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Gradient design of Pt/C ratio and Nafion content in cathode catalyst layer of PEMFCs

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ABSTRACT

In order to increase the utilization of Pt, reduce mass transfer loss and improve the performance of polymer electrolyte membrane fuel cells (PEMFCs) under low humidity and high current densities, the cathode catalyst layers with two layers of different Pt/C ratio and Nafion content are fabricated and evaluated. Polarization curves (IVs), cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) are employed to characterize and compare the effects of Pt/C ratio and Nafion content gradient on the performance of PEMFCs under different humidification conditions. The results indicate that the performance of the membrane electrode assembly (MEA) can be significantly improved via allocating more Nafion and Pt/C in the sublayer near the membrane in cathode catalyst layer. The MEA with optimal gradient cathode catalyst layer results in improved catalysts utilization compared to MEA with single cathode catalyst layer, 0.403 g kW_{rated}^{-1} and 0.711 g kW_{rated}^{-1} under 80 RH% and 20 RH%, respectively. The areal power density of the optimal MEA is 28.4% and 135.7% higher than the conventional single-layer catalyst layer MEA under high and low humidity, respectively.

Introduction

Proton exchange membrane fuel cell (PEMFC) exhibits a multitude of merits including high energy density, high efficiency and environmentally friendliness, thus is attracting intensified attention in stationary and transportation applications [1]. Membrane electrode assembly (MEA), consisting of gas diffusion layer (GDL), catalyst layer (CL) and proton exchange membrane, is the core component of PEMFC. Reduction in the MEA's cost, one of the most urgent issues in commercialization of fuel cell vehicles, can be accomplished by improving the Pt utilization which at present is generally restricted by impeded mass transfer processes [2] and sluggish kinetics of oxygen reduction reaction (ORR) [3]. Given today's electrocatalysis in PEMFCs, tailoring the structure of the MEA to facilitate the mass transport and extend the triple-phase boundary is practically significant [4–16]. The approaches

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Table 1 – Specifications of SCL cathode and DCL cathodes.

Sample	Pt/C ratio(C_{in}/C_{out})/wt%	Nafion content(N_{in}/N_{out})/wt%
MEA-1	60/60	24.5/24.5
MEA-2	40/70	24.5/24.5
MEA-3	70/40	24.5/24.5
MEA-4	60/60	33/23
MEA-5	40/70	33/23
MEA-6	70/40	33/23
MEA-7	60/60	40/20
MEA-8	40/70	40/20
MEA-9	70/40	40/20

include introducing pore former [4], optimizing ionomer content [5,6] and fabricating a multi-layer structured CL [7–16]. The multi-layer structured cathode has been proved to be an effective method to reduce the mass transfer loss and improve Pt utilization in the cathode. The key parameters for the multi-layer structured CL are the gradients of Nafion ionomer content [7–11] and Pt loading.

It has been reported that PEMFCs performance can be improved through fabricating multi-layer structured CL with graded distribution of Nafion ionomer. Specifically, Nafion ionomer content at CL/GDL interface should be lower compared to that at CL/membrane interface in cathode [7–11]. The reason can be attributed to the facilitated oxygen supply and water removal process in cathode without sacrificing the proton conductivity [7–9].

Regarding Pt loading in the cathode, in-plane [12,13] and through-plane [14–16] gradients have been studied. The reactant gas concentration and current density will gradually decrease from the gas inlet to outlet, thus leading to a non-uniform utilization of the active area, which may have negative effects on the PEMFC lifetime. The current density uniformity and therefore the PEMFC performance can be improved through introducing an in-plane Pt gradient by increasing the Pt loading from cathode inlet to outlet [12,13]. In terms of through-plane gradient, increasing the Pt loading from the outer side (near the gas diffusion layer, GDL) to inner side (near the PEM) is beneficial to the performance [14–16] [17]. Furthermore, the PEMFC performance can be significantly improved through combined optimization of both Pt and Nafion ionomer distribution in the cathode CL [15,16]. Moreover, hydrophilic gradient catalyst layer with different kind of catalyst support improves water management of PEMFC in low-humidity conditions and at high current density [18]. In some cases the hydrophilic gradient design improved Pt utilization (above 50%), lowered CL/gas diffusion layer interfacial resistance and decreased mass transportation, thus yielding an output power density up to 0.76 W cm-2 with cathode Pt loading as low as 0.28 mg cm-2 [19].

In this paper, a double catalyst layer (DCL) cathode with both gradient Pt/C ratio and Nafion content is designed. The performance of DCL with different Pt/C ratio gradient direction and Nafion gradient span is compared to that of a single catalyst layer (SCL) in the cathode MEAs. Enhanced performance has been obtained by optimizing the gradient of Pt and Nafion in the DCL. We also investigated the effects of Pt/C ratio gradient and Nafion gradient under low humidity. With optimal gradient cathode the poor performance of the MEA under low humidity improved significantly due to the better catalysts utilization, mass transfer and water management.

Experiment

MEA fabrication

In this study, the catalyst slurry was prepared by disperse Pt/C catalyst Johnson Matthey (JM) in isopropanol with 5 wt% Nafion solution (DuPont, USA). Nafion 211 (DuPont, USA) was

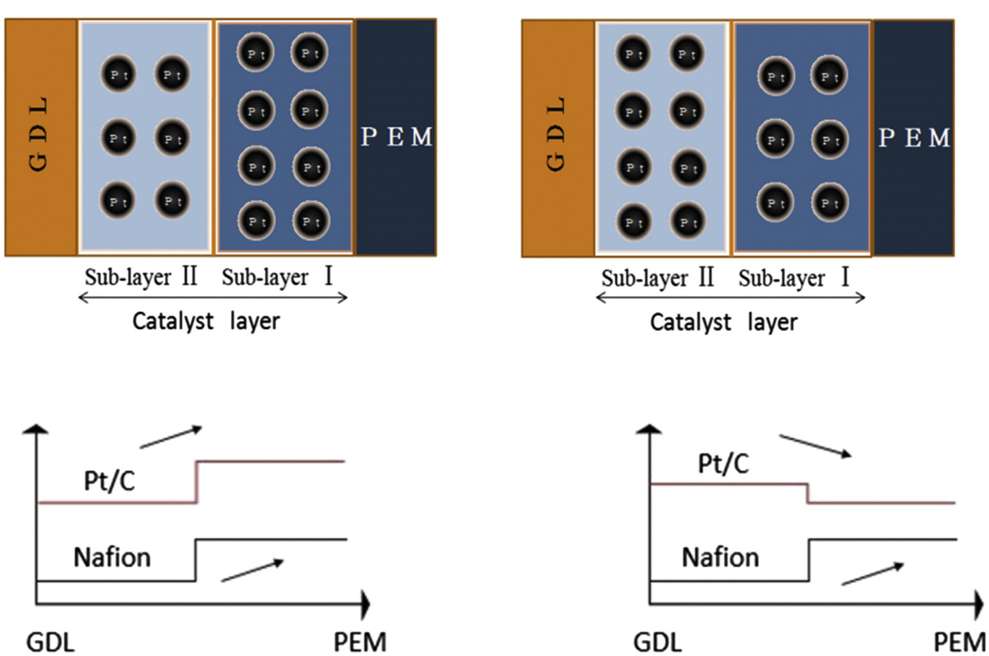

Fig. 1 – Schematic diagram of Pt/C ratio and Nafion content gradient in DCL cathode.

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