Electronic Supplemental Information (ESI)

Performance-improved Li-O₂ battery with Ru nanoparticles supported on binder-free multi-walled carbon nanotube paper as cathode

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Experimental

The MWCNTP is prepared according to References 29 and 30. Ru nanoparticles were prepared in an ethylene glycol solution containing RuCl₃, of which pH was adjusted to 11 using 0.1 M of NaOH in ethylene glycol, at 165 °C under flowing N₂ for 3 hrs.31,32 The MWCNTP without further treatment was pressed onto an Al mesh, and immersed into the neutralized solution containing Ru nanoparticles under continuous stirring overnight. The Ru@MWCNTP on Al mesh was taken out from the solution and washed with de-ionized water and ethanol, and dried in a vacuum oven at 105 °C for at least 12 hrs.

The MWCNTP and Ru@MWCNTP were used as cathodes in Li-O₂ batteries assembled in coin cells (CR2032) with 7 holes exposed to pure O₂ (purity of 99.9 %). An Al mesh with a diameter of 10 mm was employed as the current collector, a lithium foil of 10 mm in size was applied as an anode, a piece of glass fiber filter of 12 mm in size was used as a separator, and lithium
bis(trifluoromethanesulfonyl)amide (LiTFSA, Wako) and triglyme (G3, Wako) with a molar ratio of 1:5 was employed as the electrolyte. The amount of electrolyte used in each cell is 50 μL. All the procedures were conducted in an argon filled glove box with O₂ < 5 ppm and H₂O < 1 ppm. The assembled coin cells were stored in a closed glass container which was flushed with O₂ for 3 hrs before each test.

The Li-O₂ batteries were discharged and charged at a galvanostatic current density of 500 mA/g on a Hokuto discharging/charging system between 2.3 and 4.6 V (vs. Li⁺/Li). The specific capacity of Ru@MWCNT is based on the total mass of MWCNT and Ru nanoparticles. The cycling performance of Li-O₂ batteries was evaluated by continuous discharge and charge with a limiting capacity of 5000 mAh/g at 500 mA/g. All electrochemical measurements were carried out at room temperature. The discharged and charged MWCNT and Ru@MWCNT were disassembled from coin cells, washed with dried dimethoxyethane, and dried in a glove box. The electrodes, enclosed by transparent Kapton films to reduce their exposure to air, were then subjected to X-ray diffraction (XRD) on a Bruker D8 Advanced diffractometer with Cu Kα (λ = 1.5406 Å) radiation with a continuous 2θ scan between 30° and 80° at 0.02°/s. Scanning electron microscopy (SEM) and high-resolution transmission electron microscopy (HRTEM) were obtained on S4800 (Hitachi) and JEOL2010 with an accelerating voltage of 200 kV.
**Scheme S1** Flowchart of the preparation process of MWCNTPs.

(1) Carrier gas $\text{H}_2$, (2) flowmeter, (3) carbon source: benzene with $\sim 0.5\%$ of thiophene, (4) first tube furnace where ferrocene is vaporized, (5) tube furnace reactor where carbon nanotubes are synthesized, (6) gas outlet, (7) valves.

**Fig. S1** Photograph of a free-standing MWCNTP.
Fig. S2 TEM image of Ru@MWCNTP.

Fig. S3 HRTEM image of Ru@MWCNTP (a) and FFT (b) of the selected area by the red square in (a).
Fig. S4 EDS of the Ru@MWCNTP.

Fig. S5 TGA curves of MWCNTP and Ru@MWCNTP.