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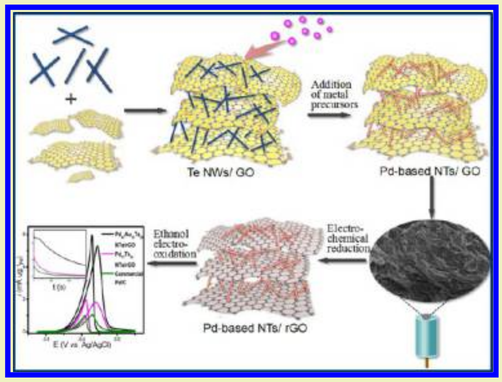
Graphene Oxide as a Stabilizer for "Clean" Synthesis of High-Performance Pd-Based Nanotubes Electrocatalysts

Huan Zhang, Kai Cai, Pan Wang, Zhao Huang, Jiawei Liu, Long Wu, Mohamed F. Foda, and Heyou Han*

State Key Laboratory of Agricultural Microbiology, College of Science, Huazhong Agricultural University, Wuhan 430070, China

Supporting Information

ABSTRACT: By using graphene oxide (GO) as the stabilizing and dispersing agent in aqueous solution at room temperature, Pd-based (PdTe, PdPtTe, and PdAuTe) nanotubes (NTs)/GO nanocomposites were readily prepared. The obtained GO-supported one-dimensional (1D) Pd-based NTs have relatively uniform morphology and good dispersity. The catalytical properties of PdAuTe NTs/reduced GO (rGO) nanocomposites with different metal ratios were systematically investigated after electrochemical reduction process. $Pd_{47}Au_{33}Te_{20}$ nanotubes/rGO nanocomposites possess the optimal mass activity (5.31 mA $ug^{-1}{}_{Pd}$), which is 5.16-fold that of commercial Pd/C catalyst. The study provides a new strategy for clean synthesis of Pd-based NTs/GO nanocomposites, which are efficient catalysts for ethanol electrooxidation. It will probably inspire more appropriate utilization of graphene to design new hybrid materials with better properties applied in fuel cells or other related fields.



KEYWORDS: *Graphene oxide, Pd-based nanotubes, Stabilizing agent, Electrocatalyst, Ethanol oxidation*

INTRODUCTION

Development of electrochemical energy conversion devices is an effective route to meet the increasing demand for energy.[1−3] Direct ethanol alkaline fuel cells (DEAFCs) have received intense research attention in recent years due to the special advantages of ethanol, such as higher theoretical energy density (8 kW·h kg^{-1}) than methanol and formic acid and easier large-scale production directly from the fermentation of biomass.[4−6] In industrial and commercial applications of DEAFCs, the exploration of efficient and low-cost catalysts for ethanol electrooxidation should be given full consideration.[7]

It has been well-documented that the components, size, shape, and morphology as well as the supports are determinants of the catalytic activity of nanostructures.[8−11] Pt and Pt-based nanomaterials have been used as superior catalyst for DEAFCs due to the ultrahigh catalytic activity of Pt to break carbon bonds, but their inherent drawbacks hinder the sustainable development of DEAFCs.[7,12,13] Pd has been regarded as a competitive alternative of Pt when taking the reserve, cost, resistance to CO-poisoning, and reaction kinetics into account.[14] However, Pd has intrinsically low activity of cleaving carbon bond (C−C) and relatively weak stability when catalyzing ethanol electrooxidation.[15] To overcome this problem, other metals were incorporated into Pd materials to form Pd-based catalyst. So far, various Pd-based multi-component catalysts, including the low-Pt nanostructures such as PtPd- and PdPt-containing nanocrystals,[16−18] and non-Pt nanocatalysts such as PdAu, PdCu, PdFe, and PdRuTe nanostructures, have exhibited remarkably higher electro-catalytic activity and durability than their corresponding Pd counterparts.[12,19−21] Additionally, the one-dimensional (1D) nanostructures, especially hollow nanotubes, endow the catalysts with remarkable structure stability, catalytic property, and durability in contrast to zero-dimensional (0D) nano-particles.[6,22−26]

Moreover, to relieve the corrosion of common carbon supports and the resulting damage to the catalyst structure, promising support materials are an urgent need.[24,27,28] Graphene is single-layered and consists of carbon atoms with close-packed hexagonal lattices, possessing high electrical conductivity, large specific surface area, and mechanical flexibility.[29−32] It has been demonstrated that graphene or graphene oxide (GO) can be used as a superior support for electrocatalysis.[33,34] For example, graphene- or GO-supported Pd nanoparticles show high electrocatalytic ability for ethanol and formic acid oxidation.[35−38] Generally, there are two kinds of methods to prepare metal/graphene or metal/GO nano-composites: in situ growth and assembly after the preparation of metal nanoparticles. However, both methods have some defects. In the former, the morphology of nanoparticles can not be controlled well, and their distribution is usually not uniform.[36] In the latter, surfactant or stabilizing agent is often used in the preparation, which is generally difficult to be removed and will result in decrease of catalytic activity.[39]

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