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In-situ Reaction-Growth of $PtNi_X$ Nanocrystals on Supports for Enhanced Electrochemical Catalytic Oxidation of Ethanol via Continuous Flow Microfluidic Process

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Abstract

An *in-situ* composite methodology was first proposed to prepare carbon supported PtNix (X=1, 3, 1/3) via online reaction, nucleation and growth of nanocrystals (NCs) on carbon support along the microfluidic channel. Uniform dispersed ultra-small $PtNi_X$ NCs (1 nm~3 nm) with high surface exposed Pt atoms and enhanced nanocrystal-carbon interface interaction can be constructed. The annealed nanocomposites with a Pt/Ni ratio of 1/3 exhibit a mass catalytic activity of 7.6 times higher than the commercial Pt/C nanocatalysts in the ethanol oxidation reaction. The chronoamperometry measurement indicates that their mass activity still retains 3.6 times of the commercial Pt/C nanocatalysts at 4000 s. Moreover, $PtNi_3$/C nanocatalysts have the highest tolerance to CO poisoning according to CO stripping measurements.

Keywords Pt-based electrocatalysts; Ethanol electro-oxidation; Microfluidic; *In-situ* reaction; Ultra-small nanocystal

1. Introduction

It is well known that direct alcohol fuel cell (DAFC) has been considered as one of the most promising power sources owning to their high calorie value and low environmental pollution.[1-3] Catalysts with high activity and long durability are the key to the commercialization of fuel cells.[4, 5] At present, there are three types of fuel cell catalysts, or pure platinum[6, 7, 3] platinum alloys [8-13] and non-platinum materials [14, 15]. Considering the catalytic activity, power density and durability, platinum alloys are still the preferred material up to now. Many affords have been devoted into improving the catalytic activity, durability and anti-poisoning ability of platinum alloy catalysts, such as alloying with transition metals and heterogeneity at nano-scale. The improved electrochemical catalytic activity and durability is attributed to the abundant d electrons of transition metals and the unique d-d, d-p and d-s orbital hybridization.[12, 16-18] It also has been demonstrated that the atomic composition and microstructure of surface and interface directly determine the electrochemical catalytic activity and durability of the Pt-based catalysts, [8, 16, 19-24] particularly at smaller sizes.[25, 26] In addition, the opportune interaction between NCs and supports favors efficient charge transfer between NCs and chemicals for greatly enhanced catalytic performance.[27, 28]

Ethanol as fuel in DAFC has attracted great attention due to its low toxicity, low cost