

## Supporting Information

### Compressible graphene aerogel supported CoO nanostructures as binder-free electrode for high-performance lithium-ion batteries

Yanfeng Dong, Shaohong Liu, Zhiyu Wang,\* Yang Liu, Zongbin Zhao,\* and Jieshan Qiu \*

*Carbon Research Laboratory, Liaoning Key Lab for Energy Materials and Chemical Engineering, State Key Lab of Fine Chemicals, School of Chemical Engineering, Dalian University of Technology, Dalian 116023, China Email: zywang@dlut.edu.cn; zbzhaoy@dlut.edu.cn; jqiu@dlut.edu.cn*

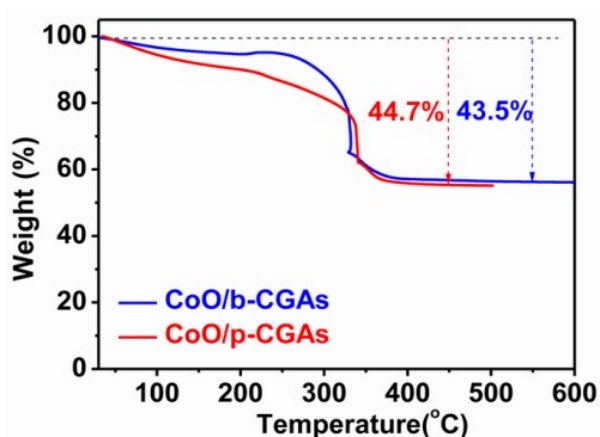
#### Experimental

**Material synthesis.** GO was firstly synthesized by modified Hummers method.<sup>21</sup> Polypyrrole functionalized CGAs (denoted as p-CGAs) were prepared by heating the mixture solution of 30 mL GO suspension (0.5 mg mL<sup>-1</sup>) and pyrrole solution (5.0 vol. %) at 180 °C for 12 h under hydrothermal conditions, followed by freeze drying for 48 h. For the comparison, bare CGAs (denoted as b-CGAs) were produced by hydrothermal reaction of GO under the same conditions, followed by freeze drying for 48 h and annealing the products in N<sub>2</sub> flow at 800 °C for 1 h. For the growth of CoO nanostructures, p-CGAs or b-CGAs were immersed into the mixture solution of Co(NO<sub>3</sub>)<sub>2</sub> (5 mmol), NH<sub>4</sub>F (10 mmol) and CO(NH<sub>2</sub>)<sub>2</sub> (25 mmol) in 50 mL of distilled water in Teflon-lined stainless steel autoclaves. After heating at 120 °C for 5 h, CoO/p-CGAs or CoO/b-CGAs marcoassemblies were handled out with tweezers, washed with distilled water for several times and were annealed at 350 °C in N<sub>2</sub> flow for 2 h.

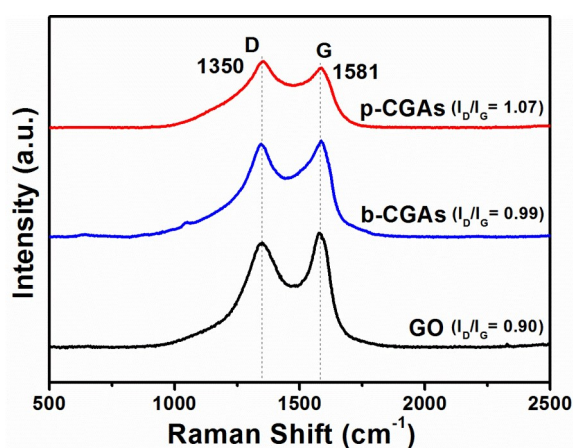
**Material characterization.** All samples were characterized by field-emission scanning electron microscopy (SEM, QUANTA 450, 20KV), transmission electron microscopy (TEM, FEI Tecnai G20, 200 KV), X-Ray diffraction (XRD, D/Max 2400 diffractometer, Cu K $\alpha$ ,  $\lambda$ =1.5406 Å) and nitrogen adsorption/desorption (Micromeritics ASAP 2020 instrument). The surface characteristics of the samples were investigated using a Nicolet-20DXB Fourier transform infrared spectrometer (FTIR). The weight ratio of CoO in the architectures was estimated by thermogravimetric analysis (TGA, TA-Q50).

**Battery performance test.** CR2026 coin cells were assembled with CoO/p-CGAs or CoO/b-CGAs marcoassemblies as binder-free anodes and pure lithium foil as the counter and reference electrode at room temperature in glove box. For comparison, the electrodes composed of the powders of CoO nanostructures, carbon black and polyvinylidene difluoride (PVDF) in a weight ratio of 7:2:1 were

also made. For all the tests, the electrolyte used are 1.0 M LiPF<sub>6</sub> in a mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) with volume ratio of 1:1. The galvanostatic charge/discharge tests were performed on a LAND CT2000 battery tester at different current densities within a cut-off voltage window of 0.01-3.00 V. The calculation of the specific capacity is based on the total mass of the composite. The cyclic voltammograms (CVs) were conducted using a CHI660D electrochemistry workstation between 0.01-3.0 V at a scan rate of 0.1 mV s<sup>-1</sup>. The electrochemical impedance spectroscopy (EIS) measurements were carried out using a CHI660D workstation by applying the AC amplitude of 5 mV over the frequency range of 100 kHz to 0.01Hz.



**Fig. S1** TGA curves of CoO/p-CGAs and CoO/b-CGAs macroassemblies, which were measured in air between room temperature to 600 °C with a ramp rate of 10 °C min<sup>-1</sup>. The loading amount of CoO is calculated assuming it is completely converted to Co<sub>3</sub>O<sub>4</sub> after annealing in air.



**Fig. S2** Raman spectra of p-CGAs, b-CGAs and GO.

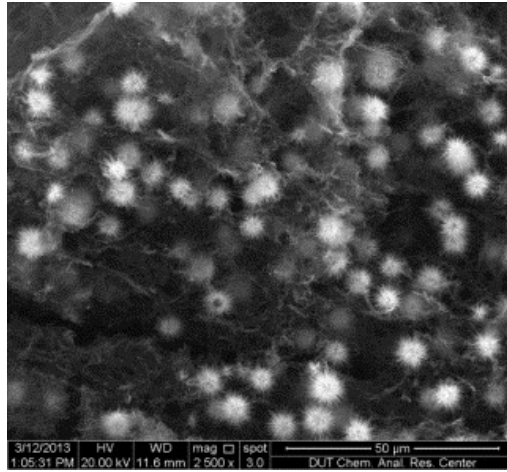


Fig. S3 SEM image of CoO/b-CGAs macroassemblies.

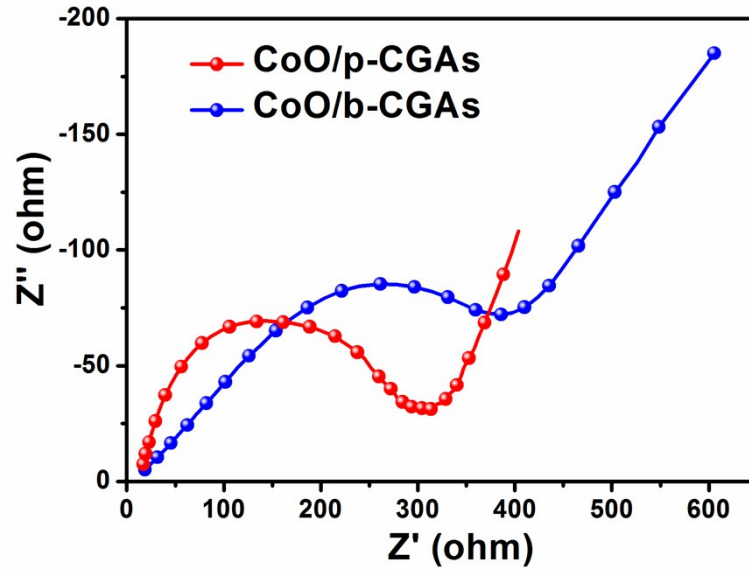


Fig. S4 Nyquist plots of CoO/p-CGAs and CoO/b-CGAs electrodes.