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Short Communication

Graphitized carbon as an efficient mesoporous layer for unitized regenerative fuel cells



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ABSTRACT

To enhance the electrochemical performance of membrane electrode assemblies (MEAs) for unitized regenerative fuel cells (URFCs), we prepared and introduced graphitized-carbon (Gr-carbon) of high crystallinity as a mesoporous layer (MPL) in MEAs. The round-trip energy conversion efficiencies (ϵ_{RT} (%)) of non-MPL, typical amorphous-carbon (Am-carbon) MPL and Gr-carbon MPL containing MEAs at the current density of 1 A cm^{-2} were 36.6, 41.8, and 43.8, respectively. The overall round trip energy efficiency was considerably higher for the graphitized form of mesoporous carbon. For Gr-carbon, a high stability of the round-trip energy conversion efficiency was achieved even after 20th cycles (42.3%) owing to the enhanced electrical conductivity and high crystallinity of Gr-carbon after thermal treatment. Therefore, Gr-carbon considerably enhances the ORR and OER performances and prevents carbon corrosion and surface oxidation during the fuel cell and water electrolyzer modes in the URFC system.

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Introduction

The unitized regenerative fuel cell (URFC) is one of the most rapidly emerging electrochemical technologies for the conversion of chemical energy into electrical energy [1,2].

Typically, the URFC system can be operated either as a fuel cell (FC) or as a water electrolyzer (WE) mode using a single membrane electrode assembly of unit cell (MEA). This system shows great potential for use in spacecraft, zero-emission vehicles and residential power sources owing to its higher power density (packaged specific energy density of 400–1000 Wh/kg),

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lighter weight, longer-term operation, and cleaner energy than other conventional energy devices [2–8], which are all desirable properties for energy-efficient systems. Despite these favorable properties, the electrochemical energy performance is low during the cyclic process due to degradation of the MEA and inefficient electron transport and fuel supply in the oxygen reduction reaction (ORR) and the oxygen evolution reaction (OER). The degradation of carbon-based materials is the suspected cause of inefficient electron transfer and fuel supply.

Fundamentally carbon structured materials have been used in the fuel cell technology as an effective material for electrocatalyst support, gas diffusion packing (GDP), and bipolar plate (BP). The URFC unit cell performance has significantly been affected by the corrosion of conventional carbon black [3,9–11] in support materials, GDP and BP due to higher applied potential (~1.5 V) in WE mode. At present, many kinds of corrosion resistant properties of novel carbon and/or metal structures have been widely used as electrocatalyst support materials to overcome the carbon corrosion issues [12–18] in FC applications. In fuel cell technology, gas diffusion packing (GDP) is known as the serious alignment of meso or micro porous layer (MPL) and gas diffusion layer (GDL) in the unit cell. This GDP layer is more helpful for the fuel distribution and water management during the unit cell operations. Additionally, it provides an easy path for electrical conductivity between the electrodes and BP. Over the last few decades, amorphous carbon materials with high electrical conductivity were used as an optimum MPL to improve the performance of FC and URFC systems [19–22]. Among the different kinds of carbon materials, graphitized carbon (denoted as Gr-carbon) can be considered as a promising support material for the MPL due to its chemical stability, corrosion resistance, superior electrical conductivity, and low electrical resistivity [18,23–25]. The objective of the present investigation is to enhance the electrochemical performance of URFC systems through the addition of a novel structured MPL in the electrodes. Here, we have incorporated Gr-carbon as a MPL in the electrode instead of mesoporous amorphous carbon (denoted as Am-carbon). The sophisticated electrochemical properties of an URFC unit cell comprising a Gr-carbon MPL were demonstrated through the cyclic voltammetry (CV), FC, and WE performances.

Materials and methods

Gr-carbon was prepared by heat-treatment from commercially obtained mesoporous carbon (Sigma–Aldrich) using a high-temperature furnace. To prepare highly ordered Gr-carbon, Am-carbon was heat treated at 2600 °C for 2 h under an inert atmosphere. The Platinum:Iridium (PtIr) electrocatalyst for the bifunctional oxygen electrode (BOE) was prepared by mixing of Pt black (85 wt.%) and Ir black (15 wt.%). To prepare the catalyst ink, 10 mg of PtIr catalyst was suspended in 1 ml of de-ionized water in an ultrasonic bath. In order to study the structure of Am-carbon and Gr-carbon, X-ray diffraction (XRD) analysis was performed using a Rigaku X-ray diffractometer with CuK_α radiation. The electrochemical properties of Am-carbon and Gr-carbon were characterized by cyclic voltammetry using a Pine Potentiostat-Model AFCBP1

instrument. Gr-carbon ($100 \mu\text{g cm}^{-2}$) was loaded on an Au electrode, and the electrochemical potential was analyzed in a 0.5 M H_2SO_4 electrolyte solution at scanning rate of 5 mVs^{-1} . The cathode MPL was made by the coating process on carbon cloth (ELAT1400). LT140W ELAT® (E-TEK) was used as a gas diffusion layer (GDL) for the electrode. The PtIr catalyst was loaded on a homemade MPL (80% carbon black with 20% PTFE) for the cathode. The electrodes were hot-pressed with a Nafion™ 112 membrane at the optimum conditions of 140 °C under 800 psi for 3 min. The MEA performance was analyzed in a single cell in the FC and WE modes. In the FC mode, hydrogen and oxygen were supplied to the anode and cathode, respectively. In the WE mode, heated de-ionized water was supplied to the BOE. The fuel gas and water were supplied to the cell at constant flow rates of $150 \text{ cm}^3 \text{ min}^{-1}$ and $60 \text{ cm}^3 \text{ min}^{-1}$ for the FC and WE modes, respectively. The operating temperature of the URFC unit cell was 75 °C and 70 °C during the FC and WE modes, respectively [26].

Results and discussions

URFC performance of amorphous carbon MPL

The URFC performances were measured in the FC and WE modes in order to evaluate the function of MEAs with and without MPL in the cathode. The determined polarization curves of the ORR and the OER are shown in Fig. 1 (a, b). Compared to non-MPL MEA, an enhanced performance was obtained for the cathode containing MPL during operation in the FC mode. At 1 A cm^{-2} , the electrochemical potential of the cathodes with and without MPL were 0.71 and 0.62 V, respectively. A high voltage was measured in the cathode of the homemade MPL due to an enhancement of the electrical conductivity and homogenous distribution of fuel in the Am-carbon MPL. In the OER (WE mode) polarization curves, a similar performance was observed in both of the MEAs.

Fig. 1 (c) represents the cyclic performance in the FC mode for the cathode containing the PtIr electrocatalyst with an Am-carbon MPL. The unit cell performance of the MEA gradually decreased as the number of cycles increased. Moreover, the cycle performance of the unit cell was measured for 2 h in the FC and WE modes. The performance in the FC mode significantly decreased in the 17th cycle compared to the initial performance under identical operating conditions. In the 10th cycle of the FC mode, the potential value considerably decreased from 0.62 V (1st cycle) to 0.47 V at 1.5 A cm^{-2} . The potential of the unit cell in the FC mode significantly decreased after operation in the WE mode due to poor electron conductivity caused by carbon corrosion of the electrode's MPL in the WE mode. Thus, a novel structured MPL is required to enhance the electrochemical performance of the unit cell and suppress the degradation of the carbon MPL in the WE mode.

X-ray diffraction and CV analyses

The XRD patterns of the Am-carbon and Gr-carbon are shown in Fig. 2. For the as-received Am-carbon, two peaks were observed at 25.19° and 43.90° in 2θ . The diffraction peaks corresponded to the graphite lattice planes of (002) and (101).

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