Characterization of symmetrical SrFe$_{0.75}$Mo$_{0.25}$O$_{3-\delta}$ electrodes in direct carbon solid oxide fuel cells

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ABSTRACT

Symmetrical solid oxide fuel cells (SOFCs) that feature nanostructured SrFe$_{0.75}$Mo$_{0.25}$O$_{3-\delta}$ (SFMO) electrodes which are impregnated into porous La$_{0.9}$Sr$_{0.1}$Ga$_{0.8}$Mg$_{0.2}$O$_{3-\delta}$ (LSGM) skeletons by solution infiltration technique are prepared. The symmetrical SOFCs are investigated in detail by operating with 5 wt% Fe-loaded activated carbon as fuel and ambient air as oxidant. A typical single cell can give a maximum power density of 405 mW cm^{-2} at 850 °C, which is comparable to the results with Ni-based anode-supported SOFCs. Also, the direct carbon SOFC (DC-SOFC) with reloaded fuel can recover to its initial performance. The microstructures and XRD of the electrodes after test are also examined and analysed. The results show that the SFMO-LSGM composite anode remains porous and maintains its structure stability, implying that the symmetrical SFMO electrodes can be applied into DC-SOFCs.

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1. Introduction

Recently, solar power, wind, tidal energy, nuclear energy, hydrogen and other clean energy sources are attracting more and more attentions with increasingly severe energy and environment crisis, they are still far from meeting the energy demand for human beings, however, because of high cost, limited resources and immature technology. Hence, for a long period of time, coal, petroleum, natural gas and other fossil fuels are still dominant in world's energy structure. High efficient and clean energy utilization of fossil fuels has become a focus in all aspects.

Direct carbon fuel cell (DCFC) which can directly convert the chemical energy of carbon into electric energy (electricity for

terminal application) through an electrochemical conversion process has obvious advantages [1]: high theoretical conversion efficiency (the theoretical electrical conversion efficiency of the complete oxidation of carbon: $C + 2O^{2-} = CO_2 + 4e^-$, is slightly higher than 100%, according to the second law of thermodynamics) and low pollution. Hence, it is worthwhile to develop such a promising technology of high efficient and clean coal utilization.

Referring to DCFCs, there are different kinds of DCFCs based on various electrolytes used, such as molten hydroxides and carbonates [2—4] which are both liquid electrolyte, and solid oxide [5—7]. Among these kinds of electrolytes, solid oxide is a uniquely completely solid state electrolyte which can avoid the problem of liquid leaking and corrosion at high temperature [5]. Therefore, direct carbon solid oxide fuel cells (DC-SOFCs) are highly attractive recently, which is a solid oxide fuel cell directly fed by solid carbon as fuel without any liquid metal or feeding gas as medium [8—13].

In 1988, Nakagawa and Ishida [5] first proposed this kind of whole solid state DCFC which used YSZ as electrolyte, platinum porous electrodes both as anode and cathode and charcoal as raw

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fuel. Then, Gür et al. [7] reported a helium fluidized bed fuel cell which converted solid carbon directly into electricity with a peak power density of about 22 mW cm^{-2} at 900 °C. Since then, there have been a few attempts on the development and research of DC-SOFC. Shao et al. [14] reported Ni-ScSZ anode-supported SOFC with ScSZ electrolyte and LSM cathode, which achieved peak power density of 297 mW cm^{-2} at 850 °C by applying catalyst-loaded activated carbon as fuel. After that, Wang et al. [15] reported a conventional tubular Ni-YSZ anode supported SOFC with Ni-ScSZ anode functional layer, ScSZ electrolyte film and LSM-ScSZ cathode with carbon black as fuel. The cell generated a maximum power density of 104 mW cm^{-2} at 850 °C. Our groups have also made research on DC-SOFC [8—13]. Tang [8] fabricated YSZ tubular electrolyte-supported SOFCs with Ag-GDC as anode and Ag as cathode operated on Fe-loaded carbon fuel. The cell showed a significantly improved performance of 45 mW cm^{-2} than the cell without any catalyst. Bai et al. [9] have demonstrated the potential of high performance battery of DC-SOFC with high volumetric power density and operation safety. The mechanism of anode reaction in DC-SOFCs have been verified by Xie et al. [10]. It shows that the reactions at the anode chamber involve the Boudouard reaction and the electrochemical oxidation of CO.

Hitherto, the mostly commonly electrolyte used in DC-SOFCs is zirconia-based material, such as yttria-stabilized-zirconia (YSZ) or scandium-stabilized zirconia (ScSZ). The strontium- and magnesium-doped lanthanum gallate (LSGM) has been considered as a promising alternative electrolyte material with higher oxygen ion conductivity than that of YSZ [16—18]. Recently, symmetrical SOFCs have gained pretty much attention using the same material as both the anode and cathode [19]. This will bring many attractive advantages, for instance, simplified fabrication process and cell structure, reduced materials and interfaces numbers and also better sulfur and coking tolerance. Among the symmetrical electrode materials, the perovskite molybdenum doped strontium ferrite (SFM) shows high electrical conductivity in both oxidizing and reducing environments and excellent redox stability, and exhibits promising performance as electrodes in symmetrical SOFCs [20—22]. As there are few reports on symmetrical electrodes and thin film LSGM electrolyte applied in DC-SOFCs, it is worthwhile to probe the feasibility of symmetrical SFM electrode operated directly on carbon fuel.

Here we report the electrochemical properties of symmetrical solid oxide fuel cells that feature nanostructured $SrFe_{0.75}Mo_{0.25}O_{3-\delta}$ (SFMO) electrodes which are impregnated into porous $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) skeletons by solution infiltration technique. The SOFCs are investigated in detail by operating with hydrogen and Fe-loaded activated carbon as fuel, respectively, including the output performance, AC impedance, discharge testing. The microstructures and XRD of the electrodes before and after test and degradation reasons are also examined and analysed.

2. Experimental

2.1. Preparation of the symmetrical fuel cells

Thin $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) electrolyte SOFCs with symmetrical $SrFe_{0.75}Mo_{0.25}O_{3-\delta}$ (SFMO) electrodes were fabricated by a two-step process in our previous work [21,22]. First, a porous |dense| porous LSGM tri-layer structure — a 17 μm thick dense layer sandwiched between two 160 μm thick porous layers — was produced by laminating three tape-casting ceramic green tapes, with 40 wt% starch as the fugitive materials for the two porous layers followed by co-sintering at 1450 °C to densify the LSGM electrolyte layer. Second, the SFMO electrode catalysts were coated on the

porous LSGM layers by solution infiltration technique. Nitrate solutions containing $Sr(NO_3)_2$, $Fe(NO_3)_3 \cdot 9H_2O$, $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$ and citric acid in a molar ratio of 1: 0.75: 0.0357: 2 was infiltrated into the porous LSGM skeletons and calcined at 850 °C for 2 h. The final loading of the infiltrated SFMO catalysts in the porous LSGM layers was estimated by weight difference before and after each infiltration/calcination cycle, and appropriate numbers of cycles were used to introduce a sufficient amount of SFMO catalysts into the LSGM skeletons.

2.2. Preparation of activated carbon fuel with Fe-loaded catalyst

Activated carbon (Taishan Yueqiao Reagent Plastic Co. Ltd; A. R.) was adopted as the fuel and 5 wt% Fe catalyst for the boudouard reaction was loaded on the activated carbon by impregnation method [8]. $Fe(NO_3)_3 \cdot 9H_2O$ and activated carbon powder with a mass ratio of Fe:C = 1:19 were mixed in deionized water. Then the mixed solution was uniformly stirred at 80 °C for 12 h to evaporate the water and get a colloid. The colloid was thoroughly dried at 120 °C, followed by calcining at 800 °C for 30 min under argon stream of 30 mL min^{-1} to decompose the nitrate.

2.3. Single cell assembling

To avoid the short circuit of the symmetric electrodes, the single cell disk was attached to one end of a corundum alumina tube by using insulated ceramic sealing agent as sealing and jointing material. Silver paste (Shanghai Research Institute of Synthetic Resins, Shanghai, China) was used to make current collector on the electrode surfaces and silver wires were used as the voltage and current leads, as shown in Fig. 1. The effective cathode area was 0.64 cm^2 for the single cell.

2.4. Characterization

The phase composition of the as prepared composite electrode was analysed by X-ray diffraction (XRD) with a Rigaku D/max-IIIA diffractometers (Cu-Kα radiation, operated at 35 kV, 30 mA, λ = 0.15418 nm) at 293 K. The selected 2θ range was from 20° to 80°, scanning in increments of 0.02°. The cell performances including current-voltage curves and electrochemical impedance spectra were measured by a four-probe method using Iviumstat electrochemical analyzer (Ivium Technologies B.V., Netherlands). The electrochemical impedance spectra were measured with voltage amplitude of 10 mV over a frequency range of

Fig. 1. The picture of the assembled single cell.

0.1 Hz−100 kHz [23,24]. The cells were heated and tested at temperatures ranging from 650 °C to 800 °C with the anode exposed to humidified (3% H_2O) hydrogen at a flow rate of 100 mL min^{-1} and cathode to ambient air. After the hydrogen fuel testing, the cells were cooled down to room temperature for the next carbon fuel testing. As shown in Fig. 2, 0.5 g of Fe-loaded carbon fuel was fed into the anode side through the ceramic tube with some ceramic cotton stuffed into the tubes to fix the carbon fuel. The open end of the ceramic tube was sealed by a gas-guide tube to lead the produced gases out. The DC-SOFC single cells were heated and the electrochemical performances were tested at 850 °C. The microstructures of the cells after testing were characterized by scanning electron microscope (SEM, Zeiss Suppra 55VP) equipped with an energy dispersive X-ray spectroscopy (EDX).

3. Results and discussion

3.1. XRD analysis and microstructures of the symmetrical fuel cells

Fig. 3 shows the XRD pattern of the as prepared SFMO-LSGM composites by infiltration technique. It confirms that the predominant phase of the perovskite oxides forms in the infiltrates, simultaneously, the infiltrated SFMO electrode and LSGM electrolyte backbone are chemical compatible, as their independent phase can be found in the XRD. The microstructure and morphology of the symmetric fuel cells are shown in Fig. 4. As can be seen in Fig. 4(a), a dense LSGM layer with a thickness of 17 μm sandwiched between two 160 μm thick porous layers are successfully fabricated. In the porous layers, the average pore size is about 3 μm and the porosity is about 45% measured by the Archimedes method. Fig. 4(b) shows a higher magnification image of the infiltrated composite SFMO-LSGM electrodes, displaying SFMO catalyst of nano-scale structure with average particle size of ~70 nm. The infiltration technique which involves no high-temperature processing steps makes the electrodes a high surface area with nanostructures which is beneficial to the triple phase boundaries of the electrodes.

3.2. Cell performances operated on humidified hydrogen fuel

To evaluate the characteristics of the novel symmetrical electrode material, electrochemical performance tests were carried out on the symmetrical fuel cells with humidified hydrogen as fuel and ambient air as oxidant at 650−800 °C. Fig. 5(a) presents the typical I-V-P curves of the cell. The open circuit voltage (OCV) decreases from 1.051 to 1.024 V in the increasing temperature range of 650−800 °C, which is lower than the calculated theoretical value due to the gas leakage from the cell and ceramic sealings. The presented symmetrical fuel cell can provide a maximum power density of 0.63, 0.54, 0.44 and 0.33 W cm^{-2} at 800, 750, 700 and 650 °C, respectively. This result is even better than that of the previous symmetrical fuel cell with 18 μm thick LSGM electrolyte and $La_{0.6}Sr_{0.4}Fe_{0.9}Sc_{0.1}O_{3-\delta}$ (LSFSc) electrodes, which gives a
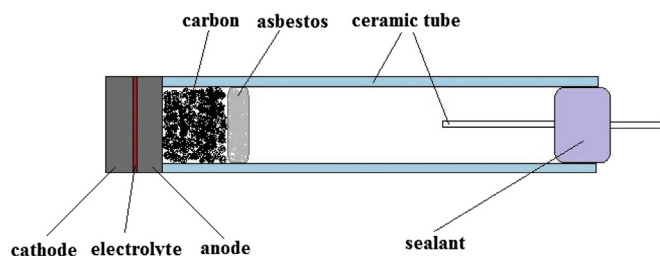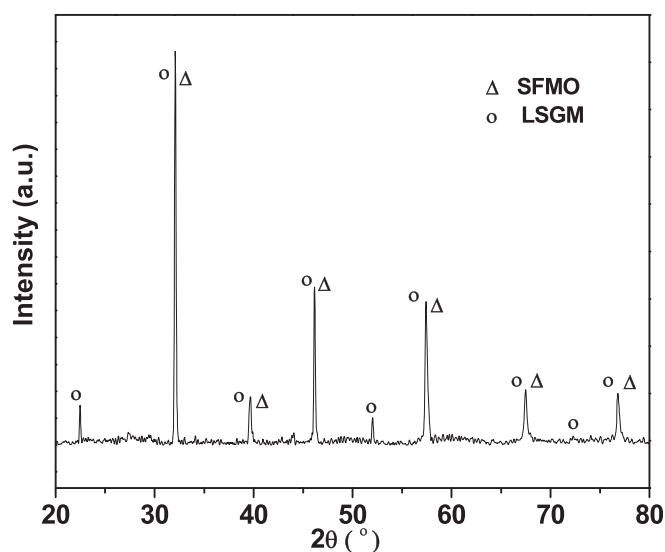

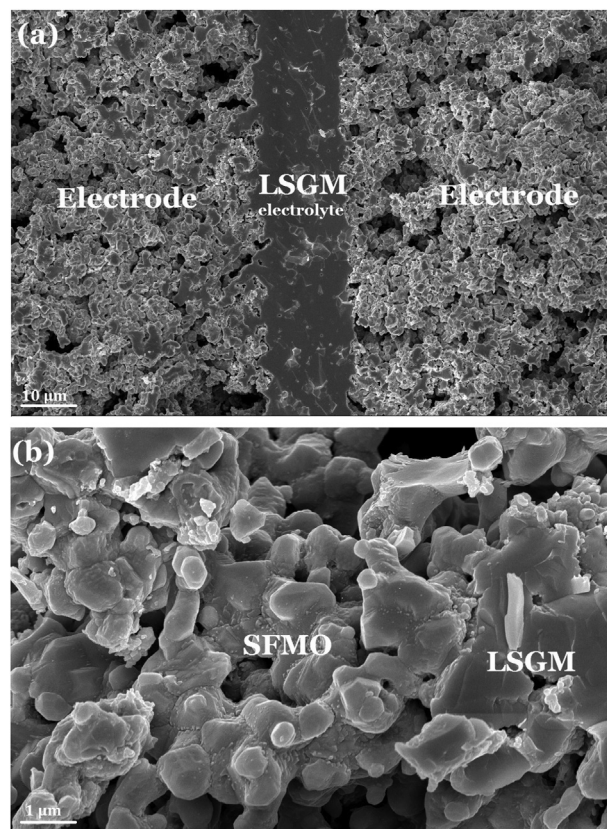**Fig. 3.** The XRD pattern of the prepared SFMO-LSGM composite electrode.

Fig. 4. SEM images of cross section of the symmetrical fuel cells: (a) a low magnification image of tri-layer structure and (b) a higher magnification image of the electrode.

Fig. 2. Schematic illustration of DC-SOFC testing.

maximum power density of 0.56 W cm^{-2} at 800 °C [21]. However, the output performances are still lower than the prior results [22], 0.97 W cm^{-2} at 800 °C. That's because the oxidant of cathode in this study is fed by ambient air versus dry air at 100 mL min^{-1} for the prior study, resulting in an increased concentration polarization. Usually, the concentration polarization is observed in high current density region of the I-V curves. As shown in Fig. 5(a), at 800 °C,
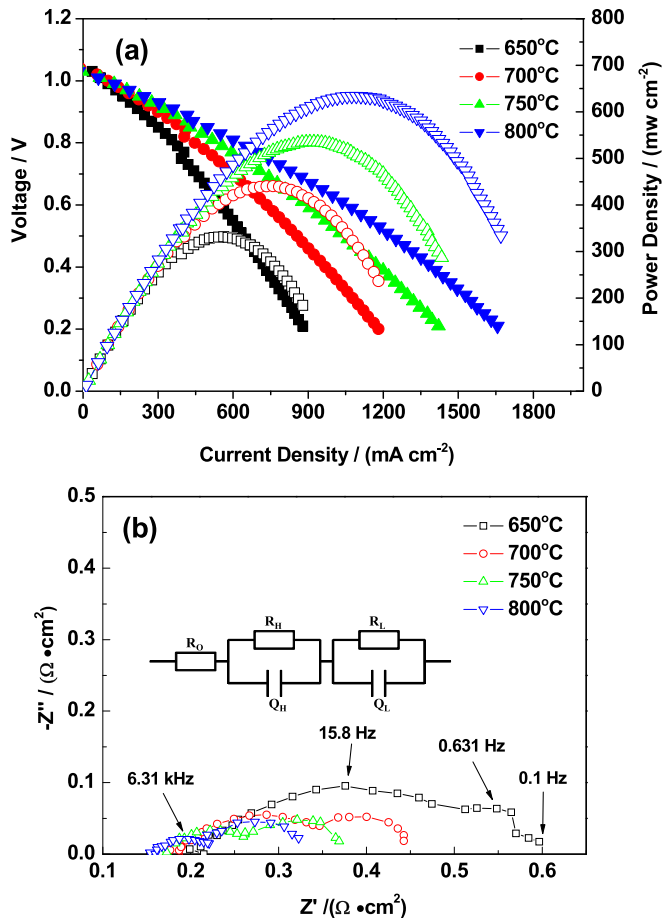
Fig. 5. (a) Typical I-V-P characteristics of the symmetrical cell operated at different temperatures using humidified hydrogen as fuel and ambient air as oxidant. (b) Electrochemical impedance spectra of the symmetrical cell measured under open circuit voltage.

when the current density is smaller than 1000 mA cm^{-2}, the I-V relation is almost a straight line. The I-V curve deviates from the line more and more seriously as the current increases when I >1200 mA cm^{-2}, which is attributed to the concentration polarization.

The representative electrochemical impedance spectra of the symmetrical fuel cell measured under open circuit voltage are shown in Fig. 5(b). Also, the equivalent circuit for the impedance spectra, $R_O(R_HQ_H)(R_LQ_L)$, is given. Here, R_O is the ohmic resistance. R_H and R_L are the polarization resistances corresponding to the high- and low-frequency arcs, respectively, Q_H and Q_L are the corresponding constant phase elements. As can be seen, two depressed arcs appear at all temperature. Table 1 lists the exact data obtained from the impedance measurements, containing the ohmic resistance R_O, the resistances respectively fitted form the high and low frequency arcs, R_H and R_L, and the total resistance R_T. As can be seen, the low-frequency arc remains almost constant, which is

probably associated with gas transport through the porous LSGM skeleton or gas adsorption on the catalyst surface with the nature of temperature-independent. On the contrary, the high-frequency arc follows an Arrhenius dependence with temperature due to the thermally-activated nature of charge transfer reactions or surface diffusions [22]. The combination of anode and cathode interfacial polarization resistance were 0.17, 0.20, 0.27 and 0.40 Ω cm^2 at 800, 750, 700 and 650 °C, respectively.

3.3. Cell performances operated on activated carbon

It has been well understood that the reactions occur in a DC-SOFC operated at high temperature contain the electrochemical oxidation of CO at the anode/electrolyte interface

$$CO + O^{2-} = CO_2 + 2e^- \tag{1}$$

and the Boudouard reaction on the surface of carbon fuel

$$C + CO_2 = 2CO \tag{2}$$

In a DC-SOFC, CO (the carbon near the electrolyte reacts with the oxygen ions to produce CO and electricity) diffuses to the anode/electrolyte interface to produce CO$_2$ and electricity for reaction (1). Afterwards, the CO$_2$ diffuses to the surface of the solid carbon fuel for reaction (2) to produce sufficient CO. To keep continuous operating of the cell, the rate of the reaction (1) and (2) must match well so that enough CO produced from reaction (2) can be provided for reaction (1) to maintain required current. Once the balance of the cell reaction cycle is broken, the cell performance will degrade rapidly.

Fig. 6 shows the stability test result of symmetrical SFMO electrode-based DC-SOFC operated at a constant current of 128 mA (0.2 A cm^{-2}) at 850 °C. As can be seen, a relatively slow drop in voltage occurs to the cell in the first 2 h, then the cell experiences a

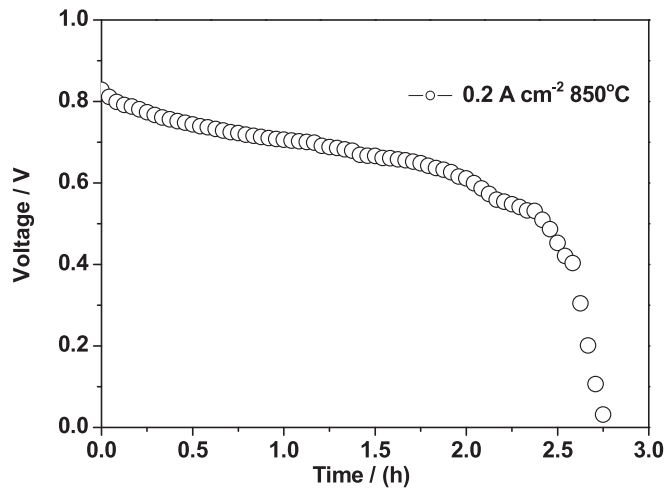**Fig. 6.** Stability test of symmetrical SFMO electrode-based DC-SOFC operated at a constant current of 128 mA (0.2 A cm^{-2}) at 850 °C.

Table 1
Impedance data (Ω cm^2) for symmetrical fuel cell operated on humidified hydrogen at different temperatures.

Temperature	Ohmic resistance R_O	High frequency R_H	Low frequency R_L	Total resistance R_T
650 °C	0.215	0.297	0.085	0.597
700 °C	0.177	0.170	0.096	0.443
750 °C	0.166	0.094	0.109	0.369
800 °C	0.154	0.067	0.102	0.323

rapid degradation to zero in voltage. From reaction (1) and (2), we know that the overall reaction at the anode chamber is $C + O^{2-} = CO + 2e^-$ which is a two electrons-reaction. Through Faraday's law, the fuel utilization of DC-SOFC using 5 wt% Fe-loaded activated carbon as fuel with constant current testing is proportional to the released electric quantity and can be calculated by the formula below:

$$\eta = ItM_C/(95\%nFm) \tag{3}$$

Here, η is fuel utilization, I, t, M_C, n, F and m are running current, running time, molar mass of carbon, molar number ($n = 2$), Faraday constant and the mass of the loaded carbon fuel. In the entire operation life of 2.77 h, the released electric quantity is calculated as 1276 C according to the coulomb law, which corresponds to 79.4 mg carbon consumed. Then, we can calculate the carbon fuel utilization through electrochemical oxidation is about 17% (excluding the amount of Fe catalyst). That is to say about 17% of the carbon has been consumed in DC-SOFC mode during the discharge test.

After the stability test, we opened the cell and found that the unconsumed carbon fuel remained in the anode chamber. As we know, the main reaction of carbon fuel consumption or utilization is the Boudouard reaction on the surface of carbon fuel: $C + CO_2 = 2CO$. There are several factors causing the rate decrease of Boudouard reaction, thus the cell performance degradation: First, the reactive surface area of carbon fuel reduces with the carbon fuel being gradually consumed; Second, the sintering of the carbon fuel during operation at high temperature, which also causes a decrease of reactive surface area; Third, the degradation of the Fe catalyst. Some proper additives, such as chemical stable ceramic powder, may be mixed with the carbon fuel to prevent it from sintering.

Fig. 7 shows the typical curves of voltage and output power density versus current density for the symmetrical fuel cell operated at 850 °C using 5 wt% Fe-loaded activated carbon as fuel and ambient air as oxidant before and after stability test. As can be seen, the cell with Fe-loaded activated carbon as fuel can give an OCV of 1.0 V at 850 °C which is lower than the theoretical expectation due to the probable gas leakage from the cell and ceramic sealings. Also, the maximum power density of symmetrical SFMO electrode-based DC-SOFC is encouraging, 405 mW cm^{-2}, which is much higher than the results reported by other groups [14,15] and is comparable to the results with Ni-based anode-supported SOFCs, with yttria-

stabilized zirconia (YSZ) electrolyte and La$_{0.8}$Sr$_{0.2}$MnO$_3$ (LSM) cathode reported by Bai et al. (424 mW cm^{-2} at 850 °C) [9]. Also, the cell performance after stability test is shown in Fig. 7(b). We can observe that serious deterioration occurs to the cell after the stability test: the open circuit voltage of the cell declines from 1 V to about 0.47 V and the maximum power density reduces from 405 mW cm^{-2} to 21 mW cm^{-2}. We attribute the a lower OCV and rapid performance degradation of the cell after stability test to the decrease of CO concentration (rate decrease of the reaction (2)) which might be caused by the consumption and the sintering of the carbon fuel. Because the OCV of a DC-SOFC is proportional to logarithm of the ratio of partial pressure of CO to CO$_2$ (P_{CO}/P_{CO2}). With the carbon fuel being gradually consumed, the reactive surface area of carbon fuel reduces, which leads to the rate decrease of the reaction (2) and the reduction of CO production. As demonstrated by Xie et al. [10] that the anode reaction of a DC-SOFC operated at high temperature is the electrochemical oxidation of CO, the output performance of the cell is strongly affected by the lower CO production at the anode.

To clarify the reason of performance degradation of DC-SOFC, the initial cell was cooled down to room temperature and reloaded with the same amount of fresh 5 wt% Fe-loaded activated carbon fuel to investigate the restarting ability of the cell. As can be seen in Fig. 7(c), the DC-SOFC with reloaded fuel can recover its initial performance, including OCV (1.01 V), maximum power density (404 mW cm^{-2}) and current density (1209 mA cm^{-2} at 0.2 V), which is almost the same as the initial data. These results indicate that the cell performance degradation is mainly caused by carbon fuel consumption rather than the quality of the symmetrical fuel cell itself. This conclusion is also supported by some previous research [25,26].

Fig. 8 shows the electrochemical impedance spectra of the cell operated on carbon fuel at 850 °C under open circuit voltage before and after stability test. As can be seen, the ohmic resistance and the polarization resistance of the cell both increases after stability test. Generally, the ohmic resistance of a SOFC with YSZ electrolyte and traditional Ni-based anode, especially of electrolyte supported SOFC, comes mainly from the electrolyte. However, in the present work, the electrolyte is very thin compared to the electrodes, and the electronic conductivity of the ceramic electrode material (SFMO) is lower than that of the Ni metal. Therefore, the electrodes
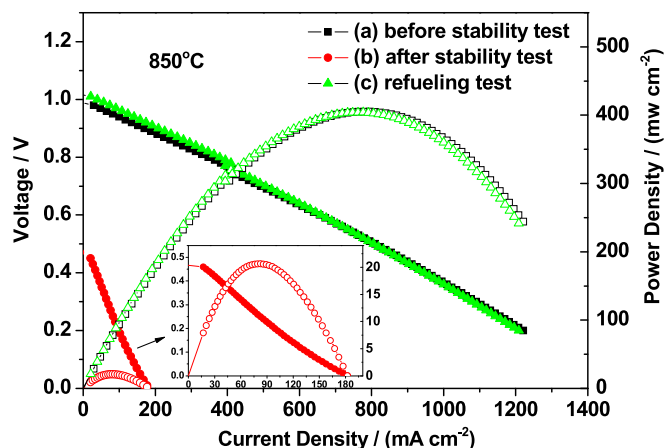

Fig. 7. The typical electrochemical performance of the symmetrical fuel cell operated at 850 °C using 5 wt% Fe-loaded activated carbon as fuel: (a) before stability test; (b) after stability test and (c) refueling test.
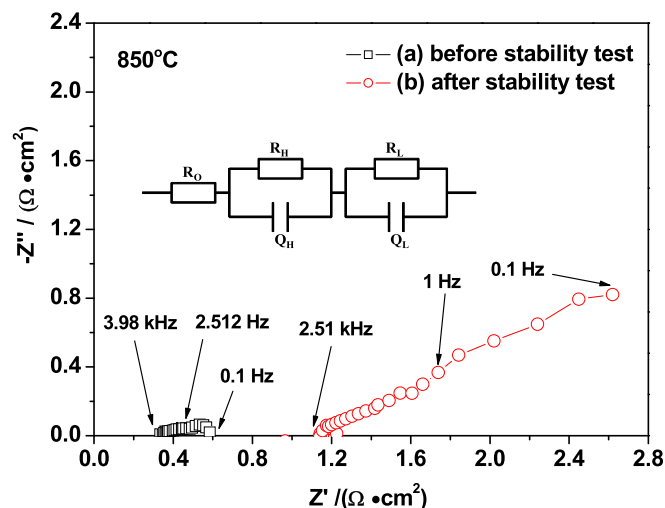

Fig. 8. Electrochemical impedance spectra of the symmetrical fuel cell operated on carbon fuel at 850 °C under open circuit voltage: (a) before stability test and (b) after stability test.

may contribute much to the overall ohmic resistance of the SOFC. As the conductivity of the SFMO varies with gas atmosphere, the ohmic resistance of SOFC with SFMO-based anode will change with fuels. The results shown in Fig. 8 suggest that the electronic resistance of SFMO in CO (DC-SOFC) is larger than that in H$_2$. Comparing to the increase of ohmic resistance, the polarization resistance increases much more, which is twice as much as the increase of ohmic resistance. This indicates that the deterioration of the symmetrical fuel cell after stability test is mainly caused by the electrode polarization, including electrode or electrode-electrolyte

interface degradation (the consumption of carbon fuel or the degradation of Fe catalyst).

After stability test with carbon fuel, the cross-sectional SEM image, the EDX spectra and the XRD pattern of the anode are shown in Fig. 9. As presented in the SEM image, there's nearly no change with the microstructure of the SFMO-LSGM anode. The anode remains porous and the catalyst particles don't coarsen over stability test time. From Fig. 9(a), the EDX spectra indicate that all the elements of the $SrFe_{0.75}Mo_{0.25}O_{3-\delta}$-$La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ anode still keep in stoichiometric composition, implying this kind of electrode

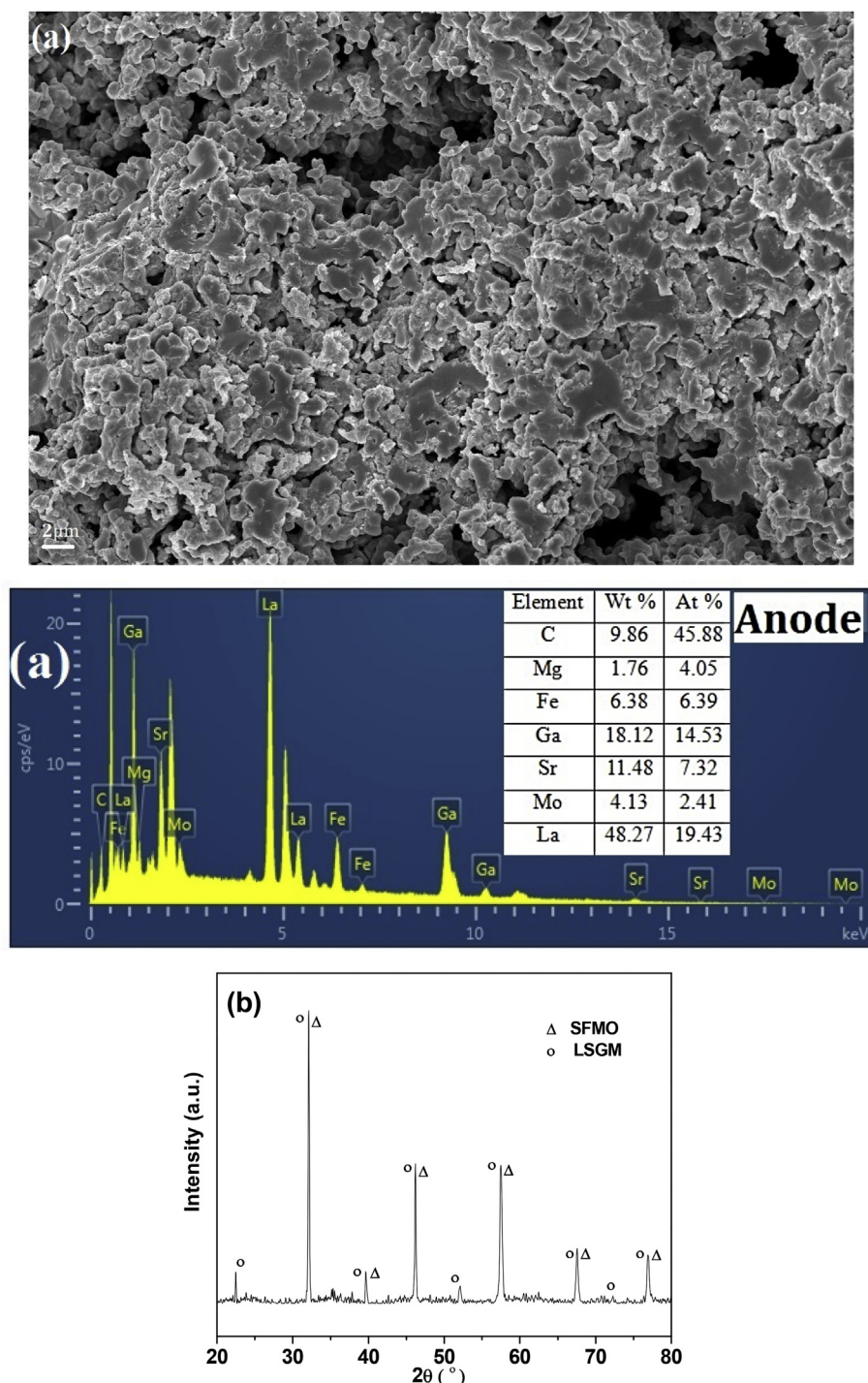| Element	Wt %	At %
C	9.86	45.88
Mg	1.76	4.05
Fe	6.38	6.39
Ga	18.12	14.53
Sr	11.48	7.32
Mo	4.13	2.41
La	48.27	19.43

Fig. 9. (a) SEM image and the EDX spectra; (b) XRD pattern of cross section of the SFMO anode operated on carbon fuel after stability test.

will maintain its structure stability very well after lifetime testing operated on carbon fuel. Note that there is still a carbon peak in the EDX, it may attribute to contamination of carbon fuel in the anode chamber as SFMO-LSGM composites showed good coking tolerance at elevated temperatures in the previous studies [22]. To further verify the stability of the SFMO-LSGM anode after test, XRD characterization was measured, as shown in Fig. 9(b). Obviously, there is no impurity phase arising except the perovskite oxides phase of SFMO and LSGM. Combining with the reload ability of the symmetrical fuel cell in Fig. 7(c), it demonstrates that the SFMO-LSGM anode keeps its stability well and has shown the promise of serving as electrode materials for DC-SOFC. Conclusively, there is no issues arisen from the SFMO-LSGM composite electrodes in DC-SOFCs. The reason of degradation of DC-SOFCs is mainly from fuel situation, which can be solved by continuous add of carbon fuel with larger anode chamber spaces from external anode. This work is being carried out in out lab.

4. Conclusions

In this paper, thin $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-\delta}$ (LSGM) electrolyte SOFCs with symmetrical $SrFe_{0.75}Mo_{0.25}O_{3-\delta}$ (SFMO) electrodes were fabricated by laminating three tape-casting ceramic green tapes (a porous|dense| porous LSGM tri-layer structure), followed by infiltrating SFMO electrode catalysts into the porous LSGM layers. The SOFCs are operated with hydrogen and Fe-loaded activated carbon fuel, yielding a maximum power density of 630 mW cm^{-2} at 800 °C and 405 mW cm^{-2} at 850 °C, respectively, which is excellent and comparable to the results with Ni-based anode-supported SOFCs. The DC-SOFC with reloaded fuel can recover to its initial performance and the SEM-EDX and XRD analysis of the anode after test shows that the SFMO electrode remains porous and maintains its structure stability, implying the prospect of applying symmetrical SFMO electrodes as electrode materials for DC-SOFCs.

Acknowledgements

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