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# A Review: Electrochemical Performance of LSCF Composite Cathodes – Influence of Ceria Electrolyte and Metal Elements

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**Abstract** – *Solid oxide fuel cells (SOFC) are efficient and clean power generation devices. Low-temperature SOFC (LTSOFC) has been developed since high-temperature SOFC (HTSOFC) is not feasible to be commercialized due to cost. Lowering the operation temperature reduces its substantial performance resulting from cathode polarization resistance and overpotential of cathode. The development of composite cathodes regarding mixed ionic-electronic conductor (MIEC) and ceria-based materials for LTSOFC minimizes the problems significantly and leads to an increase in electrocatalytic activity for the occurrence of oxygen reduction reaction (ORR). Lanthanum-based materials such as lanthanum strontium cobalt ferrite ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$) have been discovered recently, which offer great compatibility with ceria-based electrolyte to be applied as composite cathode materials for LTSOFC. Cell performance at lower operating temperature can be maintained and further improved by enhancing the ORR. This paper reviews recent development of various ceria-based composite cathodes especially related to the ceria-carbonate composite electrolytes for LTSOFC. The influence of the addition of metallic elements such as silver (Ag), platinum (Pt) and palladium (Pd) towards the electrochemical properties and performance of LSCF composite cathodes are also discussed.* ***Copyright © 2014 Penerbit Akademia Baru - All rights reserved.***

**Keywords:** Ceria-based composite cathode, low-temperature solid oxide fuel cell (LTSOFC), oxygen reduction reaction (ORR), power density, polarization resistance

## 1.0 INTRODUCTION

Solid oxide fuel cells (SOFC) can generate power efficiently. For the past few decades, SOFC have emerged as one of the new alternative energy resources. SOFC offer highly efficient power generation, clean energy conversion with low emission and low noise. Because of its high operating temperature (800-1,000˚C), SOFC have several problems such as difficulties in construction, material and process selection, thermal expansion mismatch, lower durability and less opportunities to mass produce. Much research has been conducted to lower the SOFC's operating temperature, which aims to improve their life expectancy and reliability [1]. However, the reduction of operating temperature resulted in the inefficiency of the cathode elements as the increase of cathode polarization resistance, and consequently reduces substantial performance [2]. Rahman [3] reported that designing a high performance electrode with low polarization resistance such as using a composite cathode can potentially solve the problems. Recently, the addition of electrolyte material to the cathode material increased the triple phase (air/electrode/electrolyte) regions where electrochemical reactions occur. Mixed ionic and electronic conductors (MIEC) such as $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) is one of the candidates that have been applied with this approach since LSCF has been recognized as one of the promising cathodes for SOFC [3].

Previous research also shows that electrolyte materials such as samarium-doped ceria (SDC) and gadolinium-doped ceria (GDC) are mixed with LSCF to enhance the ionic conductivity of cathode [5,6,11,12,15,16]. Higher ionic conductivities of SDC and GDC have made them suitable to be combined with LSCF as they are ionic conductor materials. Moreover, LSCF and cerium electrolyte do not react with each other since they have the same thermal expansion coefficient (TEC). Thus, the problems of thermal mismatch can be avoided. This combination will enlarge the area of electrochemical activity and enhance the electrical conductivity of SOFC [4].

## 2.0 ELECTROCHEMICAL PROPERTIES AND PERFORMANCE OF LSCF COMPOSITE CATHODE

### 2.1 Influence of SDC ceria-electrolyte

For the past decade, much research has been conducted worldwide regarding the development of composite cathode in order to determine the most suitable materials as the cathodes for SOFC systems. Table 1 lists the electrochemical properties and performances of LSCF-SDC composite cathodes that have been developed. Zhang et al. [5] mixed LSCF powder with 30 wt.% SDC and 30 wt.% of Ag via grinding method. The implementation of composite cathode concept was able to overcome the polarization of cathode-electrolyte and from their experiment, the addition of SDC reduced the electrode-electrolyte interface polarization while the mixed conduction of LSCF enhanced cathode reactions.

**Table 1:** Properties of LSCF-SDC composite cathodes for intermediate to low temperature (800-400°C) SOFC

| Composite cathode | Polarization resistance, $R_P$ ($\Omega$ cm$^2$) | Activation energy, $E_a$ (eV) | Open-circuit voltage, OCV (V) | Max. Power density (mW/cm$^2$) | Reference |
|---|---|---|---|---|---|
| LSCF-30% SDC-Ag | - | - | 0.76 | 174 | [5] |
| LSCF-40% SDC | 0.29-0.18 | - | 0.89-0.92 | 317-348 | [6] |
| LSCF-50% SDC | 0.26-0.53 | - | - | - | [7] |
| LSCF-50% SDC | 0.23-0.07 | 1.49 | - | - | [8] |
| LSCF-30% SDCC | - | - | - | 75.33 | [9] |

Recently, Harris et al. [6] developed composite cathode consists of 60% LSCF and 40% SDC using axial-injection plasma spraying, in which the LSCF and SDC were expected to be more finely mixed. Nano-structured composite plasma spray feedstock was used to obtain fine mixing, in which each particle contained an agglomerate of LSCF and SDC phases. At 750°C, an open circuit voltage was 0.89-0.92V with power densities of 317-348 mW/cm$^2$. Their aim of getting finely mixed LSCF and SDC in order to obtain a large number of potential reaction sites for oxygen reduction reaction (ORR) was achieved, but in opposite, the low porosity of composite cathode became the limitation.

Another research regarding LSCF-SDC composite cathode was reported by Lee et al. [7] that focused on the size of composite powder. This is because in considering the performance and durability of cathode, the starting powder materials and processing conditions become the crucial aspects to be concern off [10]. Nanocrystalline materials with small particle size offer high surface area, which contributes to an increase of oxygen reduction reaction (ORR) at cathode, higher catalytic activity and directly enhanced the electrochemical performance of SOFCs [11]. Core-shell type of LSCF-SDC composite cathode was fabricated, in which the LSCF as the shell was attached on the SDC as the core. The size of SDC core was varied from 2 μm up to 500 nm in order to optimize the microstructure, specifically particle connectivity, pore size and pore distribution. The result for SDC core of 500 nm gave the lowest interfacial polarization resistance of 0.265 Ω $cm^2$ at 650˚C. The core-shell composite cathode developed an ideal microstructure when the LSCF particles completely surrounded the SDC core with no unattached particles, resulting in the improvement of phase contiguity and homogeneity, and maximized the density of composite cathode for triple phase boundary (TPB) [7].

Fu et al. [8] studied the electrochemical performance of composite cathode LSCF-SDC on the SDC electrolyte. The polarization resistance and activation energy were 0.23-0.07 $\Omega cm^2$ and 1.49 eV respectively. In their case study, the LSCF-SDC composite cathode sintered at 1,100˚C showed larger grain size, smaller porosity and good adhesion to electrolyte compared to electrodes sintered at 1,000˚C. Moreover, the addition of 50% SDC to LSCF was observed to suppress the grain growth and maintain the surface area from decreasing, and at the same time it filled the pore of the original LSCF microstructure, forming a rather dense microstructure. This condition resulting in increased TPB for ORR and decreased polarization resistance [8].

Recently, the development of composite cathode for LTSOFC utilizing carbonate salt composite electrodes has been embarked due to its high prospect in material properties and suitable characteristic to optimize the performance of cathode during low operating temperature [1,3,9,10]. Jarot et al. [9] produced composite cathode combination of LSCF with 30 wt.% SDC carbonate electrolyte and showed the maximum power density of 75.33 $mW/cm^2$ at 650˚C. The excitatory performance was from the result of the molten salt carbonate in the composite cathode that enhanced the ionic conductivity of the electrode. The polarization resistance between cathode-electrolyte interfaces was reduced. Moreover, as the composite cathode was constructed from electrolyte materials, they were chemically joined at the interfaces and extended the TPB to the whole cathode surface [7,11].

Rahman et al. [3] also furthered the study on the effects of carbonate composition in the LSCF cathode, calcination and sintering temperature, as well as morphology and microstructure of carbonates cathodes towards its electrochemical properties and the performance to be developed as potential cathodes for LTSOFC. Another aspect of composite carbonate cathode can be further studied, such as long-term stability of LTSOFC at a certain period of working time in order to make them stable for commercialization in the future.

## 2.2 Influence of ceria-electrolyte GDC

The development of LSCF composite cathode combined with ceria-electrolyte GDC has the same aim as combining LSCF with SDC. It is beneficial in reducing the polarization resistance of conventional pure cathode. The list of the electrochemical properties and performances of LSCF-SDC composite cathodes that have been developed are shown in

Table 2. Kim et al. [12] developed LSCF-GDC composite cathode using impregnation method with a ratio of 15 wt.% LSCF and 85 wt.% GDC with LSCF size less than 20 nm was impregnated on the GDC surface. The polarization resistance gave the result of 0.015-0.370 Ω cm$^2$ at 800-650˚C. Meanwhile, the composite cathode that was mixed only using a three roll mill with the composition of 50wt.% LSCF and 50 wt.% GDC showed 0.05-1.20 Ω cm$^2$ at 800-650˚C. The maximum power density for 15 wt.% LSCF and 85 wt.% GDC was in the range of 800-1,450 mW/cm$^2$ at 680-780˚C. The small portion of LSCF (15 wt.%) in GDC-supported LSCF may be effective to represent the good path of electrochemical reaction [12].

**Table 2:** Properties of LSCF-GDC composite cathodes for intermediate to low temperature (800-500˚C) SOFC

| Composite Cathode | Polarization resistance, $R_P$ (Ω cm$^2$) | Activation energy, $E_a$ (eV) | Open-circuit voltage, OCV (V) | Max. Power density (mW/cm$^2$) | Reference |
|---|---|---|---|---|---|
| LSCF-85% GDC | 0.015-0.370 | - | - | 800-1450 | [12] |
| LSCF-50% GDC | 0.33-0.01 | 1.65 | - | - | [13] |
| LSCF-50% GDC | 0.19-0.03 | 1.20 | - | - | [14] |
| LSCF-50% GDC | - | - | ~0.88 | 625 | [15] |
| LSCF-60% SDC | 0.27 | - | - | 562-139 | [16] |
| LSCF-YSZ-GDC | 0.075 | - | - | 500-1240 | [17] |

A study regarding LSCF-GDC composite cathode of the same composition was reported by Murray et al. [13] and Wang and Mogensen [14], where the composition of LSCF over GDC was set to 50:50 (in weight %). In both experiments, LSCF-GDC composite cathode was applied to yttria-stabilized zirconia (YSZ) electrolyte. Wang and Mogensen [14] achieved low polarization resistance mainly due to the nanosized particles they used with the size of 100 nm, which significantly improved the TPB in cathode as compared to Murray et al. [13], where they used particle with size in the range of 100-200 nm, which was slightly larger.

Although the composite cathode from both experiments has the same composition, but they differ in polarization resistance and activation energy. The difference in the significant result is due to the morphology of the resulting microstructure of the composite cathodes. Another factor affecting the result is the good adhesion between cathode and electrolyte, which decreases the polarization at the interface between the two elements [14]. In the experiment conducted by Leng et al. [15], the composition of LSCF-GDC was also set to 50:50, but other factors influenced the result. The factor was the powder making process itself, which was roll milling rather than the size of particles used. Moreover, the use of GDC dense thin film electrolyte ~10 μm was the supporting factor to achieve a satisfactory performance. The maximum power density recorded was 625 mW/cm$^2$ with open circuit voltage ~0.88V [15].

Leng et al. [16] reported that the addition of GDC to LSCF cathode further reduced the polarization resistance compared to pure LSCF cathode. The polarization resistance of pure LSCF cathode and LSCF-SDC (40:60 wt.%) composite cathode were 1.20 Ω cm$^2$ and 0.17 Ω cm$^2$ at 600˚C respectively. For a single fuel cell consisted of LSCF-GDC (40:60 wt.%) as the cathode, GDC film as the electrolyte and Ni-GDC as the anode gave the polarization resistance lower than 0.27 Ω cm$^2$ at 600˚C and the maximum power density of 562-139 mW/cm$^2$ at 650-500˚C. Qiang et al. (2007) also reported that the addition of GDC to LSCF as the composite cathode reduced the polarization resistance. 40 wt.% of GDC was added to LSCF, which resulted in lower polarization resistance of 0.07-0.22 Ω cm$^2$ at 800-700˚C and its activation energy was between 0.86 and 1.10 eV.

Recently, Ko et al. [15,17] introduced dual composite cathode based on LSCF-YSZ-GDC, in which LSCF and YSZ phases were placed on the GDC core particles. LSCF-YSZ-GDC based cathode was introduced after LSM-YSZ-GDC because of its high catalytic activity. The role of YSZ material added on LSCF-GDC cathode was to increase the interfacial coherence and interconnectivity with electrolyte. The outstanding results of the maximum power density was achieved, which were 500 and 1,240 mW/cm$^2$ at 650 and 800˚C respectively. On the other hand, the polarization resistance was 0.075 Ω cm$^2$ at 800˚C.

## 2.3 Influence of silver (Ag)

Research on SOFC convinced that composite cathodes such as LSCF-SDC and LSCF-GDC can be improved by the addition of metallic elements such as silver (Ag), platinum (Pt) and palladium (Pd) [2]. In order to enhance the conductivity of SOFC, Ag is added based on the following considerations: (1) silver is relatively low in cost [19], (2) silver is a fine electronic conductor and it should improve the electronic conduction of the cathode, (3) the melting point of Ag is 961˚C and it can be used for the purpose of low-temperature SOFCs (LT-SOFCs)(400-600˚C), and (4) as its melting point is low, it might aid the sintering behaviour of the cathode at lower temperature [18].

There are numerous researches regarding the addition of Ag to cathode in order to boost up the conductivity of SOFC. Sakito et al. [19] conducted experiments regarding porous LSCF that was infiltrated with $AgNO_3$ solutions in citric acid and ethylene glycol using polymerized complex method. The polymerized complex method is one of the effective techniques to obtain homogenous distribution of Ag over the LSCF matrix. They fabricated two types of SOFC with the LSCF-Ag cathode, Ni-YSZ/YSZ/LSCF-Ag and Ni-GDC/GDC/LSCF-Ag, and the operating temperature examined was in the range of 530-730˚C. The infiltration of about 18 wt.% Ag into porous LSCF resulted in enhancement about 50% of the power density. The maximum power density of Ni-YSZ/YSZ/LSCF-Ag was enhanced from 0.16 W/cm$^2$ to 0.25 W/cm$^2$ at 630˚C while Ni-GDC/GDC/LSCF-Ag gave the maximum power density of 0.415 W/cm$^2$ at 530˚C. The enhancement in power density was significantly resulted from the addition of Ag.

The research on LT-SOFC reported by Fan et al., [2] again stated that single cells with cathode containing Ag present better electrochemical performances than those of Ag-free cell. $Pr_2NiO_4$ was dispersed onto $AgNO_3$ and the weight percentage of Ag to $Pr_2NiO_4$ was fixed at 10 wt.% in obtaining $Pr_2NiO_4$-Ag composite cathode They achieved the maximum power density of 696 mW/cm$^2$ at 600˚C using $Pr_2NiO_4$-Ag composite cathode. In another experiment conducted by Huang et al. [20], the infiltration of 0.3 mg/cm$^2$ Ag into composite cathode LNF-SDC improved the electronic conductivity and ORR, and thereby reduced the area specific resistance (ASR) on the cathode. The ASR value of the LNF-SDC-Ag composite cathode was 0.46 Ω cm$^2$, approximately five times lower than that those obtained without silver-infiltrated cathode at the same operating temperature of 650˚C.

Zhang et al. [18] reported that the addition of Ag in composite cathode improved the cathode performance compared to the composite cathode without Ag. Based on their experiment, which focused on IT-SOFC, where a single cell with NiO-SDC anode, SDC electrolyte and LSCF-SDC with and without the addition of Ag as the cathode (NiO-SDC/SDC/LSCF-SDC-Ag and NiO-SDC/SDC/LSCF-SDC) was fabricated. Ag was added directly as the metal powder to the composite cathode with Ag content of 30 wt.%. XRD analysis showed that the addition of Ag in the composite cathode resulted in no chemical reaction between other components. From the SEM images shown in Figure 1(a), the interface section between composite cathodes LSCF-SDC-Ag with SDC electrolyte gave much better adhesion with no gap between the cathode and electrolyte. Compared to the composite cathode without Ag in Figure 1(b), there was a contradicted line at the LSCF-SDC-electrolyte interface, and the adhesion of the cathode on the electrolyte was poor as the two sections (cathode and electrolyte) were separated at many parts around the interface area, which caused the polarization loss at the interface. SEM images also showed that the microstructure of the cathode with Ag was more uniform than that of without Ag, which contributed to the ability of enhancing sintering behaviour of Ag since Ag acted as a sintering aid in this case.

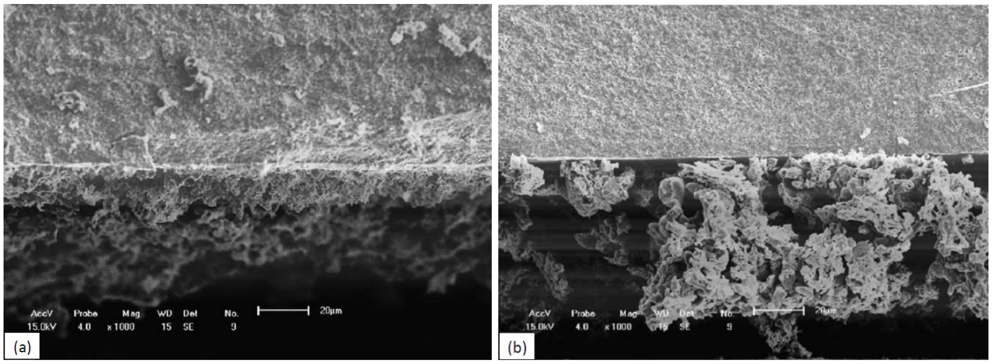

**Figure 1:** SEM images of the cathode-electrolyte interfaces of LSCF-SDC-Ag/SDC (a) and LSCF-SDC/SDC (b) [5]

The performance of SOFC with LSCF-SDC-Ag cathodes at 650˚C and 750˚C showed the maximum power density of 173.8 mW/cm$^2$, which was higher than SOFC, with LSCF-SDC cathode gave the value of 124.6 mW/cm$^2$. They concluded that Ag played the role as a fine conductor, resulting in the enhancement of the electronic conductivity of cathodes and worked as a sintering aid that contributed to the microstructure improvements in cathodes.

## 2.4 Influence of palladium (Pd)

Only few studies have been identified regarding the addition of Pd in LSCF composite cathode. Sahibzada et al. (1998) reported that porous LSCF cathode with SDC electrolyte were impregnated with a small amount of Pd. Ac impedance spectroscopy gave the result that the addition of Pd to porous LSCF cathodes resulted in the reduction of cathodic impedance 3-4 times in the temperature range of 400-700˚C. The overall cell resistance of single-cell SOFCs was found to decrease by 15% at 650˚C and 40% at 550˚C with the addition of Pd. From this result, they claimed that the SOFCs became more limited as observed by the performance of the cathode at lower temperature with the presence of Pd [21].

Haanappel et al. [22] also conducted experiments on the addition of several types of Pd on SOFC cathodes. Two routes were used to add Pd metals. The first one was infiltration of the cathode with Pd nitrate ($Pd(NO_3)_2$) solution and the other one was by mixing $La_{0.65}Sr_{0.30}MnO_3$(LSM) and YSZ cathode with Pd black and Pd on activated carbon (Pd-C) to obtain the Pd amount of 2 and 0.1 wt.% respectively. Two types of cells without the addition of Pd, conventional cells and Pd-free cells were prepared as reference cells. The results showed no catalytic enhancement with single cells containing Pd black, whereas Pd-C containing cells showed an improvement in electrochemical performance in particular at temperatures 750 and 800˚C. Infiltration of the cathode with Pd solution also did not result in a positive effect as well [22].

### 2.5 Influence of platinum (Pt)

Similar to Pd, there are a small number of studies about the addition of Pt on composite cathode that can be found. The addition of Pt in composite cathode gives no significant enhancement in electrical conductivity of SOFC. Besides, Pt involves high cost. The effect on the addition of Pt in LSCF was studied by Sasaki et al. [23]. The obtained cathode activity was only 0.5S/cm$^2$ at 700˚C. The cathode had poor porosity and narrow surface area due to high sintering temperature applied to electrode, but Pt may still enhance the sintering of LSCF. The use of Pt was also reported by Haanappel et al. [22]. Pt was added by mixing $La_{0.65}Sr_{0.30}MnO_3$ (LSM) and YSZ cathode with Pt black. There was no electrocatalytic enhancement with the addition of Pt. This was due to the high melting point of Pt (1768˚C), which did not induce the sintering process, led to weak bonding and compatibility of LSM-Pt cathode and gave no significant influence on the electrochemical performance. Therefore, the study about sintering temperature should be concerned regarding the case study involving Pt.

## 3.0 SUMMARY

This paper reviews the influence of ceria electrolytes and metallic elements on the electrochemical performance of LSCF composite cathode. From previous researches, it has been shown that SDC and GDC give a significant improvement in electrochemical performance of LSCF composite cathodes for LTSOFC. Among various candidates of metallic elements, Ag has been widely studied for its superior electronic conductivity, high catalytic activity and low in cost compared to Pd and Pt. The addition of ceria electrolyte (SDC and GDC) and Ag significantly improves the electrochemical performance of composite cathode and accommodate the polarization resistance and overpotential of cathode.

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