Supporting Information for

Lithium Cell Assisted Low-Overpotential Li-O_2 Batteries by In Situ Discharging Activation

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Experimental Section

Electrode preparation.

Li-O_2 battery performances were tested using prototype cell including CR2032 type coin cells with holes on cover (**Figure S1**). Carbon paper (CP) was pre-treated in acetone and ethanol in 10 mins respectively, dried in a vacuum oven overnight at 120 ºC. Then the CP electrode was cut with a diameter of 15 mm. Carbon nanotubes and graphene electrode Carbon materials cathode was made by mixing CNTs or graphene and PVDF in weight ratio of 8:2, the mixture was pressed into the stainless steel net (d＝15 mm)，and then dried in a vacuum oven overnight at 120 ºC. We constructed the lithium batteries that contained an electrolyte composed of 1 M lithium bis(trifluoromethanesulfonyl)imide (LITFSI) in tetraethylene glycol dimethyl ether (TEGDME) and a CP cathodes. Pure lithium slice was used as the counter electrode. For electrode activation, the battery was assembled in an argon-filled glove box. The assembled battery was initially discharged at different currents of 0.27, 0.54, 0.81 and .108 mA with cut-off voltage of 0.01, 0.1, 0.5, and 1 V on a Neware CT−3008 battery testing system, Electrochemical studies of Li-O_2 batteries were also performed at room temperature (25 ºC). After filled with oxygen in the prototype cells, discharge-charge curves were tested at a current of 0.03 mA between 2.2 and 4.5 V with a cut-off capacity of 1 mAh. More information is provided in Supporting Information.

Characterizations:

The crystal structure was characterized by X-ray diffraction (XRD, D/MAX 2500V) on an instrument equipped with Cu Kα radiation that was operated over a 2θ range of 20º-35º in a continuous scan mode with a scan rate of 2ºmin^{-1}. The morphology of the carbon paper was examined using field emission scanning electron microscopy (FESEM, SU8020). Raman spectroscopy was performed using a Raman spectrometer (Horiba Jobin Yvon, HR Evolution). X-ray photoelectron spectroscopy (XPS) experiments were carried out on a Thermo ESCALAB 250XI instrument using aluminum Kα X-ray radiation. XPS spectral analysis was conducted using XPS Peak-

fit software.

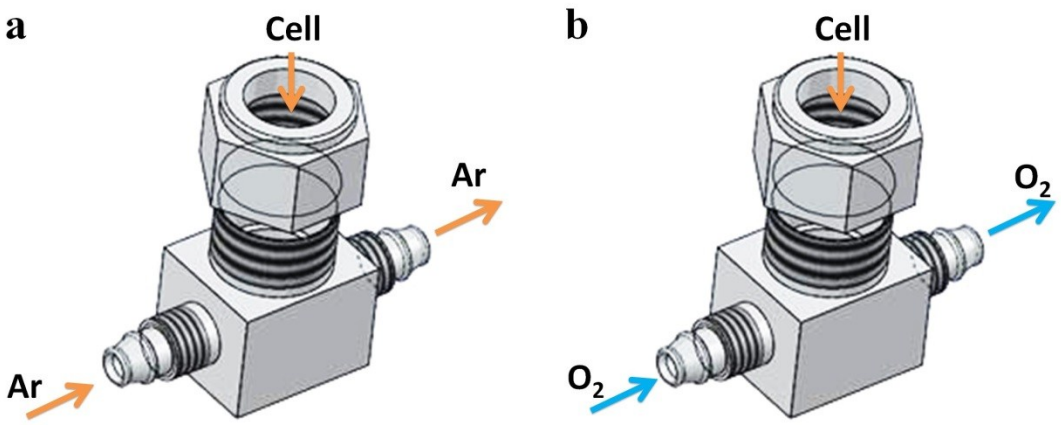

Figure S1. Prototypes of bifunctional tester for lithium ion cell (a) and lithium oxygen cell (b).

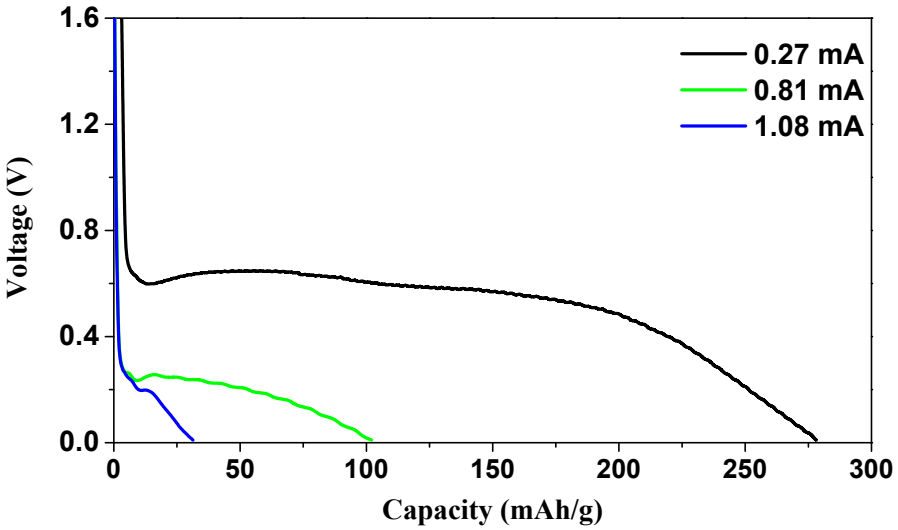

Figure S2. Discharging curves of Li/GCP cells with different current of 0.27, 0.81 and 1.08 mA respectively.

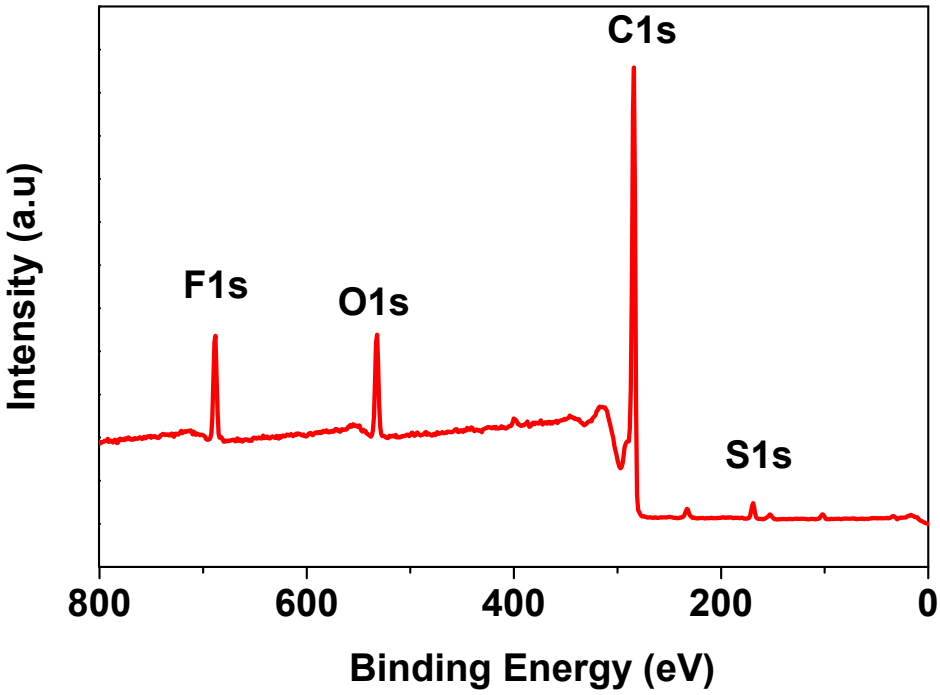

Figure S3. The survey XPS curve of activated CP with discharging to 0.01V.

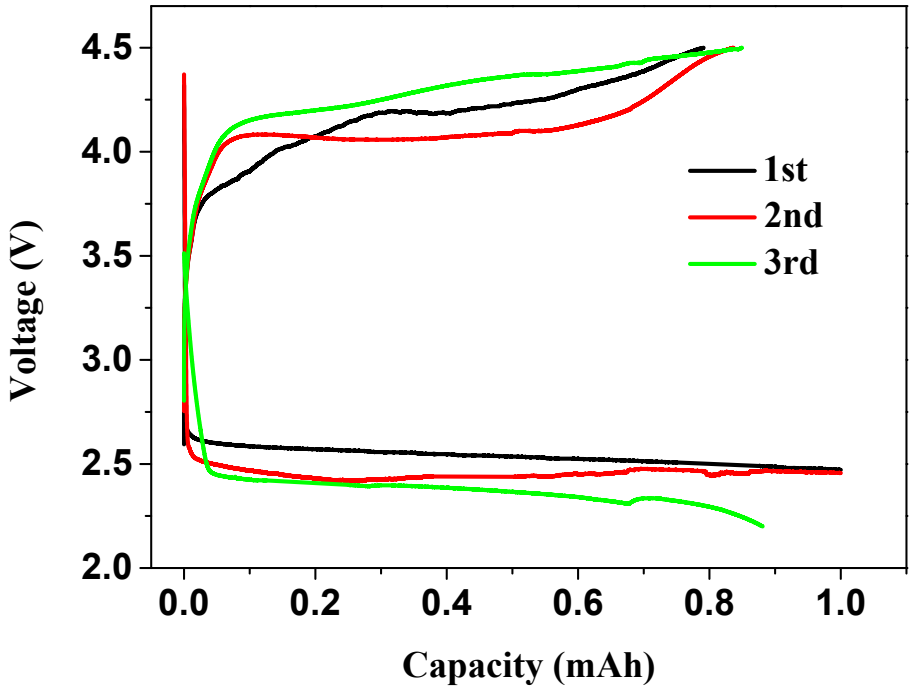

Figure S4. Discharge/charge curves of LOB with CPs activated at a rate of 0.54 mA

with cut-off voltage of 1V (GCP-1V-0.54mA).

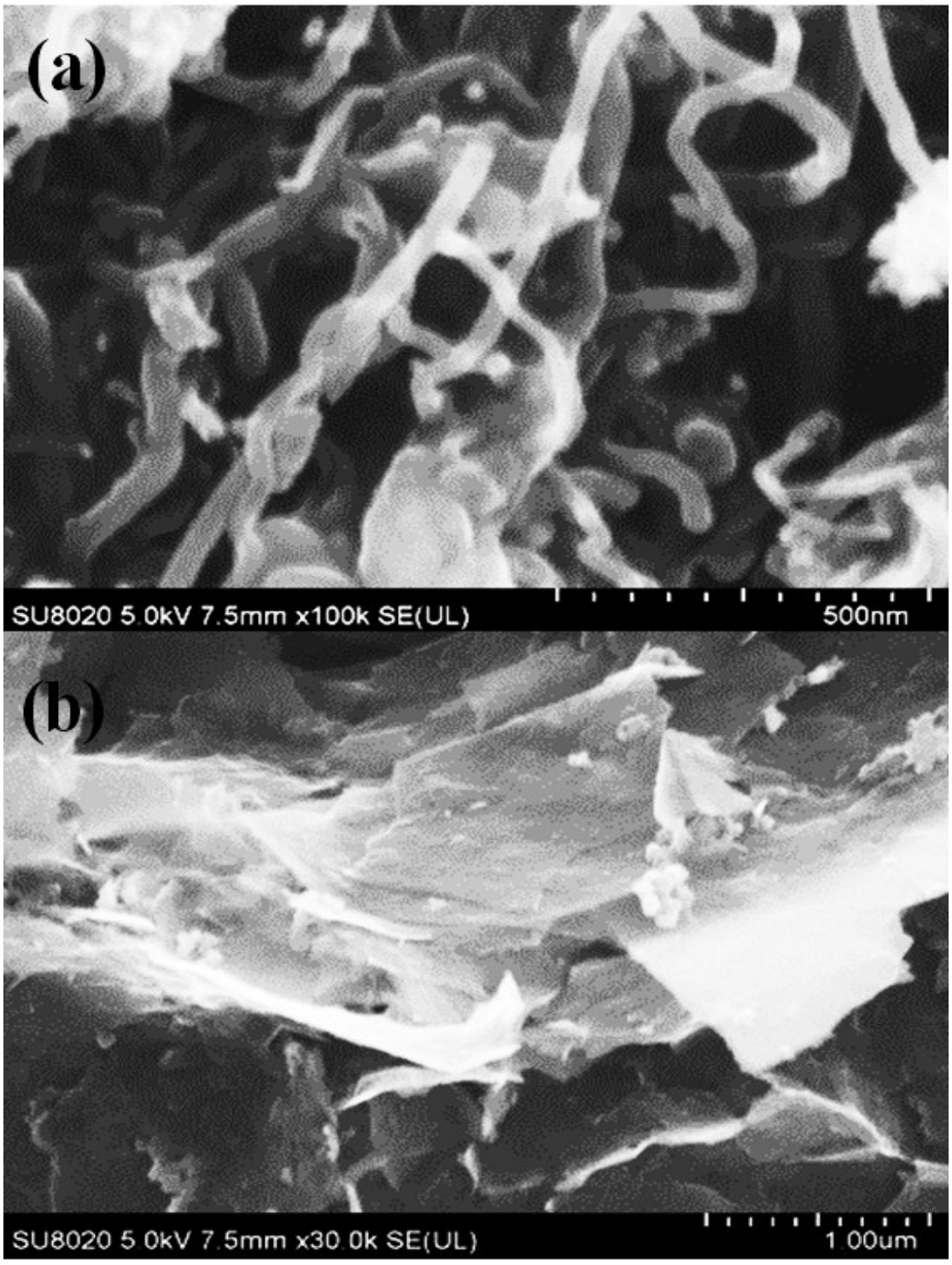

Figure S5. FESEM images of CNTs (a) and graphene (b).

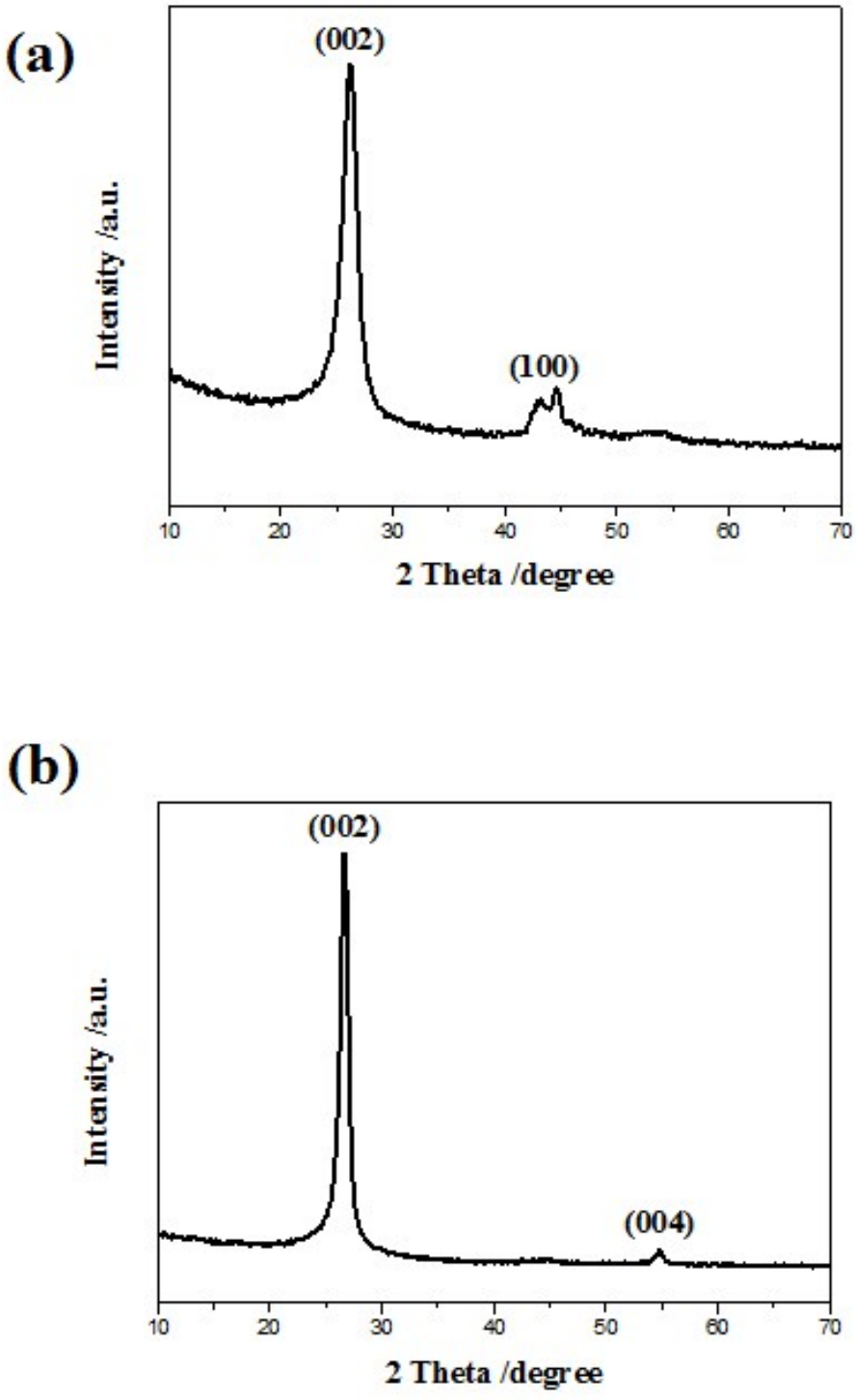

Figure S6. XRD patterns of CNTs (a) and graphene (b).

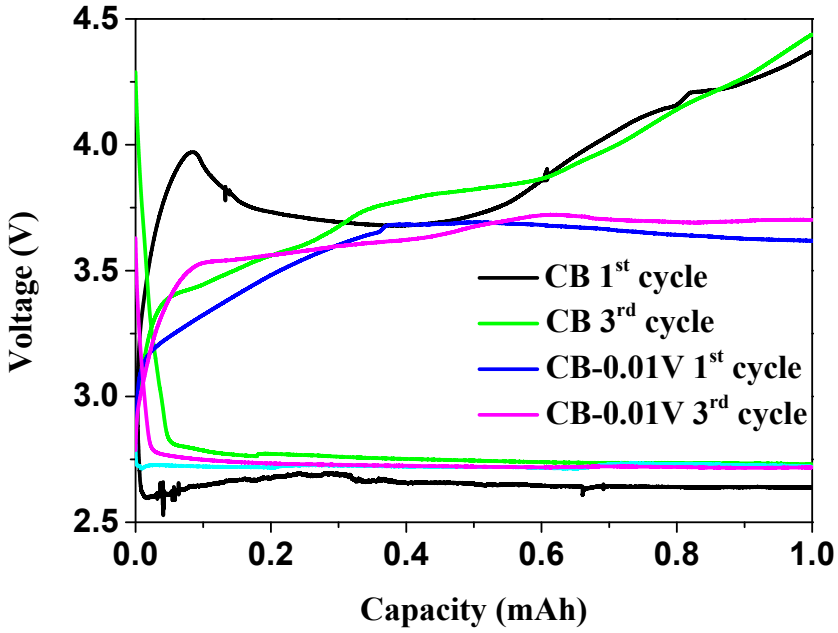

Figure S7. The discharge-charge profiles of carbon black at a current of 0.54 mA.

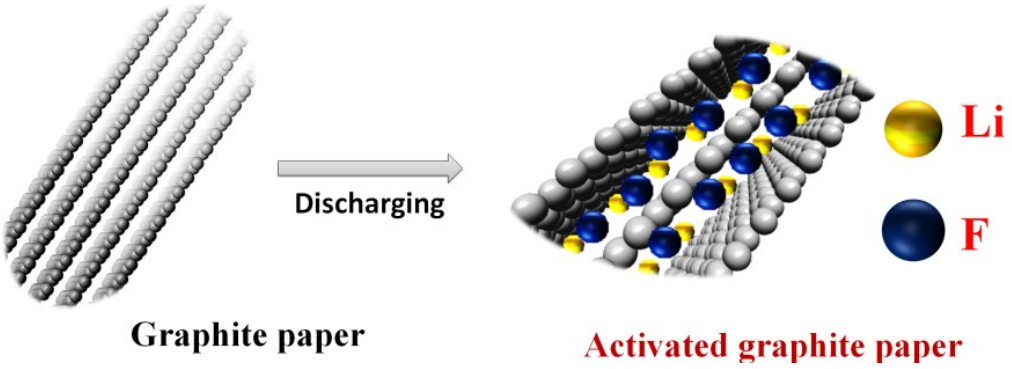

Figure S8. Scheme of the proposed activated method for graphite based materials.