Controllable synthesis of heteroatom-doped carbon nanotubes as efficient catalysts for electrochemical detection of dopamine

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Recent theoretical [1] and experimental [2] studies have suggested that heteroatom-doped carbon nanomaterials including carbon nanotube (CNTs) and graphene as novel materials with exceptional electrochemical property due to the heteroatom doping. For example, heteroatom-doped CNTs have been demonstrated as efficient catalysts for the simultaneous electrochemical detection of glucose [3], peroxide hvdrogen [4], L-cysteine [5]. and dihydronicotinamide adenine dinucleotide (NADH) [6], making them particularly attractive from both scientific studies and innovation applications.

Here we show that boron-doped CNTs (BCNTs) with tunable boron atomic concentration ranging from 0.4 to 21.1 atomic percentage (at%) can be synthesized by an atmospheric-pressure carbonthermal reaction. The asproduced BCNTs exhibit superior electrocatalytic activity for dopamine (DA) sensing than the pristine CNTs and conventional glassy carbon electrodes. Mixtures of pristine CNTs and boron oxide (boron precursor) were heated at elevated temperatures (900~1200 °C) in argon (Ar) and ammonia (NH₃) atmospheres. We found that boron atomic concentrations in the nanotubes could be tuned by simply controlling the NH₃ concentrations in the reaction flows.

High-resolution transmission electron microscopy (HRTEM) characterization reveals the BCNT sidewalls are essentially free of amorphous carbon over coating [Fig. 1(a)]. Detailed electron energy loss spectroscopy (EELS) characterization indicates that boron substitution doping mainly occurred on the nanotube surface, suggesting the formation of electrocatalytic activation sites [Fig. 1(b)]. Extensive X-ray photoelectron spectroscopy (XPS) characterization confirms that boron atoms were successfully doped into the sp² graphene lattice of CNTs, and boron atomic concentrations in CNTs can be tuned from 0.4 to 32.2 at% by simply controlling the NH₃ concentration from 0 to 15%, respectively (Fig. 2).

Electrochemical characterization indicates a significant enhancement of 25% in the amperometric response for BCNTs with 2.1 at% boron atomic concentration than pristine CNTs, suggesting its potential as efficient catalysts for electrochemical detection of DA (Fig. 3). It is also noteworthy from a practical point of view that the developed atmospheric-pressure synthesis method is amenable to industrial-scale production since it avoids the need for a vacuum system.



Figure 1. (a) HRTEM micrograph and (b) boron qualitative elemental mapping of as-produced BCNTs synthesized using 10% NH₃ concentrations (synthesis conditions: 4 hrs and 1200° C).



Figure 2. XPS spectra of as-produced BCNTs synthesized with different NH_3 concentrations. (synthesis conditions: 4 hrs and 1200°C).



Figure 3. The oxidation peak current densities of DA using pristine CNTs and BCNTs with different boron atomic concentrations.

Reference

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