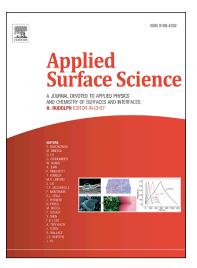
## Accepted Manuscript

## Full Length Article

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## Hierarchical PorousN-P-CoupledCarbonsasMetal-freeBifunctional Electro-catalysts for Oxygen Conversion

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Abstract: Hierarchical porous N and P co-doped carbons were facilely synthesized by employing phenoxycycloposphazenes (PCPZs) as multi-source of nitrogen, phosphorus and carbon. The pyrolysis of PCPZs that supported on monodispersed silica spheres produced high-surface graphitized carbons (N,P-GCs) with abundant surface N-P coupling sites. The N,P-GCs were demonstrated as efficient metal-free electro-catalysts for both oxygen reduction reaction (ORR) and oxygen evolution reaction (OER). The pore structures of these carbons for exposing more active sites could be adjusted by importing varisized SiO<sub>2</sub> spheres (10-50 nm) as hard templates. Typically, N, P-GC-1000 using 30 nm SiO<sub>2</sub> as templates exhibits the ORR half-wave potential of 0.85 V in 0.1 M KOH solution, with a loading density of 0.35 mg cm<sup>-2</sup> on the electrodes, which is comparable to commercial Pt/C catalyst (0.1 mg cm<sup>-2</sup>, 20 wt%, JM). Moreover, the OER over N, P-GC-1000 electrodes (0.65 mg cm<sup>-2</sup>) could also reach 10 mA cm<sup>-2</sup> current density at a lower potential of 1.56 V, close to typical  $RuO_2$  electrode (1.59 V, 0.2 mg cm<sup>-2</sup>, Premetek). This work not only suggested a simple and effective pathway to prepare N-P-doped carbon with N,P-coupled sites, but also provided a very efficient metal-free bifunctional carbon electro-catalyst for ORR and OER.

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