Supporting Information:

Synthesis of graphite oxide:

Graphite oxide was synthesized from natural graphite (SP-1) by a modified Hummer's method.[1-3] In brief, 1.5 g graphite powder was added into a mixture of 10 mL 98% H_2SO_4, 1.25 g $K_2S_2O_8$, and 1.25 g P_2O_5, and the solution was maintained at 80 ^oC for 4.5 h. The resulting pre-oxidized product was cleaned using water and dried in a vacuum oven at 50 ^oC. After it was mixed with 60 mL 98% H_2SO_4 and 7.5 g $KMnO_4$ was slowly added at a temperature below 20 ^oC, then 125 mL H_2O was added. After 2 h, additional 200 mL H_2O and 10 mL 30% H_2O_2 were slowly added into the solution to completely react with the excess $KMnO_4$. After 10 minutes, a bright yellow solution was obtained. The resulting mixture was washed with dilute HCl aqueous (1/10 v/v) solution and H_2O. The graphite oxide was obtained after drying in a vacuum oven at 30 ^oC.

Synthesis of ferrocene nanocluster/rGO:

In a typical procedure, 50 mg graphite oxide (GO) was dispersed into 70 mL 99.9% ethanol by sonication. Then, 50 mg ferrocene was added into the solution under sonication. The mixture was sealed in a Teflon-lined autoclave and maintained at 170 ^oC for 4 h. After it was cooled down to room temperature, the precipitate, e.g. ferrocene/rGO, was collected and washed by ethanol.

The pure rGO samples were obtained under the same synthesis conditions without the addition of ferrocene.

Materials Characterization:

The morphology of the samples were investigated by using a field-emission scanning electron microscopy (FESEM) system (JEOL, Model JSM-7600F), and the nanostructures of the samples were characterized by using a transmission electron microscopy (TEM) system (JEOL, Model JEM-2100) operating at 200 kV. To investigate the samples via TEM, a suspension of the ferrocene/rGO nanohybrids in ethanol was drop-casted onto carbon-coated copper grids and dried under ambient condition. Crystal phases of samples were identified using a Scintag PAD-V X-ray diffractometer with

Cu Kα irradiation. Thermogravimetry analyses (TGA, Q500) were carried out in the temperature range of 30 to 700 °C at a heating rate of 10 °C min^{-1} in Air. Here, the samples was weighted and then treated by 69% HNO_3. This process has to be taken very carefully as the addition of HNO_3 into the sample may ignite fire. After the treatment, the remaining black powder was washed and then used for TGA measurements after drying them. Finally, the weight of ferrocene in the composite was calculated based on the TGA results. Raman spectra were obtained with a WITec CRM200 confocal Raman microscopy system with a laser wavelength of 488 nm and a spot size of 0.5 mm. Matrix-assisted laser desorption/ionization time-of-flight mass spectroscopy (MALDI-TOF-MS) spectra were performed on an Axima TOF2 mass spectrometry (Shimadzu/Kratos, Singapore). Each mass spectrometer uses a nitrogen laser at 337 nm (Pulse 10 ns, 10 Hz) for desorption of analytes. The spectrum was recorded by adding 100 laser shots using a laser energy of 56.80 μJ. The accelerating voltage is 20 kV, and the operating pressure is 10^{-6} mbar. Electrochemical impedance spectroscopy (EIS) measurements were carried out in the frequency range from 10 kHz to 0.1 Hz at open circuit potential with an ac perturbation of 10 mV with the help of an impedance spectrum analyzer (Solatron, SI 1255B Impedance/grain-phase analyzer and computer software ZView).

Electrochemical Measurements:

80 wt% active material (rGO or ferrocene/rGO), 10 wt% acetylene black (Super-P), and 10 wt% polyvinylidene fluoride (PVDF) binder were mixed into N-methyl-2-pyrrolidinone (NMP). The obtained slurry was coated onto Cu foils, which were finally dried in vacuum at 50 ^{o}C for 12 h to remove the solvent. Electrochemical measurements were carried out on CR2032 (3 V) coin-type cells with lithium metal as the counter/reference electrode, Celgard 2400 membrane as the separator, and electrolyte solution obtained by dissolving 1M LiPF6 into a mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC) (EC/DMC, 50 : 50 w/w). The coin cells were assembled in an Ar-filled glovebox with concentrations of moisture and oxygen below 1.0 ppm. The amount of active material

used for one sample is around 2 mg and the thickness is around tens of micrometers. The diameter of cathode (Li foil) is 18 cm. The charge/discharge tests were performed with a NEWARE battery tester at a voltage window of 0.05-3.0 V for rGO and ferrocene/rGO samples. Cyclic voltammetry (0.05-3 V, 0.5 mV s^{-1}) was performed with an electrochemical workstation (CHI 660C).

Reference:

1.W. S. Hummers and R. E. Offeman, Journal of the American Chemical Society, 1958, 80, 1339-1339.
2.Y. Xu, H. Bai, G. Lu, C. Li and G. Shi, Journal of the American Chemical Society, 2008, 130, 5856-5857.
3. J. X. Zhu, T. Zhu, X. Z. Zhou, Y. Y. Zhang, X. W. Lou, X. D. Chen, H. Zhang, H. H. Hng and Q. Y. Yan, Nanoscale, 2011, 3, 1084.

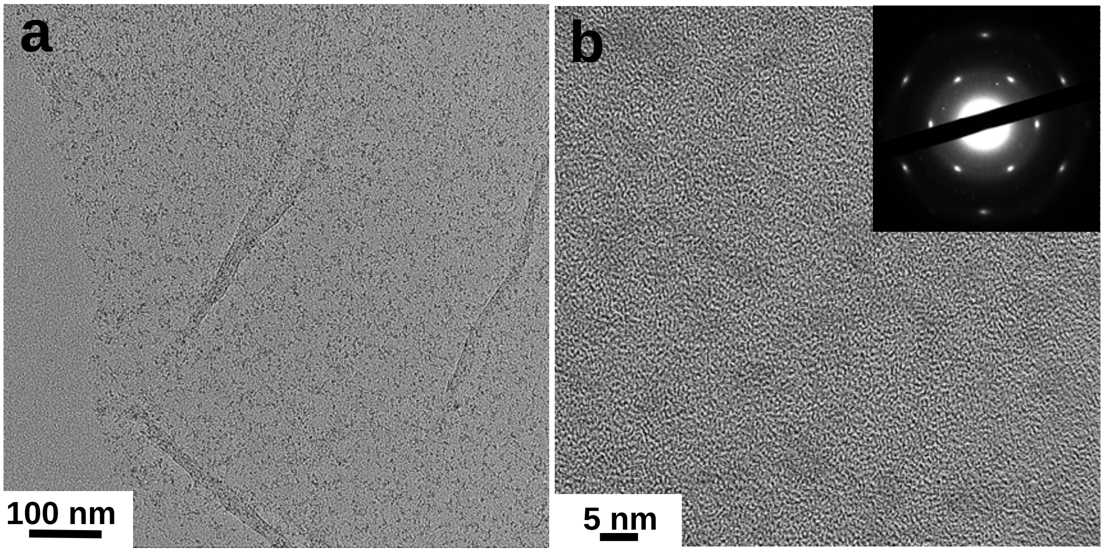

Figure S1 The enlarged version of the images of Figure 1a-b in the manuscript

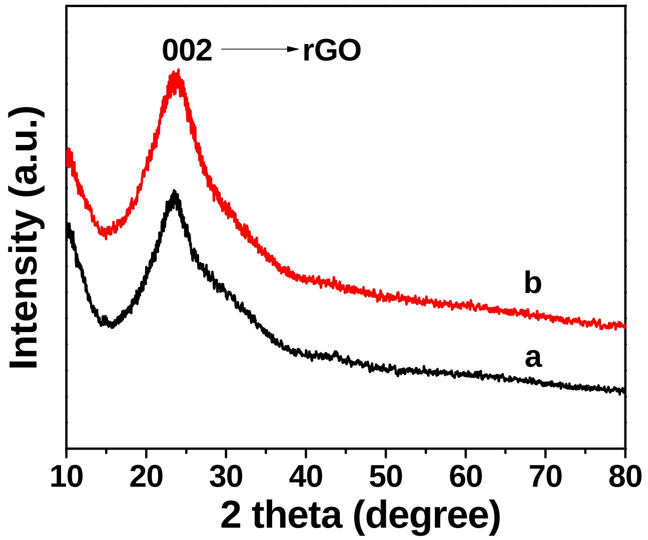

Figure S2. X-ray diffraction pattern (XRD) of the as-prepared samples with (a) $R_{ferrocene:GO}$ =1:1 and (b) $R_{ferrocene:GO}$ =2:1.

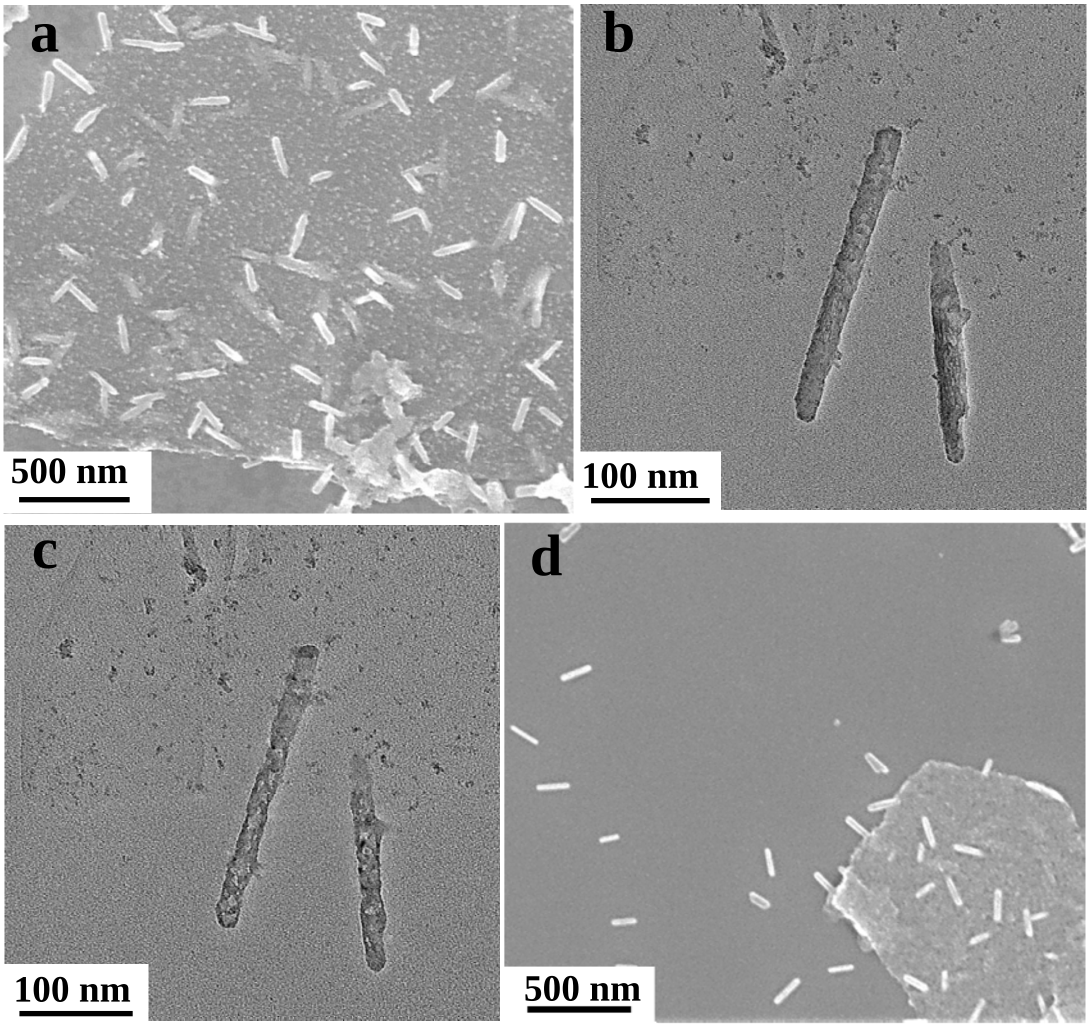

Figure S3 SEM image of the ferrocene/rGO samples prepared with $R_{ferrocene:GO}$ =2:1; (b-c) TEM images of the same sample (b) before and (c) after electron beam irradiation for 10 seconds; (d) SEM image of the ferrocene/rGO samples prepared with $R_{ferrocene:GO}$ =2:1 to show that there are free-standing ferrocene nanorods not attached to the rGO sheets.

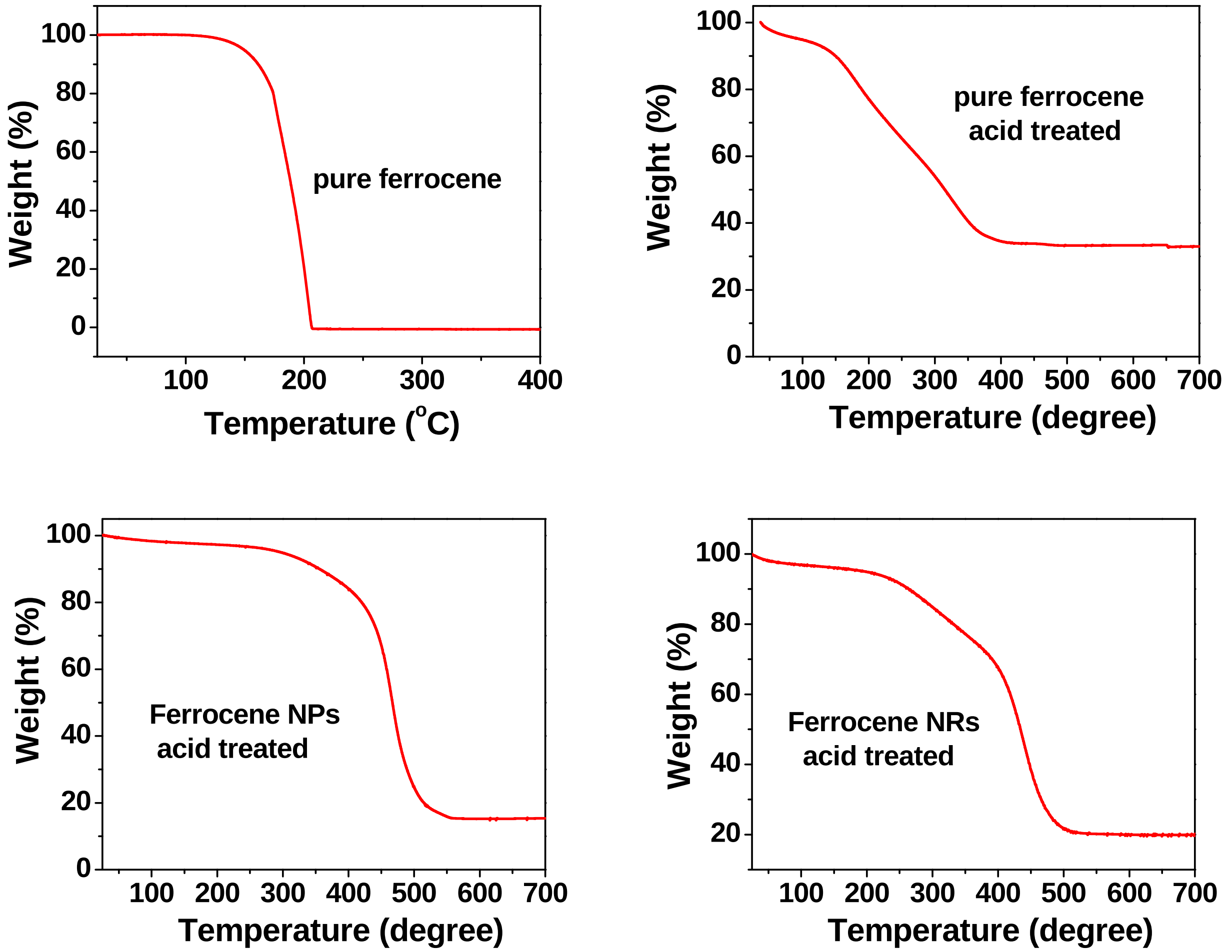

Figure S4 Thermogravimetry analyses (TGA) of the samples prepared with (a) pure ferrocene without HNO_3 acid treatment; (b) pure ferrocene after HNO_3 acid treatment; (c) ferrocene/rGO sample with $R_{ferrocene:GO}$=1:1 after HNO_3 acid treatment, and (d) ferrocene/rGO sample $R_{ferrocene:GO}$=2:1 after HNO_3 acid treatment.

Here, TGA was measured from 25 to 700 °C at a heating rate of 10 °C/min in Air, where the weight loss before 100 °C was attributed to the loss of water, the residual is treated as Fe_2O_3.

Note: The residual powder after the TGA process is verified to be Fe_2O_3. The TGA on pure ferrocene after HNO_3 treatment is used at the controlled experiment to verify that the Fe in the initial ferrocene sample is 43 wt%. Based on the weight of the residual Fe_2O_3 and the weight of the initial ferrocene/rGO samples, the weight percentage of ferrocene in ferrocene/rGO obtained with $R_{ferrocene:GO}$ =1:1 is around 39 wt% while the weight percentage of ferrocene in ferrocene/rGO obtained with $R_{ferrocene:GO}$=2:1 is around 54 wt%.

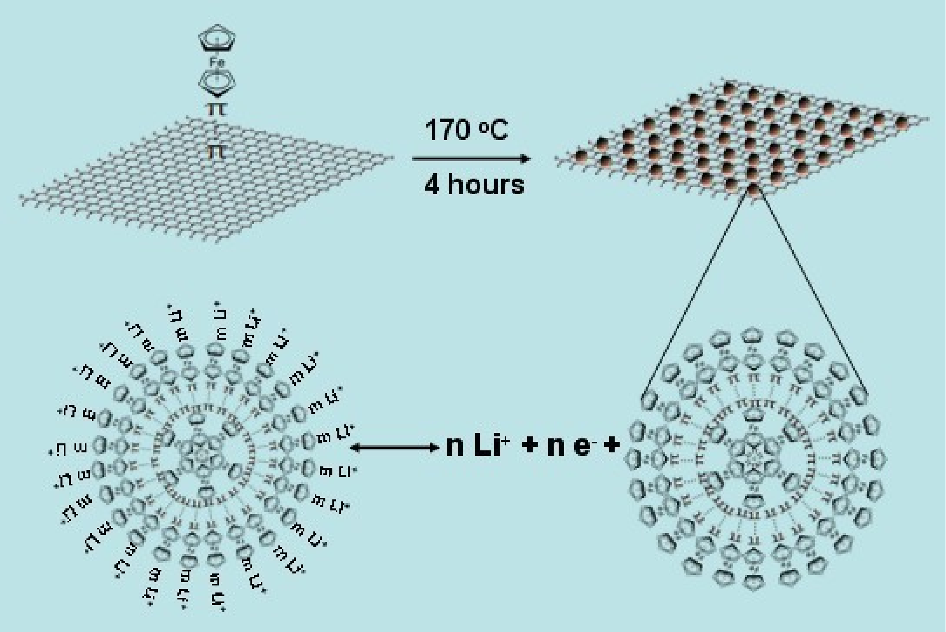

Figure S5 Scheme illustrating the proposed mechanism on the grafting of ferrocene nanoclusters onto rGO and the proposed redox reaction.

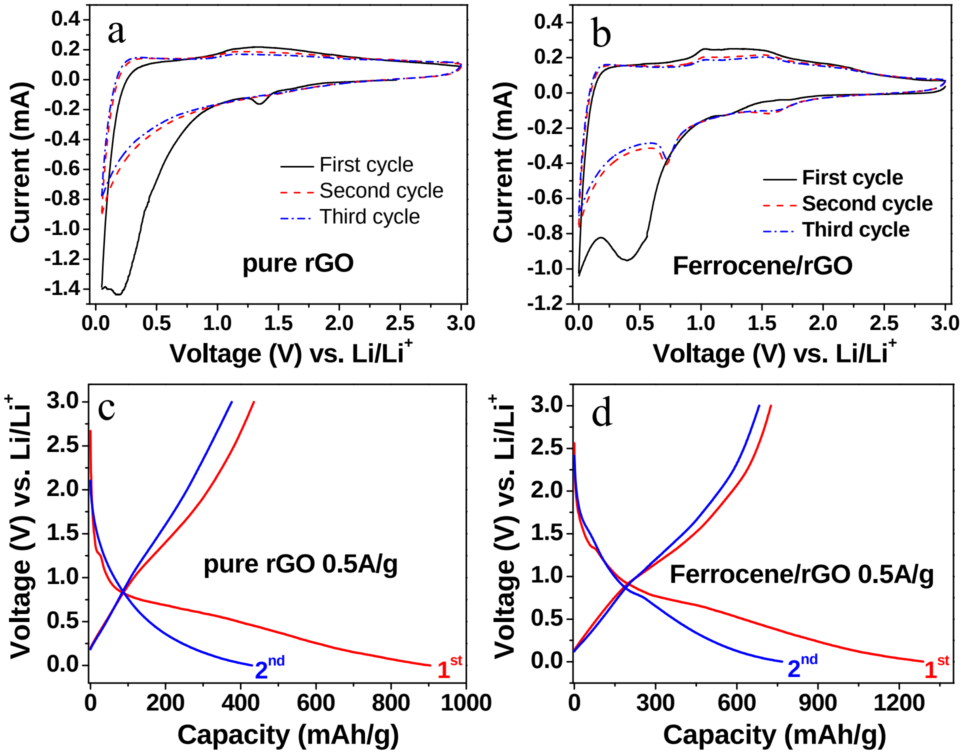

Figure S6 Representative CV profiles of (a) pure rGO electrode and (b) ferrocene/rGO electrode with $I_{ferrocene}$ = 39 wt% at a scan rate of 0.5 mV/s for the first three cycles; Charge-Discharge voltage profiles of (c) pure rGO electrode and (d) ferrocene/rGO electrode with $I_{ferrocene}$ = 39 wt% for the first two cycles at 0.5 A/g.

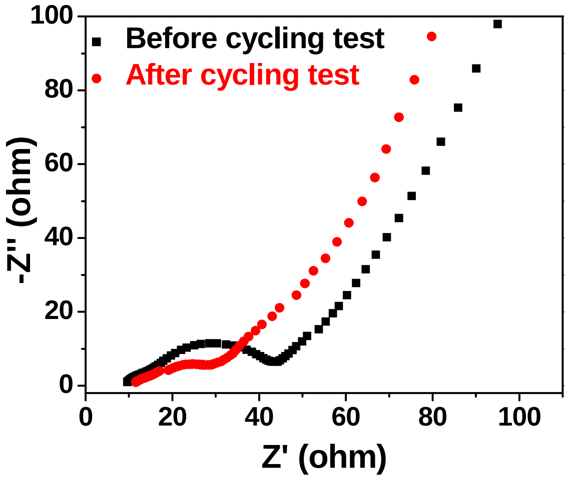

Figure S7 Nyquist plots of ferrocene/rGO with $I_{ferrocene}$ = *39 wt%* electrodes before and after cycling testing obtained by applying a sine wave with amplitude of 10.0 mV over the frequency range 10 kHz – 0.1 Hz. In impedance spectroscopy, high frequency activity is attributed to charge transfer phenomenon whereas low frequency region of the spectrum is ascribed to the mass transfer process. The Nyquist plots for the cell before and after cycling testing were similar except for the diameters of the semicircles, and thereby the associated impedance values. This indicates the impedance in ferrocene/rGO was reduced after cycling processes.

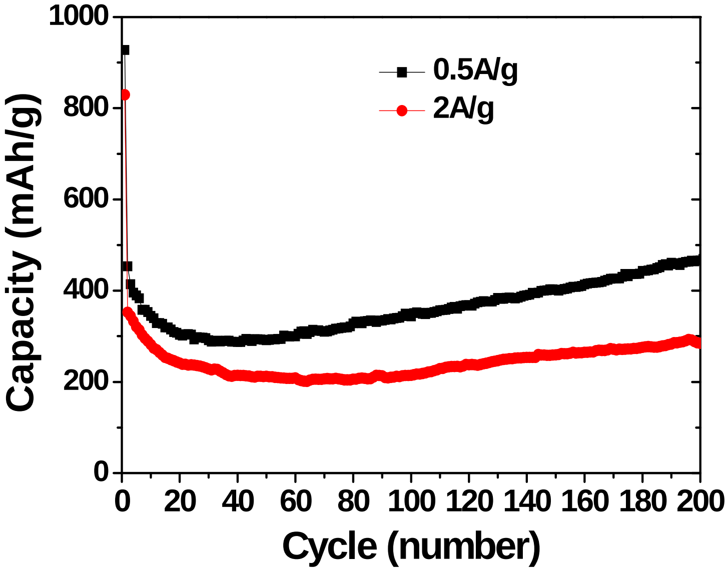

Figure S8 Cycling performances of the ferrocene/rGO sample with $I_{ferrocene}$ = *54 wt%* at different current densities within a voltage window of 0.05-3.0 V.