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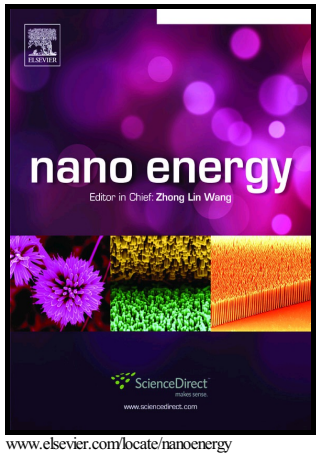

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Facile Approach for Synthesis of Doped Carbon Electrocatalyst from Cellulose Nanofibrils toward High-Performance Metal-Free Oxygen Reduction and Hydrogen Evolution

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Abstract

Multi-element doped metal-free carbon nanomaterials with a 3D architecture were prepared from inexpensive and naturally abundant cellulose nanofibrils. Compared to the most graphene-based carbons reported in the literature that show only good oxygen reduction reaction (ORR) activity, our N,S-doped carbon nanofiber network coated with N,P-doped carbon nanoparticles showed good bifunctionality toward ORR and hydrogen evolution reaction (HER) activities. The HER performance with doped carbon material exhibited an onset potential of 233 mV (*vs*. RHE), a current density of 10 mA cm^{-2} at 331 mV (*vs*. RHE), and Tafel slope of 99 mV $decade^{-1}$. The same material was also used for ORR, which could deliver an onset potential 10 mV more negative than commercial Pt/C and a cathodic peak of 0.84 V (*vs*. RHE). These values are better than most reported metal-free electrocatalysts. The superior performance of carbon hybrid may be attributed to the combination of a more exposed highly active N,P-doped carbon, a good integration of N,P-doped carbon with N,S-doped carbon nanofibers, and an accelerated electron transport through N,S-doped carbon nanofibers as good conductivity channels.

Keywords: carbon nanomaterials; electrocatalysts; cellulose nanofibrils; oxygen reduction; hydrogen evolution