## **Electronic Supplementary Information**

## **Transferring Waste Papers to Multifunctional Graphene-Decorated**

## **Carbon Papers: From Trash to Treasure**

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Fig. S1 Large-area SEM image of typical waste paper. The papers are composed of cellulose fibers with the average diameters of  $15-30 \ \mu m$ .



**Fig. S2** Synthesis procedure for GCCP composites. Typically, a piece of waste paper was directly loaded into a cubic crucible with upper and lower floors covering urea and then pyrolyzed at 1000 °C under nitrogen gas atmosphere.



**Fig. S3** SEM image of graphene@carbon nanofiber/g- $C_3N_4$  composite obtained at 600 °C. The carbonized cellulose fibers were confined in the interlayer voids of g- $C_3N_4$ .



**Fig. S4** XRD pattern of  $g-C_3N_4$  and graphene@carbon nanofiber/ $g-C_3N_4$  composite obtained at 600 °C. The typical layered structure of  $g-C_3N_4$  can be observed, indicating the well development of polymeric plane of carbon nitride.



**Fig. S5** Large-area (a) and amplified (b-c) SEM images of GCCP (precursor: A4 paper). Graphene sheets formed in-situ on the surface of carbon nanofibers without disturbing the primary structure of paper fibers too much.



**Fig. S6** Raman spectra (a) and corresponding 3D mapping (b) of graphene 2D band (2680 cm<sup>-1</sup>) in GCCP. Raman spectrum (a) of GCCP with a typical 2D-band of graphene centered at 2680 cm<sup>-1</sup>. From the 3D mapping images (b) of graphene 2D band, the uniform distribution of graphene across over the surface of the whole paper was confirmed.



**Fig. S7** Large-area (a) and amplified (b-c) SEM images of carbonized paper without the assistance of urea (Precursor: A4 paper). The carbonized paper maintained the original fiber structure of the cellulose without formation of graphene sheets on the surface.



**Fig. S8** High-magnification TEM images of the surface graphene integrated carbon nanofiber composite. The layer number of the graphene was estimated to be 1-7.



Fig. S9 Nitrogen adsorption–desorption isotherms (a) and pore size distribution (b) of GCCP. The specific surface area of GCCP is  $63.4 \text{ m}^2 \text{ g}^{-1}$  and the pore volume is  $0.24 \text{ cm}^3 \text{ g}^{-1}$ .



**Fig. S10** High-magnification SEM images of GCCP samples obtained from various waste papers. The hierarchical structure of graphene-carbon fiber composite paper can be seen in all these samples.

Precursor	Images	Contact angle
Filter paper		154.3±2.8°
Kraft paper		158.3±2.4°
Napkin		152.7±2.1°
Newspaper		154.3±3.0°
Calendar paper		154.6±3.3°

 Table S1 Wettability list of GCCP samples obtained from various waste papers.



Fig. S11 The cycle stability of the separation efficiency of carbon tetrachloride/water mixtures through GCCP. No obvious decrease in the separation efficiency was observed in the following five runs.



**Fig. S12.** Polarization curves of GCCP-Pt sample on glassy carbon electrodes in  $O_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup> at different RDE rotation rates (in rpm).



**Fig. S13.** Koutecky–Levich plots (J<sup>-1</sup> versus  $\omega^{-1/2}$ ) of GCCP-Pt sample at different electrode potentials vs RHE, showing the nearly four electron transfer process.



**Fig. S14.** Electron transfer numbers as a function of the overpotential of GCCP-Pt sample.



Fig. S15 Polarization curves of GCCP-Pt and trash derived carbon in  $O_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup>.



**Fig. S16** Polarization curves of GCCP-Pt and trash derived carbon in  $N_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup>.



Fig. S17 Polarization curves of GCCP-Pt and trash derived carbon in  $N_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup>.



Fig. S18 Tafel slopes of GCCP-Pt and CP-Pt electrocatalysts for ORR.



Fig. S19 Tafel slopes of GCCP-Pt and CP-Pt electrocatalysts for HER.



Fig. S20 Tafel slopes of GCCP-Pt and CP-Pt electrocatalysts for OER.



**Fig. S21** Polarization curves of GCCP-Pt before and after 38,000s in  $O_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup>.



Fig. S22 Polarization curves of GCCP-Pt before and after 38,000s in  $N_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup>.



Fig. S23 Polarization curves of GCCP-Pt before and after 38,000s in  $N_2$ -saturated 0.1 mol L<sup>-1</sup> KOH solution with a sweep rate of 5 mV s<sup>-1</sup>.