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Performance improvement by metal deposition at the cathode active site in solid oxide fuel cells

Sung Min Shin^a, Byoung Young Yoon^a, Jung Hyun Kim^b,
Joong Myeon Bae^{a,*}

^a Department of Mechanical Engineering, Korea Advanced Institute of Science and Technology, 373-1, Guseong-Dong, Yuseong-Gu, Daejeon 305-701, Republic of Korea

^b Department of Advanced Materials Science and Engineering, Hanbat National University, 125, Dongseo-daero, Yuseong-gu, Daejeon, Republic of Korea

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ABSTRACT

In this study, the performance improvement of the SOFC single cell and its underlying mechanism was investigated. Furthermore, an application of the identified electrochemical mechanism is proposed and tested experimentally. The deposition of Platinum (Pt) at electrochemically active sites for the oxygen reduction reaction is determined to be responsible for the improved performance. Pt migration from a current collector to the cathode active sites originates from the oxygen partial pressure difference between current collector and triple phase boundary, and the electrochemical reduction reaction. It is supported by the confirmation of Pt particles at the cathode active sites by scanning electron microscopy (SEM), transmission electron microscopy (TEM) and calculations of the thermodynamic equilibrium partial pressure values. In addition, correlation of the initial performance change and the quantities of Pt deposition are investigated. This selective Pt deposition mechanism at the active sites is applied to the LSCF cathode, as well.

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1. Introduction

Solid oxide fuel cells (SOFCs) that operate at high (600–1000 °C) temperatures have attracted worldwide attention as next-generation power plants. They have several advantages such as high electricity generation efficiency, fuel flexibility and favorability for cogeneration plants. For the development of SOFC, research of cathode material is considered to be the most crucial because of its significant effect on cell performance. So far, ABO₃ perovskite-structured lanthanum strontium manganite (LSM) and lanthanum strontium cobaltite ferrite (LSCF) are developed as the representative cathode materials and their optimal composition as well as their optimal manufacturing and operating conditions have been

investigated [1–3]. LSM and LSCF, respectively, have the following advantages and disadvantages. First, LSM is chemically stable with Yttria-Stabilized Zirconia (YSZ) serving as the electrolyte, and its thermal expansion compatibility is similar to that of YSZ. Furthermore, LSM is stable for long-term operation at high temperatures (1000 °C). However, LSM does not exhibit high oxygen ionic conductivity, so the reaction sites are limited to the LSM/YSZ/air interface (Three Phase Boundary: TPB) due to oxygen ions cannot move through the cathode. By contrast, the reaction site of LSCF which is a mixed ionic-electronic conductor (MIEC) has a remarkably enlarged active cathode region that contacts air. However, LSCF has the chemical reactivity with YSZ, so additional buffer layer is required to prevent this undesirable reaction.

* Corresponding author. Tel.: +82 42 350 3045; fax: +82 42 350 3210.

E-mail address: jmbae@kaist.ac.kr (J.M. Bae).

Nomenclature			
E_{OCV}	open circuit voltage	GDC	gadolinium-doped ceria
F	Faraday constant, $96,485 \text{ C mole}^{-1}$	LSCF	lanthanum strontium cobaltite ferrite
$P(O_2)_{\text{cathode}}$	oxygen partial pressure at the cathode	LSM	lanthanum strontium manganite
$P(O_2)_{\text{anode}}$	oxygen partial pressure at the anode	OCV	open circuit voltage
$P(O_2)_{\text{air}}$	oxygen partial pressure in air	SEM	scanning electron microscope
$P(O_2)_{\text{TPB}}$	oxygen partial pressure at the three-phase boundary	SOFC	solid oxide fuel cell
R	universal gas constant, $8.3145 \text{ kJ kmole}^{-1} \text{ K}^{-1}$	TEM	transmission electron microscope
T	absolute temperature, K	2PB	two phase boundary
V_{cell}	operating cell voltage	TPB	three phase boundary
BSE	back scattered electron	YSZ	Yttria-Stabilized Zirconia
EDS	energy dispersive spectroscopy	<i>Greek symbols</i>	
EIS	electrochemical impedance spectroscopy	η_{cathode}	cathodic polarization loss
		$\eta_{\text{electrode}}$	electrode (anode + cathode) polarization loss
		η_{ohmic}	ohmic loss

Improved initial performances of SOFC that incorporate these cathode materials have been observed by several groups [4–14]. The phenomenon known as current conditioning or activation lasts between a few hours and several weeks. Several factors have been suggested to explain this enhanced the initial performance. For the LSM cathode, the microstructure of the YSZ surface changes during current flow, which increases the length of the TPB where the oxygen reduction reaction occurs [4,5]. Excess lanthanum oxide within the perovskite reacts with YSZ to form dense isolator ($\text{L}_2\text{Zr}_2\text{O}_7$) layers that then become dissociated during current flow [6–8]. Reduction and oxidation of Mn ions promote the formation of oxygen vacancies near the TPB and lead to oxygen-ion migration [9–11]. The current flow removes the passive species (MnO_x , SrO) that are formed during the sintering process on the surface of LSM and block gas diffusion [12,13]. The initial performance improvement of the cell that incorporate LSCF cathode was lower than that achieved with the LSM cathode. According to the other groups, however, the cell performance improvement was mainly due to passive

species such as SrO on the surface of the cathode blocking the gas diffusion [14–16]. Then, the passive species were dissociated under the current flow, which resulted in improved performance. Ohmic heating increases the mean cell temperature, which thermally activates the electrochemical process at the electrode–electrolyte interface [17].

These factors collectively affect the cell performance; however, none dominate the performance because their individual effects are relatively insignificant. Moreover, it is difficult to explain the long term enhancement with the factors announced above. In this paper, we confirmed the possibility of other causes of this phenomenon and suggested another cause that is presumed as a main contributor.

2. Experimental

To manufacture the anode support, NiO/YSZ (6:4 wt %) mixed powder was pressed into pellets of 32 mm in diameter and approximately 1.2 mm in thickness at 45 kg fcm^{-2} . The pellets

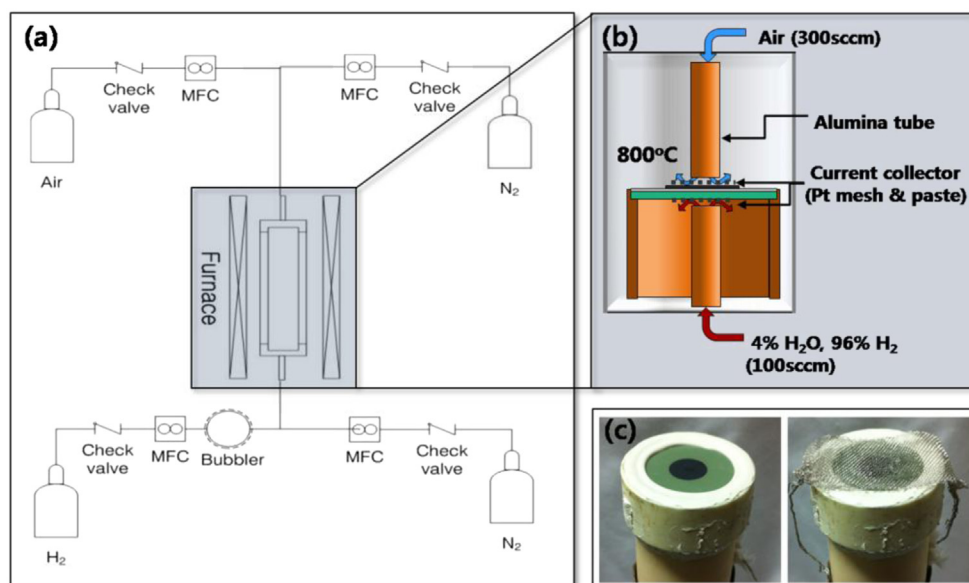


Fig. 1 – Schematic diagram for test setup and the real picture of tested single cell: (a) P&ID of experimental facility; (b) test setup and the operating conditions; (c) real pictures of single cell sealed with sealant and covered with current collector.

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