

Supplementary Information

Microwave-assisted shock synthesis of diverse ultrathin graphene-derived materials

*Xianjue Chen, Xin Bo, Wenhao Ren, Sheng Chen and Chuan Zhao**

School of Chemistry, University of New South Wales, Sydney, NSW 2052, Australia

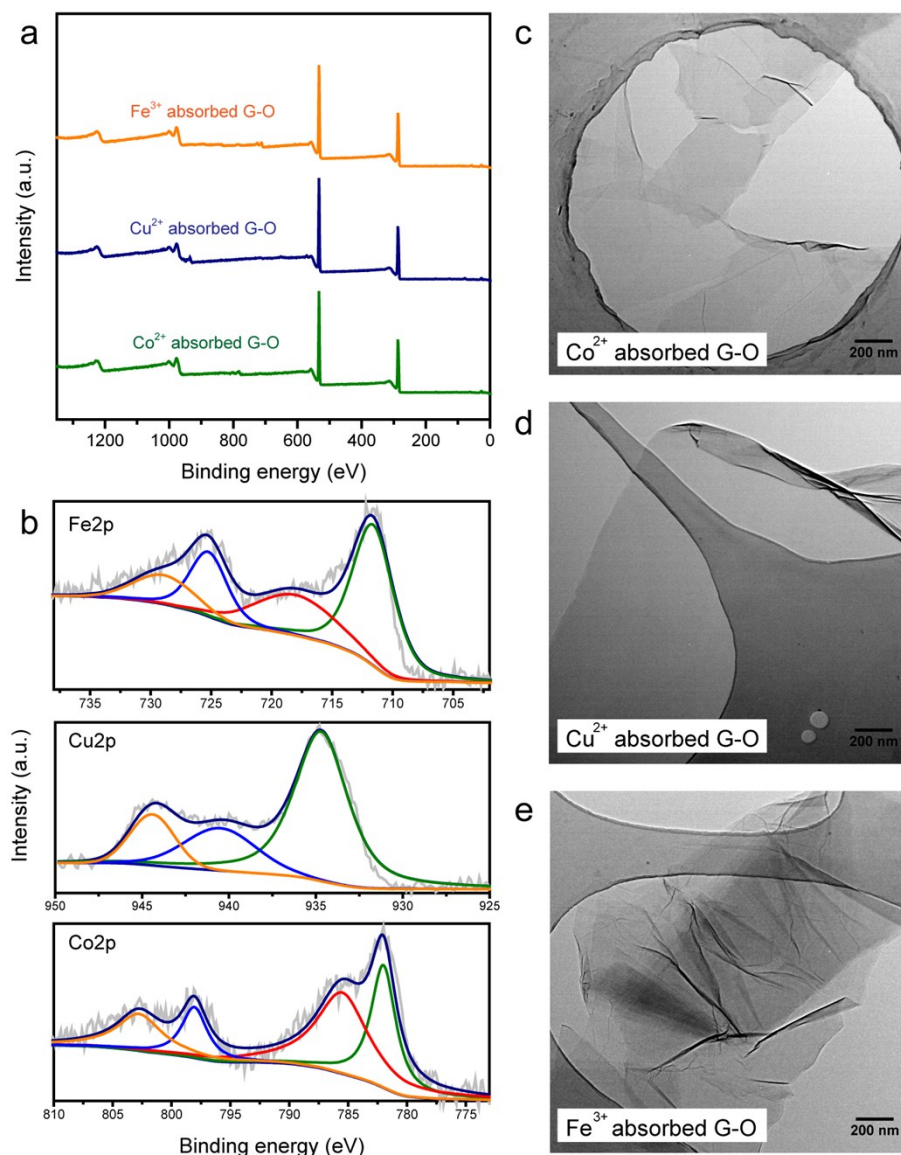


Fig. S1 (a) XPS full surveys of metal ion absorbed G-O. (b) XPS spectra of the metal ion absorbed G-O at corresponding Co2p, Cu2p and Fe2p regions. The presence of metal elements: 0.58 at% of Co in Co²⁺ absorbed G-O; 0.88 at% of Cu in Cu²⁺ absorbed G-O; 0.53 at% of Fe in Fe³⁺ absorbed G-O. The C/O ratios extracted from the XPS are 2.01, 2.04 and 1.99 for Co²⁺, Cu²⁺ and Fe³⁺ absorbed G-O, respectively. The increased C/O ratio of metal ion modified G-O in comparison to that of the as-prepared G-O (*i.e.* 1.71) can be attributed to a lower amount of intercalated water, as a result of insertion of metal ions into the interlayer regions. TEM images of (c) Co²⁺, (d) Cu²⁺ and (e) Fe³⁺ absorbed G-O platelets, showing the absence of nanoparticles.

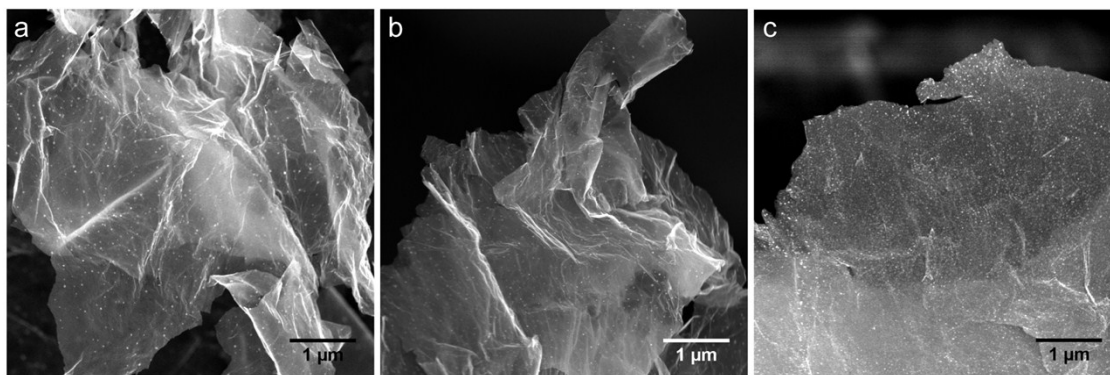


Fig. S2 SEM images of (a) Co, (b) Cu, (c) Fe nanoparticle decorated rG-O platelets.

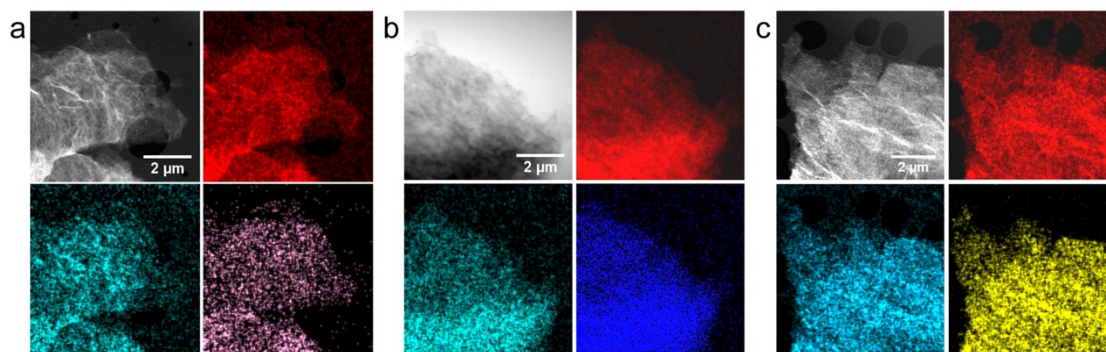


Fig. S3 TEM energy dispersive analysis showing the presence and uniform distribution of (a) Co, (b) Cu and (c) Fe nanoparticles on the rG-O. Elemental maps: C (red), O (cyan), Co (pink), Cu (blue) and Fe (yellow).

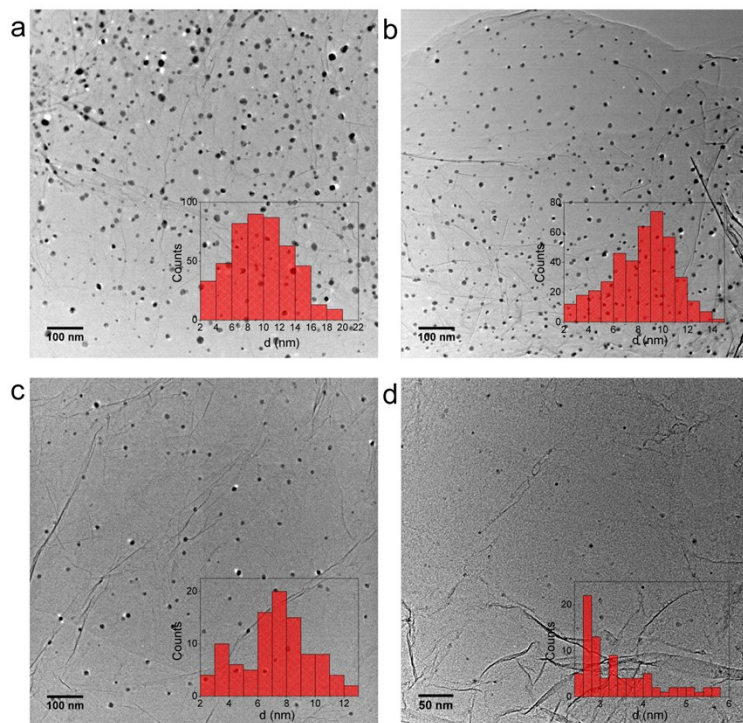


Fig. S4 TEM images and corresponding nanoparticle size distribution analysis of Co nanoparticle decorated rG-O prepared by soaking G-O in salt solutions of different concentrations: (a) 0.1 M (~10 nm), (b) 1 mM (~9 nm), (c) 0.1 mM (~7 nm) and (d) 0.01 mM (~3 nm). The size and distribution of the decorated nanoparticles are dependent on the concentration of the metal salt solution.

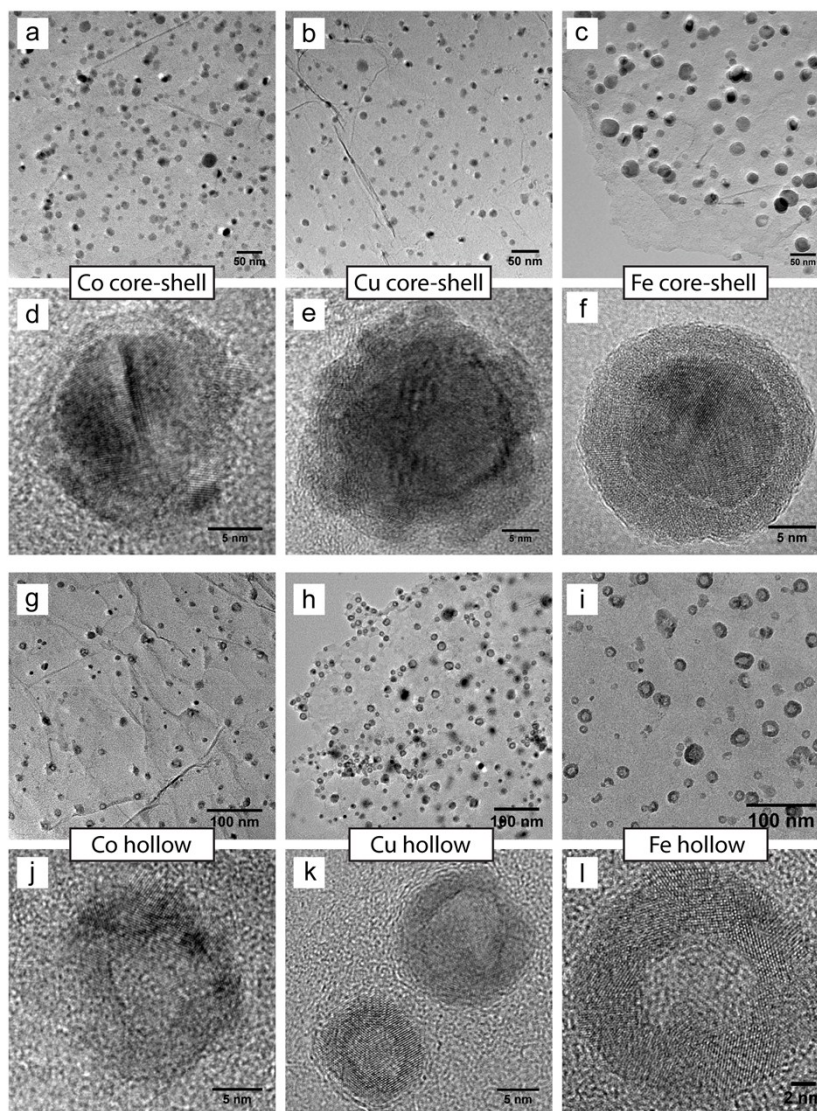


Fig. S5 TEM images of (a,d) Co, (b,e) Cu and (c,f) Fe “core-shell” nanoparticle decorated rG-O. The samples were exposed to air for 1 week at ambient conditions prior to the TEM analysis. The “core” regions show lattices corresponding to Co, Cu and Fe metals, while the “shell” regions show features for CoO, Cu₂O and Fe₂O₃, respectively. TEM images of (g,j) Co, (h,k) Cu and (i,l) Fe “hollow” nanostructures made by mildly heating the “core-shell” samples at 150 °C in air for 6 h. HRTEM images indicate that the “hollow” structures are made of CoO, CuO and Fe₂O₃, respectively.

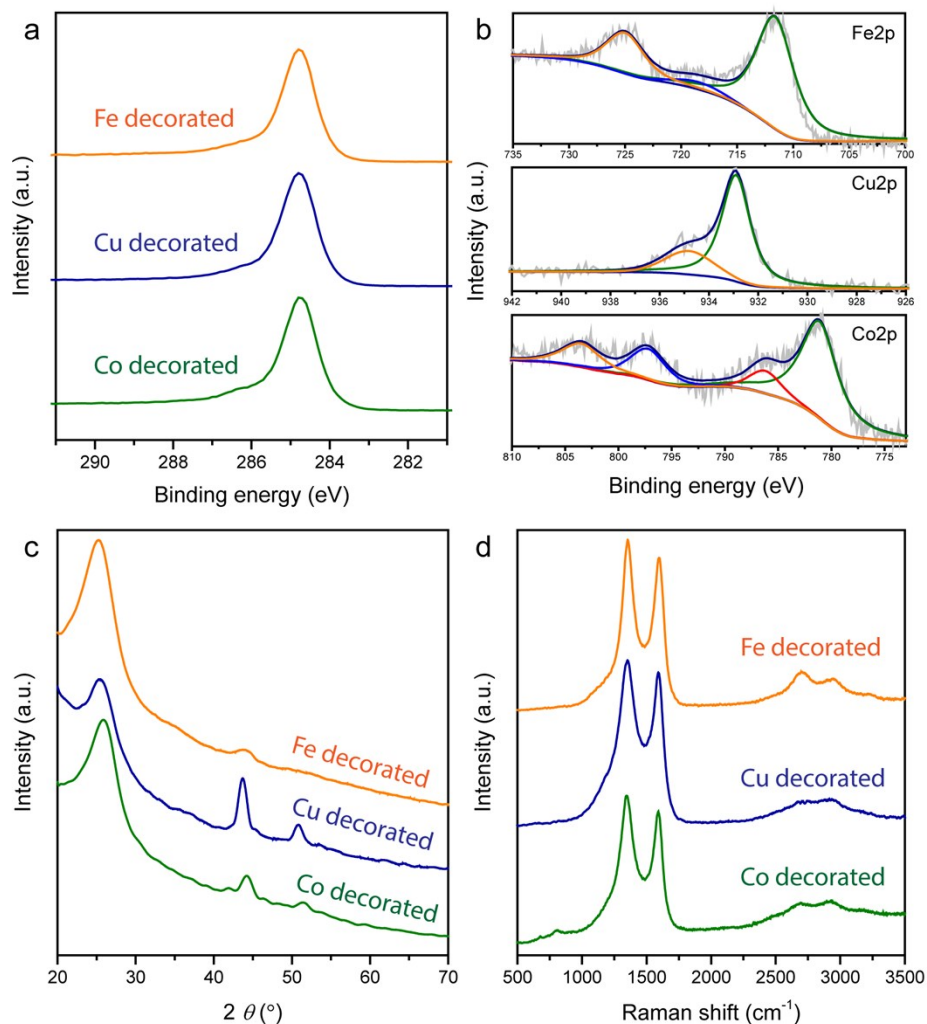


Fig. S6 XPS (a) C1s and (b) metal 2p regions, (c) XRD patterns and (d) Raman spectra of Co, Cu, Fe nanoparticle decorated rG-O. The metal 2p regions indicate the presence of corresponding metal oxides. A shoulder peak originating from CuO (~935 eV) can be observed in the case of Cu-decorated rG-O, which indicates the Cu₂O formed on the surface of Cu metal nanoparticles can be further oxidised to CuO at ambient conditions. XRD patterns reveal peaks that can be indexed to metallic Co, Cu, and Fe metals.

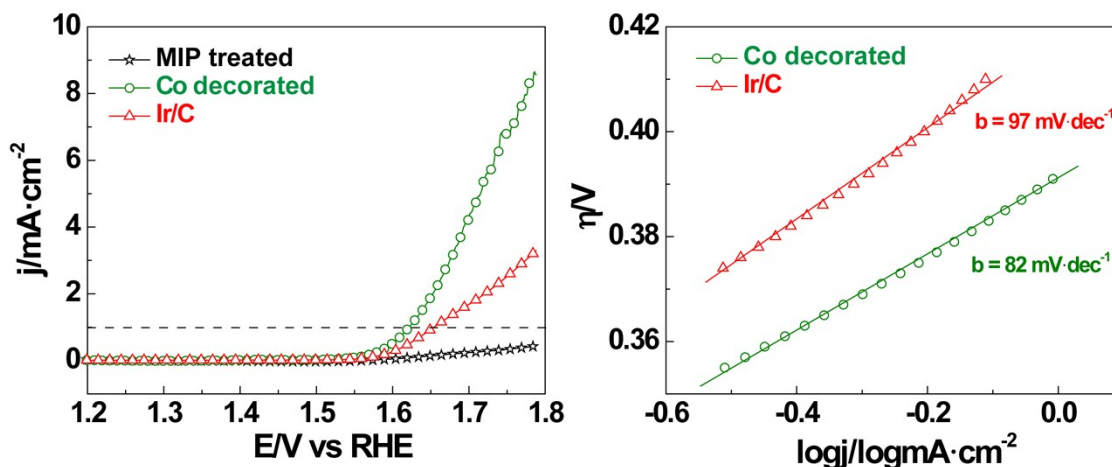


Fig. S7 Linear sweep voltammetry of nanoparticle-rG-O hybrids in electrocatalytic water oxidation. Co-decorated rG-O shows enhanced water oxidation performance in 0.1 M KOH media, where the onset potential at 1 mA cm⁻² is 1.62 V vs RHE, 30 mV lower than that of commercial Ir/C catalyst. The result is expected given that Co-based materials have been extensively studied as efficient OER electrocatalysts. The MIP treated rG-O, as a control sample, shows inert catalytic activity. Tafel slopes, related to the rate-determining step (RDS), were derived from the corresponding polarisation curves. The lowest Tafel slope value of 82 mV·dec⁻¹ was acquired from the Co-decorated rG-O. In theory, the Tafel slope values of 120 and 60 mV·dec⁻¹ correspond to the first and the second charge-transfer RDS of $M + OH \rightarrow M-OH + e$ and $M-OH + OH^- \rightarrow M-O + H_2O$ as RDSs.¹ The Tafel slope value indicates the RDS of the first and the second charge transfer processes have been accelerated.

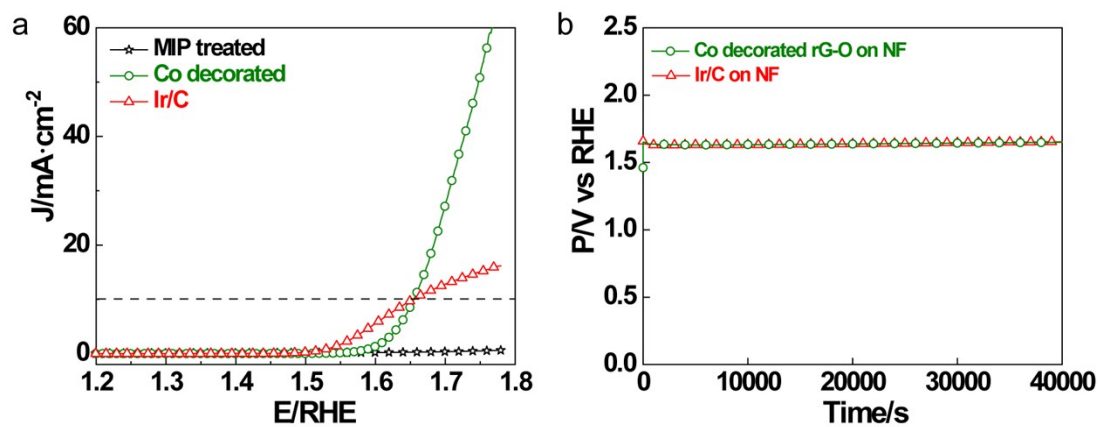


Fig. S8 The OER performance (a) and chronopotentiometry (b) under a current density input of 10 $\text{mA} \cdot \text{cm}^{-2}$ in 1 M KOH for more than 10 h.

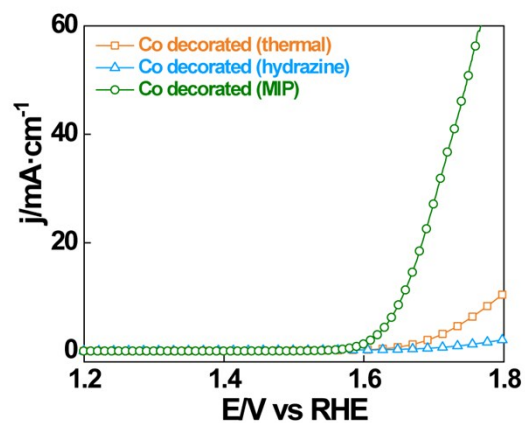


Fig. S9 The OER performance of Co decorated rG-O prepared using different reduction methods.

Table S1. An OER performance comparison of the catalyst in this work with previously reported catalysts.

Catalyst	Electrolyte	Onset potential (vs. RHE)	Overpotential under a certain current density (vs. RHE)	iR comp.	Reference
Co decorated rG-O	1 M KOH	1.58 V	1.65 V at 10 mA cm ⁻²	No iR	This work
Co decorated rG-O	0.1 M KOH	1.58 V	1.62 V at 1 mA cm ⁻²	No iR	This work
Co ₃ O ₄ NPs on N-doped graphene	0.1 M KOH	1.54 V	1.54 V at 10 mA cm ⁻²	No iR	2
Co ₃ O ₄ NPs on mMWCNT	0.1 M KOH	1.51 V	1.62 V at 10 mA cm ⁻²	No iR	3
Co ₃ O ₄ NPs on graphene	1 M KOH	1.45 V	1.73 V at 10 mA cm ⁻²	95%	4
Co ₃ O ₄ nanorod on mMWCNT	1 M KOH	1.52 V	1.54 V at 10 mA cm ⁻²	95%	5
Co ₃ O ₄ NPs on N-rGO	0.1 M KOH	1.54 V	~1.9 V at 10 mA cm ⁻²	No iR	6
CoO _x NPs on B,N-doped graphene	0.1 M KOH	~1.52 V	1.53 V at 10 mA cm ⁻²	No iR	7
Ir/C	1 M KOH	1.55 V	1.65 V at 10 mA cm ⁻²	No iR	Tested in this work
Ir/C	0.1 M KOH	1.58 V	1.65 V at 1 mA cm ⁻²	No iR	Tested in this work

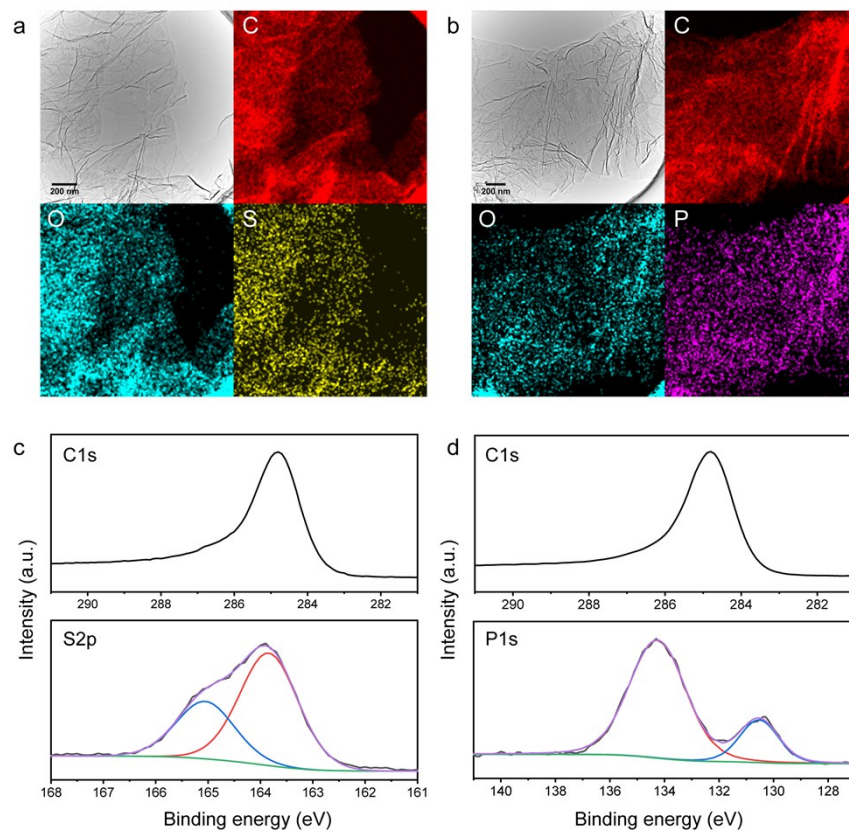


Fig. S10 TEM elemental mapping and XPS analysis of S or P doped rG-O.

Reference

1. J.-Y. Zhang, L. Lv, Y. Tian, Z. Li, X. Ao, Y. Lan, J. Jiang and C. Wang, *ACS Appl. Mater. Interfaces*, 2017, **9**, 33833
2. Y. Liang, Y. Li, H. Wang, J. Zhou, J. Wang, T. Regier and H. Dai, *Nature Mater.*, 2011, **10**, 780.
3. X. Lu and C. Zhao, *J. Mater. Chem. A*, 2013, **1**, 12053.
4. Y. Zhao, S. Chen, B. Sun, D. Su, X. Huang, H. Liu, Y. Yan, K. Sun and G. Wang, *Sci. Rep.*, 2015, **5**, 7629.
5. Y.-X. Zhang, X. Guo, X. Zhai, Y.-M. Yan and K.-N. Sun, *J. Mater. Chem. A*, 2015, **3**, 1761.
6. T. Zhang, C. He, F. Sun, Y. Ding, M. Wang, L. Peng, J. Wang and Y. Lin, *Sci. Rep.*, 2017, **7**, 43638.
7. Y. Tong, P. Chen, T. Zhou, K. Xu, W. Chu, C. Wu and Y. Xie, *Angew. Chem.*, 2017, **129**, 7227.