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Carbon-supported Pd-Ag catalysts with silica-coating layers as active and durable cathode catalysts for polymer electrolyte fuel cells

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ABSTRACT

Carbon-supported Pd catalysts (Pd/CBs) for oxygen reduction reaction (ORR) in polymer electrolyte fuel cells (PEFCs) were modified with transition metals to enhance their catalytic activity. Pd-M/CB(N) (M = Co, Cu, Y, and Ag) was prepared by the reduction of metal precursors with $NaBH_4$ in the presence of ethylene glycol (EG) without heat treatment. Pd-Ag/CB(N) showed the highest catalytic activity for the ORR among all the Pd-based catalysts tested in the present study, and it had twice higher activity than Pd/CB. The Pd-Ag/CB(N) also had the higher ORR activity than the Pd-Ag/CB prepared by a conventional impregnation method (Pd-Ag/CB(I)) due to the formation of smaller Pd-Ag alloy particles with 1–2 nm diameters. Pd-Ag/CB(N) was covered with silica layers (SiO_2/Pd-Ag/CB(N)) in order to improve their durability under severe cathode conditions. The SiO_2/Pd-Ag/CB(N) had high catalytic activity for the ORR during the durability test because silica layers prevented the diffusion of metal species from the catalysts.

Introduction

Polymer electrolyte fuel cells (PEFCs) are promising alternative devices because of their high energy conversion efficiency and low emission of CO, CO_2 and NO_x. The PFECs produce electricity from electrochemical oxidation of hydrogen with oxygen [1]. Pt metal catalysts have been used as the active catalysts for hydrogen oxidation reaction (HOR) at anode and oxygen reduction reaction (ORR) at cathode. A large amount of Pt metal catalysts are consumed at the cathode compared with the anode due to the sluggish rate of the ORR. Moreover, Pt metal is too expensive and limited resource, which results in the inhibition of full commercialization of the PEFCs. Thus, many studies on new cathode catalysts for reducing the Pt metal loading or replacing Pt metal have been reported. The Pt-based alloy catalysts, including transition metals such as Co, Cu, Fe, and Pd showed the higher activity for the ORR than pure Pt metal catalyst. Therefore, the Pt metal loading in PEFC cathode can be reduced when the Pt-based alloy catalysts are used [2–6]. It is eventually required to develop non-Pt catalysts for full commercialization of the PEFCs. However, most metal species in the cathode catalysts are dissolved due to severe cathode conditions such as low pH, high temperature,

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