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Review:

Development of lanthanum strontium cobalt ferrite composite cathodes for intermediate- to low-temperature solid oxide fuel cells*

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Abstract: Solid oxide fuel cells (SOFCs) offer high energy conversion, low noise, low pollutant emission, and low processing cost. Despite many advantages, SOFCs face a major challenge in competing with other types of fuel cells because of their high operating temperature. The necessity to reduce the operational temperature of SOFCs has led to the development of research into the materials and fabrication technology of fuel cells. The use of composite cathodes significantly reduces the cathode polarization resistance and expands the triple phase boundary area available for oxygen reduction. Powder preparation and composite cathode fabrication also affect the overall performance of composite cathodes and fuel cells. Among many types of cathode materials, lanthanum-based materials such as lanthanum strontium cobalt ferrite ($La_{1-x}Sr_xCo_{1-y}Fe_yO_{3-\delta}$) have recently been discovered to offer great compatibility with ceria-based electrolytes in performing as composite cathode materials for intermediate- to low-temperature SOFCs (IT-LTSOFCs). This paper reviews various ceria-based composite cathodes for IT-LTSOFCs and focuses on the aspects of progress and challenges in materials technology.

Key words: Ceria-based composite cathode, Lanthanum strontium cobalt ferrite, Intermediate- to low-temperature solid oxide fuel cells (IT-LTSOFCs)
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1 Introduction

Solid oxide fuel cells (SOFCs) available today are similar to other fuel cell concepts because they generate electricity directly from the reaction between fuels and an electrode (Huang *et al.*, 2011). This type of fuel cell is called an SOFC because of the solid electrolyte used, which is commonly made from ceramics (Menzler and Tietz, 2010). An SOFC is an environmental-friendly device and is preferentially used for its high efficiency in energy conversion (Timakul, 2004). Similar to other types of fuel cells, hydrogen gas is supplied at the anode and oxygen gas at the cathode side. Fig. 1 shows a schematic of an SOFC. Electrochemical reactions occur at the triple-phase boundary (TPB), where the anode/cathode meets the electrolyte (Timurkultuk, 2007). Earlier studies on SOFCs were conducted at high operating

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temperatures exceeding 1000 °C (Minh, 2004). Reducing the operating temperature to below 800 °C can save more energy and therefore has become the new aim in SOFC development. In addition, most materials that can withstand high temperatures tend to be more expensive. Thus, the selection of new materials to operate SOFCs in a lower temperature zone will proportionally cut the cost (Steven, 2005). Each SOFC component (anode, cathode, and electrolyte) requires different types of materials; considering the type of material will affect the overall cell performance. The selection of a suitable cathode material for an intermediate- to low-temperature SOFC (IT-LTSOFC) can also enlarge the TPB area (Viswanathan and Scibioh, 2007). The TPB between a cathode and an electrolyte helps determine the effectiveness of an electrochemical reaction. Therefore, studying cathode component materials offering higher electron conductivity is important.

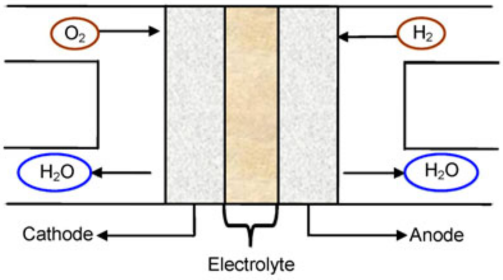

Fig. 1 Schematic of a solid oxide fuel cell

SOFC cathode materials must have certain characteristics to help develop and maintain cell performance. Lanthanum strontium manganite (LSM) was first introduced as an SOFC cathode material for its good electronic conductivity (Kuharuangrong, 2004). However, this is only applicable when LSM is operated at high temperatures. Using IT-LTSOFCs requires cathode materials that have good electronic and ionic conductivities at temperatures below 800 °C. Thus, mixed ionic-electronic conductors (MIECs) such as perovskite-based materials were suggested. MIECs may help develop larger TPBs and increase the performance of IT-LTSOFCs (Fergus *et al.*, 2009).

LSCF is a perovskite-based material suitable for IT-LTSOFC applications (Mai *et al.*, 2005). A composite cathode is introduced to improve cathodic polarization. Electrolyte materials such as samarium-doped ceria (SDC) and gadolinium-doped ceria (GDC) are mixed with LSCF to enhance the ionic conductivity of the cathode (Zhang *et al.*, 2005; Huang and Chou, 2009). Fig. 2 illustrates the mechanism of electronic and ionic conduction in the composite cathode. The electrolyte materials in the composite cathode act as an ionic conductive phase. The metal-oxide phase in the cathode serves as an electronic conductive phase. The construction of this dual-phase composite allows both electrons (e^-) and oxygen ions (O^{2-}) to be transferred simultaneously through the cathode.

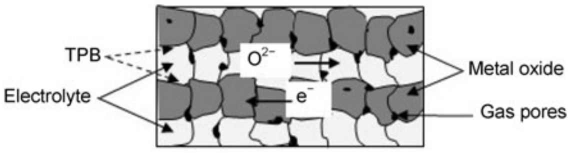

Fig. 2 Simplified schematic of the mechanism of electronic and ionic conductions in a composite cathode

2 Fabrication of composite cathodes

2.1 Composite cathode powder preparation

The Pechini and sol-gel methods are commonly used for the preparation of LSCF. The Pechini method offers various types of preparation processes, including the glycine-nitrate process. Known as the self-sustaining combustion process, the glycine-nitrate process includes glycine as fuel and metal nitrates as oxidizers. Huang and Chou (2009; 2010) used the glycine-nitrate process to prepare LSCF powders. Both studies used industrial-grade metal nitrates $La(NO_3)_3{\cdot}6H_2O$, $Sr(NO_3)_2$, $Co(NO_3)_2{\cdot}6H_2O$, and $Fe(NO_3)_3{\cdot}9H_2O$. This composition and glycine were dissolved in de-ionized water separately before being mixed together with a glycine-to-nitrate (NO_3) ratio of 1:0.8. The mixture was heated to 110 °C and stirred until combustion occurred. The LSCF powders obtained after the grinding process underwent two stages of calcinations at a heating rate of 5 °C/min. The LSCF powders were calcined at 500 °C (held for 2 h), heated to 900 °C (held for 4 h), then slowly cooled down to room temperature (Huang and Chou, 2009). In addition to being easy and speedy, the glycine-nitrate process offers great compositional uniformity and allows nano-sized particles to be obtained (Thomas *et al.*, 2009). Some changes have been made to the process to fulfill various research needs. Instead of dissolving the glycine in de-ionized water, glycine in solid state has been added to an aqueous nitrate solution (Leng *et al.*, 2008). Using this approach, the precursor solutions need to be heated first to eliminate excess water, after which they form a viscous liquid. Black ash produced after rapid self combustion is calcined at 800 °C for 2 h holding time. Calcinations are extremely important in removing carbon residue and obtaining a better LSCF crystalline perovskite structure (Leng *et al.*, 2008). Despite the fact that calcinations in the glycine-nitrate process are important in producing ample-sized particles, replacing this process with the sol-gel method allows the control of more parameters to obtain a fine particle size.

In terms of particle size obtained after calcinations, the sol-gel method provides researchers another alternative in preparing LSCF. With the sol-gel method, the final particle size obtained does not depend only on the calcinations. During the synthesis of the sol itself, parameters such as porosity and particle size can be controlled through process conditions. Process conditions reflect the external parameters (concentration precursor, solvent, and temperature) selected during synthesis. LSCF ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$) was produced using the sol-gel method (Ghouse *et al.*, 2010). The solution of various metal nitrates was mixed with citric acid, ammonia, and ethylene glycol and then heated and stirred until a chocolate-colored gel was observed.

The heating process was continued until the gel was burnt completely and became ash. The ash was then calcined at 650 °C for 6 h. Using this method, LSCF powders with a particle size of 50 nm to 200 nm have been produced (Ghouse *et al.*, 2010). Ammonia and ethylene glycol were used together with metal nitrates but ethanol can also be used for the sol-gel method. Ethanol (100 ml) is dissolved with metal nitrates and then refluxed and stirred for 4 h in a water bath. After cooling to room temperature, the mixture is concentrated to about 1/3 of the original volume. The evaporation of ethanol for 4 h brings the mixture to the desired concentration, thus obtaining the sol precursors. The sol precursors are directly applied onto electrolyte components through screen printing (Liu *et al.*, 2006). In both studies (Liu *et al.*, 2006; Ghouse *et al.*, 2010), the sol-gel method required more steps compared with the glycine-nitrate process. Both processes have unique advantages. The glycine-nitrate process is simpler and consumes less time, whereas the sol-gel method opens up more options for controlling the synthesis. LSCF powders can be prepared successfully using either method. Table 1 summarizes the characteristics of the two types of LSCF powder preparation method.

Other materials such as GDC or SDC can be combined with LSCF to produce composite cathodes. Ball milling has always been used to manufacture composite powders. In this case, GDC or SDC is milled together with LSCF along with a medium such as propanol. The mixture is milled for about 24 h, then dried (Leng *et al.*, 2008). Subsequently, calcinations take place to refine the size of composite cathode particles. Although the process is helpful in

Table 1 Preparation methods for LSCF powder

Item	Glycine-nitrates method	Sol-gel method
Fuels	Glycine	More options for fuels such as a combination of citric acid, ammonia, and ethylene glycol, or ethanol only
Calcination	Two stages	Once
Particles-size controlling parameters	Calcinations	Precursor concentration, solvent, and temperature
Advantages	Compositional uniformity, nanosized particles, and simple and not time-consuming	More parameter options for controlling the resulting powders (precursor concentration, solvent, and temperature); suitable method for particle size studies; more solution options for preparing the mixture (ammonia, ethylene glycol, or ethanol)
Disadvantages	Limited number of operational parameter (calcinations) that can be controlled	Wide operational parameter scope because of more required steps

achieving a good particle size for the composite cathode, sizing still depends on the raw powder preparation. LSCF and ceria-based powders should be prepared well before both are mixed to form composite powders.

2.2 Fabrication of composite cathode components

LSCF usually does not alone comprise the cathode; it must be combined with electrolyte powders to obtain a composite cathode. Due to the Fe^{3+} doping of the LSCo cathode material, a lower LSCF thermal expansion coefficient (TEC) is produced. However, this process decreases the ionic conductivity between LSCF cathodes and the electrolyte (Fu *et al.*, 2007). Lowering the operating temperature leads to a higher polarization resistance of the LSCF cathode, requiring the addition of an ionically conducting second phase that can be obtained from a technical oxide ceramic such as ceria. GDC and SDC are widely used to obtain higher ionic conductivity of electrolytes. Hence, amounts of these materials were mixed together with LSCF to achieve better ionic conductivity for cathodes at a lower temperature (Fu *et al.*, 2007). In addition, GDC and SDC are usually used for IT-SOFCs. Composites of LSCF with yttria-stabilized zirconia (YSZ) were found to perform better than others for high operating-temperature SOFCs. However, the combination of LFSC and YSZ is considered a mismatch because the purpose of using LSCF is to reduce the operating temperature. Thus, the use of YSZ is excluded. Nevertheless, two options can be taken to use YSZ at lower temperatures: an applied interlayer that contains doped ceria, or an advanced fabrication method such as applied impregnation of LSCF slurry into presintered YSZ on YSZ electrolyte (Chen *et al.*, 2008). The simpler method is always preferred by researchers. Thus, GDC or SDC combined with LSCF to produce composite cathodes for IT-LTSOFCs are the prevailing methods. Several fabrication methods such as dry-pressing, screen printing, tape casting, slurry painting, and deposition methods are used to produce composite cathodes (Tietz *et al.*, 2006; Nie *et al.*, 2010a). Factors such as cost, handling difficulty, and time are considered before undertaking production (William, 2001).

The fabrication of composite cathodes such as LSCF-GDC/SDC is similar to that of producing cathodes containing LSCF alone. Screen printing, sputtering, tape casting, and electrophoretic deposition (EPD) are methods that can be applied. Tietz *et al.* (2006) reported fabrication of a 45 μm thick LSCF cathode using screen printing. Nielsen *et al.* (2011) also used screen printing to fabricate LSCF-CGO composite cathodes to study the impedance of the porous composite cathode component. The thickness of the composite cathode was found to be a factor influencing resistance and cathode polarization.

Reducing the cathode thickness was taken into consideration to improve cathode performance. Cathode thin films can also be fabricated using tape casting, as demonstrated by Nie *et al.* (2010b) in producing a porous LSCF cathode. Layers of LSCF cathode were fabricated to reduce the interfacial resistance of the cathode. Three layers of LSCF cathode with an overall thickness of 60 μm were obtained. Sputtering is another method for fabricating thin layers or LSCF cathode films. Radio frequency (RF)-magnetron sputtering was used to develop an LSCF cathode film with a thickness of about 200 nm (Lee *et al.*, 2009).

Whether the product is an LSCF cathode or an LSCF composite cathode, controlling the thickness of the cathode is important. EPD has been considered a potential method for producing a thin layer or film of SOFC cathode component because of its simple application and low cost (Besra *et al.*, 2010; Li *et al.*, 2011; Rahman *et al.*, 2011). Parameters such as deposition time, pH of the suspension, type of dispersion agent, and suspension concentration need to be studied to ensure enough LSCF is deposited onto the electrode. All these fabrication methods have been used to fabricate LSCF cathodes and LSCF composite cathodes. Although the type of fabrication method used affects the performance of the cells, selecting suitable materials to fabricate the other components (anode and electrolyte) is also important.

3 Material selection of other components for use together with LSCF composite cathodes in IT-LTSOFCs

Numerous materials have been selected to match the LSCF cathode. However, selection is clearly dominated by two main types of material, zirconia-based and ceria-based materials. Zirconia-based materials commonly used in SOFC applications are YSZ and scandia-stabilized zirconia (SSZ). For ceria-based materials, GDC and SDC have been used in most experiments. Each material is selected based on certain requirements. A general description of materials and methods used for the combination of LSCF with each of these base materials will be discussed in this section.

3.1 Zirconia-based materials (YSZ and SSZ)

In the early stages of using LSCF as a cathode material, YSZ was always used to complete the cell. High-temperature SOFCs have been produced using YSZ as one of the material options. YSZ is a commonly used electrolyte material because of its excellent oxygen ion conductivity and chemical stability properties at high temperatures (Kawada and Yokokawa, 1997). LSM with YSZ is the most commonly used cathode material. Given the requirement to reduce the temperature, LSCF was introduced as a cathode (Dutta *et al.*, 2009). However, early in IT-LTSOFC development, proposals using YSZ together with LSCF were common because researchers were more familiar with these materials. Combining YSZ with LSM produced good results in terms of the electrochemical performance of the cell operating at high temperatures (Jiang, 2002). Introducing LSCF as an alternative material for IT-LTSOFC required ample research to confirm the good results at lower temperatures. Using LCSF in lower-temperature applications slightly decreases the ionic conductivity of YSZ, thus affecting cell electrochemical performance.

Dutta *et al.* (2009) added Gd-doped ceria (GCO) as an interlayer between the LSFC cathode and YSZ electrolyte to increase the ionic conductivity of the cell. Furthermore, the TEC of LSCF is generally higher than 17×10^{-6} K^{-1}, whereas that of YSZ (about $10.8\times17\times10^{-6}$ K^{-1}) is considerably smaller compared with LSCF (Mai *et al.*, 2006). This difference will cause stresses between the electrolyte and cathode layers. Thus, the GCO layer is required as a buffer layer to avoid stress formation at the layer interface. DiGiuseppe and Li (2011) reported that the buffer layer can be used as an interlayer between the two components to avoid the occurrence of undesired reactions. They used doped ceria as a buffer layer material. Anode-supported NiO-YSZ SOFC was fabricated using LSCF as a cathode, and then testing was conducted at different temperatures to obtain parameters such as the voltage, current, and impedance for the cells. Overall, using YSZ together with LSCF will require the addition of an interlayer. Irrespective of increasing ionic conductivity or acting as a protector of the interface, using YSZ with a buffer layer will enhance electrochemical performance of the cell. Thus, using an LSCF cathode with YSZ at lower temperatures is not simple because of the addition of an interlayer, which will add to the parameters to be controlled in obtaining better ionic conductivity in the cells.

Ionic conductivity determines the efficiency of the cells. Previously, a combination of a Ni-YSZ anode with a YSZ electrolyte produced high ionic conductivity. Carbon was formed after operating at high temperature for a certain time using hydrocarbon fuels. The carbon was deposited on the Ni-YSZ anode, which decreased fuel utilization and led to anode deactivation (Ke *et al.*, 2006). However, for the Ni-SSZ electrode, carbon deposition decreased with increasing temperature. According to a thermodynamic theory, carbon formation is directly proportional to the temperature. However, adsorbed H_2O (H_2O_{ad}) enhanced the effect from anodic current. The enhanced effect of Ni-SSZ is stronger than that of Ni-YSZ. Thus, at higher temperatures, Ni-SSZ performs better than Ni-YSZ (Ke *et al.*, 2006). This phenomenon increases the likelihood of SSZ being selected as a material, considering its capability to operate at intermediate temperatures, and rivaling YSZ even at higher temperatures.

With regards to lowering the temperature of SOFCs to an intermediate range, new materials have been suggested aside from the proposed interlayer applied in between cell components. In lowering the temperature, the ionic conductivity must remain high. SSZ is selected not only for this reason, but also because of its chemical stability during the oxidation and reduction processes (Sillassen *et al.*, 2010). In addition, Sc^{3+} has an ionic radius of 0.087 nm which is similar to that of Zr^{4+}. The closer the size of the dopant is to Zr^{4+}, the better. This small gap will affect electrical transport, thus influencing ionic conductivity (Sillassen *et al.*, 2010). Even though SSZ performed very well in substituting YSZ, the operating temperature remains inflexible in the intermediate region. Ceria-based materials were suggested to optimize cell performance at lower temperatures. A ceria-based electrode offers many advantages compared with the zirconia-based YSZ electrode given its higher conductivity at lower operating temperatures and compatibility with high-performance cathode materials such as LSCF.

3.2 Ceria-based materials (GDC and SDC)

An IT-LTSOFC supposedly operates at temperatures below 800 °C. When YSZ was introduced as an electrolyte, it was found to operate at 1000 °C and to give an ionic conductivity of only about 0.1 S/cm. Much work is needed to increase the ionic conductivity of YSZ at lower temperatures. Aside from reducing the thickness of the YSZ electrolyte and applying a buffer layer in between the components, changing the material was also considered. The use of an SOFC offers the choice of numerous ceria-based electrolyte materials for intermediate- to lower-temperature operations. The addition of ceria to SOFC anodes and cathodes enhances electro-catalytic activity and provides a viable path towards internal reforming and direct utilization of hydrocarbon fuels. Two types of well-known ceria-based materials are GDC and SDC.

GDC is one of the alternative materials for IT-LTSOFCs. GDC gives an ionic conductivity of about 0.12 S/cm at 800 °C (Jadhav *et al.*, 2007). Materials are prepared in situ and then sintered at various temperatures (1200, 1300, 1400, and 1500 °C). A conductivity of 0.12 S/cm was obtained from a sample sintered at 1400 °C (Jadhav *et al.*, 2007). Even though the conductivity obtained is already satisfactory, the research must continue. A suitable conductivity is not enough to confirm cell capability. The interaction between the GDC electrolyte and the LSCF cathode at the interface must be considered.

Problems at the interface of the cells occur for certain reasons. From sample preparation to the fabrication of the complete cell, much effort is required to overcome these problems. Interdiffusions are the main problem occurring at the interface because of interactions between two species (in this case, the GDC electrolyte and LSCF cathode) that result in the formation of new layers or impurities at the interface. A small amount of impurities will decrease the durability of the cells. Interdiffusions can occur for several reasons, but most of the time, the sintering temperature of the cell has been considered the main factor (Li *et al.*, 2011). Several methods have been proposed to study interdiffusions in detail. Investigations on this issue have been reported by Li *et al.* (2011) in which transmission electron microscopy, energy dispersive X-ray spectroscopy (EDX), and electron energy-loss spectroscopy (EELS) were used. Half-cell substrates were prepared with GDC as the electrolyte and LSCF as the cathode. Using those methods, two types of diffusions were found at the interface, namely, diffusion along grain boundaries and mutual diffusion. The diffusion of elements from the LSCF cathode into the GDC electrolyte along grain boundaries was observed under EELS, whereas mutual diffusion was observed under EDX. Mutual diffusion differs from diffusion along grain boundaries because it involves the diffusion of cations. Cations in GDC and LSCF diffuse into one another, neglecting the ionic radius of the diffusing cation (Li *et al.*, 2011). Mutual diffusion or bulk diffusion was found to be five times slower than diffusion along the grain boundaries (Li *et al.*, 2011). Interdiffusions also reduce cell conductivity. GDC was selected as an electrolyte because of its good conductivity.

In addition, GDC has been added to LSCF to produce composite cathodes. Among higher-temperature SOFCs, the LSM/YSZ composite exhibited high conductivity because of the similar TECs of both materials. The combination of these two materials as a composite cathode also produced larger TPBs. In other words, the addition of YSZ to LSM will enlarge the area for the ionic reaction to occur. Thus, the same concept was applied in IT-LTSOFCs. The composite cathode LSCF/GDC facilitated the occurrence of electrochemical reactions (Izuki *et al.*, 2011). Furthermore, GDC will help maintain the TEC for both components without reducing the ionic conductivity of the electrodes. Through AC impedance spectroscopy, the addition of GDC to the LSCF cathode can enhance the rate of oxygen diffusion along the component interface. Thus, in the composite, an increase in the electrochemical reaction zone has been achieved (Qiang *et al.*, 2007).

Despite all the advantages offered by GDC, Singh *et al.* (2006) demonstrated that SDC performs better than GDC in lower-temperature SOFC applications. Both materials were prepared using glycine-nitrate combustion synthesis. GDC and SDC were tested using differential thermal analysis and thermogravimetric analysis (TGA) to obtain the thermal decomposition. TGA was conducted in the temperature range between 30 and 1000 °C at a heating rate of 20 °C/min. As a result, GDC and SDC were crystallized nearly perfectly at temperatures above 800 °C. However, when it came to the sintering temperature, SDC was found to have a higher sinterability than GDC. Impedance analysis also proved that SDC has a higher ionic conductivity than GDC (Singh *et al.*, 2006).

SDC has been used in most studies over GDC because of ionic conductivity. SDC has a better chemical compatibility with cobalt-based cathodes such as LSCF (Zuo *et al.*, 2012). This latest research on LSCF and SDC involved pallets made from LSCF-SDC composites (weight ratio of 40:60), which were tested from 400 to 600 °C. A maximum power density of 797 mV/cm^2 was obtained at 600 °C using hydrogen and air as fuel. An open-circuit voltage of 0.94 V was also obtained from the same operating temperature (Zuo *et al.*, 2012). The polarization resistance of an LSCF-GDC composite at an operating temperature of 750 °C was 0.11 $\Omega\cdot\text{cm}^2$, whereas that of LSCF-SDC was 0.074 $\Omega\cdot\text{cm}^2$ (Qiang *et al.*, 2007; Nie *et al.*, 2010a). This small difference still proves that SDC is more suitable in forming composite cathodes with LSCF, which explains the current trend of using SDC in IT-LTSOFCs.

4 Performance of LSCF composite cathodes

4.1 Factors affecting the performance of LSCF composite cathodes

Generally, the factors or parameters affecting the performance of LSCF composite cathodes can be classified into three groups. The first involves the parameters related to the properties of the powder or starting materials, such as particle size, surface area, microstructure, and thermal expansion. The next group involves the composition of the composite cathode and geometrical parameters such as layer thickness. Finally, the third group involves the variables in the performance test, which include the testing atmosphere, the current or applied voltage, and the amount of fuel supplied (Tietz *et al.*, 2008; Fergus *et al.*, 2009). All these factors are interrelated in terms of determining the optimum performance of SOFCs.

The starting powders have a large effect on the performance and durability of the cathode as well as the SOFCs. The starting particulate material size can be in micro- or nano-scale. Smaller particles will possess a larger surface area, which will increase the TPB within the electrode for a higher catalytic reaction (Li *et al.*, 2006; Baqué *et al.*, 2008; Tietz *et al.*, 2008). Baque *et al.* (2008) showed that smaller particles (130 nm) give lower area-specific resistance with a difference of up to ±30% compared with larger particles (180 nm). The use of electrolyte powders with different particle sizes in the composite cathode mixture and variations in the sintering temperature can exert a significant effect on the microstructure of the cathode, which in turn determines cathode performance. Lee *et al.* (2010) reported that the medium-size (500 nm) SDC core particles used in the LSCF-SDC composite cathode give the lowest polarization resistance (0.265 $\Omega\cdot\text{cm}^2$ at 650 °C) compared with smaller (130 nm) and larger (2 μm) particles sintered at 850 °C (Fig. 3). The increased polarization resistance (R_P) of the LSCF-SDC cathode using smaller and larger SDC particles can be attributed to a decrement in the TPB resulting from the coarsening of particles (Lee *et al.*, 2010). The vital relationship between the particle size of starting powders and the final microstructure of the electrode, which affects performance, has prompted extensive studies on modifying and enhancing the properties of the starting powder used in SOFCs.

In applying the cathodes onto the electrolyte substrates, the electrode microstructure plays an important role in cell performance (Lee *et al.*, 2010; Sun *et al.*, 2010). The electrode microstructure can be manipulated by varying the cathode composition. Many studies have investigated the effect of cathode

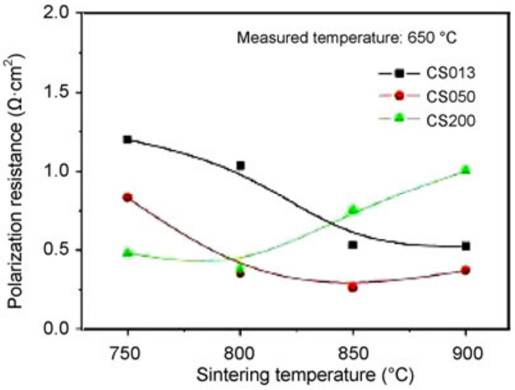

Fig. 3 Cathode polarization resistance of a symmetric cell (core-shell cathode/SDC/core-shell cathode) as a function of the sintering temperature and SDC core sizes of 130 nm (denoted CS013), 500 nm (CS050) and 2 μ m (CS200) (Lee *et al.*, 2010)

composition on cell performance (Dusastre and Kilner, 1999; Wang and Mogensen, 2005; Liu *et al.*, 2007; Fan and Liu, 2009; Jarot *et al.*, 2011b). The addition of 50% (in weight) GDC to an LSCF cathode produces lower R_P at low temperatures compared with a cathode with 30% (in weight) GDC (Wang and Mogensen, 2005). In addition, a mixture of 40% (in volume) LSGM with LSCF offers lower overpotential and R_P (0.24 Ω·cm^2) compared with LSCF mixtures with higher percentages of LSGM. An excessively high amount of electrolyte or ionic conducting materials in the composite cathode can cause a crossover and thermal mismatch between an electrode and an electrolyte, leading to higher R_P and lower SOFC cell power output (Murray *et al.*, 2002; Liu *et al.*, 2007; Fan *et al.*, 2011). However, composite cathodes with a matching amount of electrolytes can offer different cathode properties. A composite cathode LSCF-55% (in volume) GDC produced an R_P of 0.6 Ω·cm^2 at 590 °C (Dusastre and Kilner, 1999), whereas another with the same composition produced a lower R_P value (0.2 Ω·cm^2) at 600 °C (Wang and Mogensen, 2005). The latter composition had nano- to submicron-sized particles that helped to increase the TPB and in turn improve cathode performance. Furthermore, additional geometrical parameters such as cathode layer thickness and the addition of a current-collecting layer need to be investigated efficiently to obtain the optimum conditions for attaining maximum power output (Tietz *et al.*, 2008).

Another group of parameters that needs to be considered when studying the performance of SOFCs is the variables involved in the performance test. Lin and Barnett (2008) studied the effect of O_2 concentration in the atmosphere through impedance analysis. The cathode gave a higher impedance magnitude under 1% O_2 (balance Ar) at 650 °C compared with that under 20% O_2 (balance Ar). The effect of fuel flow rate on LSCF-GDC performance has been studied by Leng (2004). At low current densities, the air flow rate exhibited no clear effects on I-V characteristics. However, at higher current densities, a slight increase in the maximum power density occurred. A gain of 625 mW/cm^2 was achieved at an air flow rate of 1093 sccm (standard-state cubic centimeter per minute) compared with 578 mW/cm^2 at an air flow rate of 559 sccm (Leng *et al.*, 2004). In relation to SOFC durability for long-term operation, the current density and operating temperature significantly affect the degradation rate (Mai *et al.*, 2006; Tietz *et al.*, 2008). Mai *et al.* (2006) found that cells operated at 800 °C showed a doubly higher degradation rate than those operated at 700 °C.

All the parameters discussed play important roles and affect cell performance. Careful optimization and compatibility among the parameters are needed to obtain the maximum power output from SOFCs cells. However, among all the parameters, the composition and microstructure of the composite cathode are the main factors affecting the performance of SOFCs.

4.2 Electrochemical properties and performance of LSCF composite cathodes

Combining LSCF with ionic conductor materials such as SDC and GDC will broaden the area of electrochemical activity because of their higher ionic conductivities compared with perovskite materials. LSCF does not react with the cerium electrolyte and has the same TEC. This allows the cathode to operate at lower temperatures (Yamamoto, 2000). For the past decade, composite cathode performance has gained increasing attention from SOFC researchers (Fig. 4). Research on the development of these composite cathode materials has made significant discoveries and achievements and should be carried on to determine the materials most suitable for demonstrating optimum performance for the cathodes of SOFC systems. Table 2 lists the electrochemical properties and performances of LSCF composite cathodes that have been developed.

Murray *et al.* (2002) and Wang and Mogensen (2005) developed a composite cathode 50% LSCF-50% GDC that was applied to YSZ electrolyte substrate. Although the cathodes developed from both studies have the same composition, different activation energy (E_a) and resistance polarization (R_P) values were obtained. These differences may be attributed to the morphology of the resulting microstructure of the cathodes. Wang and Mogensen (2005) succeeded in obtaining a microstructure of nano-sized to submicron-sized particles with a size of 100 nm to help improve the TPB, compared with the cathode obtained by Murray *et al.* (2002) which had a larger particle size range (100 to 200 nm). Good adhesion between the cathode and the electrolyte also decreases the R_P value (Wang and Mogensen, 2005). Therefore, the particle size and morphology of cathode materials need exceptionally careful control and observation during their development.

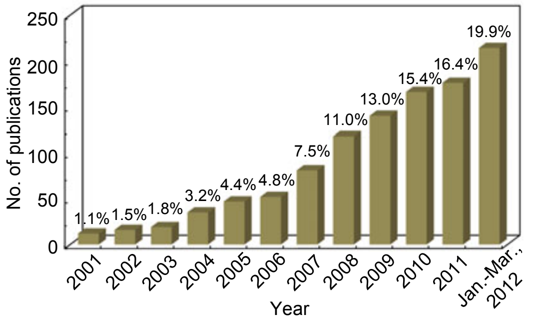

Fig. 4 Increasing number of journal publications related to composite cathode SOFC (search keywords "solid oxide fuel cell cathode or SOFC-composite cathode" in the ISI Web of Knowledge, Thomson Reuters http://apps.isi-knowledge.com.www.ezplib.ukm.my, Mar. 20, 2012)
The total number of publications from 2001 to March 2012 is 1076 journals. Therefore, the stated percentage refers to the number of publications for each year divided by 1076

The $La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ and SDC electrolyte composite has been proven to produce a composite cathode with good performance. The addition of 50% SDC has resulted in an LSCF composite cathode with a fine, porous microstructure that helps improve the TPB, enabling a more efficient chemical reaction to occur (Fu *et al.*, 2007; Lee *et al.*, 2010). Leng *et al.* (2004) also showed the capability of the $La_{0.8}Sr_{0.2}Co_{0.2}Fe_{0.8}O_3$-GDC composite cathode in providing high cell performance with a maximum power density of 578 mW/cm^2 at 600 °C. The $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$-LSGM composite produced by Lin and Barnett (2008) was able to offer the best electrochemical performance with an LSCF content between 40% and 60% (in volume) (the volume and the weight percentages were approximately the same as the theoretical densities of LSCF and LSGM are very close to each other). However, an LSCF content

Table 2 Properties of LSCF composite cathodes for intermediate- to low- temperature (800–500 °C) SOFCs

Composite cathode	Polarization resistance, R_p ($\Omega\cdot cm^2$)	Activation energy, E_a (eV)	Open-circuit voltage, OCV (V)	Maximum power density (mW/cm^2)	Reference
LSCF-30% SDC-Ag	–	–	0.76	174	(Zhang *et al.*, 2005)
LSCF-50% SDC	0.23–0.07	1.49	–	–	(Fu *et al.*, 2007)
LSCF-50% SDC	0.26–0.53	–	–	–	(Lee *et al.*, 2010)
LSCF-GDC	–	–	~0.80	60	(Liu *et al.*, 2007)
LSCF-30% GDC	0.60	~1.45	–	–	(Dusastre and Kilner, 1999)
LSCF-(40%–44%) GDC	0.40–2.40	–	~1.10	855	(Fan and Liu, 2009)
LSCF-50% GDC	0.33–0.01	1.65	–	–	(Murray *et al.*, 2002)
LSCF-50% GDC	0.19–0.03	1.20	–	–	(Wang and Mogensen, 2005)
LSCF-50% GDC	–	–	~0.88	625	(Leng *et al.*, 2004)
LSCF-60% GDC	0.27	1.40	~1.09	562	(Leng *et al.*, 2008)
LSCF-40% LSGM	0.24	–	0.88	209	(Guo *et al.*, 2009)
LSCF-LSGM	1.72–8.76	1.46	1.04	272	(Chang *et al.*, 2011)
LSCF-(40%–60%) LSGM	0.24–0.18	1.63	0.98	570	(Lin and Barnett, 2008)
LSCF-YSZ	0.54–0.05	–	–	–	(Chen *et al.*, 2008)
LSCF-37% YSZ	0.65	–	1.22	473	(Chen *et al.*, 2010)

exceeding 60% (in volume) results in higher polarization resistance values. Studies have shown that LSCF-cerium electrolyte composite cathodes have very good compatibility and can exhibit high performance given the appropriate cathode composition.

The overall performance of an SOFC can also be improved by using nano-structured perovskite cathodes. Nano-porous LSFC-SDC composite cathodes with long-term stability and low R_P values were obtained by manipulating the initial particle size of the SDC and LSCF powders. Nano-scale composite powders in the form of core-shell structures were used as starting materials to maintain a constant interfacial polarization resistance with a well-controlled microstructure (Lee *et al.*, 2010). The excellent performance of LSCF-GDC composite cathodes produced by Wang and Mogensen (2005) was attributed to the nano-submicron structure of the cathode. The presence of nano to submicron particles with small open pores has helped increase the TPB, thus broadening the electrochemical active region (Fig. 5). The use of nano-sized cathode powders has successfully improved the performance of the cathode. Two approaches using SOFC nano-structured perovskite cathodes to improve performance have been used by researchers. The first involves the synthesis of mesoporous particles and the precipitation of particles to form a macro-porous film using coating techniques such as screen printing. The second method is through the formation of nano-structured films on the surface of the electrolyte from the electrocatalysis of nanoparticle materials (Serra *et al.*, 2006).

Improvement in the power output, performance stability, and operational life of SOFCs can be achieved via the infiltration or impregnation technique (Jiang, 2006; Chen *et al.*, 2010; Jiang *et al.*, 2010). This technique involves the addition of ionic conductors to the electronic backbones, or vice versa, to increase the TPB length. An LSCF-SDC composite cathode fabricated via infiltration has shown a significant reduction in polarization, which yielded a lower interfacial resistance compared with a blank LSCF (Nie *et al.*, 2010a). The findings from the same study also confirmed that an enhancement of cell stability was obtained (Fig. 6). A nano-structured LSCF-YSZ cathode has been developed by infiltrating LSCF into YSZ (Chen *et al.*, 2008; 2010). Both findings showed a significant effect of the infiltration technique on increasing cell performance. The ability to apply the infiltration process of LSCF in cells with large active areas (81 cm^2) without using any buffer layer was demonstrated when a good thermal matching was achieved between LSCF and YSZ (Chen *et al.*, 2010). Low polarization resistance of cathodes can also be attained by controlling the firing conditions of the ionic electrolyte and the cathode-infiltrated particles. A low firing temperature (800 °C) for LSCF infiltrated into the GDC backbones has produced a nano-scale LSCF network with lower polarization resistance (0.24 Ω·cm^2) at 600 °C (Shah and Barnett, 2008).

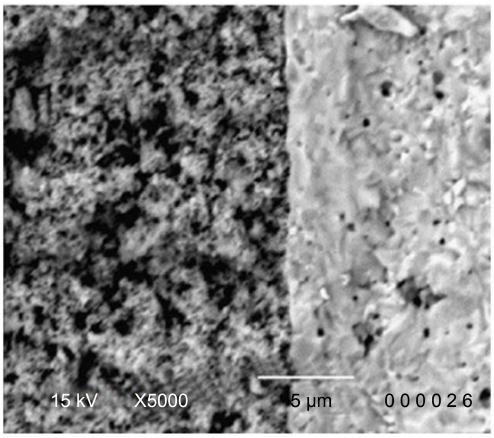

Fig. 5 SEM picture of the cross-section of a fractured symmetric sample with a 70% LSCF-30% CGO (in weight) cathode on CGO electrolyte (Wang and Mogensen, 2005)

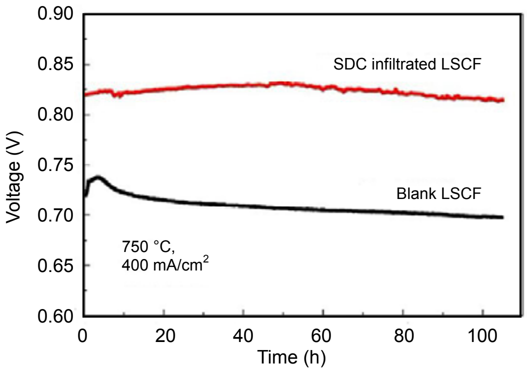

Fig. 6 Performance stability of anode-supported SOFCs with a blank LSCF cathode and an SDC infiltrated LSCF cathode (0.25 mol/L, 10 μm) operated at a constant current density of 400 mA/cm^2 at 750 °C for 100 h (Nie *et al.*, 2010a)

Fuel cells based on carbonate salt composite electrolytes and electrodes have gained considerable attention in the development of LTSOFCs because of their highly suitable characteristics and optimal performance (Zhu, 2003; Jarot *et al.*, 2011a; Zhao *et al.*, 2011; Rahman *et al.*, 2011; 2012). A single cell composed of an LSCF-GDC carbonate cathode, a GDC carbonate electrolyte, and a Ni-GDC anode has given a maximum power density of 238 mW/cm^2 from about 530 to 540 °C (Zha *et al.*, 2003). The combination of a composite cathode LSCF with 30% (in weight) SDC carbonate electrolyte has produced a single cell with 75.33 mW/cm^2 maximum power density at 650 °C (Jarot *et al.*, 2011b). The satisfactory performance is achieved with salt molten carbonate present in the LSCF composite cathode, which enhances the ionic conductivity of the electrode close to the electrolyte interface and extends the TPB to the whole cathode surface (Simner *et al.*, 2003; Lee *et al.*, 2010; Amar *et al.*, 2011). Therefore, detailed studies on the effects of carbonate composition in the LSCF cathode, the calcination or sintering temperature of carbonate cathodes, and the microstructure evolution of the LSCF carbonate composite cathode, on its electrochemical properties and performance require serious attention for further development of LTSOFCs.

Thus, many aspects of cell performance still require further study, especially for LSCF composite cathode carbonate SOFC systems. Such aspects include the degradation of the fuel cell, the stability of the electrode in the long run in relation to the use of nano-sized powders in the production of a composite cathode, the effect of microstructure, and the fabrication method.

5 Conclusions

We have reviewed the development of the LSCF composite cathode in this paper with a primary focus on the fabrication process, the compatibility of LSCF with electrolyte materials, and overall performance. The powder properties, microstructure, cell design, fabrication process, and performance test parameters of the LSCF composite cathode are all closely related in obtaining the optimum power output of SOFC cells. LSCF shows good compatibility with ceria-based electrolytes. Nano-scale cathode particles, thin layer electrolytes, and carbonate salt electrolytes have been used to enhance the cathode's properties. Recently, the LSCF carbonate composite cathode has gained significant attention because of its promising characteristics as cathode material for LTSOFCs. Nevertheless, the effects of the carbonate phase on the LSCF composite cathode properties, such as its oxygen stoichiometry and ionic conductivity, require thorough research.

Research on LSCF composite cathodes for LTSOFCs is rapidly expanding. LSCF composite cathode materials show high performance and potential for use in IT-LTSOFCs. Many other related aspects of the LSCF composite cathode material need to be studied to improve performance of the cell. These include the long-term stability of the composite LSCF when combined with other fuel cell components, the degradation mechanism in performance, and the optimization of processing parameters in forming the cathode layer. The scope of the research outlined above provides an opportunity and a challenge to researchers to develop and improve the ability of LSCF composites as cathode materials for IT-LTSOFCs.

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