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IMPEDANCE STUDIES ON THE SYMMETRICAL CELL OF PROTON CONDUCTOR Y^{3+} -DOPED $Ba(Ce,Zr)O_3$ LAYERED WITH $LaSrCoO_3$ CATHODE MATERIAL

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ABSTRACT

The metal nitrate salts of Ba, Ce, Zr, and Y, are used as inorganic precursors for the preparation of Y^{3+} doped $Ba(Ce,Zr)O_3$ (BCZY) electrolyte, and La, Sr, Co, and Fe for the LaSrCoFe (LSCF) cathode. The electrolyte powder is pressed to fabricate a pellet with a 25 mm diameter and a thickness of 1.3 mm. The pellet is coated on both sides with the LSCF slurry using the spin-coating technique to form a symmetrical cell of LSCF|BCZY|LSCF. The cell is subjected to an electrochemical impedance measurement (EIS) in the air at two different temperatures. As the temperature increased, the size of the impedance spectrum decreased indicating that the cell showed thermally activated behavior. The area-specific resistance (ASR) of the cell is recorded to be 0.40 Ωcm^2 and 1.08 Ωcm^2 at 750 °C and 700 °C, respectively.

Keywords: Solid electrolyte, EIS, ASR, polarization resistance

INTRODUCTION

A fuel cell's electrolyte material is typically used to categorize the type of cell. There are five major types of fuel cells: polymer electrolyte membrane fuel cells, alkaline fuel cells, phosphoric acid fuel cells, molten carbonate fuel cells, and solid oxide fuel cells (SOFCs) [1]. The SOFCs are a type of fuel cell that can generate high-efficiency

electricity from a wide range of fuels. Due to these characteristics of high efficiency, low pollution, and fuel versatility, the SOFCs technology is widely regarded as a promising solution for addressing the rising global energy demand and the unavoidable impact of climate change. Research is being performed to reduce the operating temperature of SOFCs to improve their life expectancy and dependability. One type of the SOFCs that can operate at reduced temperature is the proton ceramic fuel cells (PCFCs). However, lowering the operating temperature causes inefficiency in the conventional cathode elements, as the cathode polarization resistance (R_p) rises, resulting in a significant drop in the PCFCs performance.

One of the approaches to reduce R_p is by introducing the mixed ionic-electronic conductor (MIEC) based on perovskite LSCF, which is widely studied and used in labs and industries. The perovskite shows high mixed ionic-electronic conductivity, excellent oxygen reduction reaction (ORR) catalytic activity, thermal and chemical compatibility with most electrolytes materials [2][3]. Furthermore, the LSCF has been reported to exhibit satisfactory cathode performance in PCFCs when coupled with doped $Ba(Ce,Zr)O_3$ electrolyte at an intermediate temperature range [3][4].

METHODOLOGY

The powders of $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF) and $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$ (BCZY) were prepared by a sol-gel method using nitrate salts of Ba, Ce, Zr, Y, La, Sr, Co, and Fe in aqueous solution at room temperature with the addition of complexing agents like EDTA, citric acid, and ethylene glycol as previously reported [5]–[7]. A 25-mm symmetrical cell of LSCF|BCZY|LSCF was fabricated following these procedures (a) the BCZY powder was hydraulicly pressed and sintered using a Two-Step Sintering (TSS) profile [7], and (b) a slurry of LSCF was then deposited on both sides of BCZY pellet using a spin-coating process. The symmetrical cell was subjected to a sintering temperature of 600 °C and 900 °C to burn out the organic binders and form good contacts between the electrode layer and the electrolyte surface, respectively.

The impedance measurement of the cell was carried out in the air using ZIVE SP2 Electrochemical Workstation (ZIVE LAB WonATech) with a perturbation amplitude of 10 mV at the frequency range of 10 mHZ – 1 MHz. A ZMAN**™** 2.2 f3 (ZIVE LAB) software was used to extract the responses from the spectrum.

RESULTS AND DISCUSSION

Figure 1 shows the pattern of the impedance spectrum of the symmetrical cell in the air at temperatures of 700 °C and 750 °C, respectively. The spectrum consists of two semicircles and became smaller in size as temperature increased from 700 ^oC to 750 ^oC indicating that the electrochemical process was thermally activated.

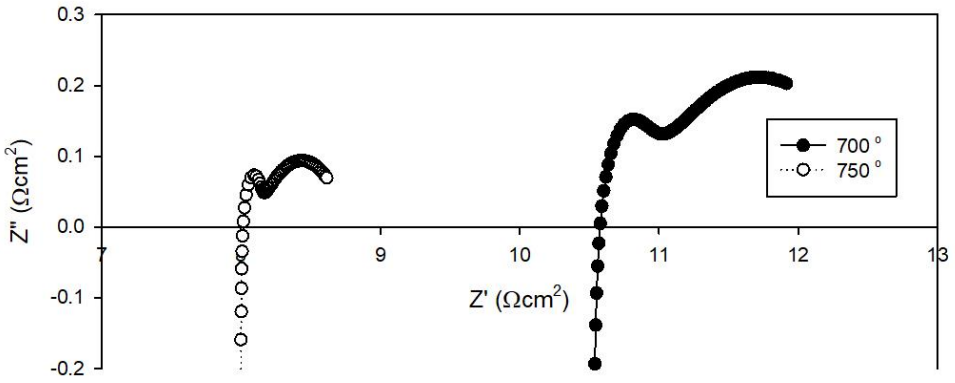

Figure 1: The pattern of impedance plot for the LSCF|BCZY|LSCF symmetrical cell at 700 °C and 750 °C, respectively

An example of the fitting procedure to extract the processes is shown in Figure 2. The processes that correspond to the two kinetics reactions were resolved by an equivalent circuit consisting of ohmic resistance (R_s) and electrode resistance of a *series connection* between *two parallel* RQ circuits as shown in inset Figure 2. The arc summit frequency (f^o) was applied to confirm the presence of these two arcs in the middle and low-frequency regions. The first arc (R_1Q_1) was associated with an electronic charge transfer that occurred at the cathode surface and the second arc (R_2Q_2) was referred as the ionic charge transfer reaction at the cathode-electrolyte interphase [8]. The calculated capacitance ranged from 3.36 x 10^{-4} to 2.00 x 10^{-2} Fcm^2 for the R_1Q_1 and R_2Q_2 circuits of 750 °C and from 4.03 x 10^{-3} to 7.45 x 10^{-1} Fcm^2 for the R_1Q_1 and R_2Q_2 circuit of 700 °C, that was representing to the respective cathode process. The ASR values obtained were 1.08 Ω.cm^2 and 0.40 Ω.cm^2 at 700 ^{o}C and 750 ^{o}C, respectively. The calculated values of capacitance (C), arc summit frequency (f^o), cathode resistance (R_1, R_2) and area-specific resistance (ASR) of the fabricated symmetrical cell are presented in Table 1.

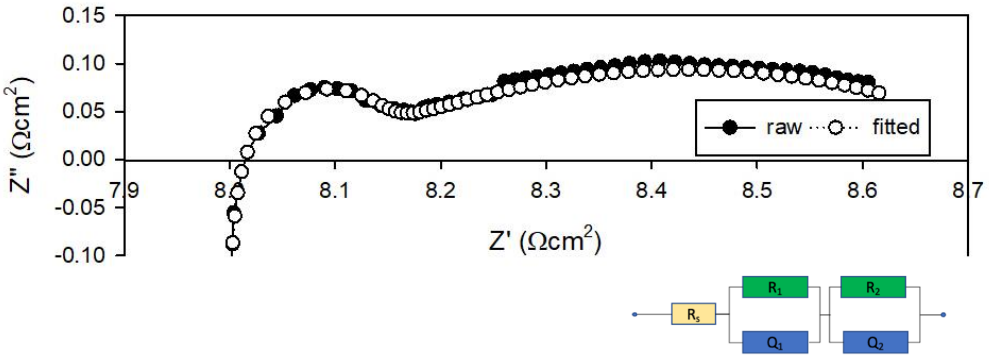

Figure 2: Experimental and fitted data of the symmetrical cell at T=750 ^{o}C. The inset is the equivalent circuit used for the fitting procedure

Table 1: Extracted parameters from the equivalent circuit of the symmetrical cell at temperatures of 700 ^{o}C and 750 ^{o}C

T/°C	C/Fcm^{-2}	f^{o}/Hz	R_1/Ω	R_2/Ω	ASR/Ωcm^{-2}
700	$C_1 = 4.03 \times 10^{-3}$ $C_2 = 7.45 \times 10^{-1}$	$f_1 = 9.68 \times 10^{2}$ $f_2 = 9.33 \times 10^{-1}$	0.12	0.65	1.08
750	$C_1 = 3.36 \times 10^{-4}$ $C_2 = 2.00 \times 10^{-2}$	$f_1 = 7.99 \times 10^{2}$ $f_2 = 1.26 \times 10^{-2}$	0.04	0.25	0.40

CONCLUSION

The 25-mm symmetrical cell with configuration LSCF|BCZY|LSCF was fabricated using dry-pressing and spin-coating techniques, accordingly. The cell showed the lowest ASR value of 0.48 Ωcm^2 at 750 °C. The constructed cell is considered to be viable and has the potential to be enhanced and implemented for practical usage.

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