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Abstract: This study reports the micro-structural and electrochemical properties of metal-supported solid oxide fuel cells (MS-SOFCs) with an  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-d}$  (LSM) /8 mol % yttria-stabilized zirconia (8YSZ) composite cathode, fabricated at room temperature using the aerosol deposition process (ADP). The composite cathode fabricated with the ADP technique shows uniform distribution of components and pores and the interface between the cathode and the electrolyte displays excellent joining properties. The area specific resistance (ASR) of the ADP-LSM/8YSZ sample is approximately  $1.50 \Omega \cdot \text{cm}^2$  at 800 °C, so this sample shows a significantly lower ASR value than the values usually reported for samples fabricated by the in-situ treatment method for MS-SOFCs. The power density of the cells with the ADP-LSM/8YSZ cathode coated on MS-SOFCs shows a maximum value of  $0.38 \text{ mW} \cdot \text{cm}^{-2}$  at 800 °C and stable performance in the severe thermal durability test. Therefore, these research results can broaden the opportunities for adoption of the ADP coating processes to fabricate cathode materials in MS-SOFCs.

## Detailed Responses to Reviewers

Ref. No.: CERI-D-15-04553

Title: Metal-supported SOFC with an aerosol deposited in-situ LSM and 8YSZ composite cathode

Dear editor and reviewers of Ceramics International:

I am Jung Hyun Kim, a corresponding author of the manuscript “Metal-supported SOFC with an aerosol deposited in-situ LSM and 8YSZ composite cathode.” Above all, I would like to thank you for your comments. Revised expressions or explanations from reviewers were written with [blue color](#) and minor corrections from professional proofreader were written with [red color](#) in the modified manuscript and “Detailed Response to Reviewers” file. The revisions with respect to the comments from each reviewer are as follows:

Reviewer #1:

1) Figure 1 - Are there possibly secondary peaks in the XRD pattern for the ADP sample at approximately 28° and 54° ?

→ Minor XRD peaks at approximately 28° and 54° can be observed in Fig. 1(a). However, they were not found in Fig. 1(b).

When comparing the XRD patterns measured at 28° and 54°, shown in Fig. 1(a) of this manuscript, with patterns found in the literature for  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-d}$  ([20] D. Grossin , J.G. Noudem, Synthesis of fine  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  powder by different ways, Solid State Sciences 6 (2004) 939-944, [21] V. S. Reddy Channu, R. Holze, E. H. Walker, Synthesis and characterization of  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-\delta}$  nanostructures for solid oxide fuel cells, New Journal of Glass and Ceramics, 3 (2013) 29-33) and 8YSZ ([23] C.W. Kuo, Y. H. Shena, I. M. Hung, S.

B. Wenc, H. E. Lee, M. C. Wang, Effect of  $Y_2O_3$  addition on the crystal growth and sintering behavior of YSZ nanopowders prepared by a sol-gel process, *Journal of Alloys and Compounds* 472 (2009) 186-193), the peak at approximately  $28^\circ$ , shown in Fig. 1(a), came from the background noise effect; the peak in the vicinity of  $54^\circ$ , shown in Fig. 1(a), originated from  $La_{0.8}Sr_{0.2}MnO_{3-d}$ . Therefore, an open circle (o) on the  $54^\circ$  peak was also added in Fig. 1(a) of the modified manuscript.

In addition, the peak in the vicinity of  $78^\circ$  was also assigned to  $La_{0.8}Sr_{0.2}MnO_{3-d}$  and an open circle was added to Fig. 1(a).

Literature references [20, 21, 22] that show LSM and 8YSZ single phases were also added in blue color in the modified manuscript.

2) Decomposition of PVdF is not the only reason for the generation of porosity. If possible, could you explain microstructural changed in the point of particle sintering effect?

→ The porous structures shown in Figs. 4(a) and (b) were caused by the decomposition of PVdF organic material.

When increasing the heat treatment temperature from  $800^\circ C$  to  $1100^\circ C$ , the sintering effect of the composite cathodes comprised of LSM and 8YSZ was also measured; significantly, the relatively dense structure shown in Fig. 4(b), compared to that shown in Fig. 4(a), is believed to be caused by the sintering effect at high temperature.

Therefore, the sentence “In addition, when increasing the heat treatment temperature from  $800^\circ C$  to  $1100^\circ C$ , the sintering effect of the composite cathodes comprised of LSM and 8YSZ was also measured; significantly, the relatively dense structure shown in Fig. 4(b), compared to that shown in Fig. 4(a), is believed to be caused by the sintering effect at high temperature.” was added in blue color in the modified manuscript.

3) When discussing thermal cycle (Fig. 8), the size of the cell or stack should be described because they significantly affect on the characteristics.

→A button cell was used to measure the current (I) - voltage (V) - power density (P), long term stability and thermal cycling stability of the ADP-LSM/8YSZ coated MS-SOFC. The final cathode surface area and thickness were about 1 cm<sup>2</sup> and 12 μm. The thickness of the anode-supported ceramic cell without cathode and metal support was 1 mm.

In addition, the sentence describing the size of the cell “The final cathode surface area and thickness were about 1 cm<sup>2</sup> and 12 μm. The thickness of anode-supported ceramic cell without cathode and metal support was 1 mm.” was also added in section 2.3 Fabrication process of MS-SOFC of experimental section in blue color.

#### Other minor modifications in the revised manuscript

- 1) In abstract section, minor modifications were written with red color.
- 2) In introduction section, minor modifications were written with red color.
- 3) In experimental section, minor modifications were written with red color.
- 4) In results and discussion section, minor modifications were written with red color.
- 5) On page 7, the phrase of “XRD patterns can be assigned to either YSZ or a LSM” was changed to “XRD patterns can be assigned to either an LSM or to an 8YSZ”
- 6) Reference numbers of [20], [21], [22], [23], [24] in the previous manuscript were changed to [23], [24], [25], [26], [27] in the revised manuscript.
- 7) In conclusions section, minor modifications were written with red color.
- 8) In acknowledgements section, minor modifications were written with red color.
- 9) In references section, minor modifications were written with red color.
- 10) In figure captions section, minor modifications were written with red color.
- 11) The legend color of Fig. 6 was changed to black and the summarized fitting ASR values

were changed to a black dash line.

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4 Metal-supported SOFC with an aerosol deposited  
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6 *in-situ* LSM and 8YSZ composite cathode  
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## 11 12 13 14 **Abstract** 15

16  
17 This study reports the micro-structural and electrochemical properties of metal-supported  
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19 solid oxide fuel cells (MS-SOFCs) with an  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-d}$  (LSM) /8 mol % yttria-stabilized  
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21 zirconia (8YSZ) composite cathode, fabricated at room temperature using the aerosol  
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23 deposition process (ADP). The composite cathode fabricated with the ADP technique shows  
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25 uniform distribution of components and pores and the interface between the cathode and the  
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27 electrolyte displays excellent joining properties. The area specific resistance (ASR) of the  
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29 ADP-LSM/8YSZ sample is approximately  $1.50 \Omega \cdot \text{cm}^2$  at  $800 \text{ }^\circ\text{C}$ , so this sample shows a  
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31 significantly lower ASR value than the values usually reported for samples fabricated by the  
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33 *in-situ* treatment method for MS-SOFCs. The power density of the cells with the ADP-  
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35 LSM/8YSZ cathode coated on MS-SOFCs shows a maximum value of  $0.38 \text{ mW} \cdot \text{cm}^{-2}$  at  
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37  $800 \text{ }^\circ\text{C}$  and stable performance in the severe thermal durability test. Therefore, these research  
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39 results can broaden the opportunities for adoption of the ADP coating processes to fabricate  
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41 cathode materials in MS-SOFCs.  
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50 *Keywords:* metal-supported solid oxide fuel cells (MS-SOFCs), aerosol deposition process  
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52 (ADP), cathode, sintering process, electrochemical property  
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## 1. Introduction

Metal-supported solid oxide fuel cells (MS-SOFCs) have been investigated as high-potential energy devices because they exhibit high mechanical strength, excellent sealing efficiency, and improved thermal resistance, as well as **having** a quick start up performance when compared to **that of** conventional SOFCs [1-6]. In spite of the many advantages of MS-SOFCs, there are many limitations in the fabrication process caused by the direct contact between **the** metal and ceramic components [7-10]. For example, sintering the cathode material under oxidizing conditions is impossible if the cathode is attached to the substrate, **and combined** with a metal part, **because** this combination has to be fabricated under reducing conditions. The use of high temperature sintering is also confined by the oxidation of the metal substrate used for the interconnector, the oxidation of the Ni-Cr-Fe material used for the cohesive layer and the oxidation of the anode substrate comprised of NiO-8YSZ [7, 11].

To solve the problems mentioned above, many studies have reported on relevant topics like atmospheric plasma spray coating [12], pulsed laser deposition (PLD) [2], wet ceramic processes [2], high temperature sintering in a reducing condition [13] and vacuum plasma spray processing [14].

Recently, an aerosol deposition process (ADP) method to form ceramic films has been developed; **because ADP can form either a thick and dense ceramic coating layer or induce the growth of the deposited phase in the bulk regime at room temperature**, this ADP technique has been applied to fabricate cathode layers and electrolyte films for lower temperature-operating solid oxide fuel cells (LT-SOFCs) as well as for the fabrication of an oxidation resistance coating layer on top of the metallic interconnectors in SOFCs [11,15,16].

In this work, the microstructural and electrochemical properties of **an** LSM/8YSZ composite cathode material prepared using the ADP technique without any subsequent high temperature treatment are investigated, especially for the use of this material in MS-SOFCs.



1 The developed ADP-LSM/8YSZ composite cathode was applied to a metal-supported cell  
2 and the electrochemical properties and thermal durability of this cell were investigated.  
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## 7 **2. Experimental section**

### 8 *2.1. Sample preparation process using the ADP technique*

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10 The sample preparation procedure for ADP treated samples has been described in details  
11 elsewhere [11, 17]. The preparation procedures for composite cathodes via the application of  
12 the ADP technique are as follows:  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-d}$  (LSM, Praxair) and 8 mol % yttria-  
13 stabilized zirconia (8YSZ, Tosoh) powders were used as the composite cathode materials and  
14 polyvinylidene fluoride (PVdF, Aldrich) was added as a pore former. To obtain an advanced  
15 triple phase boundary (TPB), the LSM powder was physically mixed with the 8YSZ powder  
16 for 6 h in a nylon jar ball mill using zirconia balls and ethanol as a solvent; this was followed  
17 by a drying step at 80 °C. The dried LSM-8YSZ-PVdF mixture was transported to a 25 x 0.8  
18 mm<sup>2</sup> rectangular shaped nozzle through a tube with compressed air as carrier gas. The aerosol  
19 flow was accelerated by a rotary pump with a mechanical booster and a nozzle; aerosol was  
20 sprayed onto the dense 8YSZ electrolyte surface with high energy impact. The gap between  
21 the substrate and the nozzle was set to 5 mm. Following the ADP technique as described, the  
22 composite cathode system comprised of LSM-8YSZ was deposited on top of the dense 8YSZ  
23 electrolyte without heat treatment.  
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### 48 *2.2. Sample preparation process for a symmetrical half cell*

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50 The cathodic polarizations displayed as Area Specific Resistance (ASR) were measured  
51 using symmetrical half cells. The 8YSZ electrolyte for the half cell measurement was  
52 fabricated using the powder compression technique; fabricated material was subsequently  
53 sintered at 1500 °C for 4 hours. This process produced a dense pellet (~95% of the calculated  
54 density) with a diameter of 26.0 mm and thickness of 1.5 mm.  
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1 In order to compare ASRs with respect to different fabrication processes such as ADP,  
2 screen printing and *in-situ* composite cathodes, symmetrical half cells were fabricated  
3 utilizing these different techniques. The term “*in-situ*” in this paper indicates that the coating  
4 process was carried out by screen printing and subsequent drying, but without additional heat  
5 treatment, so the cathodes prepared according to this method did not benefit from a sintering  
6 process.  
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14 For the ADP treated symmetrical half cell, the processes were carried out on 8YSZ  
15 electrolyte using the same experimental sequences as can be found in the literature [11, 17]  
16 with the experimental apparatus explained in chapter 2.1. In order to prepare the screen  
17 printed cathode, cathode inks for screen printing were fabricated using LSM (50 wt%)-8YSZ  
18 (50 wt%) with an acetone and a binder system comprised of  $\alpha$ -terpineol and KD-1. These  
19 were coated onto the dense 8YSZ electrolytes using screen printing to fabricate the  
20 symmetrical half cells, which was sintered for 1 h at 1100 °C in order to form a porous  
21 electrode structure well bonded to the electrolyte. For the *in-situ* test cell, cathode layers were  
22 screen printed on both sides of the dense 8YSZ electrolyte and dried at 200°C for 12 h  
23 without a subsequent sintering step. The final surface area of the symmetrical cell was about 1  
24 cm<sup>2</sup>.  
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### 43 2.3. Fabrication process of MS-SOFC

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45 A fully sintered anode-supported ceramic cell consisting of NiO-8YSZ as the anode  
46 substrate and 8YSZ as the electrolyte, without a cathode layer was joined to a Stainless steel  
47 430 (STS430, POSCO) metal support using a functional anode layer with a bonding property.  
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51 The functional anode layer used to connect the stainless steel with the anode supported  
52 SOFC single cell is composed of Ni-Cr-Fe powder (325-mesh, Alfa Aesar), 8YSZ, NiO (J.T.  
53 Baker) and graphite. STS430, with a plate thickness of 500  $\mu\text{m}$ , was used as the metal support  
54 for the MS-SOFC application. The single serpentine channel required for the fuel gas  
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diffusion was fabricated by wire cutting [18, 19].

The functional anode layer was directly casted onto the metal support and sintered for 10 h at 1400 °C to promote adhesion between the interface of the metal part and the functional layer (anode supported SOFC cell without cathode layer). This step was carried out in a 4 % H<sub>2</sub>/96 % Ar atmosphere to prevent the oxidation of the metal part. Afterwards the composite cathode was applied directly on top of the electrolyte using the ADP coating technique. The final cathode surface area and thickness were about 1 cm<sup>2</sup> and 12 μm. The thickness of anode-supported ceramic cell without cathode and metal support was 1mm. The MS-SOFC single cell fabricated in this way was used to measure the power density and impedance characteristics.

#### 2.4. X-ray diffraction and microstructure analysis

X-ray diffraction (XRD) results were obtained in a RIGAKU D/MAX-IIIC (3 kW) using Cu K $\alpha$  radiation ( $\lambda=0.15418$  nm) operated under conditions of 40 kV and 45 mA. The data were collected at 0.06 ° steps with a counting time of 1 second per step, in the 2 $\theta$  range from 20 ° to 80 °.

The microstructures of the symmetrical half cells were investigated using a field emission scanning electron microscope (FE-SEM, S-4200, Hitachi) combined with energy-dispersive spectroscopy (EDS).

#### 2.5. Electrochemical characterization

Measurements of electrochemical properties and ASRs of the cathodes in air were conducted at open circuit voltage (OCV) as a function of temperature between 500 and 850 °C, with an increment of 50 °C. An AC four-probe method using a Solartron 1287/ Solartron 1260 (electrochemical interface impedance, gain-phase analyzer) was used to measure the electrochemical properties. The impedance measurements were conducted in a frequency

1 range of 5 MHz to 100 mHz; the amplitude of the applied voltage was 20 mV under OCV.  
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3 The ASRs, measured from the differences between the first intercept in the vicinity of the  
4 high frequency and the last intercept at low frequency, were divided in two because the tested  
5 cells had two symmetrical electrodes.  
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10 Power densities were measured using the same Solatron with a four probe configuration.  
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12 3% H<sub>2</sub>O humidified H<sub>2</sub> was supplied by bubbling H<sub>2</sub> fuel through de-ionized water to the  
13 anode chamber with a flow rate of 30 sccm. At the same time, compressed air was fed into the  
14 cathode side with a flow rate of 30 sccm.  
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### 22 3. Results and discussion

23  
24 Figs. 1(a) and (b) show the X-ray diffraction (XRD) patterns of the La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3-d</sub>  
25 (LSM) /8 mol % yttria-stabilized zirconia (8YSZ) composite cathode prepared using the  
26 aerosol deposition process (ADP) on a dense 8YSZ electrolyte at room temperature and of the  
27 LSM/8YSZ composite cathode sintered at 1100 °C after screen printing onto a dense 8YSZ  
28 electrolyte. In detail, in Fig. 1, open circles (○) and closed squares (■) respectively indicate  
29 peaks of the XRD patterns originating from LSM and 8YSZ. Each of the peaks observed in  
30 the XRD patterns can be assigned to either an LSM or to an 8YSZ single phase perovskite  
31 with stabilized cubic structure [20-22]. After the screen-printed sample is sintered at 1100 °C,  
32 no secondary phases or unknown phases are observed additional to the deposited LSM and  
33 the YSZ. Significantly, the broad peaks shown in Fig. 1(a) changed to sharp peaks after  
34 sintering in all ranges tested; comparing the XRD results of the sintered and the unsintered  
35 cathodes, this observation implies that the cathodes of the LSM/8YSZ prepared using ADP  
36 without heat treatment show relatively smaller particle sizes than do the cathodes prepared  
37 using heat treatment.  
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57 Figs. 2 and 3 show scanning electron microscopy (SEM) images of the ADP-LSM/8YSZ  
58 cathode and of the *in-situ* LSM/8YSZ composite cathode, both on a dense 8YSZ electrolyte.  
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1 The thickness of the deposited LSM/8YSZ layer fabricated with various coating techniques is  
2 approximately 12~14  $\mu\text{m}$ . The cross-view images of the ADP-LSM/8YSZ and of the *in-situ*  
3 LSM/8YSZ composite cathode seem to show that these materials have similar structures, as  
4 can be seen in Fig. 2(a) and Fig. 3(a). However, when comparing the enlarged images that  
5 show the microstructural properties, as provided in Fig. 2(b) and Fig. 3(b), the sample using  
6 ADP treated LSM/8YSZ can be seen to have a melted surface shape and the *in-situ* sample  
7 shows the existence of randomly distributed LSM and 8YSZ phases, having various shapes  
8 and sizes without any certain tendency.

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19 Top views of the two samples are provided in Fig. 2(c) and Fig. 3(c) under larger  
20 magnification. The grain boundaries and the interfaces between the particles of the ADP  
21 treated cathode can be distinctively seen in Fig. 2(c); in fact, they can be seen, much more  
22 clearly than those of the *in situ* cathode, which are shown in Fig. 3(c).

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29 Fig. 4 provides a comparison of the cross-view microstructures of the ADP fabricated  
30 LSM/8YSZ composite cathode after sintering at 800  $^{\circ}\text{C}$  (Fig. 4(a)) and 1100  $^{\circ}\text{C}$  (Fig. 4(b))  
31 and the *in-situ* treated LSM/8YSZ composite cathode sintered at 1100  $^{\circ}\text{C}$ . The compact  
32 structure of the sample shown in Fig. 2(a) changes to a porous microstructure after heat  
33 treatment, as shown in Fig. 4(a) and (b), because the PVdF organic material used in the ADP  
34 coating process was decomposed and then removed at 1100  $^{\circ}\text{C}$ . In addition, when increasing  
35 the heat treatment temperature from 800  $^{\circ}\text{C}$  to 1100  $^{\circ}\text{C}$ , the sintering effect of the composite  
36 cathodes comprised of LSM and 8YSZ was also measured; significantly, the relatively dense  
37 structure shown in Fig. 4(b), compared to that shown in Fig. 4(a), is believed to be caused by  
38 the sintering effect at high temperature. If the compact structure shown in Fig. 2(a) does not  
39 change, it can result in a lack of gas diffusion which would increase the cathodic polarization.  
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1 When considering the differences in adhesion properties between the ADP-treated cathode  
2 **shown** in Fig. 2 and the sintered cathode **shown** in Fig. 4(c), the ADP-treated composite  
3 cathode shows better contact properties at the interface and the bulk. For example, the  
4 sintered sample was easily peeled off **using** 3M tape, but the sample prepared using the ADP  
5 technique showed stronger adhesion properties, **proven** by its getting stuck on 3M adhesive  
6 tape.  
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14 **To compare the electrochemical properties with respect to the fabrication processes,** Fig. 5  
15 displays the impedance spectra of the symmetrical half cells measured at 700 °C. For example,  
16 Figs. 5(a) and (b) **provided** the impedance results **for** an ADP-LSM/8YSZ composite cathode  
17 fabricated at room temperature and an LSM/8YSZ composite cathode sintered at 1100 °C. In  
18 addition, Fig. 5(c) **gives** the impedance results **for** a half cell with an *in-situ* coated  
19 LSM/8YSZ cathode layer. The ASRs of the ADP-LSM/8YSZ sample **shown** in Fig. 5(a), the  
20 LSM/8YSZ sample **sintered** at 1100 °C **shown** in Fig. 5(b), and the *in-situ* treated LSM/8YSZ  
21 sample **shown** in Fig. 5(c) are approximately 1.50, 1.80, and 11.0  $\Omega \cdot \text{cm}^2$ , **respectively**, at 700  
22 °C. Comparing the results of the ADP treated sample **shown** in Fig. 5(a) and the *in-situ*  
23 sample **shown** in Fig. 5(c), the ADP-LSM/8YSZ **sample** shows a significantly lower ASR  
24 value than that of the sample prepared by the *in-situ* method, which is commonly used for  
25 conventional MS-SOFCs.  
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43 Considering the shape of the impedance arcs, the impedance plot of the *in-situ* treated  
44 LSM/8YSZ sample, **shown** in Fig. 5(c) appears to be a single merged circle, very similar to  
45 the shape of the impedance plot of the ADP-LSM/8YSZ sample **shown** in Fig. 5(a). An  
46 important result of the two impedance plots shown in Fig. 5(a) and (c) is that the plot of the  
47 ADP-treated sample consists of a single merged circle, which is different from the shape of  
48 the plot of the sintered sample, **which** shown in Fig. 5(b), **and which displays** two different  
49 arcs separated by a local minimum at a frequency of 1000Hz.  
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1 It is notable that, when considering the impedance arcs with respect to the frequency  
2 ranges, the high frequency resistance ( $R_H$ ) associated with frequencies above 1000 Hz **can be**  
3 **seen** at the left side of the arc in the impedance plot of the ADP-LSM/YSZ half cell, but at the  
4 middle of the arc in the impedance plot of the half cell with the *in-situ* LSM/YSZ cathode  
5 fabricated without any sintering process after screen printing. The reason for this is that the  
6 interfacial state of the *in-situ* processed cathode is not connected to the electrolyte, **rather, the**  
7 **cathode is** deposited without any physical **or** chemical processes. According to the literature  
8 related to the behavior of impedance plots, the characteristic frequency of arcs in an  
9 impedance spectrum **can give** insight into the cathode reactions [23]. The resistance  
10 associated with high frequency ( $R_H$ ) is generally related to charge transfer between particles.  
11 In particular, the resistance caused by charge transfer at the interface of **an** electrolyte and **an**  
12 electrode is presented as a high frequency (HF) semicircular region above 1000 Hz [23-26].  
13 Considering **this**, the main differences between Figs. 5(a) and (c) can be explained as follows:  
14 the different positions of  $R_H$  in the arcs of the ADP-treated cathode in Fig. 5(a) and of  $R_H$  of  
15 the *in-situ* treated cathode in Fig. 5(c) **are** directly related to interfacial adhesion. The *in-situ*  
16 cathode with very poor interfacial properties suffers from a high ASR, which, **due to** the  
17 characteristic frequency of the related arc in the  $R_H$  range, can be related to interface contact  
18 problems. As **can be** seen in Fig. 2, the ADP-LSM/YSZ cathode shows highly enhanced  
19 interfacial adhesion between the cathode particles and between the cathode and the electrolyte.  
20 This advanced microstructure, caused by the high impact energy of ADP, obviously improved  
21 the electrochemical performance.

22 Fig. 6 shows summarized logarithmic ASR results **for** the ADP-LSM/8YSZ cathode layer  
23 without any sintering process as well as **for** the sintered LSM/8YSZ cathode layer prepared  
24 using screen printing; **results are** plotted against the inverse temperature. The ASRs of the  
25 ADP-treated sample **are** observed **to be** 1.50, 0.49 and 0.13  $\Omega \cdot \text{cm}^2$  at 700, 800, and 900 °C,  
26 respectively. The ASRs of the sintered samples are 1.12, 0.73, and 0.22  $\Omega \cdot \text{cm}^2$  at 700, 800,  
27 and 900 °C, respectively.



1 and 900 °C, respectively. With respect to these measured results, it is important to note that  
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3 the ADP sample shows smaller ASR values than those of the screen printed and sintered  
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5 sample at measuring temperatures of 750 °C or higher. This indicates that the ADP technique  
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7 can be directly applied to the cathode material of the MS-SOFC fabricated without a cathode  
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9 sintering process because MS-SOFCs are generally operated at high temperature ranges (over  
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11 800 °C).  
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14 Fig. 7 provides a cross-sectional image of the anode in the developed MS-SOFC. The  
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16 developed MS-SOFC consists of metal and ceramic parts in a layered formation of ADP-  
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18 LSM/8YSZ composite cathode / 8YSZ electrolyte / 8YSZ-NiO anode (Anode layer 1) / Ni-  
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20 Cr-Fe powder-8YSZ-NiO functional anode layer (Anode layer 2) / STS430 metal support.  
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22 The electrode consist of an anode part comprised of NiO-8YSZ for the fuel oxidation, and a  
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24 functional adhesive anode layer made of a Ni-Cr-Fe based powder, which connects the 8YSZ-  
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26 NiO part to the STS metal support. The STS430 metal support layer, displayed at the bottom  
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28 of Fig. 7, has 400 µm sized holes working as channels to allow gas diffusion.  
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33 Fig. 8(a) shows the current (I) - voltage (V) - power density (P) curve of an MS-SOFC  
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35 with the ADP-LSM/8YSZ cathode at 800 °C. The open circuit voltage (OCV) value of the  
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37 MS-SOFC cells comprised of the ADP-LSM / 8YSZ composite cathode / 8YSZ electrolyte /  
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39 8YSZ-NiO anode / Ni-Cr-Fe powder-8YSZ-NiO functional layer / STS metal support was  
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41 found to be approximately 1.07 V, which is satisfactorily close to the theoretical value  
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43 obtained from the Nernst equation [27]. Most importantly, the high OCV value indicates that  
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45 the dense structure of the 8YSZ electrolyte has not been destroyed and is well formed by the  
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47 many processing steps adopted in the fabrication of the MS-SOFCs. From the electrochemical  
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49 properties shown in Fig. 8(a), the maximum power density of the ADP-LSM/8YSZ coated on  
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51 an MS-SOFC is 0.38 W·cm<sup>-2</sup> at 800 °C. Fig. 8(b) shows the long term stability of the ADP-  
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53 LSM/8YSZ applied as a cathode material of an MS-SOFC. The cell performance is slightly  
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55 degraded during the initial 12 h experiment time; this trend was found to continue until 30 h  
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1 after the start. The initial degradation of the cell may be caused by the created porous  
2 structure in the ADP-LSM/8YSZ cathode because the experimental temperature of 800 °C  
3 can affect the structure of the composite cathode shown in Fig. 4(a). Thereafter, the cell  
4 resistance does not significantly increase for the next 70 hours.  
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10 Fig. 8(c) shows the thermal cycling stability of the ADP-LSM/8YSZ coated MS-SOFC.  
11 Thermal cycling was conducted within the very severe temperature range between room  
12 temperature and 800 °C. The cycles numbered 1 to 6 in the legend in Fig. 8(c) show the  
13 number of thermal cycles. From these impedance plots, it can be seen that the ADP-  
14 LSM/8YSZ coated metal-supported cell shows nearly equivalent impedance results regardless  
15 of the thermal shock, except for slight increases in the low frequency ranges. Therefore, the  
16 MS-SOFC with ADP-LSM/8YSZ can be said to exhibit excellent thermal shock durability.  
17 These results derive from not only the robust metal-supported cell technology but also from  
18 the robust cathode-electrolyte system, with a perfectly joined interface following ADP.  
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#### 34 4. Conclusions

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36 LSM/8YSZ cathode formation without the use of a high temperature sintering process is  
37 an important requirement for MS-SOFCs with long-term stability and high cathode  
38 performance, because the oxidation of the metal component can be successfully avoided. The  
39 interfacial properties between the cathode and the electrolyte and between the particles of the  
40 cathode composite phases should be considered because they have a significant influence on  
41 the electrochemical behavior of the cell, even though good cathode performance is primarily  
42 related to the intrinsic electrocatalytic properties of the materials. The utilization of the ADP  
43 technique induces a strong interconnection between the particles and the layers via an  
44 anchoring effect induced by the high impact energy; this anchoring effect results in enhanced  
45 microstructural properties of ADP-LSM/8YSZ. The ADP-LSM/8YSZ cathode shows a low  
46 ASR values of approximately  $1.50 \Omega \cdot \text{cm}^2$  at 800 °C; the ADP-LSM/8YSZ coated MS-SOFC  
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1 shows a maximum power density of  $0.38 \text{ mW} \cdot \text{cm}^{-2}$  at  $800 \text{ }^\circ\text{C}$ . The stable performance of the  
2 MS-SOFC with ADP-LSM/8YSZ is confirmed through a severe thermal durability test.  
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## Figure captions

Fig. 1. XRD diagrams of (a) LSM/8YSZ fabricated by aerosol deposition at room temperature and (b) LSM/8YSZ fabricated by screen printing and subsequent sintering at 1100 °C.

Fig. 2. SEM images of an ADP-LSM/8YSZ composite cathode on a dense 8YSZ electrolyte showing (a) a cross-section image, (b) an enlarged cross-section image, and (c) a top-view image.

Fig. 3. SEM images of an *in-situ* LSM/8YSZ composite cathode on a dense 8YSZ electrolyte showing (a) a cross-section image, (b) an enlarged cross-section image, and (c) a top-view image.

Fig. 4. SEM images of (a) ADP-LSM/8YSZ composite cathode sintered at 800 °C, (b) ADP-LSM/8YSZ composite cathode sintered at 1100 °C, and (c) *in-situ* LSM/8YSZ composite cathode sintered at 1100 °C.

Fig. 5. Impedance spectra of half cells fabricated using different processes: (a) ADP-LSM/8YSZ, (b) screen printed LSM/8YSZ sintered at 1100 °C, and (c) *in-situ* LSM/8YSZ cathode without heat treatment.

Fig. 6. Arrhenius plot of area specific resistances (ASRs) of the ADP-LSM/8YSZ composite cathode without sintering process and of the sintered LSM/8YSZ composite cathode.

Fig. 7. Structure of the developed metal-supported cell; Anode layer 1: 8YSZ-NiO anode, Anode layer 2: Ni-Cr-Fe powder-8YSZ-NiO functional anode.

Fig. 8. (a) Current (I) - voltage (V) - power density (P) curve of the ADP-LSM/8YSZ coated metal-supported cell measured at 800 °C, (b) impedance spectra (long term stability) of ADP-LSM/8YSZ coated metal-supported cell measured at 800 °C, and (c) impedance spectra (thermal cycles) of the ADP-LSM/8YSZ coated metal-supported cell between 800 °C and room temperature.

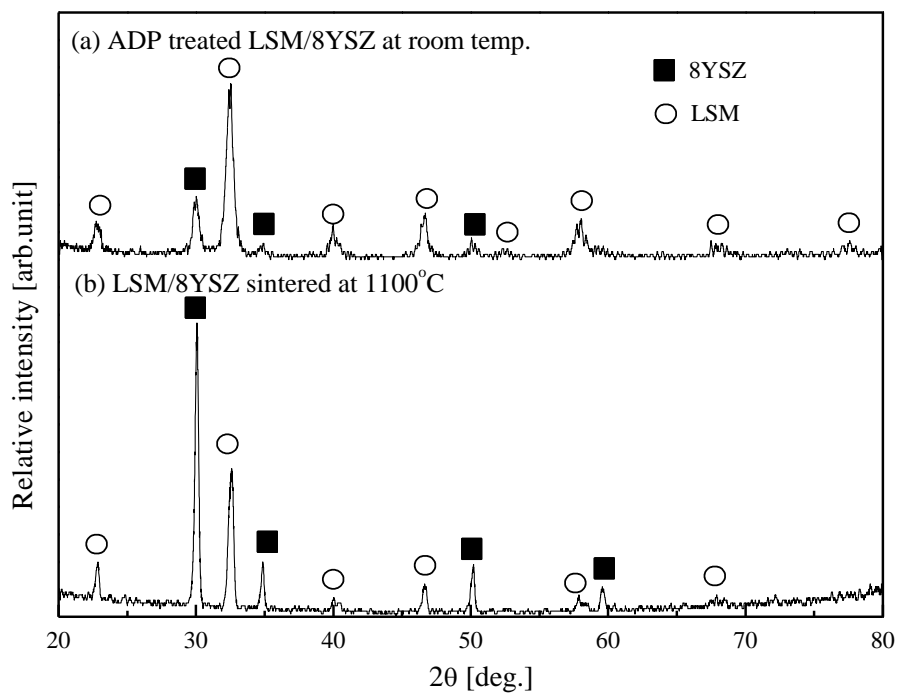


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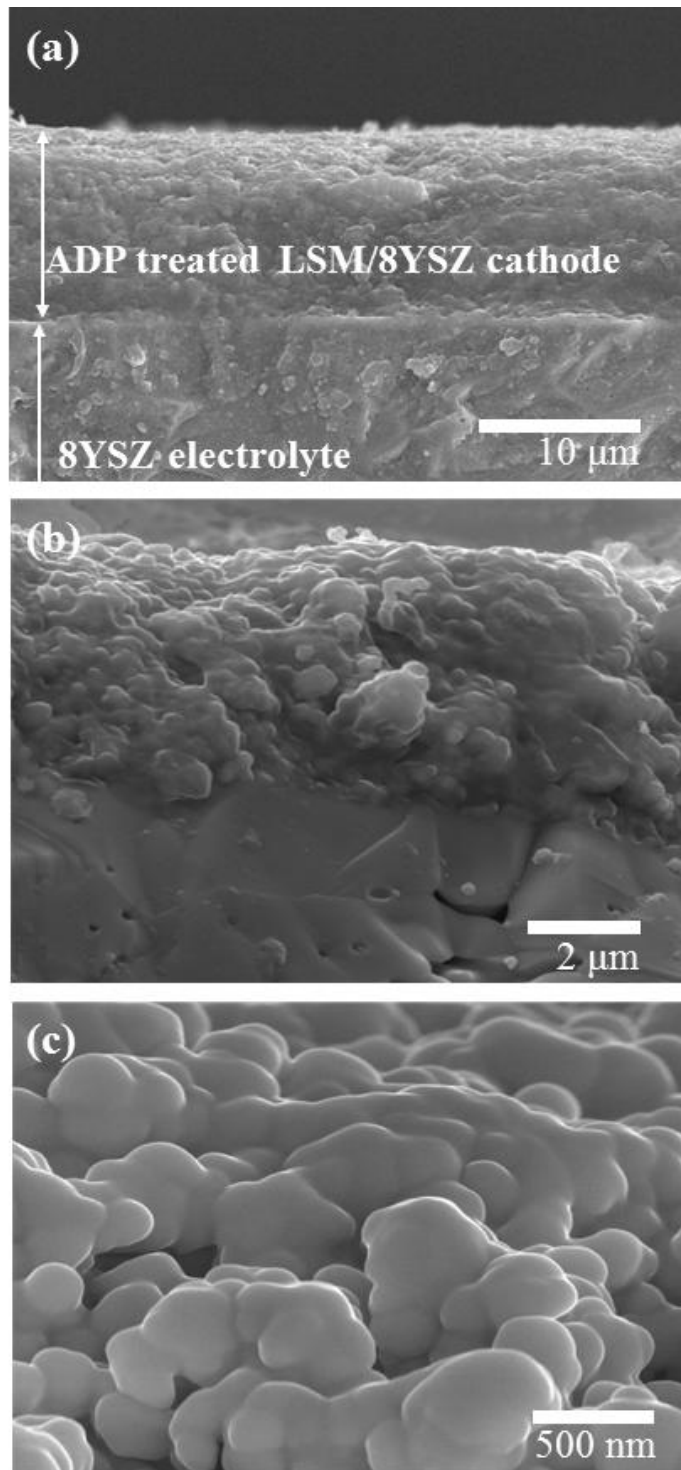


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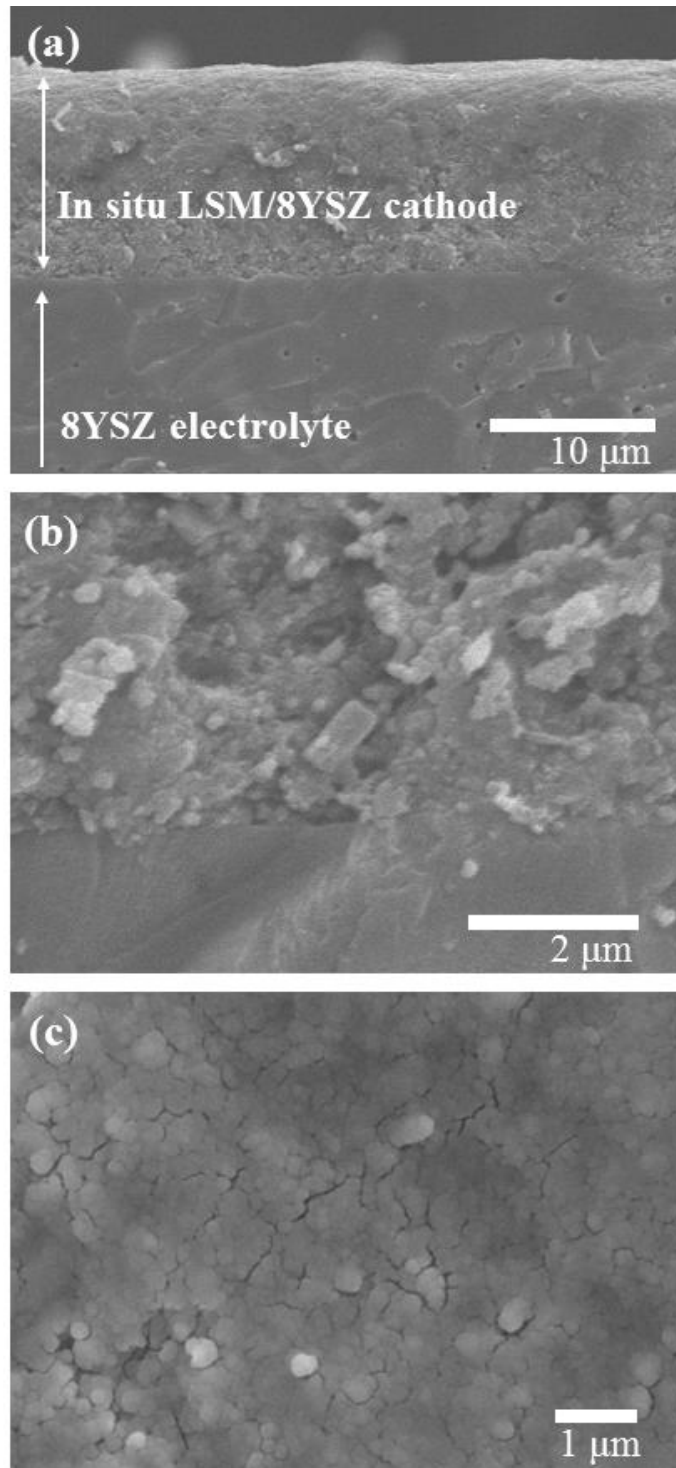


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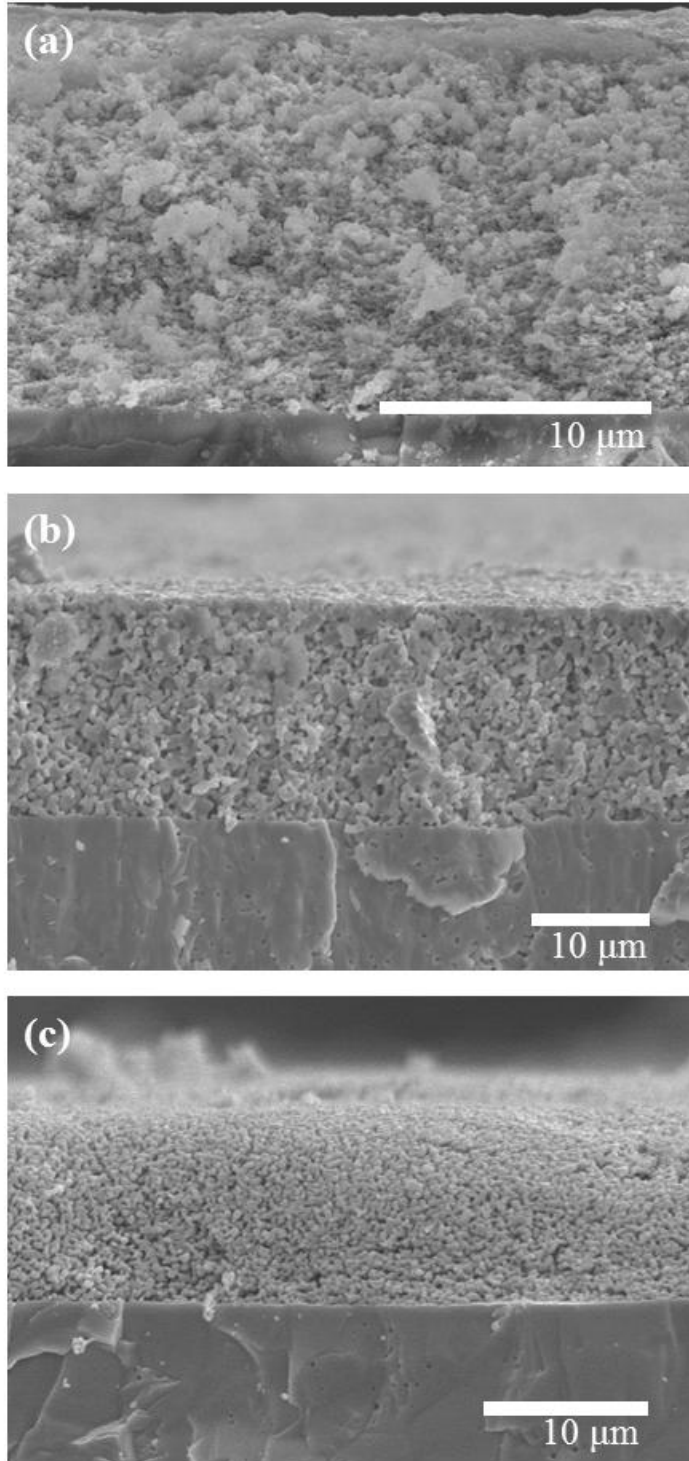


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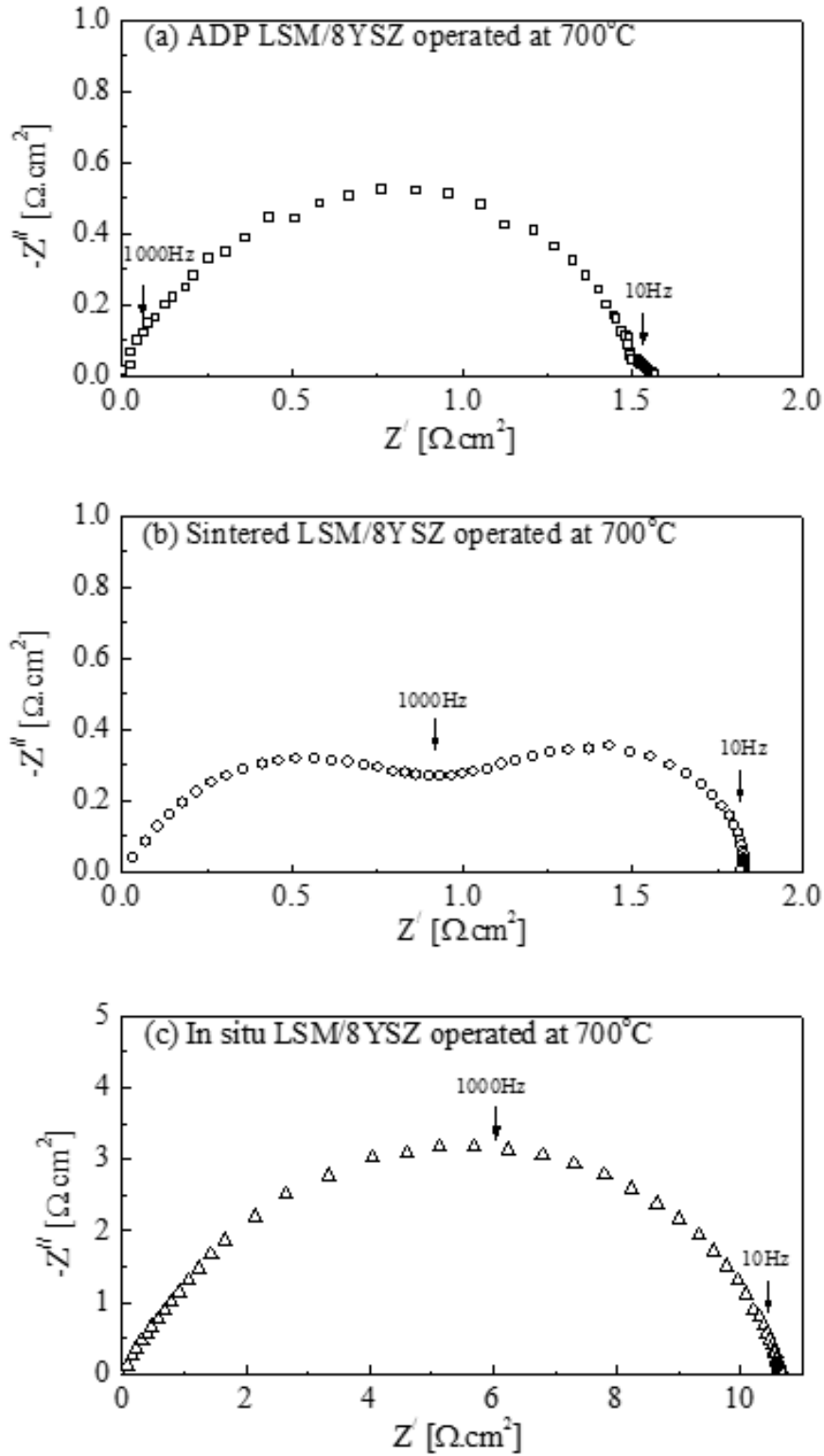


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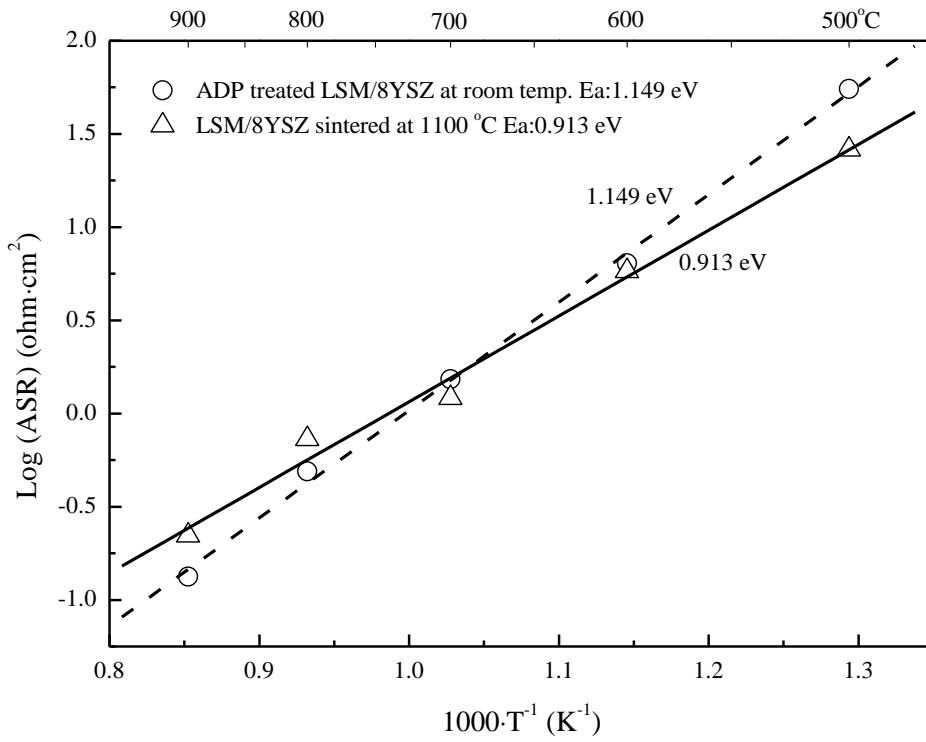


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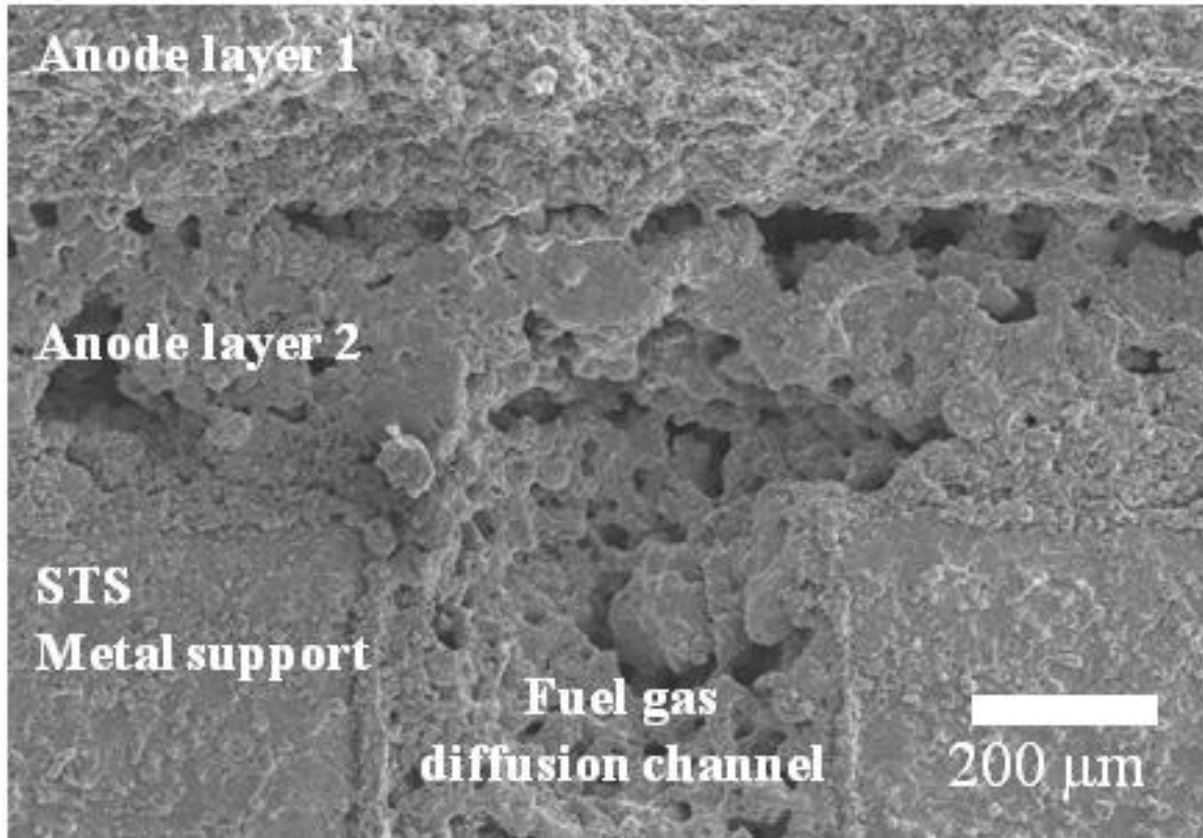


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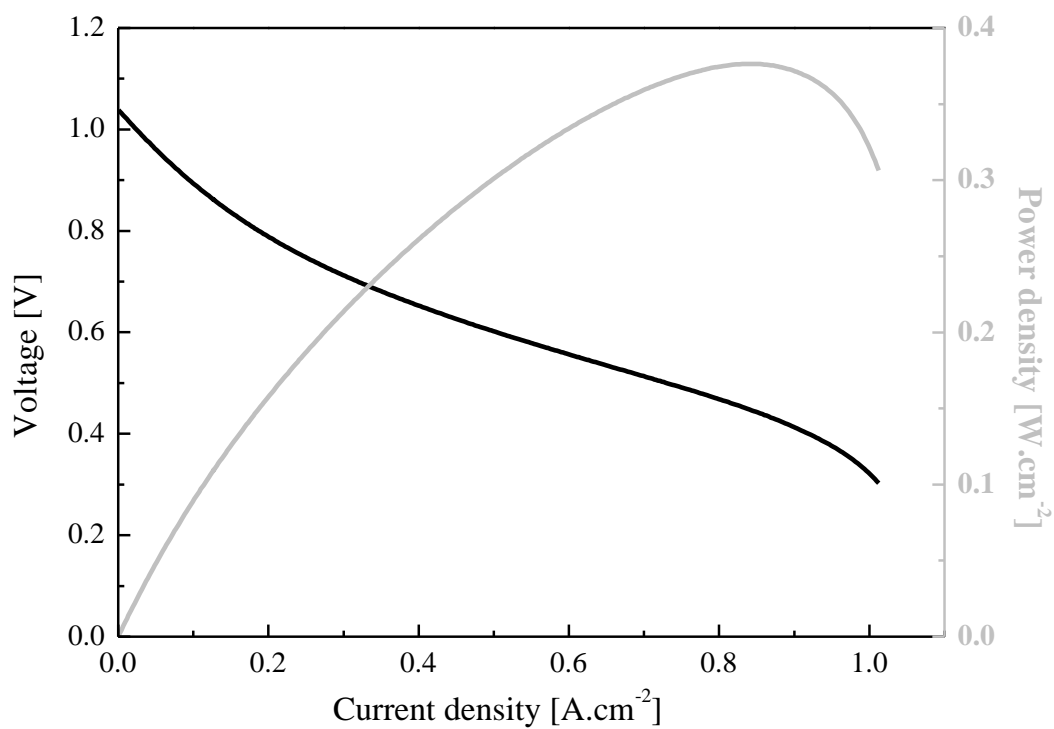


Fig. 8. (a)

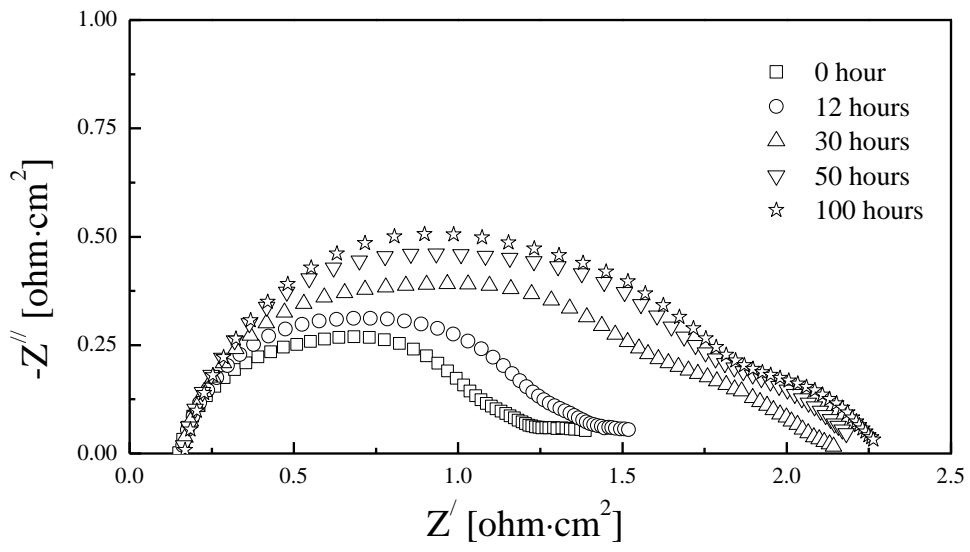


Fig. 8. (b)



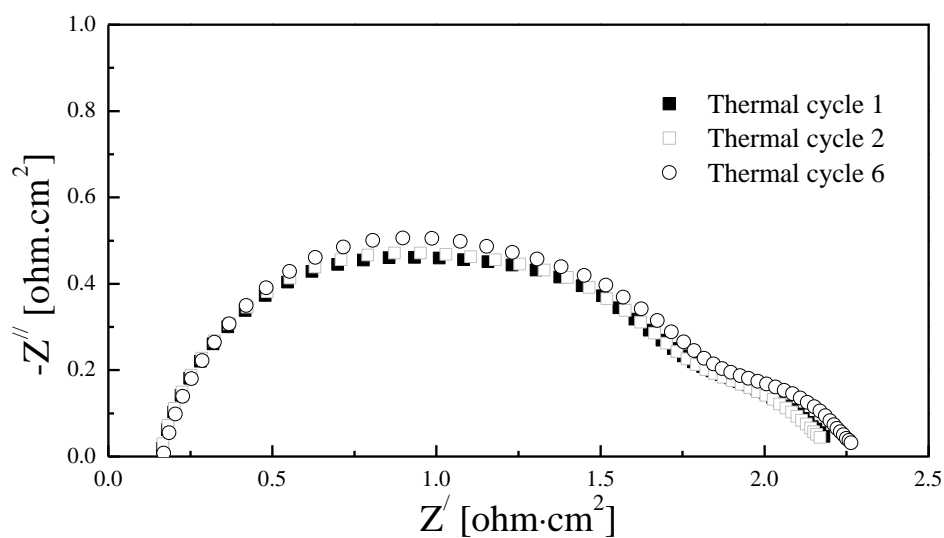


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