



Variations in performance-degradation behavior of Pt/CNF and Pt/C MEAs for the same degree of carbon corrosion

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ABSTRACT

Carbon corrosion due to high voltages encountered during the operation of fuel cells and the accompanying deterioration of cell performance are among the main issues affecting the durability of fuel cells. In this study, the carbon-corrosion durability of membrane electrode assemblies (MEAs) fabricated with Pt/CNF and Pt/C catalysts is studied using an accelerated stress test (AST) while applying high potentials. We compare the cell performance for the same degree of carbon corrosion, and the two MEAs show almost the same degradation behavior at the early stage of carbon corrosion. However, as the degree of carbon corrosion is increased, the performance-decay behavior of the two MEAs differs significantly, with there being a more serious performance decay in Pt/C than Pt/CNF, even at the same degree of carbon corrosion. The different behavior of the performance degradation between the two MEAs is due to the different catalyst layer structure, which results from the different catalyst-support morphologies between the three-dimensional (3D) high-structure Vulcan carbon (VC) and the one-dimensional (1D) structure of platelet carbon nanofibers (PCNFs). The support structure-dependent performance-degradation characteristic is analyzed in a single cell by performing an electrochemical analysis of the kinetic resistance, ohmic resistance, and diffusion resistance. In addition, the characterization of the catalyst layer structure before and after the deterioration is performed to understand the deactivation mechanism. This study is expected to provide a design strategy from a structural perspective of carbon support for the development of highly durable MEAs.

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1. Introduction

In polymer electrolyte fuel cell (PEFC) systems, the carbon-corrosion problem caused by high voltages that are generated during system operation, such as starting and stopping, is an important technical challenge that needs to be overcome in order to improve the lifetimes of fuel cells [1]. Therefore, various studies have been conducted to improve the understanding of the electrochemical oxidation reaction of carbon and to develop a highly durable material that can handle this issue [1]. Carbon corrosion can occur thermodynamically from 0.207 V, but the actual reaction, i.e., the carbon-oxide formation of the carbon support, occurs from 0.8 V or higher at 65 °C owing to its slow kinetics at low potentials

[2,3]. The electrochemical oxidation of carbon is also accelerated by the platinum nanoparticles that exist on the surface of the carbon support. The oxidation of carbon support destroys the connection site between the Pt nanoparticles and the support, leading to an agglomeration of Pt nanoparticles and Pt detachment from the carbon support. This results in the reduction of Pt active sites and oxygen reduction reaction (ORR) activity loss of the electrocatalyst [4–7]. Although carbon corrosion directly affects the reduction in the activity of the electrode catalyst, from the perspective of the membrane electrode assembly (MEA), the problem of oxygen diffusion in the catalyst layer due to the oxidation of the carbon support leads to more severe cell-performance degradation. The increase of the oxygen diffusion resistance in the catalyst layer by carbon oxidation occurs in a two-step process [8]. In the initial stage of carbon oxidation, the surface properties of the catalyst layer are hydrophilized owing to the formation of carbon oxides on the support surface, which leads to water-management problems

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