

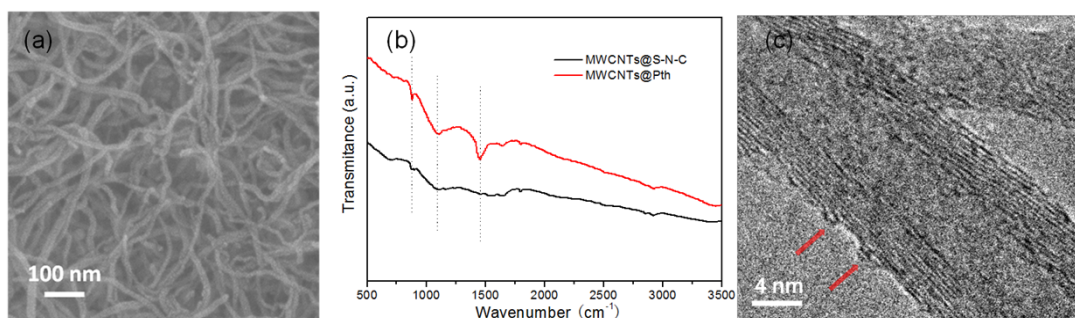
## Supporting Information

### **Carbon nanotubes/S-N-C hybrid nanomaterials as a bifunctional electrocatalyst for oxygen reduction and evolution reactions**

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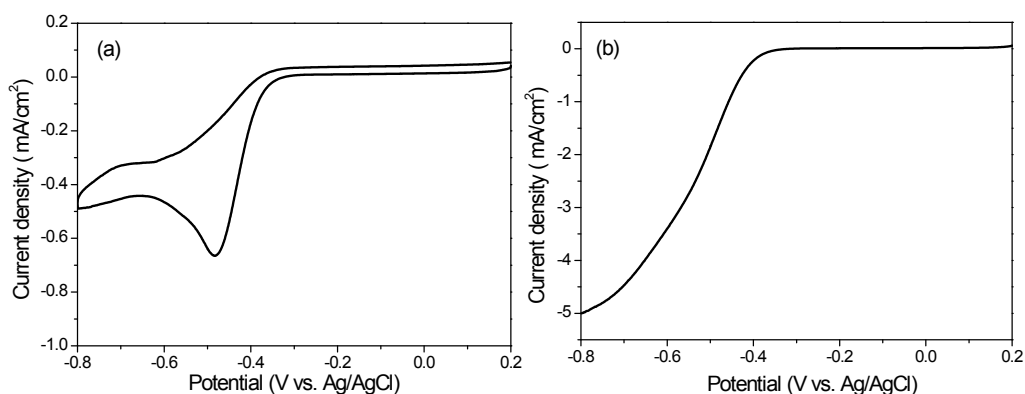
**Fig. S1** (a) SEM image of MWCNTs. (b) FTIR spectra of the MWCNTs@S-N-C and MWCNTs@Pth precursor. (c) HRTEM image of MWCNTs@S-N-C.

The number of electrons transferred ( $n$ ) during ORR was calculated by Koutecky-Levich equation at various electrode potentials and is given below:<sup>1,2</sup>

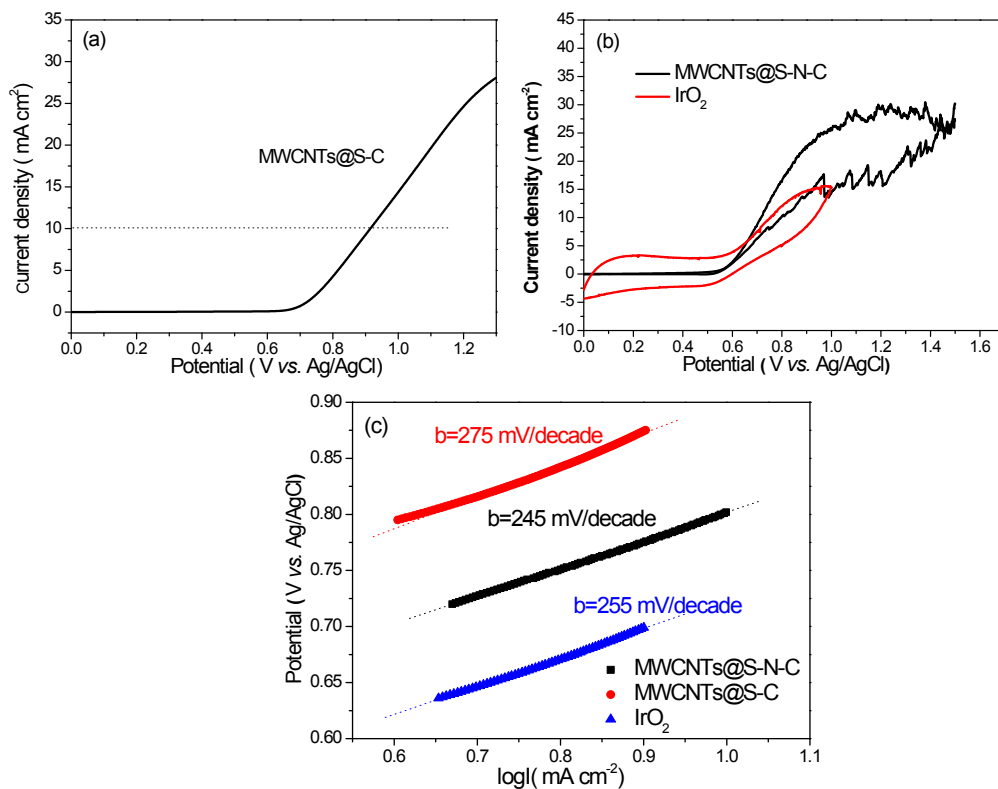
$$\frac{1}{J} = \frac{1}{J_L} + \frac{1}{J_K} = \frac{1}{B\omega^{1/2}} + \frac{1}{J_K}$$

$$B = 0.62nFC_0D_0^{2/3} \nu^{-1/6}$$

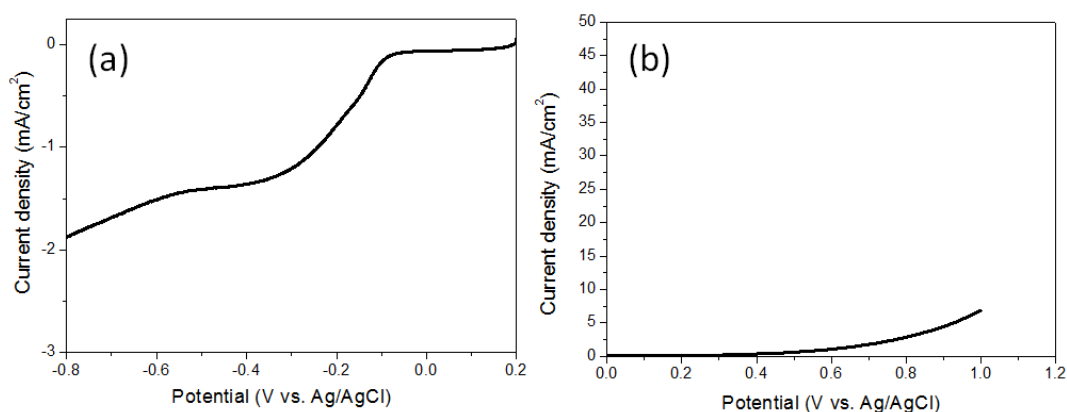
where  $J_k$  is the kinetics current density,  $J$  is the measured ORR current density,  $n$  represents the number of electrons transferred per oxygen molecule,  $F$  is the Faraday constant ( $F= 96485 \text{ C mol}^{-1}$ ),  $C_0$  is the bulk concentration of  $O_2$  ( $=1.2 \times 10^{-6} \text{ mol cm}^{-3}$ ),  $D_0$  is the diffusion coefficient of  $O_2$  in the KOH electrolyte ( $=1.9 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ ),  $\nu$  is the kinetic viscosity of the electrolyte ( $=0.01 \text{ cm}^2 \text{ s}^{-1}$ ), and  $\omega$  is the angular velocity of the disk ( $\omega=2\pi N$ ,  $N$  is the linear rotation speed).



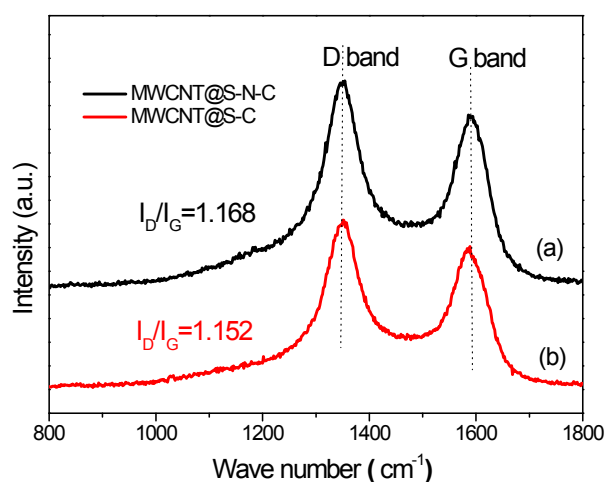
**Fig. S2** (a) Cyclic voltammograms of MWCNTs@S-C catalyst and (b) RDE voltammograms of the MWCNTs@S-C catalysts at 1600 rpm in 0.1 M KOH saturated with  $O_2$  gas.



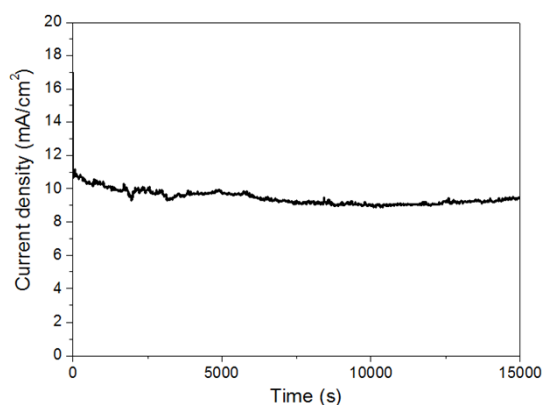
**Fig. S3** (a) linear sweep voltammograms of MWCNTs@S-C catalyst; (b) Oxygen evolution CV curves of MWCNTs@S-N-C and IrO<sub>2</sub> measured in a 0.1 M KOH with a sweep rate of 10 mV s<sup>-1</sup> and (c) Tafel plots of the IrO<sub>2</sub>, MWCNTs@S-C and MWCNTs@S-N-C hybrids derived by the mass-transport correction of corresponding RDE data.



**Fig. S4** (a) linear sweep voltammograms of the MWCNTs catalysts at 1600 rpm in 0.1 M KOH saturated with O<sub>2</sub> gas. (b) Oxygen evolution curves of MWCNTs in a 0.1 M KOH with a sweep rate of 10 mV s<sup>-1</sup>.



**Fig. S5** Comparison of the Raman spectra: (a) MWCNTs@S-N-C and (b) MWCNTs@S-C.



**Fig. S6** Chronopotentiometric response at a constant current density of  $10.0 \text{ mA cm}^{-2}$  of MWCNTs@S-N-C at 1600 rpm in 0.1 M KOH ( $E = 0.7 \text{ V vs. Ag/AgCl}$ ).

#### References:

- (1) S. Y. Wang, D. S. Yu, L. M. Dai, D. W. Chang and J. B. Baek, Polyelectrolyte-Functionalized Graphene as Metal-Free Electrocatalysts for Oxygen Reduction. *ACS Nano*, 2011, **5**, 6202–6209.
- (2) S. B. Yang, X. L. Feng, X. C. Wang and K. Mullen, Graphene-Based Carbon Nitride Nanosheets as Efficient Metal-Free Electrocatalysts for Oxygen Reduction Reactions. *Angew. Chem. Int. Ed.*, 2011, **50**, 5339–5343.