

Reducing the overpotential of an aprotic Li-O₂ battery using a conductive graphene interlayer

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Experimental Section

Synthesis of B-rGO. B-rGO was prepared by a facile freeze-drying method. The detailed preparation process can be found in our previous work.¹

Synthesis of RuO₂-B-rGO. B-rGO (200 mg) was firstly dispersed in deionized water by sonication for 2 h, followed by stirring for 6 h. 1 g RuCl₃·xH₂O was added to the as-prepared B-rGO solution and stirred for 12 h, after which ammonium hydroxide was slowly added under stirring to adjust the pH of the solution to around 10. After stirring for another 12 h, the as-obtained samples were collected by vacuum filtration, and rinsed with deionized water for several times. After drying at 80 °C for 12 h, RuO₂-B-rGO was obtained by thermal treatment at 150 °C for 2 h and 200 °C for 3 h.

Preparation of graphene interlayer. A slurry containing 80 wt% of graphene tablet (CG41814, XG Science, Inc. Lansing, MI. ChemTrec, USA) and 20 wt% of polyvinylidene fluoride (PVDF) as binder was well mixed by grinding, which was then coated on a PP/PE/PP separator (Celgard 2325) by the blading method. The graphene interlayer was dried in a vacuum oven at 60 °C for 24 h. And the loading of the graphene layer on the separator is around 0.2 mg cm⁻².

Material characterization. The transmission electron microscopy (TEM) and the energy dispersive X-ray spectroscopy (EDX) were conducted on JEM-2010 transmission electron microscopy at an acceleration voltage of 200 KV. The field-emission transmission electron microscope (FESEM, JEOL, JSM-6700F) was employed to observe the morphology of materials and discharge products. Powder X-ray diffraction patterns of samples were collected on a Bruker D2 Phaser X-Ray Diffractometer with Ni filtered Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) at a voltage of 40 kV and a current of 40 mA.

Electrochemical measurements.

A mixture containing 80 wt% of cathode material and 20 wt% of polyvinylidene fluoride (PVDF) as binder was well mixed by grinding and then pressed onto carbon paper, which served as a current collector. The cathodes were then dried at 100 °C under vacuum for 24 h before use. The loading density of the active material on the B-rGO cathodes was approximately 0.3 mg cm⁻², while that on the RuO₂-B-rGO cathode was approximately 0.5 mg cm⁻². 1 M lithium bistrifluoromethanesulfonimide (LiTFSI) in tetraethylene glycol dimethyl ether (TEGDME) was employed as the electrolyte, and prepared in an argon-filled glove box with water and oxygen contents below 0.1 ppm.

The electrochemical tests of Li-O₂ batteries were carried out using 2032 coin-type cells with the holes at the cathode side. The cells were composed of a lithium metal tablet as anode, one slice of glass microfiber separator (Whatman), 200 μ L electrolyte, and as-prepared cathode (11 mm diameter). For RuO₂-B-rGO-CGI electrode, the graphene interlayer was placed between the separator and cathode, and the graphene side faced the cathode material. The batteries were also assembled in the argon-filled glove box. Before the measurements, the batteries were flushed with pure oxygen. Each measurement was begun after a 12 h open circuit potential step to ensure the equilibrium in the cell. The electrochemical measurements were carried out using a LAND cycler (CT2001A). Cyclic voltammograms were recorded between 2.0 V and 4.3 V at 0.2 mV/s on an electrochemical workstation (CHI660D, Shanghai Chenhua).

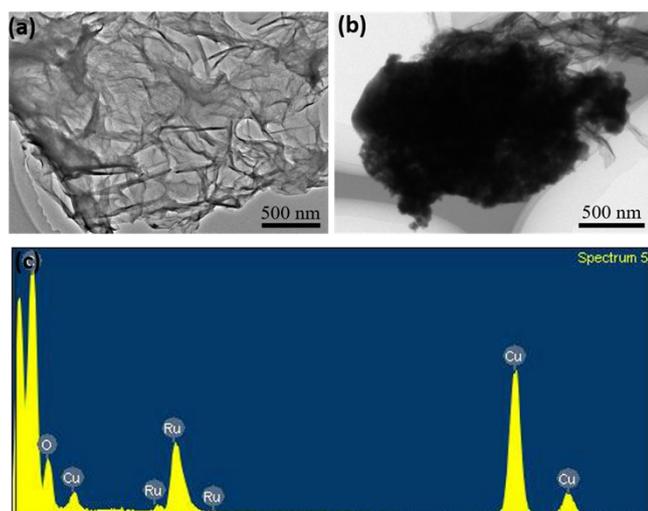


Figure S1 TEM images of (a) B-rGO and (b) RuO₂-B-rGO; (c) TEM-EDS spectrum of RuO₂-B-rGO.

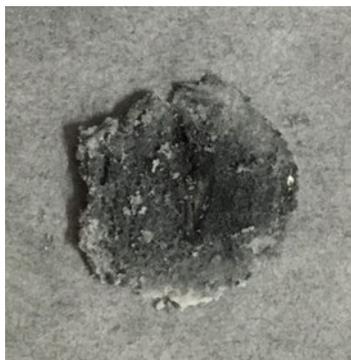


Figure S2 Lithium anode disassembled from the Li-O₂ battery with RuO₂-B-rGO-CGI as cathode after 200 cycles

References

1. F. Wu, Y. Xing, L. Li, J. Qian, W. Qu, J. Wen, D. Miller, Y. Ye, R. Chen, K. Amine and J. Lu, *ACS Appl. Mater. Interfaces*, 2016, **8**, 23635-23645.