The Role of Oxygen Functionalities for Enhanced Oxygen Reduction Activity in Carbon Nanotubes

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The advancement of non-noble metal catalysts for the electrochemical reduction of oxygen is vital to further facilitate the commercialization of fuel cells. Various approaches involving transition metal and nitrogen doping have been documented, but unfortunately, the precise mechanism with respect to carbon support, transition metal, and nitrogen doping towards oxygen reduction reaction (ORR) is still not well understood. Another approach is to use carbon materials such as carbon nanotubes without any dopants since it is also known to catalyze ORR, but this has not been well documented.

Here, we study the origin of enhanced ORR activity in nitrogen and metal-free multi-walled carbon nanotubes (MWNTs) by examining the role of oxygen functionalities and structure of MWNTs through induced defects by nano-drilling via cobalt oxide which acts as a low-temperature carbon oxidation catalyst.[1],[2] The use of nano-drilling allows for the control of surface structure and the distribution of oxygen functional groups.

Briefly, MWNTs were heat treated at 500°C in air followed by acid reflux in boiling concentrated nitric acid and 2 M sulfuric acid for 4h. The heat and acid-treated MWNTs were then impregnated with cobalt and then oxidized at 250°C in air for 25 minutes. Afterwards, any residual metals and oxides were successfully removed by acid washing.

Through thermal treatments in Ar the effects of functional group removal is examined. Oxygen functional groups, ORR activity, structural morphology, and impurities are examined with temperature programmed desorption (TPD), electrochemical characterizations, transmission electron microscopy (TEM), and inductively coupled plasma mass spectroscopy (ICP-MS) respectively.

Fig. 1 Typical transmission electron micrograph (TEM) of nano-drilled carbon nanotubes showing the induced structural defects

References