

Redox Stable Electrode Material of Symmetrical Solid Oxide Fuel Cells

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Abstract: Solid oxide fuel cells (SOFCs) technologies are considered the most efficient and environmentally-friendly option for both stationary and distributed power generation due to their advantages of low pollution, low noise and strong building blocks. Despite the potential, widespread adoption of SOFC technology has not yet been achieved. The main reason is that the high cost of manufacturing and maintenance and the insufficient operating life make SOFC technology less competitive economically. More and more people begin to pay attention to symmetrical solid oxide fuel cells (SSOFCs) because SSOFCs have many potential advantages, such as improving the thermal mechanical compatibility between electrolyte and electrode, reducing manufacturing costs, and improving the ability to resist coking and sulfur poisoning. This article reviews the development of SSOFCs redox stable electrodes in recent years, summarizes the current research hotspots, and looks forward to the future development of redox stable electrode materials.

1 INTRODUCTION

The need to shift towards a sustainable infrastructure due to the detrimental impact of greenhouse gases and environmental pollution from inefficient and excessive use of fossil fuels necessitates the development of new energy materials and technologies. Fuel cells, based on this concept, were first introduced 184 years ago.^[1] According to the use of electrolyte in the cell, fuel cells are mainly divided into the following four categories recently: proton exchange membrane fuel cells (PEMFCs), phosphoric acid fuel cells (PAFCs), molten carbonate fuel cells (MCFCs) and solid oxide fuel cells (SOFCs).^[2,3] SOFC devices have been shown to be highly efficient and clean power-generation sources, making them a preferred choice among various types of sustainable energy sources.^[4,5] However, conventional SOFCs electrodes require separately manufacturing of fuel and oxygen electrodes, which greatly increases operating and maintenance costs and is not conducive to large-scale commercial SOFCs applications. To solve this problem, such a SOFCs configuration was first proposed by Badding et al in 2001.^[6] It was named the symmetrical solid oxide fuel cells (SSOFCs). For SSOFCs, the most important structure is the symmetrical electrode, so the choice of electrode material has become the main problem in current SOFCs' researches. Some researchers have found that some materials can be used as both cathode and anode materials for SOFCs.^[7,8]

SSOFCs work in a similar way to SOFCs, except for the difference in electrode materials. (Figure 1.) At the cathode is a reduction reaction, oxygen molecules (O₂)

gain electrons to become oxygen ions (O²⁻), namely:



The electrolyte is located between the cathode and the anode. Driven by the oxygen concentration difference on both sides of the cells, the oxygen ions are generated on the cathode while being migrated to the anode through the oxygen vacancy in the electrolyte and then react with the fuel (H₂ for example here) to release electrons, namely:

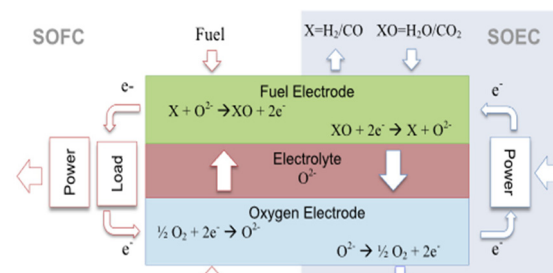
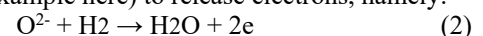


Figure 1. Principle of solid oxide fuel cells.^[9]

The connecting material separates multiple series-parallel cells with cathodes and anodes on opposite sides and prevents oxidants from being mixed with fuel.^[10] When it comes to electrode materials for such SSOFCs, they usually require the following conditions: oxidation catalytic activity for fuel (hydrogen, etc.); catalytic activity for oxygen reduction reaction; high electron-ion conductivity in cathode/anode environments; thermal expansion coefficient matching common electrolyte materials; stable chemical properties under oxidation and reduction conditions.^[11] Theoretically, most oxides used as anode materials of SOFCs can be used as electrode

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materials in SSOFCs because of their stable structure in the oxidation atmosphere (such as oxygen in the cathode). However, many cathode materials with good oxygen reduction reaction (ORR) performance are unstable in a reducing atmosphere (such as the atmosphere on the anode). So far oxide electrode materials used in SSOFCs are generally derived from anodic oxide materials that are already used in conventional SOFCs. And these electrodes are further classified into oxidation-reduction stable electrodes and oxidation-reduction reversible electrodes. The following mainly introduce oxidation-reduction stable electrodes, which commonly come from traditional anode materials and keep the structure unchanged in the reducing atmosphere.

2 REDOX STABLE ELECTRODE

2.1 Single phase stable electrode

In 2006, Irvine et al applied $(\text{La}_{0.75}\text{Sr}_{0.25})\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_3$ (LSCM), the anode material of SOFCs that he had previously studied to SSOFCs and obtained power densities of $300 \text{ mW}\cdot\text{cm}^{-2}$ and $230 \text{ mW}\cdot\text{cm}^{-2}$ at 900°C using wet H_2 and CH_4 as fuel, respectively.^[12-14] Afterwards, Kharton et al investigated the A-site defect (i.e. $(\text{La}_{0.75}\text{Sr}_{0.25})_{0.95}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$). Research has found that at $750\text{-}950^\circ\text{C}$, the ionic conductivity significantly increases, reaching $1\text{-}3 \text{ S}\cdot\text{cm}^{-1}$, but the overall conductivity is still low.^[15] Since LSCM, many electrode materials with similar components and structures have been selected as symmetrical electrode materials,^[16,17] and many ions have been doped or substituted to improve the problems of LSCM.^[18,19] Furthermore, the catalytic activity of perovskite oxides is mainly influenced by B-site cations.^[20,21] Peña-Martínez et al conducted an in-depth study on the influence of B-site cations in oxides composed of $\text{La}_{0.75}\text{Sr}_{0.25}\text{Cr}_{0.5}\text{X}_{0.5}\text{O}_3$ ($\text{X} = \text{Mn, Fe and Al}$) on electrode performance in SSOFCs.^[22] Research has found that the performance of Mn doped anode oxides is higher than that of Fe and Al. However, it is worth noting that the cathode performance of SSOFCs configured by LSCM under medium temperature conditions of 800 degrees

Celsius is not satisfactory. Although it is generally believed that the catalytic activity of perovskite is mainly determined by the B-site cation, the A-site cation can still indirectly affect the catalytic activity of perovskite oxides by changing the oxidation state of the B-site cation. Furthermore, Zhang et al investigated the effects of A-site doping with Ca, Sr, and Ba on $\text{La}_{0.7}\text{A}_{0.3}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$ respectively to investigate their impact on the electrochemical performance of SOFC.^[23] The experimental results indicated that $\text{La}_{0.7}\text{Ca}_{0.3}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$ (LCCM) doped with Ca element had the best electrocatalytic activity in both oxygen reduction reaction and fuel oxidation reaction, which indicated that LCCM may be a potential electrode material for SSOFCs. However, the PPD of LCCM is only tens of $\text{mW}\cdot\text{cm}^{-2}$ at 900°C . The performance of these materials is generally not as good as that of traditional SOFCs. The academic community expects more excellent SSOFCs electrode material system.^[24]

In 2010, Chen et al developed a new perovskite material $\text{Sr}_2\text{Fe}_{1.5}\text{Mo}_{0.5}\text{O}_{6-\delta}$ (SFM).^[25] The experimental results showed that at 780°C , the conductivity of SFM in air and hydrogen reaches 500 and $310 \text{ S}\cdot\text{cm}^{-1}$, while the anode materials required only $100 \text{ S}\cdot\text{cm}^{-1}$ in fuel air/hydrogen, respectively. Additionally, the power density of $835 \text{ mW}\cdot\text{cm}^{-2}$ and $230 \text{ mW}\cdot\text{cm}^{-2}$ could be obtained by using wet H_2 and wet CH_4 at 900°C and SFM had good cycle stability at the anode compared with LSCM, respectively. Compared with LCCM, the power density increased by more than one order of magnitude. In 2014, the study of Fernández-Ropero et al on $\text{SrFeO}_{3-\delta}$ base oxide with a nominal composition of $\text{SrFe}_{0.75}\text{M}_{0.25}\text{O}_{3-\delta}$ ($\text{M} = \text{Ti, Zr, V, Nb, Cr, Mo, W}$) in the background of SSOFCs as electrode materials, the influence of foreign ions on phase structure and phase stability was systematically studied.^[26] They studied the relationship between conductivity, temperature dependence of polarization resistance, ionic radius and polarization resistance by doping B-site elements in SrFeO_3 . (Figure 2.) After doping, the specific polarization resistance of these materials has been improved in both oxidation and reduction atmospheres, indicating their potential application as SSOFC electrode materials.

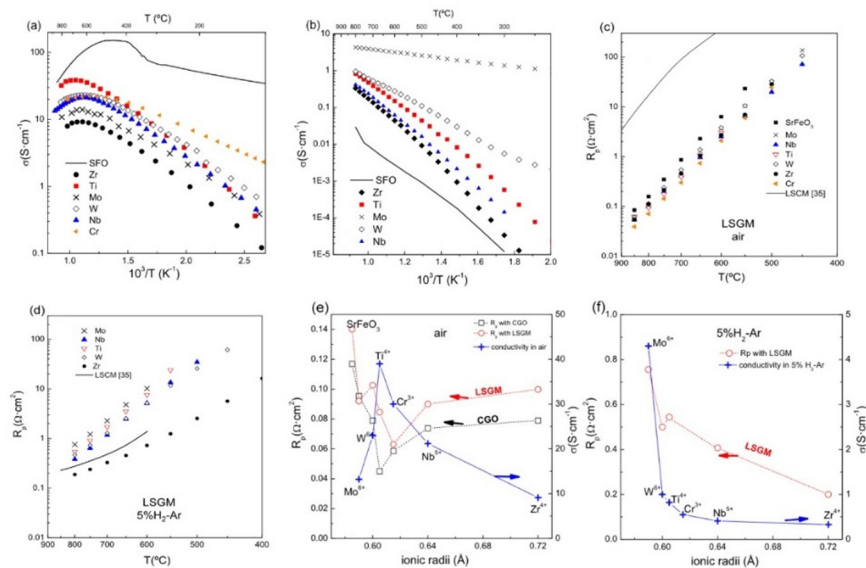


Figure 2. (a) and (b) shows Arrhenius plots of conductivity for the SrFe_{0.75}M_{0.25}O_{3-δ} series in air and 5% H₂-Ar, respectively. (c) and (d) illustrate the temperature dependence of polarization resistance for SrFe_{0.75}M_{0.25}O_{3-δ} electrode over LSGM electrolyte in air and 5% H₂-Ar at open circuit conditions, with comparison values of polarization resistance for La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O_{3-δ} (LSCM).¹² Additionally, (e) and (f) depict the correlation between area specific polarization resistance and dopant ionic radii in SrFe_{0.75}M_{0.25}O_{3-δ} with LSGM and Ce_{0.9}Gd_{0.1}O_{1.95} (CGO) electrolyte at 800 °C in air and 5% H₂-Ar.²⁶

The above LSCM and SFM are two typical single-phase stable electrode materials. In fact, before the birth of LSCM, researchers had found that LaMnO₃ exhibits higher hydrogen oxidation activity compared to LaCrO₃.^[27,28] Unfortunately, LaMnO₃ was chemically unstable in reducing atmospheres. In a related example, Zhang et al reported that La_{0.8}Sr_{0.2}MnO₃ (LSM) underwent a phase transition after undergoing hydrogen treatment at 600 °C for 1 hour.^[29] The PPD of the battery decreased by more than half, which indicating that phase reduction may lead to a decrease in battery performance. In contrast, LaCrO₃ demonstrated superior chemical stability in reducing conditions.^[27] The addition of Cr reduces the coefficient of thermal expansion of the electrode, which helps to improve stability. Therefore, Sfeir et al proposed a strategy to improve the B site of perovskite lattice by co doping Cr and Mn, so as to achieve a reasonable balance between chemical stability and hydrocarbon oxidation catalytic activity.^[30] This provide us with a new idea that we can integrate two or more materials with complementary properties together.

2.2 Mixed stabilizing electrode

In traditional SOFCs, composite electrodes are widely used as cathodes and anodes. Electrodes composed of multiple phases can acquire beneficial qualities from each phase, tackling the challenge of obtaining all necessary aspects from a single-phase material. In recent years, composite electrodes have also been used for SSOFCs. Like Ni-SDC cermet,^[31] Ni has good conductivity and catalytic capacity, while Sm_{0.2}Ce_{0.8}O_{1.95-δ} (SDC) has a considerable number of oxygen vacancies, which makes these composite materials meet the requirements of anodic reaction. At the same time, SOFC adopts the overall framework of SDC to eliminate the mismatch of coefficient of thermal expansion of different units.

Common composite methods include physical mixing method and impregnation method. The conventional physical mixing method is to physically mix two or more phases with different beneficial components in an appropriate proportion, while the filtration impregnation method is developed on the basis that it is difficult to achieve ideal microstructure and good penetration of all phases in the process of preparing composite electrodes by conventional physical mixing method.

2.2.1 Conventional physical hybrid composite electrode

As mentioned previously, LSCM is a promising SSOFCs electrode material. However, when SOFCs containing LSCM operates at intermediate temperatures, SOFCs' cathode performance remains relatively poor because LSCM exhibits a low oxygen vacancy concentration, particularly at lower temperatures. Similarly, the commonly used conventional cathode material LSM demonstrates poor oxygen ion conductivity at intermediate temperatures.^[32] Ruiz Morales et al prepared LSCM / 8% mol Y₂O₃ / ZrO₂ (YSZ) (1:1) composite symmetric electrode and used polymethyl methacrylate (PMMA) microspheres as templates to optimize the microstructure of the electrodes.^[33] The PPDs of SOFCs are 546 and 347 mW•cm⁻² under the support of YSZ electrolyte (250μm), respectively. Compared to the PPD obtained by Irvine et al.¹²⁻ using wet hydrogen and methane as fuel for LSCM at 900 °C, the PPD increased by 82% and 51%, respectively. Ruiz Morales et al further prepared LSCM / YSZ / Gd_{0.2}Ce_{0.8}O_{2-δ} (GDC0.2) SSOFCs composite electrode.^[34] They found that when the weight ratio of YSZ to GDC0.2 is 1:1, optimal electrode performance can be obtained. They manufactured a symmetrical battery configured as LSCM+YSZ+GDC0.2 | YSZ (180 μ m) |

LSCM+YSZ+GDCO. For battery performance testing, PPDs of 400 and 120 $\text{mW}\cdot\text{cm}^{-2}$ were obtained under wet H₂ and CH₄ conditions at 950 °C, respectively. Compared to LSCM and YSZ without doping GDC0.2, the reduction is 27% and 65%, respectively. In contrast, Zhou et al measured a PPD of 264 $\text{mW}\cdot\text{cm}^{-2}$ for SSOFs made of LaSr₂Fe₂CrO_{9- δ} (LSFCr) + GDC0.2 composite electrode at 800°C, using wet hydrogen as fuel, which is 18% higher than the PPD observed in batteries containing only a single LSFCr phase (224 $\text{mW}\cdot\text{cm}^{-2}$).^[35] Comparing the two experimental results, this may be due to the use of PMMA

microspheres as templates to obtain a well followed electrode microstructure in the LSCM-YSZ (1:1) study. Jung et al used a method of synthesizing nanoparticles using polymer composite resin approach to prepare a simple powder mixture of LSCM and YSZ (75:25).^[36-39] The polarization resistance and PPD were measured at 850°C in hydrogen gas containing 4% water vapor, with a polarization resistance of 1.82 $\Omega\cdot\text{cm}^{-2}$ and a PPD of 177 $\text{mW}\cdot\text{cm}^{-2}$. The following table summarizes the research results above. (Table 1.)

Table 1. the PPD of LSCM and its composites using H₂ and CH₄ as fuels at a certain temperature.

Electrode	Electrolyte	Fuel	Temperature(°C)	Peak Power density($\text{mW}\cdot\text{cm}^{-2}$)	Reference
LSCM	YSZ	Wet H ₂	900	300	Ref.
LSCM	YSZ	Wet CH ₄	900	230	12;13;14
LSCM+YSZ(1:1)	YSZ(250 μm)	H ₂	950	576	Ref. 33
LSCM+YSZ(1:1)	YSZ(250 μm)	CH ₄	950	347	Ref. 33
LSCM+YSZ(3:1)	YSZ	Wet H ₂	850	177	Ref. 39
LSCM+YSZ+GDC0.2	YSZ(180 μm)	Wet H ₂	950	400	Ref. 34
LSCM+YSZ+GDC0.2	YSZ(180 μm)	Wet CH ₄	950	120	Ref. 34
LSFCr+GDC0.2	YSZ	Wet H ₂	800	264	Ref. 35
LSFCr	YSZ	Wet H ₂	800	224	

Although the testing conditions and preparation process may vary, it can be seen from the table that the performance of composite electrode materials is significantly higher.

2.2.2 Composite electrode prepared by impregnation technology

As mentioned earlier, Ruiz Morales et al used PMMA microspheres as a template to optimize the microstructure of the LSCM and YSZ (1:1) composite symmetric electrodes, resulting in a significant improvement in the PPD value.^[33] The following figure shows the microstructure of LSCM and LSCM-YSZ composite materials. (Figure 3.)

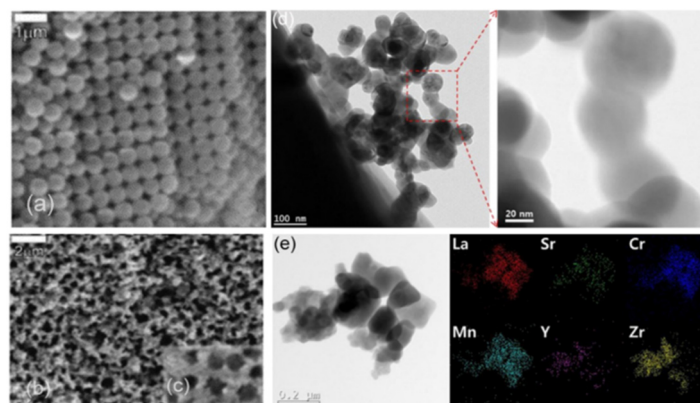


Figure 3. SEM image of (a) PMMA microspheres utilized to regulate porosity in the LSCM-YSZ composite; (b, c) display the optimized microstructure of a LSCM-YSZ composite after organic template removal.^[33] Furthermore, (d) presents a high-resolution transmission electron microscope (HRTEM) image of LSCM single phase powders calcined at 1100°C, while (e) shows LSCM-YSZ nanocomposite powder calcined at 1100°C along with its corresponding EDX elemental mapping image.^[39]

Therefore, the micro pore structure of the electrode has an important impact on the electrochemical performance of the electrode. Recently, a penetration/impregnation technology has been developed, which is a unique method for preparing composite electrodes with special electrode microstructures.^[40-42] The schematic diagram of its microscopic process is shown below. (Figure 4.)

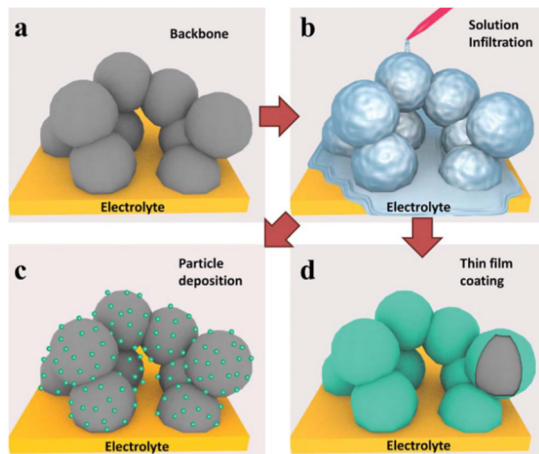


Figure 4. Illustrates a typical infiltration process, depicting (a) an as-fired electrode backbone and (b) solution drops infiltrating into the electrode backbone. After thermal treatment, two common morphologies of the infiltrated electrode are particle deposition and thin film coating, as displayed in (c) and (d) respectively.⁴²

The advantages of composite electrodes prepared by infiltration method include: optimizing the electrode structure at the micro level and improving the compatibility between the electrode and the electrolyte carrier; Macroscopically, it increases the apparent conductivity of oxygen ions and electrons, resulting in higher catalytic activity for ORR and fuel oxidation.

Due to their ion conductivity, excellent surface exchange, and oxygen storage capabilities, penetrants made of fluorite doped cerium dioxide are widely used as catalyst materials.^[43-45] What's more, the catalytic activity of $\text{La}_{1-x}\text{Sr}_x\text{Co}_{1-y}\text{Fe}_y\text{O}_{3-\delta}$ (LSCF) is limited by the surface catalytic properties. Thus, infiltration of doped ceria onto the LSCF cathode can potentially promote surface exchange of oxygen and increase the surface area of the electrode, therefore resulting in improved ORR kinetics of the LSCF cathode.^[46] This was partially confirmed through the investigation by Ding et al, who

analyzed the interfacial polarization resistance at different temperatures between blank LSCF cathodes and those with varying levels of SDC infiltration under open circuit conditions.^[42] (Figure 5(a)) They used their self-developed new catalyst coating to further improve the performance of LSCF cathodes, including $\text{Pr}_{0.75}\text{Sr}_{0.2}\text{MnO}_{3-\delta}$ (PSM) and $\text{PrSrCoMnO}_{6-\delta}$ (PSCM).^[47] These materials have a structure similar to LSCF, making it easy to form conformal coatings on the surface of LSCF. Compared with LSCF cathodes modified by LSM which exhibit higher reactive power (R_p) at open circuit voltage (OCV), LSCF infiltrated by PSM and PSCM exhibits lower R_p values. It is evident that the power output of the anode supported battery of PSM and LSCF infiltrated by PSCM is higher than that of LSCF infiltrated by LSM.^[47] (Figure 5(b)) What's even more intriguing is that the LSCF cathodes with infiltration showed a stronger activation behavior during operation. The PSCM infiltrated LSCF cathode exhibits the best performance and significant activation behavior in the battery, possibly related to changes in the surface chemistry of the thin film and the reconstruction of Mn based perovskite phase under operating conditions.^[48] Jiang et al conducted a comparative study on the power output of anode supported batteries with three types of electrodes: (I) mechanically mixed LSM-YSZ, (II) nanostructured mixed LSM-YSZ, and (III) nanostructured Pd infiltrated LSM-YSZ.^[49] (Figure 5(c)) In subsequent studies, Jiang et al reported a Pd infiltrated YSZ and LSM + YSZ cathode.^[50] They deposited LSM nanoparticles on highly porous YSZ scaffold, achieving reasonable conductivity even below the normal percolation threshold of 30%. In comparison, a Pd + LSM-YSZ cathode was created by depositing Pd nanoparticles onto a conventional LSM + YSZ backbone. It remains uncertain whether doped Pd nanoparticles possess unique catalytic properties in contrast to oxides like LSM.

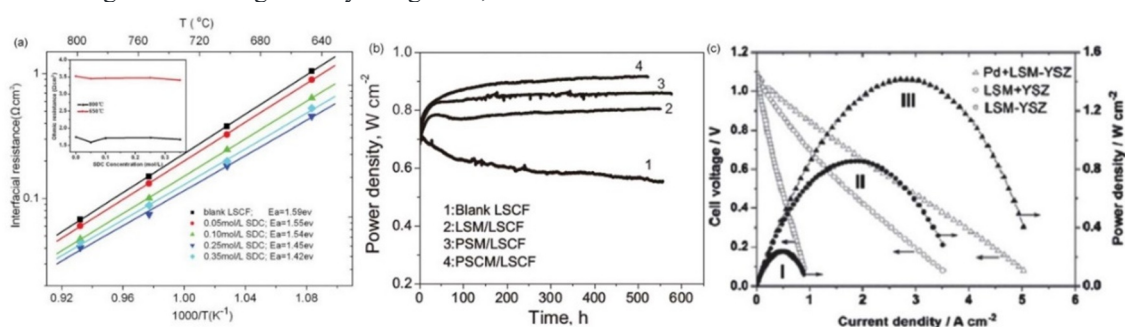


Figure 5. (a) Temperature dependence of cathodic interface polarization resistance measured under open circuit conditions (OCV).⁴² (b) Power outputs of anode-supported cells with catalyst-infiltrated LSCF cathodes at a constant cell voltage of 0.7V at 750 °C with humidified H₂ (3 vol% water vapor) as the fuel and stationary air as the oxidant.⁴⁷ (c) power output of anode-supported cells with electrode types I, II, and II, measured at 750°C in H₂/air.⁴⁹

Although progress has been made in improving the catalytic activity and durability of composite electrodes prepared by impregnation method, current research on the detailed mechanism of performance enhancement is insufficient to provide scientific basis for designing more efficient catalysts and new electrode structures. As a result, the commercial application of next-generation SSOFCs still faces significant challenges.

3 CONCLUDING REMARKS AND PROSPECTS

Compared with traditional SOFCs, SSOFCs has very significant characteristics. On the one hand, SSOFCs have the same cathode and anode materials, which can reduce production costs. On the other hand, these new electrode

materials have relatively high peak power density and low resistivity, thereby reducing electrical energy loss. However, SSOFCs require more stringent electrode material specifications to meet the needs of both anode and cathode, the electrode materials need to meet the needs of both anode and cathode. Since the development of the earliest LSCM electrode material for SSOFCs, significant improvements have been made in the performance of SSOFC electrodes. In recent years, more and more research has focused on medium to low temperature electrode materials and redox-stable electrodes that can function reversibly. This article discusses the oxidation-reduction stable electrodes of SSOFCs, which are divided into single-phase and multiphase materials. The multiphase materials are further divided into two types: simple physical mixing method and impregnation method. We have collected rich research results in recent years and summarized the PPD values of common single-phase redox stable electrodes as a reference in the table.

Despite the many advantages of SSOFCs, there are still many urgent issues to be addressed. One of the most worth solving problems is the low power density. At present, whether it is SOFC or SSOFCs, in order to achieve large-scale commercial applications, batteries need to be assembled into stacks. However, current technology is far from reaching the maximum theoretical power density. The main reason is the difficulty in thermal management of the battery pack. How to quickly transfer the heat generated by the stack to the outside of the stack and improve the management ability of the thermoelectric stack is a problem that needs to be considered. Improving the heat dissipation structure of the stack and developing electrode materials that can work effectively under medium to low temperature conditions are two feasible ideas.

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