



Direct anchoring of platinum nanoparticles on nitrogen and phosphorus-dual-doped carbon nanotube arrays for oxygen reduction reaction



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ABSTRACT

A novel and facile strategy has been developed to synthesize nitrogen and phosphorus-dual-doped carbon nanotube (N,P-CNT) array and used as Pt support for fuel cell application. In this strategy, the N,P-CNT array with well-designed nanostructure is directly grown on the cheap Ni foam using self-generated cylindrical Ni₂P as catalysts. The N,P-CNT with open ends and a large hollow channel provides large number of reactive sites in inner walls with access to reactants compared with conventional bamboo-shape nitrogen-doped carbon nanotubes (N-CNTs). The N,P-CNTs which possess abundant disordered nanostructures and reaction active sites can ingeniously anchor Pt with no need of pretreatment. The highly distributed Pt nanoparticles provide well Pt utilization and strengthen interaction between N,P-CNTs and Pt nanoparticles. The synthesized Pt/N,P-CNTs catalyst shows much better electrocatalytic performance for oxygen reduction reaction in comparison with Pt/CNTs, Pt/N-CNTs and commercial Pt/C catalysts. At 0.9 V vs. RHE in 0.1 mol L⁻¹ HClO₄, the Pt mass activity of Pt/CNTs and Pt/N-CNTs are 93.5 and 165.4 mA mg⁻¹_{Pt}, while, the Pt/N,P-CNTs catalyst shows a mass activity of 285.3 mA mg⁻¹_{Pt}, which is 3.1 and 1.7 times as high as that of Pt/CNTs and Pt/N-CNTs, respectively.

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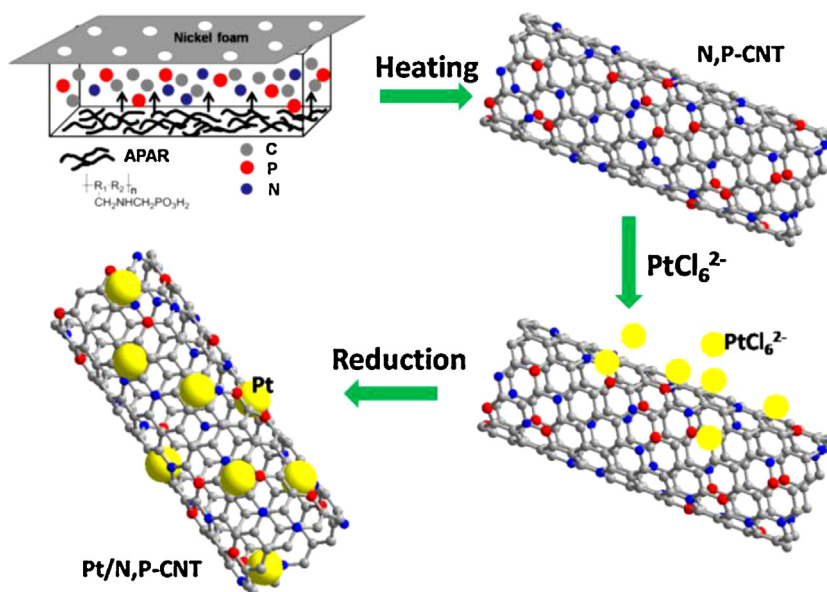
1. Introduction

Oxygen reduction reaction (ORR) has been realised existing in many devices like low-temperature fuel cells [1,2], metal-air (oxygen) batteries [3], oxygen sensors [4] and the production of hydrogen peroxide [5]. Currently, most popularly used catalysts for ORR are platinum (Pt)-based catalysts [6–8]. In fact, even the Pt-based catalysts require over 0.3 V overpotentials under reasonable operating conditions for ORR, which is about 10 times larger than that for hydrogen oxidation at the anode [2]. This is the evidence that oxygen reduction on Pt-based catalysts is extremely lower in kinetics compared with anodic reaction. Therefore, the development of catalysts with high electrocatalytic activity and stability for ORR is a critical challenge.

The manufacture of the supported catalysts is an effective approach to promote reactivity of Pt-based catalysts via synergistic effects or/and reduce the cost by increased utilization of

nanostructured platinum particles [9–11]. Carbon nanotubes (CNTs) have been extensively used as support of Pt particles and the Pt/CNTs catalysts display superior catalytic performance than that of the traditional Pt/C due to the high specific surface area, high stability and good electronic conductivity of the CNTs [12–15]. However, it still remains a formidable challenge to directly disperse Pt nanoparticles onto CNTs since CNTs generally need chemical modification [16–18] or noncovalent functionalization [19,20] before loading Pt nanoparticles on their inert surface. It also suffers from low inherent catalytic activity toward the ORR [12]. Doping with heteroatoms (N, B, P and S) in CNTs can break out the chemical inertness of the pure CNTs by creating defects, which facilitate deposition of Pt nanoparticles on the CNTs [21–23]. In addition, heteroatom-doped carbon itself has a certain electrocatalytic activity [24–26] and shows synergistically enhanced performance toward ORR when used as Pt support [21,27]. However, the conventional N-doped CNTs [21,28,29], P-doped CNTs [30] and N,P-doped CNTs [31,32] possessed bamboo-like structure and nodes, which lead to unavailable inner walls of the CNTs. Also, the nitrogen atoms seem to be preferentially localized on the nodes leading to heterogeneous distribution of nitrogen

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Scheme 1. Schematic representation of the preparation procedure for the Pt/N,P-CNTs.

atoms and reactive sites [33,34], which result in adverse effect to uniform Pt dispersion. Therefore, it is highly required to develop novel CNTs support which not only possesses inherent electrocatalytic activity but also has significant advantageous architecture.

Herein, we report that a nitrogen and phosphorus co-doped carbon nanotube (N,P-CNT) array has been successfully designed and used as a support to deposit Pt nanoparticles as shown in Scheme 1. The N,P-CNTs with open ends and large hollow channels provide a large number of available reactive sites in the inner walls,

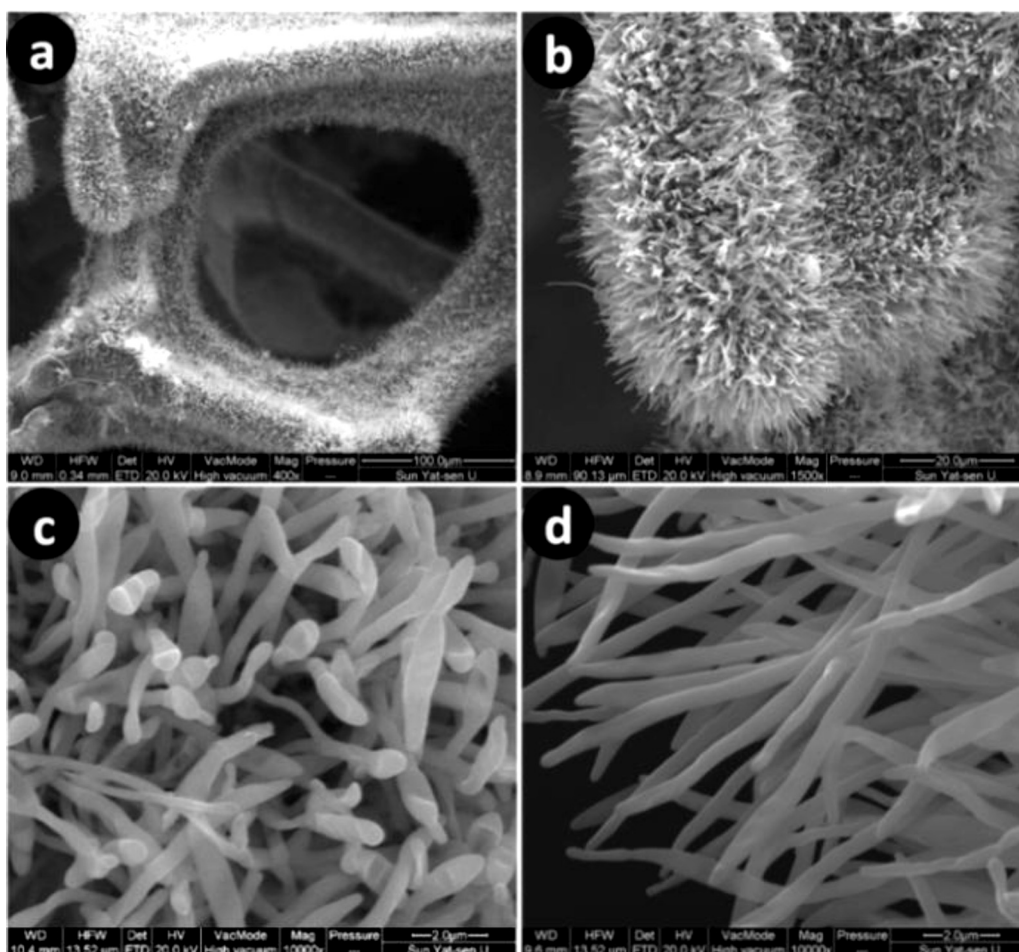


Fig. 1. SEM images of the as-synthesized N,P-CNTs array. (a) Overall view, (b, c) top view and (d) side view of the N,P-CNTs array.

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