Supporting Information

All carbon nanotube and free standing air electrodes for

rechargeable Li-air batteries

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NMP-based electrolytes: 0.1 M electrolytes were prepared by mixing N-methyl-2-pyrrolidone (Sigma-Aldrich, battery grade, $H_2O<10$ ppm) with lithium perchlorate (LiClO₄, battery grade >99.9%, Sigma-Aldrich). The electrolytes were prepared and stored in a glove box, where the moisture and oxygen content was less than 1 ppm.

Li-air cell assembling and characterization Li-O₂ cells were constructed as reported in our previously published work. A lithium foil, two celgard 2500 separators, and an all MWCNTs electrode were placed in turn into a custom-designed cell (Fig S1), then about 50 μ L electrolyte solution was added. Cells were cycled on a Land cycler (Wuhan Land Electronic Co. Ltd.) within a voltage range from 2.0 V to 4.0 V (vs. Li⁺/Li) at a constant current density. All tests are carried under 1 atm pure O₂ pressure at 25 °C. X-ray photoemission spectroscopy (XPS) measurements were performed on a K-Alpha 1063 (Thermo Fisher Scientific) spectrometer with monochromatic Al-K α as the excitation source. N₂ adsorption-desorption data was collected using a QuadraSorb SI (Quantachrome) automated gas sorption system. The pore distribution was evaluated by the Density Functional Theory (DFT) methods. The cycled cells were disassembled in an Ar-filled glove box and the cathodes were rinsed twice with dimethoxyethane (DME) and then dried under an Ar atmosphere. The morphologies of pristine and cycled electrodes were observed by using a Hitachi S-4800 filed emission scanning electron microscope (SEM). The Raman spectra of the cathodes were collected on a Bruker SENTERRA Raman spectrometer. The 532 nm line from a Krypton laser with a power of 20 mW is used as the excitation source.



Fig. S1 Photo of custom-designed cell