

Supporting online information

**Hybrid electrolyte Li-air rechargeable batteries base on  
the Nitrogen- and Phosphorus-doped graphene nanosheets**

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## Experimental

GNSs were obtained using precursor graphite as a starting material through the chemical reduction of graphene oxide in solution [1, 2]. The reduction of graphite oxide was carried out by stirring with hydrazine hydrate at room temperature for 24 h, then washed with distilled water until neutral and dried at room temperature for 24 h to obtain the GNSs.

Then GNSs were doped with nitrogen by heating in flowing  $\text{NH}_3$  ( $50 \text{ cm}^3/\text{min}$ ) in a quartz-tube reactor at  $900^\circ\text{C}$  for 2 h [3]. The P-doped GNSs was prepared by heating GNSs and triphenylphosphate ( $\text{OP}(\text{OC}_6\text{H}_5)_3$ ) in a quartz-tube reactor at  $900^\circ\text{C}$  in nitrogen atmosphere for 2 h. The catalyst paste was prepared from the N-doped GNSs and P-doped GNSs by the following procedure. The carbon materials (N-doped GNSs and P-doped GNSs,) (90%), polytetrafluoroethylene (PTFE) (7%) and acetylene black (AB) (3%) were well mixed, then roller-pressed into a sheet to form catalytic layer. The gas diffusion layer was prepared by mixing the acetylene black (60%) and PTFE (40%), and rolled to form a film. Consequently, the air catalytic layer was prepared by pressing the catalytic layer and gas diffusion layer onto nickel mesh.

The electrochemical test set up for Li-air battery with hybrid electrolyte was described in previous work by our group [4]. 1 M  $\text{LiClO}_4/\text{EC}/\text{DEC}$  ( $\text{LiClO}_4$ /ethylene carbonate/diethyl carbonate) was used as the organic electrolyte and 1 M  $\text{LiNO}_3 + 0.5 \text{ M LiOH}$  was used as the aqueous electrolyte. Then the solid state electrolyte  $\text{Li}_{1+x+y}\text{Al}_x(\text{Ti,Ge})_{2-x}\text{Si}_y\text{P}_{3-y}\text{O}_{12}$  (LISICON) film was used as a separating membrane between organic electrolyte and aqueous electrolyte to prevent intermixes between the two solutions. The cells were discharged at the current density of  $0.5 \text{ mAcm}^{-2}$  for 24 h. The charge-discharge performance was carried out in the voltage range from 2 to 4.8 V at the current density of  $0.5 \text{ mAcm}^{-2}$  for 2 h, respectively.

The N-doped GNSs and P-doped GNSs were characterized by X-ray photoelectron spectroscopy (XPS) (S-Probe ESCA Model 2803), and scanning electron microscopy (SEM) (JEOL, JEM-2010F).

A commercial 20wt%Pt/carbon black (CB) (Johnson Matthey) was also used as a reference to compare the electrode performance for Li-air batteries with hybrid electrolyte.

## Reference

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