

Electronic Supplementary Information (ESI)

**Rechargeable Li-CO<sub>2</sub> batteries with carbon nanotubes as air cathodes**

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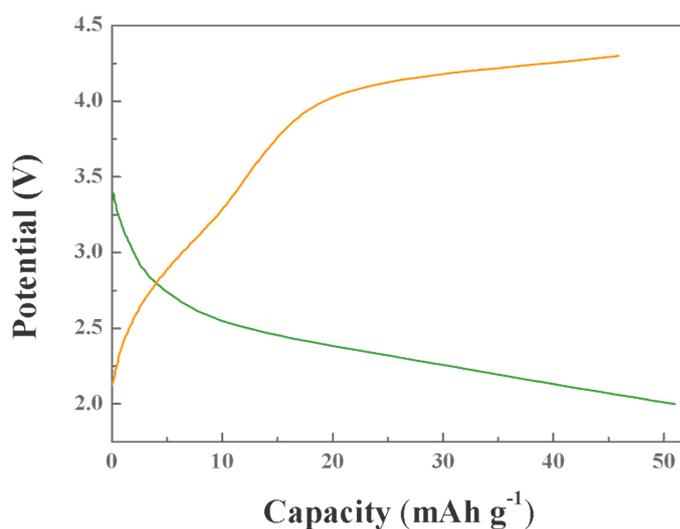
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**Experimental Section**

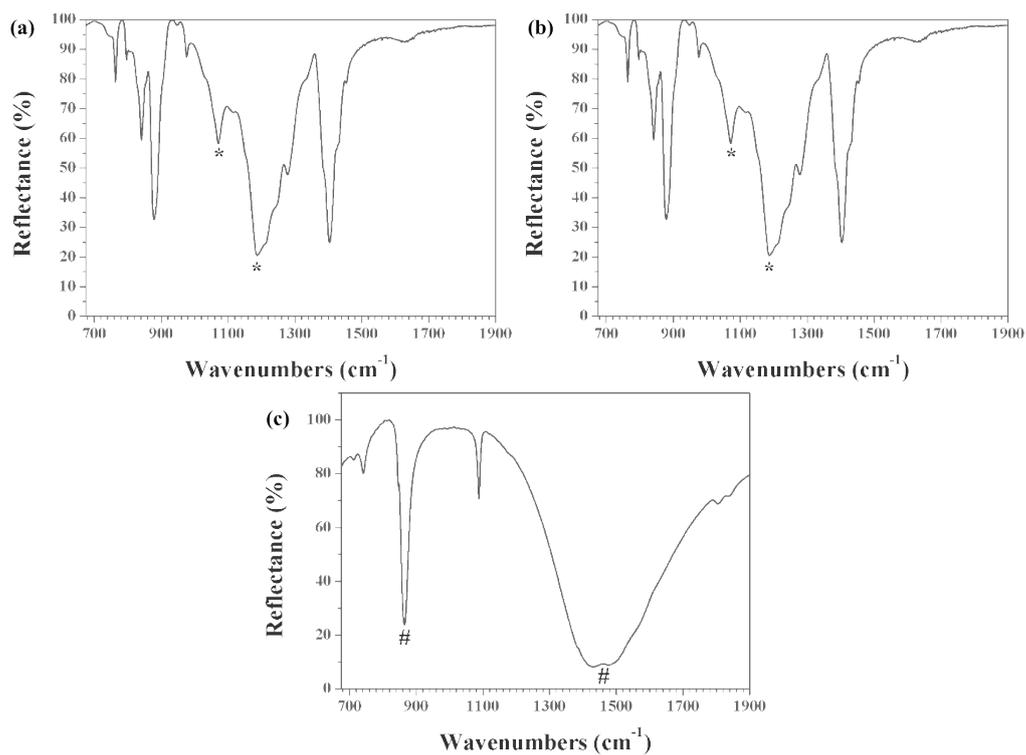
*Material Characterization:* CNTs were purchased from J&K. Field emission SEM (FESEM) images were obtained on a JEOL-JSM 7500 microscope. HRTEM and EELS were conducted on a FEI Tecnai G2F20 field emission TEM. FTIR spectroscopy was collected with a Nicolet MAGNA-560 spectrometer. XRD was performed to characterize the discharge and charge products on a D/MAX III diffractometer with Cu K $\alpha$  radiation

*Cell Assembles and Electrochemical Tests:* The cathodes were prepared as follows. A slurry containing 90 wt. % CNTs and 10 wt.% PVDF was mixed and then uniformly spread on the carbon paper. The electrochemical performances were evaluated in Swagelok-type cells. The cells were assembled in a glove box filled with high-purity argon (O<sub>2</sub> and H<sub>2</sub>O < 1 ppm). The cells composed of lithium metal anodes,

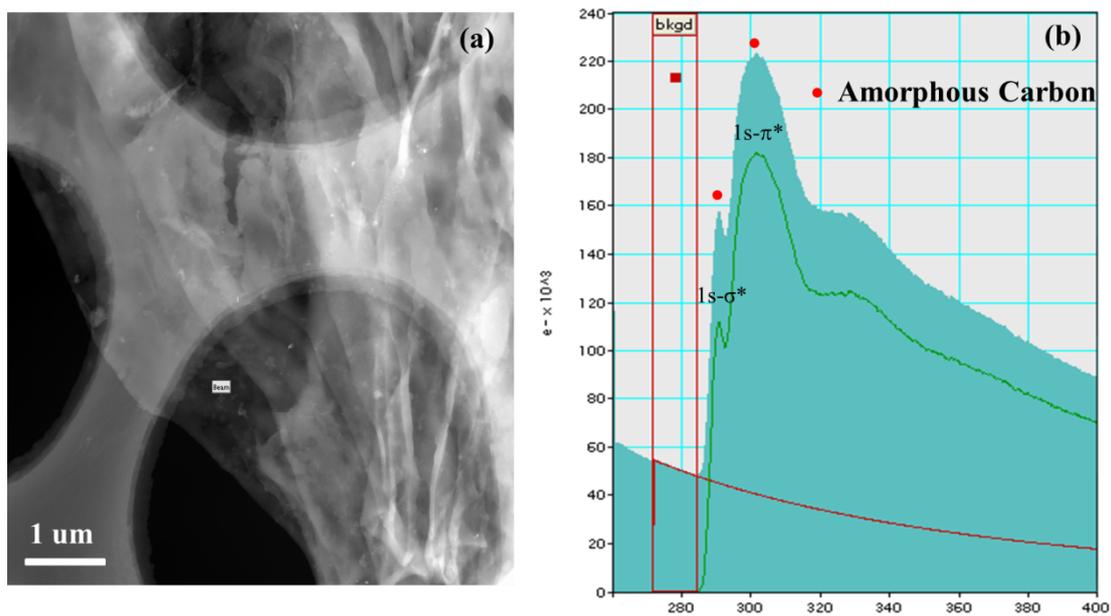
polytetrafluoroethylene (PTFE) separators, and CNTs cathodes. The electrolyte composed of 1 mol L<sup>-1</sup> LiTFSI dissolved in TEGDEM. Electrochemical measurements were performed on the LAND-CT2001A tester at room temperature. EIS was also performed for the assembled Li-CO<sub>2</sub> cells at different discharge-charge stages under an electrochemical workstation (Zahner Elektrik IM6e) in a frequency window between 100 kHz and 10 mHz.



**Fig. S1** Discharge-charge curves of Li-CO<sub>2</sub> batteries with Ar as the working gas.



**Fig. S2** FTIR spectra of (a) PVDF, (b) LiTFSI/TEGDME and (c)  $\text{Li}_2\text{CO}_3$ .



**Fig. S3** (a) TEM and (b) EELS of discharge product of  $\text{Li}/\text{CO}_2$  batteries with platinum net cathodes in the first discharge state.

**Table S1** Summary of electrochemical performance of Li-CO<sub>2</sub> batteries with different cathodes at room temperature.

Cathode	Maximum capacity	Cyclability	Ref
Super P	~0 mAh g <sup>-1</sup>	/	[16]
High surface area carbon	~750 mAh g <sup>-1</sup>	/	[16]
KB	1032 mAh g <sup>-1</sup>	7 cycles at 30 mA g <sup>-1</sup>	[17]
Graphene	14772 mAh g <sup>-1</sup>	20 cycles at 50 mA g <sup>-1</sup>	[22]
Carbon nanotubes	8379 mAh g <sup>-1</sup>	29 cycles at 50 mA g <sup>-1</sup>	This work