



# Enhancement of cell performance using a gadolinium strontium cobaltite coated cathode in molten carbonate fuel cells

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## ABSTRACT

To enhance cathode performance, gadolinium strontium cobaltite ( $\text{Gd}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ , GSC) is coated onto a porous Ni plate by a vacuum suction method, for use as the cathode in molten carbonate fuel cells (MCFCs). GSC is a mixed ionic and electronic conductor (MIEC) material, and thus has high electronic conductivity and catalytic activity at low temperatures. The electrode performance of the GSC-coated cathode is examined by various methods, such as single cell operation and electrochemical impedance spectroscopy (EIS). At  $600^\circ\text{C}$ , the performance of a single cell using a GSC-coated cathode is  $0.813\text{ V}$ . This result is very surprising given that the performance of an uncoated conventional cathode is  $0.69\text{ V}$ . Impedance analysis confirms that a dramatic decrease in the charge transfer resistance after GSC coating is primarily responsible for the cell enhancement at low temperature. The reaction orders for  $\text{O}_2$  and  $\text{CO}_2$  at uncoated and GSC-coated cathodes are also examined via a symmetric cell test, to identify the reaction mechanism of oxygen reduction. The peroxide mechanism, which is known to be a fast reaction, is predominant for the GSC-coated cathode at low temperatures, whereas the superoxide mechanism is predominant for the uncoated cathode.

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## 1. Introduction

Fuel cells have received much attention as next-generation electricity production systems, due to their high efficiency and negligible emissions. Among many types of fuel cells, molten carbonate fuel cells (MCFCs) operated at a high temperature ( $650^\circ\text{C}$ ) have been developed for decades, and will soon enter the initial stage of commercialization. These fuel cells are highly efficient, do not require expensive noble metal catalysts, and have a variety of usable fuels [1–3]. MCFCs do, however, have a number of problems that must be solved in order for these cells to compete with current commercially available power generation devices, such as diesel engines and gas turbines. One of the most serious problems is guaranteeing the long-term lifetime of MCFC stacks [4–6]. The commercial target for the lifetime of a MCFC stack is more than 40,000 h, but this goal has not yet been reached with a single stack.

The main obstacle to reaching a 40,000 h lifetime is electrolyte loss caused by the volatilization and corrosion of the metallic separators [7]. An easy way to suppress electrolyte loss is to operate the MCFC stack at a low temperature, as this reduces the volatilization of the electrolyte and makes the corrosion reaction slow, thus

suppressing the electrolyte loss. MCFCs operated at low temperatures, however, have low stack performance, primarily because of reduced ionic conductivity in the electrolyte and the slow rate of the electrode reaction [8]. It is well known that the oxygen reduction reaction (ORR) rate at the cathode slows down when the operating temperature is decreasing, and a slow cathodic reaction leads to large over-potentials at the cathode, causing the low cell performance at low temperature. In contrast, the hydrogen oxidation reaction at the anode is a relatively fast reaction at low operating temperatures. Thus, the development of many alternative materials is required to enhance the electrochemical performance of the cathode in MCFC systems. To date, however, very few alternative materials for MCFC cathodes have been reported.

In solid oxide fuel cells (SOFCs), research on reducing the operating temperature is also a major issue in enhancing the chemical, physical, thermal, and mechanical durability of components such as the interconnectors, electrodes, electrolytes, and sealants. Reducing the operating temperature also results in low electrochemical performance in the cathode. Therefore, new cathode materials for operation at  $600\text{--}800^\circ\text{C}$  have been widely studied in order to enhance the electrochemical performance of the cathode [9–16]. Among the many potential cathode materials for intermediate temperature solid oxide fuel cells (IT-SOFCs), mixed ionic and electronic conductors (MIECs), which have a perovskite structure such as  $\text{La}_x\text{Sr}_{1-x}\text{Co}_y\text{Fe}_{1-y}\text{O}_3$ ,  $\text{Ba}_x\text{Sr}_{1-x}\text{Co}_y\text{Fe}_{1-y}\text{O}_3$ , and  $\text{Gd}_x\text{Sr}_{1-x}\text{CoO}_3$ ,

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have received much attention because of their high ionic and electronic conductivities, and their high catalytic activity for electrochemical reactions [10–16]. In particular,  $\text{Gd}_x\text{Sr}_{1-x}\text{CoO}_3$  has a high conductivity at low temperatures, and a high electrochemical catalytic activity for the cathode reaction of SOFCs.  $\text{Gd}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$  has a higher conductivity, at over  $540\text{Scm}^{-1}$  at  $600^\circ\text{C}$ , than that of  $\text{La}_x\text{Sr}_{1-x}\text{Co}_y\text{Fe}_{1-y}\text{O}_3$ , which is typically used as the cathode in IT-SOFCs [17]. Thus,  $\text{Gd}_x\text{Sr}_{1-x}\text{CoO}_3$  has been studied intensively as a cathode material for IT-SOFCs in recent years. For these reasons,  $\text{Gd}_x\text{Sr}_{1-x}\text{CoO}_3$  is also expected to enhance cathode performance when it is applied to the cathode of MCFCs. In this study, a modified cathode for MCFCs was prepared by coating  $\text{Gd}_x\text{Sr}_{1-x}\text{CoO}_3$  powder onto a conventional lithiated NiO cathode, and the electrode properties of the prepared cathode in an MCFC system were tested.

## 2. Experimental

A modified cathode was prepared by coating GSC powder ( $\text{Gd}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ , Newell, Inc., South Korea) onto a conventional lithiated NiO cathode. The GSC powder was coated onto a porous Ni plate, which becomes a lithiated NiO cathode after an in-situ oxidation and lithiation process in the single cell. The porous Ni plates were prepared by a tape casting method, and were then coated with GSC powder using a vacuum suction method. The GSC suspension was prepared as follows. Disperbyk-190 (Daehan, Inc., South Korea) at 3 wt.%, a dispersion agent, was dissolved in ethanol, and the prepared solution was agitated for 30 min. Then, 10 wt.% of the GSC powder, relative to the mass of the Ni plate, was added to the prepared solution, and was dispersed by ultrasonication for 1 h. The prepared GSC suspension was slowly dropped onto the Ni plate, which was then placed on a Buchner funnel connected to a vacuum pump before being dried in an oven at  $100^\circ\text{C}$  for 2 h. The final amount of GSC coating on the Ni plate was approximately 4.7 wt.%.

The physical properties of the coated cathode were characterized by various methods. The morphology of the coated cathode was examined by scanning electron microscopy (SEM; NOVA NanoSEM200), and the phase structure of the cathode was analyzed by X-ray diffraction (XRD; RIGAKU) using Cu-K $\alpha$  radiation.

The electrode performance of the GSC-coated cathodes was evaluated in a single cell with an active electrode area of  $100\text{cm}^2$ . Conventional components, such as a Ni–5 wt.% Al anode, a  $\text{Li}_{0.62}/\text{K}_{0.38}$  carbonate electrolyte, and an  $\alpha\text{-LiAlO}_2$  matrix, were used for the single cell test, and the GSC-coated cathodes and conventional uncoated lithiated NiO cathodes were tested in single cells. The performance of each single cell was monitored. The detailed operating conditions of the single cells have been reported previously in the literature [18]. Under the standard operating conditions, a mixture of air and  $\text{CO}_2$  was used as the cathode gas, and the fuel gas was a mixture of  $\text{H}_2$ ,  $\text{H}_2\text{O}$ , and  $\text{CO}_2$ . The flow rate of the reaction gas was fixed at 0.4 for fuel and air utilization in the anode and cathode gases. The  $\text{H}_2\text{O}$  in the anode gas was supplied using a water bubbler set to  $50^\circ\text{C}$ , and the DC current was applied to the single cells using an electric loader (ELTO DC Electronics Co., ESL300Z). The cell performance was evaluated by measuring the cell voltage at various current densities. To understand the electrochemical characteristics, a symmetric cell test was also performed. A symmetric cell was assembled using two of the same cathodes, matrices, and electrolytes with an area of  $25\text{cm}^2$ . To analyze the electrode polarization, electrochemical impedance spectroscopy (EIS) analysis was carried out at different operation conditions, using a Solartron SI1287 and a Solartron 1255B. The frequency range of the present EIS experiment was from 10,000 to 0.01 Hz.

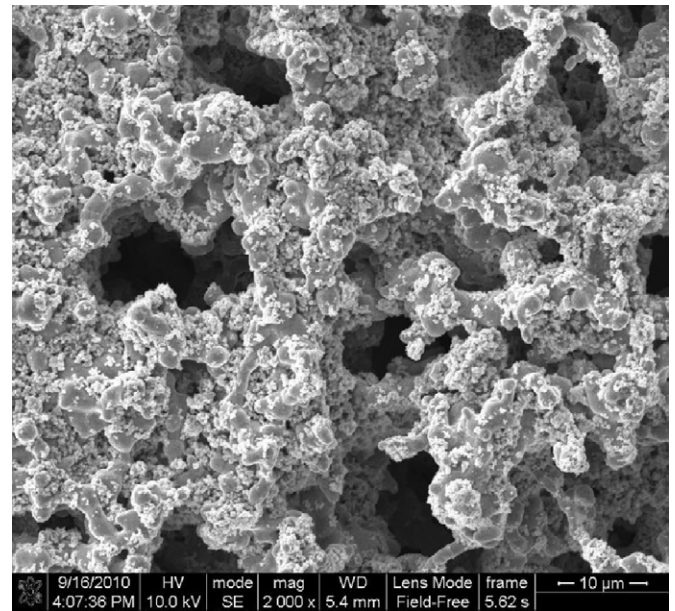


Fig. 1. SEM image of a 4.7 wt.% GSC-coated Ni plate.

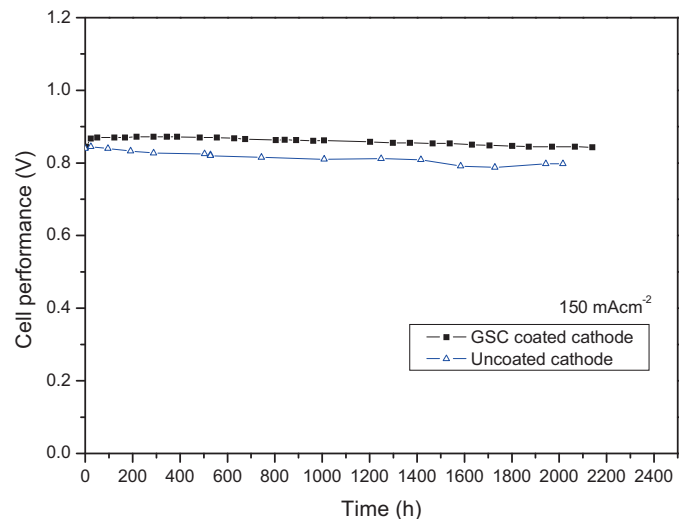


Fig. 2. The variation in the cell performance of single cells containing an uncoated cathode or a GSC-coated cathode operated at a current density of  $150\text{mAcm}^{-2}$  and a temperature of  $650^\circ\text{C}$  with standard anode and cathode gas compositions.

## 3. Results and discussion

The GSC-coated cathode was successfully prepared by a vacuum suction method. The top-view SEM image of a 4.7 wt.% GSC-coated Ni plate is shown in Fig. 1. This shows that small particles of GSC are well deposited on the top and inner surfaces of the porous Ni plate, without forming a thin layer of coating material.

The electrode performance of the GSC-coated cathode was evaluated using single cells. The performance and operating times of single cells using the GSC-coated cathode and the uncoated cathode, operated using standard cathode and anode gas compositions ( $U_f = U_o = 0.4$ ) at  $650^\circ\text{C}$ , are shown in Fig. 2. The cells operated at a current density of  $150\text{mAcm}^{-2}$  throughout. As shown in Fig. 2, the cell using the GSC-coated cathode has operated stably for approximately 2200 h, with a high cell performance averaging 0.85 V and no rapid drop off. The cell using the uncoated cathode also has operated very stably, with an average cell performance of 0.82 V for 2200 h. Thus, the GSC coating on the conventional cathode

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