Efficient Electrocatalysts Based on Nitrogen-doped Carbon Nanostructures for Energy Applications

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Abstract
Carbon nanostructures have emerged as a key material in nanotechnology and continuously find new areas of applications. Particularly, they are attractive due to their excellent properties as support for catalyst nanostructures leading to highly efficient composite materials for various electrochemical applications. The interest in these structures is further increased by the possibility to alter their electronic and structural properties by various methods. Heteroatom doping of carbon nanostructures is one of the approaches which may induce intrinsic catalytic activity in these materials. In addition, such introduction of guest elements into the hexagonal carbon skeleton provides strong nucleation sites which facilitate the stabilization of nanostructures on their surface. In this thesis we present detailed studies on the nitrogen incorporation into carbon nanostructures, particularly carbon nanotubes and reduced graphene oxide. Due to the high impact of nitrogen configuration on the intrinsic electrocatalytic properties of carbon nanostructures, we investigated the nitrogen functionalities using X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy. Based on our achievements we could assign the most electrocatalytic active nitrogen site in nitrogen-doped carbon nanotubes (NCNTs) for catalytic oxygen reduction reaction (ORR) which is an important reaction in energy conversion systems such as fuel cells. We then used nitrogen-doped carbon nanostructures as a key component to manufacture hybrid material, where the nitrogen doped nanostructures has a role of both stabilizing the nanostructures and to work as conductive additive to assist the charge transfer from the other constituents suffering from inherently poor conductivity. Our hybrid material comprising transition metal oxides (Fe_2O_3 and Co_3O_4) anchored on nitrogen-doped carbon nanostructure were used to both manufacture an exotic type of graphene nanoscrolls, as well as studied and evaluated as an electrocatalyst in various electrochemical reactions. We show that the self-assembled electrodes exhibited better performance and higher stability compared to when the same material was loaded on common current collectors such as fluorine tin oxide (FTO) coated glass and glassy carbon electrode, with both higher current densities, more efficient charge transfer and lower overpotentials for oxygen evolution and hydrogen evolution reactions, the two important processes in a water splitting device. Our NCNTs-based electrodes showed further excellent performance in lithium ion batteries with high cyclability and capacity. The thesis gives insight into processes, materials, and methods that can be utilized to manufacture an efficient water splitting device, based on earth-abundant self-assembled materials. It further represents a significant advancement of the role of nitrogen in heteroatom-doped nanostructures, both regarding their intrinsic catalytic activity, as well as their role for stabilizing nanostructures.

Keywords
Nitrogen-doped carbon nanotubes, hybrid materials, Transition metal oxides, carbon paper, Li-ion microbatteries, water electrolysis

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