

Palladium nanoparticles functionalized graphene nanosheets for Li-O₂ batteries: enhanced performance by tailoring the morphology of discharge product

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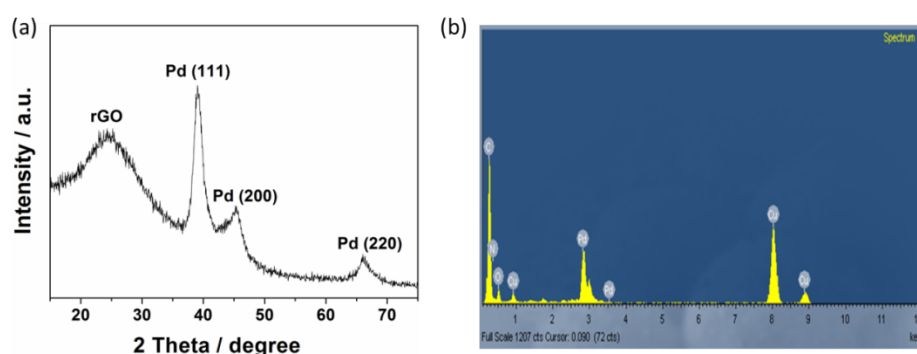


Fig. S1 (a) XRD patterns, (b) EDS spectra of Pd functionalized GNSs.

X-ray diffraction (XRD) was used to evaluate the phase purity of the final product, as shown in Fig. S1a. The diffraction patterns at $2\theta=39.2^\circ$, 45.4° , 66.2° can be assigned to the crystallized metallic palladium (PDF NO. 46-1,043) while the broad peak at around 25° can be indexed to the diffraction peak (002) of the hexagonal structure of GNSs.¹ The results indicated that the palladium precursor and GO have been chemically reduced simultaneously.

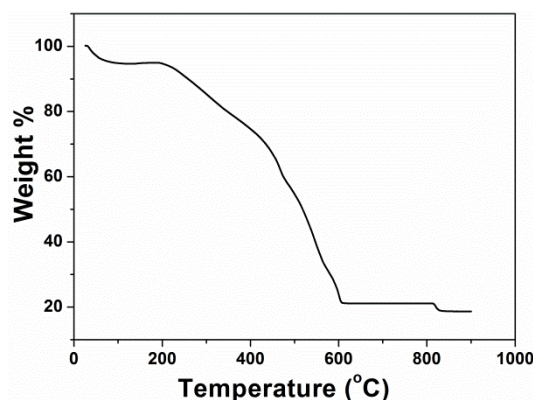


Figure S2. TGA curve of palladium functionalized graphene nanosheets catalysts.

As shown in Fig. S2, The weight loss within 100 °C is due to desorption of adsorbed water. The drop around 200 °C can be ascribed to the evolution of CO₂ and CO from the remnant oxygenated groups on graphene surface. The weight loss around 450 °C is assigned to the combustion of carbon. In the subsequent heating process, the Pd NPs in catalysts were easily oxidized to palladium oxide. So the weight loss around 820 °C is due to the decomposition of palladium oxide at high temperature.² The real Pd loading is estimated to be 18.8 wt % for palladium functionalized graphene nanosheets nanocomposites.

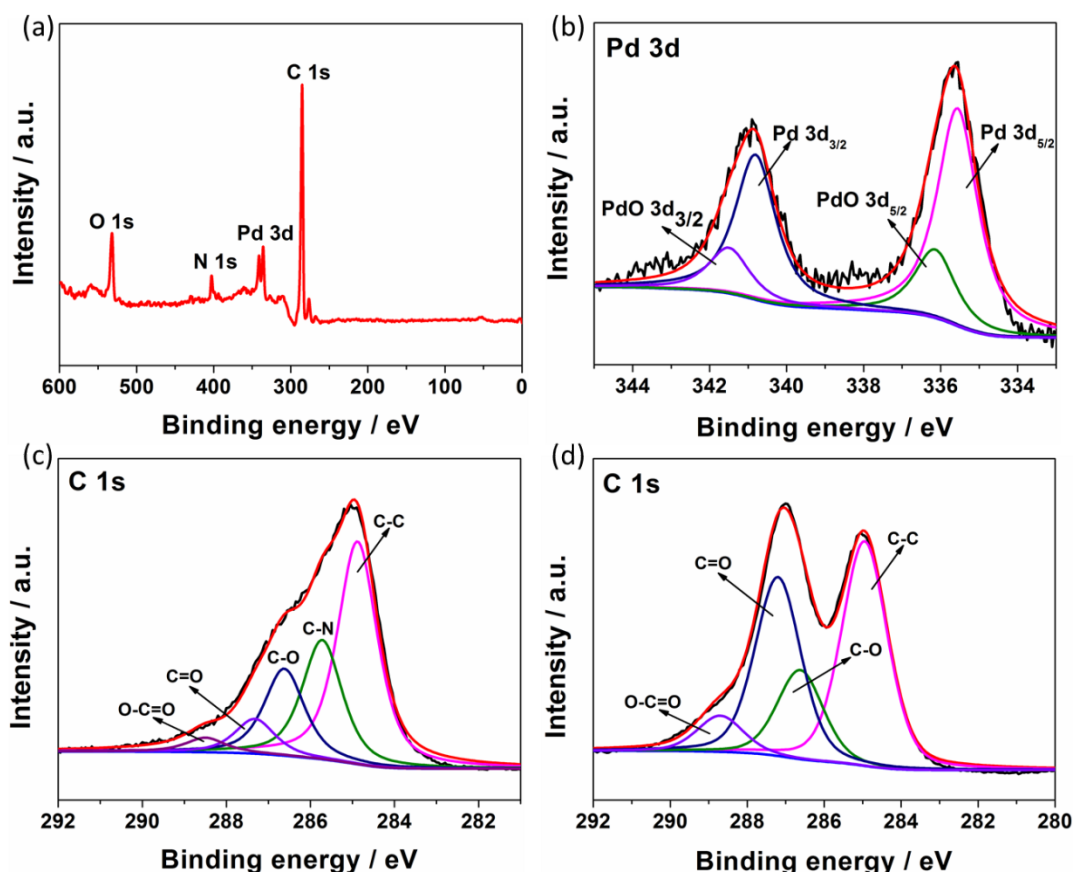


Fig. S3 (a) Survey XPS spectra, (b) Pd 3d, (c) C 1s XPS spectra of Pd functionalized GNSs. (d) C 1s XPS spectra of GO.

X-ray photoelectron spectroscopy (XPS) was employed to further investigate the chemical state in the Pd functionalized GNSs. The Pd 3d spectrum shown in Fig. S3b indicates the presence of two chemical states of Pd. The component at the lower binding energy could be assigned to the metallic Pd; while the component at the higher binding energy to PdO due to the surface partial oxidation of Pd NPs. XPS results demonstrated the Pd loading ratio was 19%, which was close to our expected value (20%). C 1s spectrum in Fig. S3c confirms the existence of five kinds of carbon components: C-C, C-O (epoxy / hydroxyls), C=O (carbonyl / ketone) and O-C=O (carboxyl).³ The additional component at 285.8 eV can be assigned to C-N bond.⁴ Compared with C1s spectrum of the pristine GO in Fig. S3d, the peak intensities of oxygen functional groups in the Pd functionalized GNSs are much weaker than that in GO, confirming the reduction of GO.

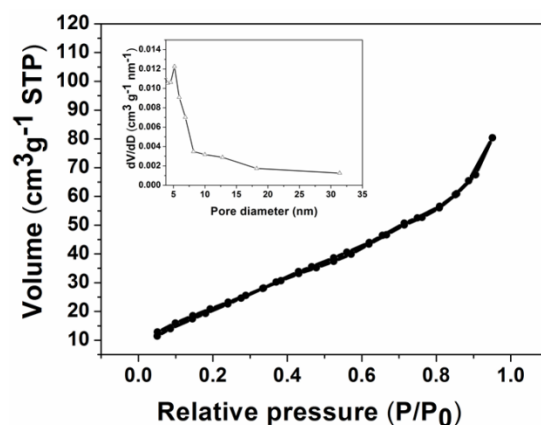


Fig. S4 Nitrogen adsorption-desorption isotherms and pore-size distribution (inset) of Pd functionalized GNSs.

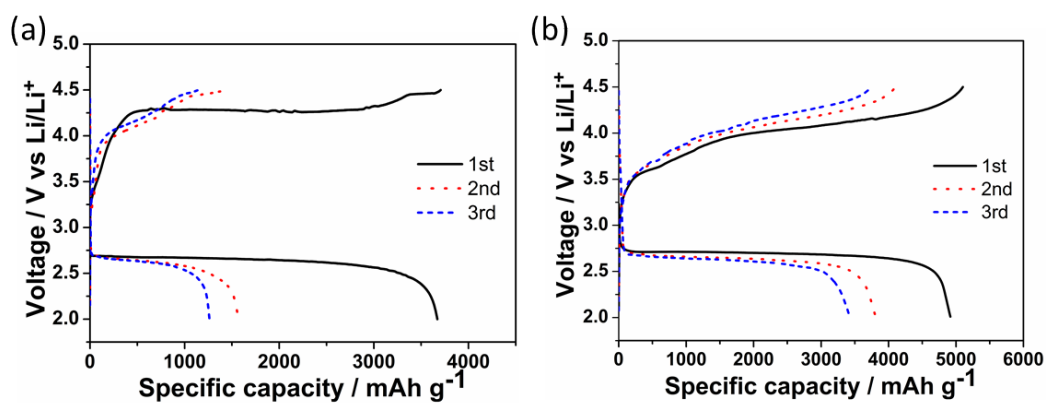


Fig. S5 First three cycle discharge-charge profiles of Li-O₂ batteries with (a) bare GNSs and (b) Pd functionalized GNSs cathode at a current density of 0.16 mA cm⁻².

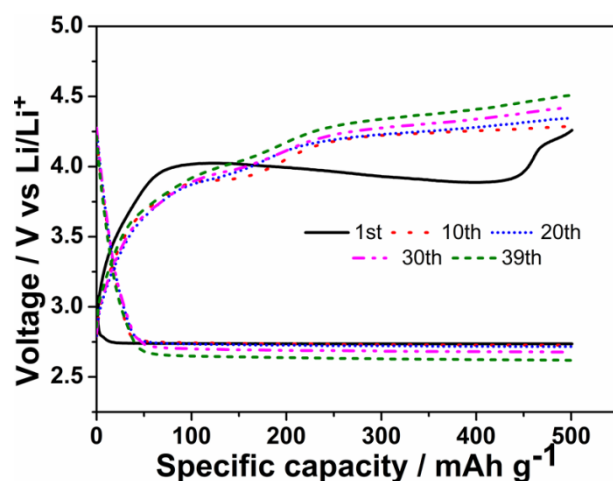


Fig. S6 Cycling performance of bare GNSs electrode at a limited capacity of 500 mAh g⁻¹.

References:

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