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Effects of porous flow field type separators using sintered Ni-based alloy powders on interfacial contact resistances and fuel cell performances

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ABSTRACT

The novel separators with a porous flow field using sintered corrosion resistant Ni-base alloy C276 (Ni–16Cr–16Mo–5Fe–4W mass%) powders or SUS316L (Fe–17Cr–12Ni–2Mo mass%) powders are investigated for proton exchange membrane fuel cells to enhance power density, which is one of the most important challenges for the widespread use of fuel cells. The developed separator with C276 powders demonstrated low ICRs (interfacial contact resistance) less than 10 mΩ cm^2 between separators and GDLs (gas diffusion layers), and it extensively enhanced power density by 90% higher than a conventional graphite separator. This is due to the superior adherence mechanism between the convex surfaces of the spherical powders and porous GDLs as well as the Ni concentration in passive oxide films in powder surfaces. Furthermore, this developed separator shows potential for using without an expensive conductive coating such as Au coating, which has been usually employed to lower ICRs for metallic separators with passive oxide films. In addition, the amount of eluted Cr, which could deteriorate catalyst and cell performance, from sintered C276 powders in a 1 mass% sulfuric acid aqueous solution is reduced by approximately 82% than SUS316L powders.

1. Introduction

Fuel cells have great potential as a clean, efficient, and environmentally friendly energy sources in diverse applications (e.g., mobile phones, household generators, automobiles, etc.) [1]. Key challenges for widespread commercialization of fuel cells are enhancing durability and reliability of fuel cell structures, realizing economical fabrication, and achieving higher power density [2–4]. In particular, separators are major components of fuel cell structures, which have important functions such as supplying hydrogen and oxygen, draining generated water, and transferring electrons; therefore, developing economical separators with higher durability, reliability, and capabilities for higher power density are extremely important [3,4].

Conventional graphite separators with groove type flow fields are the current benchmark [3,4], which show relatively lower ICR (interfacial contact resistance), important for achieving higher power density, between separators and GDLs (gas diffusion layers) because oxide films with high electric resistance are hardly formed on graphite surface. To enhance those power densities, studies of flow field designs are focused. J.G. Carton and A.G. Olabi investigated detailed effects of flow field configurations on cell performances and demonstrated superior cell performance of serpentine flow plates than parallel flow plates and maze flow plates [5].

On the other hand, there are two drawbacks in graphite separators; their brittleness with lower reliability makes reducing their thickness difficult and groove fabrication requires an expensive machining process. Because graphite polymer composite separators can be prepared economically via a stamping process, studies have been tried to improve their toughness. However, the presence of a polymer increases the internal electrical resistance, resulting in inferior electrical conductivities and a lower power density [6].

In contrast, metallic separators using stainless steel have been extensively studied due to their excellent toughness, good

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stamping formability, and high electric conductivity. However, compared to a conventional graphite separator, the passive oxide film formed on the surface of stainless steel separator results in a larger ICR between the metallic separator and GDLs, which leads to an inferior cell performance. For example, the reported target value for ICRs is 20 mΩ cm^2 [4], and other report says less than 10 mΩ cm^2 for the improvement of cell performance [7]. To lower the ICRs, electric conductive coatings (e.g., gold, carbon, and nitride) have been investigated [8–13]. Cabir Turan et al. lowered ICRs between SS316L stainless steel separator and GDL from 123 mΩ cm^2 to 10 mΩ cm^2 by TiN coating and to 23 mΩ cm^2 by CrN coating on SS316L stainless steel separators [13]. Cristopher J. et al. lowered ICRs between titanium separator and GDL to 4.75 mΩ cm^2 by gold coating [9]. However, these coatings tend to increase manufacturing costs, therefore, the other approaches without additional coatings have been studied [14,15]. Kaikai Huang et al. applied surface treatments, acid solution treatment, heat treatment, and electrochemical treatment, to SS304 stainless steel separators and lowered ICRs to 9.8 mΩ cm^2 due to the formation of Cr_2O_3/C layer on the surface of SS304 stainless steel by those treatments [15]. However, complicated treatments mentioned above are still necessary, and simplified processes are assumed to be expected.

In addition to low ICRs, metallic separators must be corrosion resistant because the conditions within a fuel cell are similar to an aqueous solution of sulfuric acid, and eluted elements from the metallic separator may deteriorate the membrane quality and reduce cell performance [3]. Therefore, expensive corrosive coatings (e.g., gold plating and carbon coatings) have been studied not only to reduce ICRs but also to improve corrosion resistance.

To overcome these challenges, we have been studying a fuel cell separator using a porous flow field that consists of sintered spherical alloy powders in which the interspaces between the sintered powders are used to feed oxygen/air and hydrogen as well as to drain generated water. The sintered alloy powders act as electron paths that correspond to the ribs in conventional separators [16] (i.e., the outer surface of the porous flow field is in direct contact with the porous GDLs made from carbon fibers).

In this study, the porous flow field type separators using sintered corrosion-resistant Ni-base alloy C276 (Ni–16Cr–16Mo–5Fe–4W mass%) powders and stainless steel SUS316L (Fe–17Cr–12Ni–2Mo mass%) powders are investigated. In particular, the influences of chemical composition of the alloy powders, powder size, and gold plating on ICRs are investigated. Furthermore, the power generation properties utilizing these porous flow field type separators are evaluated.

2. Experimental

2.1. Corrosion resistance tests of the sintered powders

Ni-base alloy C276 (Ni–16Cr–16Mo–5Fe–4W mass%) spherical powders and stainless steel SUS316L (Fe–17Cr–12Ni–2Mo mass%) spherical powders were produced by nitrogen gas atomization. To prepare sintered porous test samples with a 2-mm height and 25-mm diameter for the sulfuric acid solution immersion test, alloy C276 or SUS316L powders (210–297 μm diameter) were densely packed into an alumina case measuring 2-mm high and 25-mm in diameter. The filled cases were then vacuum sintered (1×10^{-5} Torr) at 1200 °C for 90 min. Each sintered test sample was immersed in a 1 mass% sulfuric acid solution with a volume of 500 ml at 80 °C for 336 h, and the eluted substances were analyzed by an ICP-MS (inductively coupled plasma mass spectrometer).

2.2. ICRs between sintered powders and GDLs

To evaluate the ICRs between the porous samples and the GDLs, sintered porous test samples with a 1-mm height and 25-cm^2 area (5 cm × 5 cm) were prepared. Alloy C276 powders (210–297 μm diameter), alloy C276 powders (20–69 μm diameter), or SUS316L powders (210–297 μm diameter) were densely packed into an alumina case with a 1-mm height and 25-cm^2 area (5 cm × 5 cm), and subsequently vacuum sintered (1×10^{-5} Torr) for 90 min at 1200 °C (1100 °C) for powders with 210–297 μm (20–69 μm) diameters.

Fig. 1 schematically illustrates the setup to measure the ICRs between the prepared specimens consisting of sintered powders and GDLs. Each specimen was placed between two pieces of 0.19-mm thick GDLs (Toray TGPH060), which were sandwiched by current-collecting copper plates. In step (I), the resistance with a specimen between the current-collecting plates was measured using a direct current resistance–measuring instrument (Yokogawa 275200) at room temperature. In step (II), the resistance without a specimen between two pieces of GDLs was measured using the procedure shown in Fig. 1. Then the ICRs were calculated by dividing the difference between step (I) and step (II) by two because their difference contains the ICRs for both sides of a specimen. This calculation assumed that the internal electric resistance of the metallic specimen itself and the ICRs between two GDL sheets were zero because these values were considered negligible. A compaction force ranging from 5 kgf cm^{-2} to 40 kgf cm^{-2} was applied during the ICRs evaluation. The ICRs between the SUS316L sheet (0.2-mm thick) heat treated in a reducing atmosphere with hydrogen and the same GDLs were also investigated for comparison.

Moreover, elemental distribution analysis on the surface of the atomized powders before sintering, sintered powders, and the SUS316L sheet were conducted by Auger electron spectroscopy (SAM670 Ulvac-phi) at the target sputtering depth rate of 10 nm min^{-1} to reveal the differences in the passive oxide films.

2.3. Porous flow field type separators

To prepare a porous flow field type separator, alloy C276 spherical powders with 60–106 μm (210–297 μm) diameters were densely packed into a recessed portion with a 25-cm^2 area and a 0.2-mm (0.5-mm) height in a stainless steel (Fe–17Cr–12Ni–2Mo mass%) plate. The samples were subsequently annealed for 90 min by vacuum sintering (1×10^{-5} Torr) at 1100 °C (1200 °C) for powders with 60–106 μm (210–297 μm) diameters. The average porosity in the flow field was approximately 48%. For comparison of single cell performance, a conventional serpentine groove type

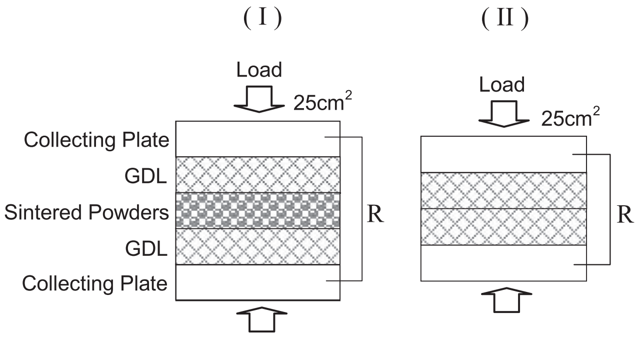

Fig. 1. Schematic of the assembly for an interfacial contact resistance (ICR) measurement.