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Improvement in stability of PtRu electrocatalyst by carbonization of in-situ polymerized polyaniline

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ABSTRACT

As well known, ruthenium is electrochemically unstable in direct methanol fuel cell (DMFC) operation shortening lifetime and deteriorating performance of DMFC device. In this work, a facile methodology for improvement in stability of PtRu electrocatalyst is described, in which PtRu alloyed nanoparticles are decorated by nitrogen-doped carbon (N_xC) originating from the carburization of in-situ polymerized polyaniline (PANI). Deceleration in Ru dissolution of N_xC protected PtRu electrocatalyst comes from the ionized Ru atoms induced by the carburization of PANI transferring electrons from Ru to nitrogen atoms evidenced by XPS measurement, which results in higher CO tolerance during potential sweeping from 0.6 to 1.0 V versus RHE for N_xC decorated PtRu electrocatalyst. Meanwhile, N_xC protected PtRu electrocatalyst only losses 10% of active sites after stability estimation; in contrast, 50% of active sites are lost for bare PtRu electrocatalyst. Moreover, fuel cell test illustrates that N_xC protected PtRu electrocatalyst shows comparable performance to bare PtRu electrocatalyst.

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Introduction

With increased environmental pollutants and serious energy crisis, searching for sustainable and clean energy sources to replace fossil fuels has attracted much attention [1–3]. Fuel cells have been widely concerned due to the advantageousness of pollution-free products and high-energy conversion efficiency. Direct methanol fuel cells (DMFC) have emerged as a promising energy supplier for portable devices owing to the low operation temperature and quick startup compared to hydrogen fuel cells [4–10]. Nevertheless, it is known that the commercial application of DMFC is still hindered by two main

bottlenecks: adsorption of carbon monoxide (CO) intermediates on the platinum (Pt) active sites and poor stability inducing poisoning of Pt active sites and serious deterioration in lifetime of DMFC devices [11–14]. Thus, it is of great scientific and practical importance to exploit highly CO-tolerant and durable electrocatalyst.

It is well known that Pt alloyed electrocatalysts for methanol oxidation reaction (MOR) have been extensively studied and exhibited higher CO tolerance compared to Pt electrocatalyst since $M(OH)_{ads}$ species ($M = Sn$ [15–17], Ru [18–21], Ni [22,23], Au [24,25], etc.) formed under lower potential enable to remove the carbonaceous adsorbents from covered Pt

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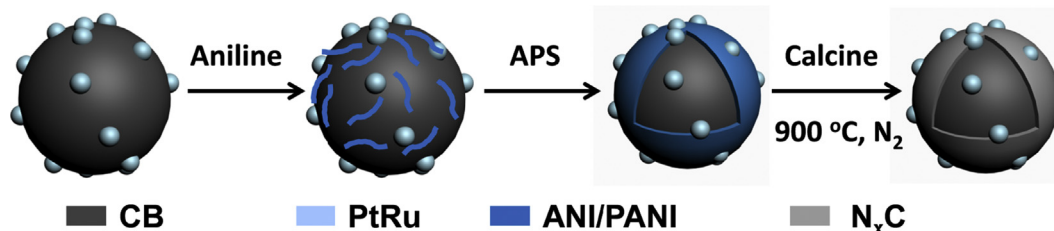


Fig. 1 – Synthetic routine of PtRu/CB@N_xC electrocatalyst.

nanoparticles. Until now, PtRu electrocatalyst is the most efficient alloyed electrocatalyst for DMFC since Ru(OH)_{ads} species are able to be formed under more negative potential [26,27]. Unfortunately, Ru oxides are extremely unstable in acidic condition leading to serious deterioration of catalytic activity [28]. Thus, some studies aimed to improve the stability of PtRu electrocatalyst have been carried out. Hsieh et al. fabricated ordered bilayer Ru–Pt core-shell nanoparticles to improve the stability of electrocatalyst [29]. Zhou et al. coated carbon nanotubes (CNTs)-supported PtRu with manganese dioxide (MnO₂) and successfully prevented the Ru dissolution resulting in improved stability [30]. However, MnO₂ layers also have enveloped the catalyst activity sites triggering low methanol oxidation reaction activity. Recently, we have reported that nitrogen doped carbon (N_xC) layer by carbonization

of poly(vinyl pyrrolidone) (PVP) and poly(2,5-benzimidazole) enhanced the stability of PtRu electrocatalyst [31,32]. N_xC originated from the carbonized polyaniline (PANI) has been considered as efficient protector for the enhancement in ORR activity as well as the stability of Pt electrocatalyst [33]. However, nitrogen doped carbon layer from PANI has not applied for PtRu electrocatalyst yet. Here, we proposed another simple methodology to significantly promote PtRu electrocatalyst's stability, in which an *in situ* polymerized PANI on the surfaces of PtRu/CB electrocatalyst was carburized to N_xC as shown in Fig. 1. During the process of polymerization, the aniline monomer was comparatively close to the carbon surface via preferential π - π conjugation [34–36]. During the carbonization of PANI, electrons from Ru was seized by the nitrogen atoms leading to partially oxidized Ru making it hard to be dissolved

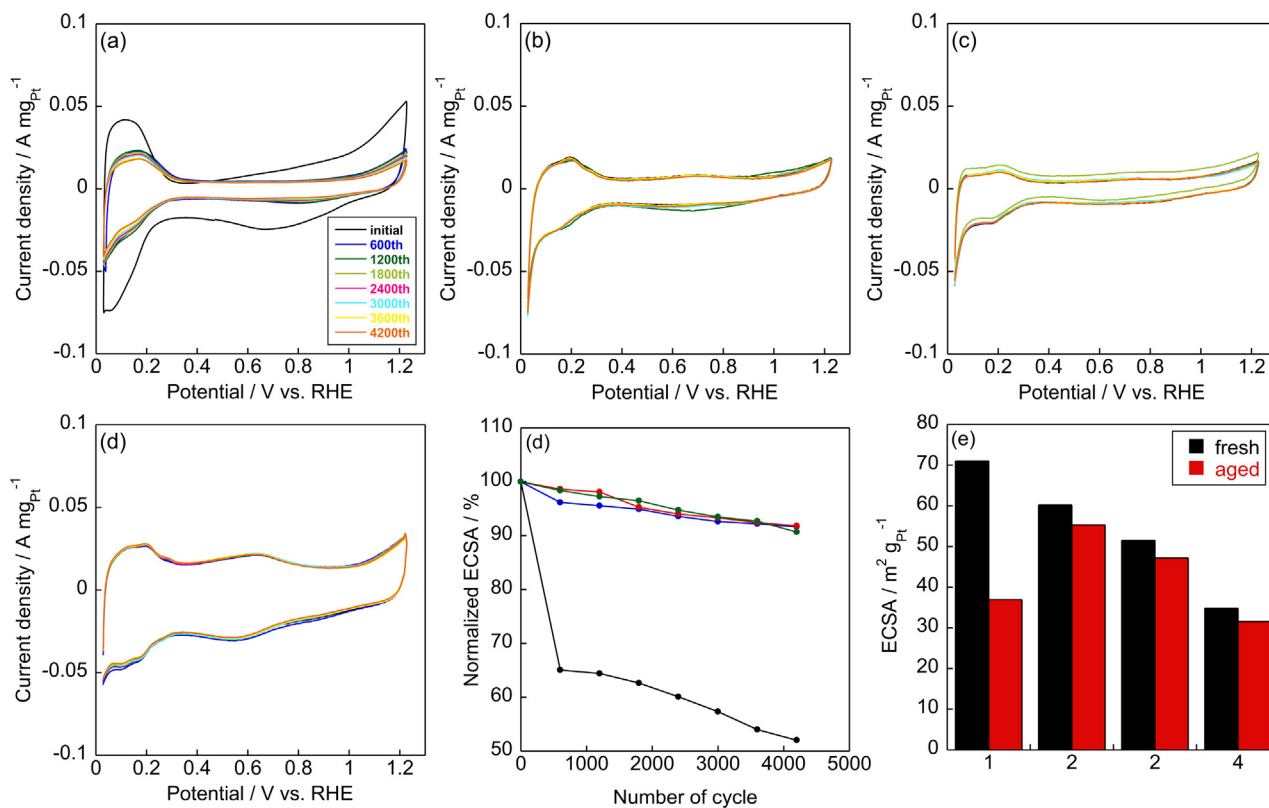


Fig. 2 – Stability results of (a) commercial PtRu/CB, (b) PtRu/CB@N_xC-20%, (c) PtRu/CB@N_xC-30% and (d) PtRu/CB@N_xC-50% after potential sweeping cycles from 0.6 to 1.0 V vs. RHE. (e) Normalized ECSA values of commercial PtRu/CB (black line), PtRu/CB@N_xC-20% (red line), PtRu/CB@N_xC-30% (blue line) and PtRu/CB@N_xC-50% (green line). (For interpretation of the references to color/colour in this figure legend, the reader is referred to the Web version of this article.)

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