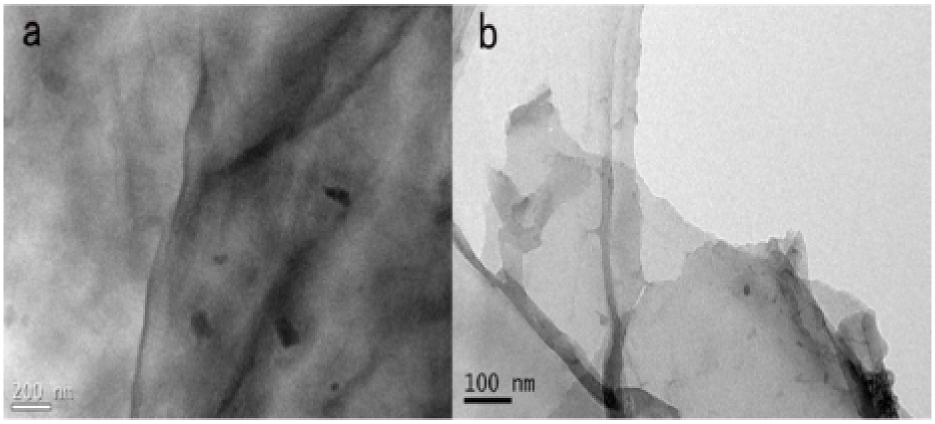

Fig.S3 SEM of the prepared GO at basal (a) and edged (b) views.

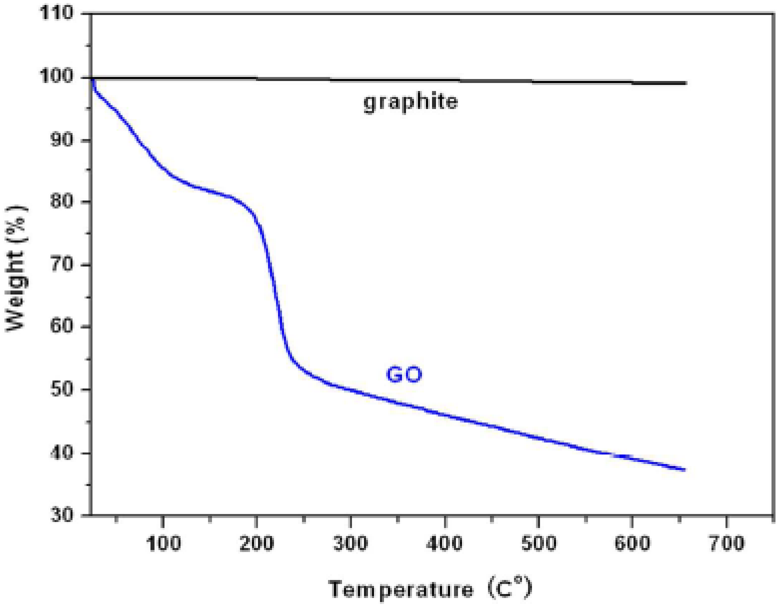

Fig.S4 TGA of the prepared GO and the pristine graphite.