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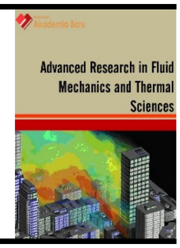

Fabrication Process of Cathode Materials for Solid Oxide Fuel Cells

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ABSTRACT

Solid oxide fuel cells (SOFCs) are advanced systems and currently a leading power source technology. SOFCs are appealing because they offer high power conversion efficiency, reliability, fuel adaptability and environmental friendliness. In general, SOFCs consist of electrodes (anode and cathode), electrolyte and interconnects. The cathode in particular was highlighted in this research to improve the performance and material compatibility of SOFCs. This paper reviewed the fabrication parameters generally involved in determining the microstructural characteristics of cathode materials for SOFCs. Several fabrication methods to produce cathode materials for SOFCs were reviewed. Finally, at the end of this paper, state-of-the-art cathode materials for SOFC applications were suggested for further study.

1. Introduction

Energy production, delivery and storage technologies are being continuously innovated to satisfy global demands for sustainable energy [1]. The demands for clean and sustainable fuel cells have led to the development of electrochemical energy conversion devices, which are able to convert the chemical energy of fuels directly into electricity through electrochemical reactions. Meanwhile, SOFCs are a type of fuel cell that operate at high temperature and thus have faster reaction kinetics [2]. SOFCs employ the advantages of high energy conversion and excellent fuel diversity [3]. SOFC applications include both mobile and stationary systems [4]. As is known, SOFCs are composed of few components: electrodes (anode and cathode), electrolyte and interconnects. However, the high operating temperatures result in limited choice of suitable materials and decrease the durability [5] of the fuel cell. SOFC cathode materials must have high electrocatalytic activity, oxygen transport ability and high electron conductivity [6].

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The current challenge is to either develop new cathode materials or improve existing materials by changing the synthetic route, surface modification or fabrication parameters. Surface modification typically causes materials to undergo structure reformation, for example, material infiltration/impregnation and composite materials. $La_{1-x}Sr_xMnO_3$ (LSM) is a traditional pure conductor cathode material that has been used for many years. However, LSM has found renewed context, in which LSM is used in various fabrication techniques, such as infiltration within a porous structure [7]. Meanwhile, Li et al. reported the enhanced electrochemical performance of a co-synthesized $La_2NiO_{4+\delta}$-$Ce_{0.55}La_{0.45}O_{2-\delta}$ composite cathode [8] and showed an example of improvement by changing the synthetic route. Finally, the performance of a fuel cell can be improved through detailed study of the fabrication parameters. Thus far, there is one review regarding fabrication parameters in SOFC applications; however, the report is far from the latest trends. Further discussion in this paper will review the effect of the fabrication parameters along with recent experimental data. The main purpose of this report is to elucidate the microstructural variables and provide a general review of the few fabrication processes for cathode materials. At the end of the review, there is a compilation of recent trends of cathode materials using various fabrication techniques.

2. Characteristic of Cathode

To ensure excellent SOFC performance, cathode materials should have the following features[9–13].

1. High electrical conductivity (~100 S/cm).
2. High compatibility between the thermal expansion coefficients (TECs) of the cathode and electrolyte (~13.1-23.8 $\times 10^{-6}$ K^{-1}).
3. Excellent chemical stability.
4. Sufficient material porosity to aid in the diffusion of oxygen gas at the interface (20-40%).
5. Good oxide ion (O^{2-}) conductivity (~1 x 10^{-3} S/cm)[14].
6. High catalytic activity in the oxygen reduction reaction (ORR).
7. Low cost.

However, the most important focuses within this research field include the following:

1. The oxygen transport mechanism in different types of cathode materials as shown in Figure 1.
2. The development of new compositions and microstructures of cathode materials.
3. Study of the relation among the structure, properties and performance.
4. The influence of various factors, such as time, temperature, thermal cycling, polarization, operating conditions, and impurities, on the cathode performance.
5. Determination of detailed processing and characterization techniques that allow the control and evaluation of the cathode microstructure.

3. Fabrication Parameters

The fabrication parameters are important because materials can undergo chemical and physical changes due to the alteration of the working environment and fabrication process. It is important to optimize microstructural variables such as the particle size, thickness and porosity distribution [15]. However, the optimization process is quite challenging due to the requirements of cathode materials, as mentioned in section 2.0. SOFC electrodes must have excellent porosity to facilitate gas diffusion and support the redox reactions [16]. For example, electrodes with a uniform distribution of pores exhibit better electrochemical performance than electrodes with randomly distributed pores due to increased pore percolation [17]. Meanwhile, the relation between the porous microstructure and small particle size elevates the catalytic properties by enhancing gas diffusion and increasing the reactive surface area [18]. The oxygen reduction reaction (ORR) can occur in two possible ways: along the triple phase boundary (TPB) and through the bulk transport of oxide ions. Therefore, the thickness and porosity of the film play an important role because cathodes with a dense thin-film structure show an improved oxygen reduction reaction (ORR) along the TPB [10]. In addition, the porosity distribution caused the electrode to become less conductive with a smaller density of reaction sites as the film thickness increased [19]. A recent study by Riazat et al. investigated the effect of the film thickness and particle size on the electrochemical reaction and mass transport. The results showed that cathode materials with smaller particle sizes simultaneously increased the electrochemical performance and deteriorated the mass transport efficiency [20].

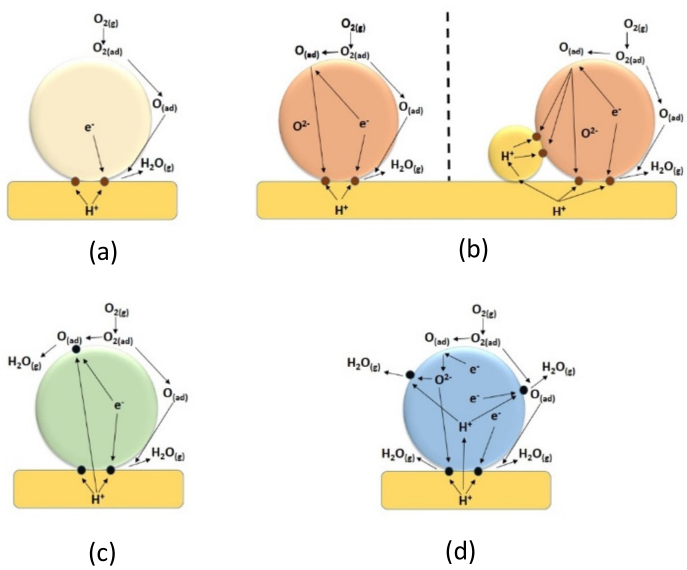

Fig. 1. Schematic diagrams of the reaction site zones in(a) pure electronic conductors (PEC), (b) mixed ionic and electronic conductors (MIEC), (c) mixed protonic and electronic conductors (MPEC), and (d) triple $H^+/O^{2-}/e^-$ (THOEC)

4. Fabrication Process

There are many techniques to fabricate fuel cells, for example, screen printing, spin coating and spray coating. What makes them different from each other are the mechanism, advantages, cost and resulting layer thickness. In this section, a variety of fabrication processes are reviewed, and recent reports for fabricating cathode materials are outlined in Table 1.

4.1 Screen Printing

Screen printing is a simple, cost-effective and commonly used technique for fabricating SOFC films within the thickness range of 10-100 μm [21]. It is a well-established technique that exhibits full two-dimensional (2D) patterning of the printed layer [22]. Thus, screen printing is a practical way to produce porous electrodes and dense electrolyte [23, 24]. However, there are limitations to screen printing, such as substrate evenness and layer thickness size of the screen, mainly due to evaporation of the solvents [25].

Table 1
Summary of fabrication processes for cathode materials in SOFC

Process	Materials	Film thickness (μm)	Particle size	Peak power density		Ref.
				Temperature (°C)	Power (W cm^{-2})	
Tape casting	$(La_{0.8}Sr_{0.2})_{0.95}MnO_3$ (LSM)	16	~1.2-2.2 μm	750 800 850	0.325 0.479 0.625	[26]
	$Mn_{1.5}Co_{1.5}O_4$ (MCO)-YSZ	25	-	-	-	[27]
	$La_xSr_{1-x}Co_yFe_{1-y}O_{3-\delta}$ (LSCF)	40	50-300 nm	700 750 800	0.371 0.744 1.075	[28]
Screen printing	$La_{1-x}Sr_xFeO_3$	20-60	500 nm	-	-	[29,30]
	$SrCo_{0.7}Nb_{0.1}Fe_{0.2}O_{3-\delta}$	40	-	700	0.208	[31]
	$YBaCo_{3.2}Ga_{0.8}O_7$	25-30	~5 μm	800	0.395	[32]
	$SmBaCo_{2-x}Ni_xO_{5+\delta}$	30	<1 μm	800	0.536	[16]
	$Sm_{0.5}Sr_{0.5}CoO_{3+\delta}$ (SSC)	-	-	700	1.09	[33]
Spray coating	BSCF-SDC	8 (SDC)	-	700	0.495	[34]
	LSC	40	20-30	-	-	[35]
Spin coating	LSM-SDC	-	80 nm (LSM) 30 nm (SDC)	-	-	[36]
	LSM-SDC	20	-	850	0.58	[37]
	$La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$	0.14	50 nm (800°C) 150 nm (1000°C) 300 nm (1200°C)	-	-	[38]
	$Sm_{0.2}Ce_{0.8}O_{1.9}$ (SDC)-$Sm_{0.5}Sr_{0.5}CoO_3$ (SSC)	7.5	200 nm	600	0.3	[39]

4.2 Tape Casting

Tape casting is another potential low-cost fabrication process, which produces thin flat layers with thicknesses of 10 μm to 1000 μm through a firing and simple shape-forming method [40]. The well-known tape casting method possesses impressive features, such as the production of layers with uniform thickness and structure, the formation of multilayer tapes per round and the ability for mass production [25]. The only disadvantage of this process is the limitation in the thickness layer [41]. Recent studies have reported that the parameters that improve the electrochemical performance and mechanical strength are the solid loading and binder content in the slurry [42]. It was determined that increasing the solid loading and binder affects the particle network. Unfortunately, increasing the solid loading and binder increase the viscosity and lead to excessive air bubbles in the slurry, which reduce the electrochemical performance [43].

4.3 Spin Coating

Spin coating is a technique to apply a thin uniform film on a flat substrate via spinning the substrate [44]. This technique can produce homogenous films over large areas with diameters over approximately 30 cm [44]. While screen printing and tape casting produce a particular range of thicknesses, the spin coating technique slightly differs because the desired thickness can be varied by adjusting the number of coating cycles. However, the typical film thickness produced is >1 μm [40]. Spin coating is a process that utilizes centrifugal force by rotating the substrate to evenly spread the coating solution over the surface [45].

4.4 Spray Coating

Spray coating is a process in which a suspension is directly sprayed on a substrate using a spray gun and is widely used in industrial applications because of its simplicity and cost efficiency [46,47]. In [35] stated that the spray coating is suitable for tubular substrates and can be considered as an efficient technique for the deposition of uniform and large-area thin films on a variety of substrates. The fabrication of electrodes and electrolytes via spray coating technique has been demonstrated in detail by She et al. and Morales *et al*., [34,35].

5. Conclusion

In this article, the relation among the microstructural variables of SOFC cathode, including the porosity, particle size and film thickness of the cathode have been discussed and related to the overall SOFC performance. Each of the variables is an important element, as cathodes require high porosity for gas transfer to occur and particle size to provide active sites for the reaction. A few fabrication processes were discussed with a focus on the basic advantages, characteristics and technical challenges of the techniques. In addition, a summary of recent works on cathode materials was presented at the end of the review. From the summary, the screen printing technique is most commonly used in recent works, considering its simplicity and ability to produced cathode films comparable to those produced by other complex techniques. However, other techniques are still being used to produce cathode materials.

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