Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Power Sources 117 (2003) 26–34
JOURNAL OF POWER SOURCES
www.elsevier.com/locate/jpowsour

Effect of characteristics of Y_2O_3/ZrO_2 powders on fabrication of anode-supported solid oxide fuel cells

Yong Jun Leng[a], Siew Hwa Chan[a,*], Khiam Aik Khor[a], San Ping Jiang[a], Philip Cheang[b]

[a]School of Mechanical and Production Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore
[b]School of Materials Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

Received 16 December 2002; accepted 5 February 2003

Abstract

A comparative study is carried out on the effect of nano-sized and micron-sized Y_2O_3/ZrO_2 (YSZ) powders on the fabrication and performance of anode-supported solid oxide fuel cells. It is found that pellets made of nano-sized YSZ powder can achieve a relative density of 96% at a sintering temperature of 1400 °C and 92% at sintering temperature as low as 1200 °C. For pellets made of micron-sized YSZ powder, densification only occurred at a sintering temperature of 1400 °C. On co-sintering the nano-sized YSZ electrolyte film with the anode support/substrate, the electrolyte is unable to sinter fully at 1400 °C, but forms a porous structure which leads to a reduced open-circuit potential and poor cell performance. This is most likely due to the nano-sized YSZ electrolyte thin film having a very low green density and there being a significant difference in the sintering behaviour of the YSZ thin layer and the Ni/YSZ cermet substrates. The sintering behaviour and the nature of YSZ powders exert a significant effect on the fabrication and performance of Ni/YSZ anode-supported thin YSZ electrolyte cells.

Keywords: Anode-supported solid oxide fuel cell; Sintering; YSZ powder; Thin-film electrolyte

1. Introduction

Conventional solid oxide fuel cells (SOFCs) based on a thick yttria-stabilized zirconia (YSZ) electrolyte as support are required to be operated at high temperature (~1000 °C) in order to have sufficient ionic conductivity. High temperature operation over a long period will, however, incur many problems such as limited material selection, high material cost and fast performance degradation of cell components [1–5]. Therefore, it is desirable to reduce the operation temperature of the fuel cell to 600–800 °C in order to widen the selection of materials for electrode, electrolyte, interconnect and cell manifolds and to improve long-term stability [6]. One of the challenges associated with lowering the operating temperature is the resulting significant increase in the resistivity of the YSZ electrolyte. There are two approaches to overcome this problem, one is to use alternative electrolyte materials with higher ionic conductivity than YSZ, such as doped ceria and doped $LaGaO_{3-\delta}$ [6–12], and the other is to reduce the thickness of the YSZ electrolyte [13–17]. There has been significant progress recently in the development of intermediate temperature anode-supported SOFCs with thin-film YSZ electrolytes. For example, de Souza et al. [13] reported that a thin-film SOFC with a YSZ electrolyte of ~10 μm, a Ni/YSZ anode and a $La_{1-x}Sr_xMnO_3$ (LSM)-based cathode can achieve a high power density of ~1.8 W cm^{-2} at 800 °C.

Most of the high-performance anode-supported SOFCs reported to date are based on thin-film electrolytes made of YSZ powders with an average particle size of ~0.5 μm (e.g. Tosoh TZ-8Y powder) [13–15]. The sintering temperature for such powders is generally in the range 1350–1400 °C in order to form a dense electrolyte and a good electrode and electrolyte interfacial contact [13,14,17]. For an anode-supported SOFC prepared by tape casting and lamination techniques, the control of the flatness of large-plate cells becomes difficult due to bending and different shrinkage of cell components at high sintering temperatures. Chen et al. [18] studied the sintering behaviour of YSZ powders with particle sizes of 50–60 nm and found that pellets made from these powders can achieve ~94% of the theoretical density at a sintering temperature as low as 1200 °C. This suggests that for a thin-film electrolyte made of nano-sized YSZ powders, it may be possible to sinter an anode-support/electrolyte bilayer at reduced temperatures. A low sintering

* Correspondig author.
E-mail address: mshchan@ntu.edu.sg (S.H. Chan).

0378-7753/03/$ – see front matter © 2003 Elsevier Science B.V. All rights reserved.
doi:10.1016/S0378-7753(03)00350-1

temperature not only improves the quality and flatness of the anode-supported thin electrolyte cells but also reduces the sintering and grain growth of the Ni phase in the Ni/YSZ anode substrate. This, in turn, increases manufacturing productivity and cell performance. In the work reported here a study is made of the characteristics of YSZ powders and their sintering behaviour on the fabrication process and performance of an anode-supported SOFC with a thin-film YSZ electrolyte.

2. Experimental

Two different 8 mol% Y_2O_3/ZrO_2 (YSZ) powders were obtained from Nextech Materials, USA, and Tosoh, Japan. The YSZ powder from Nextech has an average particle size of 50–60 nm and specific surface area of 120–130 $m^2\ g^{-1}$ with a crystallite size of less than 10 nm (termed nano-sized YSZ powder). For Tosoh YSZ powder, the average particle size is 0.5 μm, the specific surface area is 6.1 $m^2\ g^{-1}$, and the crystallite size is ∼56 nm (termed micron-sized YSZ powder). The sintering behaviour of the powder was studied on pellets made from both micron-sized and nano-sized YSZ powders. YSZ pellets were made by conventional ceramic processing, i.e. mixing and uniaxial pressing (100 MPa), followed by sintering. The pellets were divided into two batches. The first batch was sintered at 1400 °C for 2 h, and the second batch was sintered at 1200 °C for 2 h to facilitate a comparison of the sinter-ability for both powders. The heating and cooling rates were both 5 °C min^{-1}. The final dimensions of the green pellets were ∼24 mm in diameter and ∼1 mm in thickness. The density of green samples and porous sintered samples was estimated by the dimensions and the weight of the samples, while the density of dense sintered samples was measured by means of the Archimedes technique.

The 8 mol% Y_2O_3/ZrO_2 (TZ8Y, Tosoh) and NiO (J.T. Baker) were used to prepare Ni/YSZ cermet anode substrates. The powder was mixed in a composition of 56 wt.% NiO and 44 wt.% YSZ to make all anode support/substrates. The 3 wt.% of binder (polyvinyl alcohol) and 1 wt.% of surfactant (Triton X-100, Aldrich) were added to the mixture before the ball milling. The anode powder was then compacted under uniaxial pressure to form a disc. The green anode discs were subsequently baked at 900 °C for 1 h to reduce the shrinkage of the substrate. The suspensions of YSZ powder from Tosoh Corporation and Nextech Materials were prepared by mixing with suitable organic additives. The YSZ suspension was then applied on one side of the anode using a spray-coating technique. The coated Ni/YSZ cermet substrate was fired at 1400 °C for ∼2 h. Platinum paste was applied on the electrolyte side of the anode-supported cell by a screen-printing technique, to form the cathode, followed by sintering at 1100 °C for 30 min. In order to estimate the green density of the spray-coated YSZ film, YSZ films with a thickness of ∼100 μm were prepared by a tape-casting technique, using YSZ slurry prepared from the same formula as that for the YSZ suspension used in spray coating, except that the amount of solvents was different. The compositions of the suspension and slurry of YSZ powders are listed in Table 1.

Table 1
Composition of slurry used for tape-casting YSZ film and of suspension used for spray-coating YSZ thin film on Ni/YSZ cermet anode substrate

Composition	Function	Slurry used for tape-casting film (g)	Suspension used for thin film on anode support (g)
YSZ	Ceramic powder	40.00	2.00
Trichloroethylene	Solvent	24.00	4.00
Ethanol	Solvent	6.00	36.00
Corn oil	Dispersant	2.00	0.10
Benzyl butyl phthalate	Plasticizer	2.00	0.10
Polyethylene glycol	Plasticizer	2.00	0.10
Polyvinyl butyral	Binder	1.40	0.07

The ionic conductivity of YSZ pellets was measured by a frequency response analyzer (FRA, Model 1260, Solartron, UK). Each sample was held in a purpose-built holder which was fitted in a vertical tube furnace. The YSZ pellets were coated with a silver paste on both sides to serve as electrodes, and the pellets were fired at 850 °C for 30 min. Platinum (Pt) gauze was used as a current-collector. Two platinum wires were connected to each platinum gauze on the anode and cathode of the pellets to serve as separate voltage and current probes. The purpose of using separate voltage and current Pt wires is to eliminate the effect of Pt wire resistance on the cell impedance. Measurements were carried out in the temperature range of 250–600 °C and the frequency range of 10 MHz to 0.1 Hz with an amplitude of 50 mV.

The cell performance of the SOFC was evaluated using an in-house test station. Humidified H_2 (3 vol.% H_2O) was fed to the anode chamber, while the cathode was exposed to open air. The anode side was sealed with ceramic paste (Ceramabond 668, Aremco Products Inc., USA). Pt gauze was used as a current-collector for both anode and cathode. As with ionic conductivity measurements, there were separate Pt voltage and current probes. The electrochemical measurements were conducted using the Autolab PG30/FRA system (Eco Chimie, The Netherlands). The current–voltage (*i–V*) characteristics of the cells were measured using linear sweep voltammetry at a sweep rate of 1 mV s^{-1}. The impedance of the cells was recorded in the range 100 kHz to 0.1 Hz under open-circuit conditions.

The microstructure and morphology of both the green and the sintered samples and the cells were examined by a JEOL JSM-5600LV scanning electron microscope (SEM).

The particle size was measured using a Fritsch Particle Sizer Analysette.

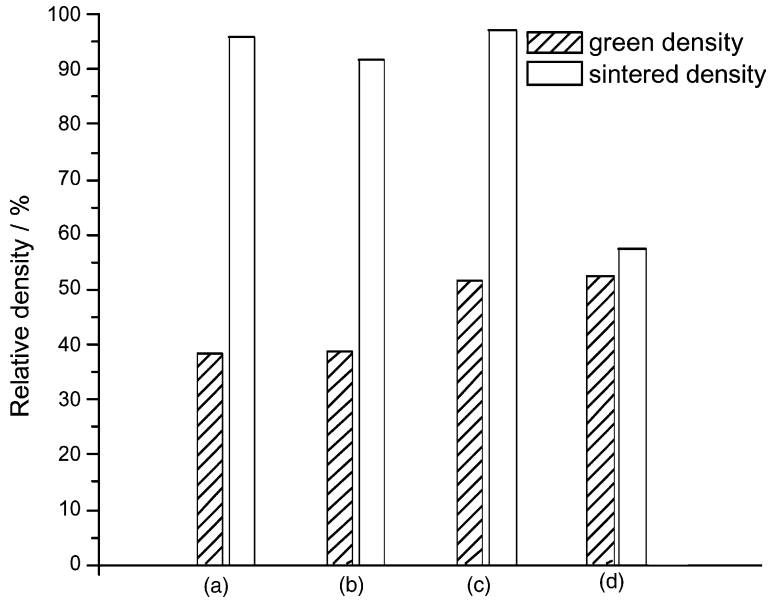

Fig. 1. Relative green and sintered density of YSZ pellets made from: (a) nano-sized YSZ sintered at 1400 °C for 2 h; (b) nano-sized YSZ sintered at 1200 °C for 2 h; (c) micron-sized YSZ sintered at 1400 °C for 2 h; (d) micron-sized YSZ sintered at 1200 °C for 2 h.

3. Results and discussion

3.1. Sintering behavior of YSZ pellets

The variation of the relative green and sintered density of the pellets of YSZ powders before and after sintering at 1200 and 1400 °C is shown in Fig. 1. Though the YSZ pellets made of micron-sized and nano-sized powders were prepared with the same procedure and under the same conditions, there is a significant difference in their green densities, namely, 52 and 39%, respectively. After sintering at 1400 °C for 2 h, both powders achieve relative sintered densities that are higher than 96% of theoretical density. When the sintering temperature is reduced to 1200 °C, however, the pellets made of the nano-sized YSZ powder still have a reasonably good relative density of about 92%, while the relative density of the pellets made of micron-sized YSZ powder was ∼57%, an increase of only ∼5% compared with the green density. The relatively high sintered density of the pellets made of nano-sized YSZ powder sintered at 1200 °C despite the low green density compared with those made of micron-sized YSZ powder indicates a superior sinter-ability of nano-sized YSZ powder. The shrinkage of the pellets made of nano-sized YSZ powder is 27 and 26% at a sintering temperature of 1400 and 1200 °C, respectively, while the micron-sized YSZ pellets experience ∼19% shrinkage at sintering temperature of 1400 °C.

Scanning electron micrographs of the surface of the YSZ pellets sintered at 1400 and 1200 °C for 2 h are presented in Fig. 2. Both nano-sized and micron-sized YSZ pellets sintered at 1400 °C are quite dense with some isolated pinholes. The average grain size of micron-sized and nano-sized YSZ pellets is ∼1.3 and ∼1.7 μm, respectively.

The grain size of micron-sized YSZ pellet is slightly smaller than that of nano-sized YSZ pellet despite the fact that the initial particle size of micron-sized YSZ is larger than that of nano-sized YSZ powder. At a lower sintering temperature of 1200 °C, the pellets made of micron-sized YSZ powder are very porous and show little sintering (Fig. 2c). For pellets prepared from nano-sized YSZ powders, it appears that sintering and densification have already taken place as indicated by the much lower porosity compared with that of micron-sized YSZ powder (Fig. 2d). The significant difference in the sintering and densification behaviour of pellets made of nano-sized and micron-sized YSZ powders can be related to the nature of the sintering behaviour of the powder. It has been shown [19,20] that the maximum shrinkage rate for micron-sized YSZ powder occurs at ∼1300 °C and sintering of YSZ electrolyte based on Tosoh powder typically takes place at 1350–1400 °C. For pellets made of nano-sized YSZ powder prepared by the hydrothermal homogeneous precipitation method with a specific surface area of 144 $m^2\ g^{-1}$, relative densities of ∼94 and ∼98% of theoretical density were achieved at a sintering temperature of 1200 and 1350 °C, respectively [21]. For YSZ powder prepared by co-precipitation, the maximum shrinkage occurred at ∼1050 °C [22], i.e. about 200 °C lower than that of micron-sized YSZ powders. This indicates that the sintering behaviour of micron-sized and nano-sized YSZ powder can be very different. It is reported that pellets made of nano-crystalline 3%Y_2O_3/ZrO_2 (15 nm) powders prepared by a co-precipitation method can achieve the same sintered density at a sintering temperature which is 150 °C lower than that for pellets made of Tosoh 3%Y_2O_3/ZrO_2 powders with a size of 0.17 μm [23].

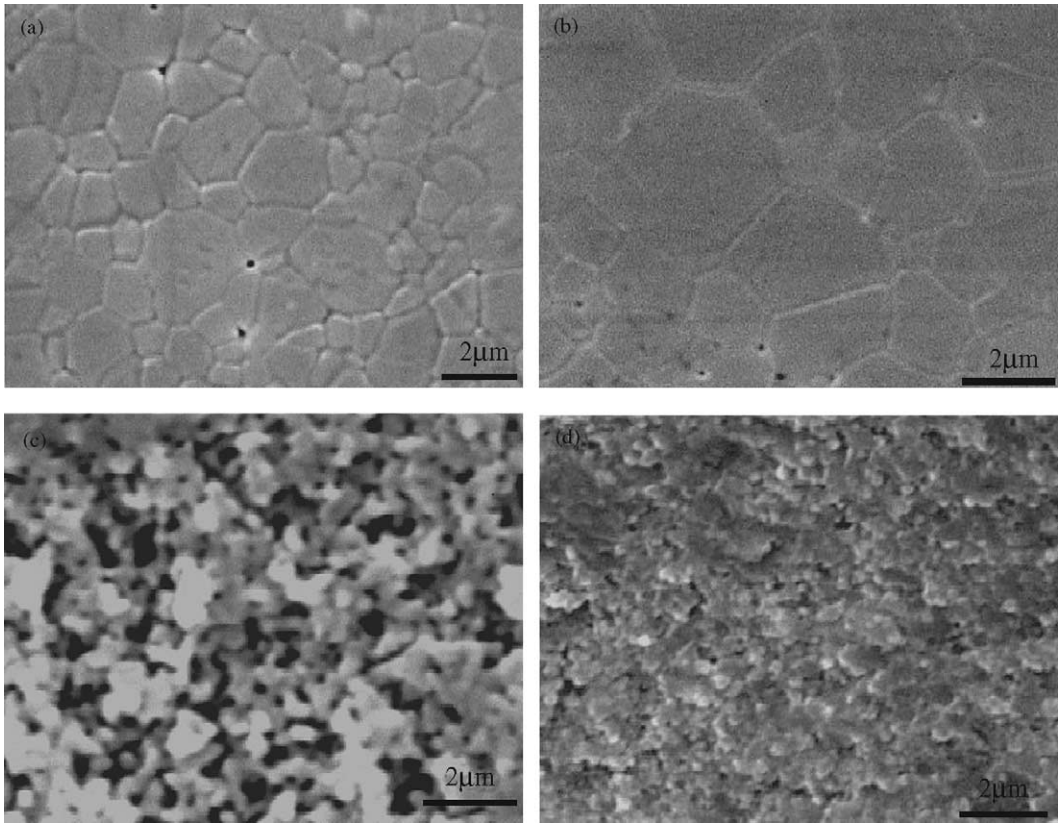

Fig. 2. Scanning electron micrographs of surface of YSZ pellets: (a) micron-sized YSZ sintered at 1400 °C; (b) nano-sized YSZ sintered at 1400 °C; (c) micron-sized YSZ sintered at 1200 °C; (d) nano-sized YSZ sintered at 1200 °C.

The impedance curves for both nano-sized YSZ and micron-sized YSZ pellets measured at 350 °C in air are given in Fig. 3. The impedance spectra of micron-sized YSZ powder sintered at 1200 °C were not measured due to the low mechanical strength of the very porous pellets. Two semicircles at high and low frequencies, which correspond to the resistance of the bulk and grain boundaries, can be clearly identified. Based on the equivalent circuit of an ionic conductor proposed by Bauerle [24], the resistance of the bulk and grain boundaries can be determined by curve-fitting the impedance data with two RC circuits in series. The bulk resistance and grain boundary resistance of micron-sized YSZ measured at 350 °C are 3.32×10^4 and 7.24×10^3 Ω cm, respectively. These values are close to those reported by Steil et al. [25] for a similar Tosoh YSZ powder. The bulk resistance of nano-sized YSZ sintered at 1400 °C is 3.72×10^4 Ω cm, which is similar to 3.78×10^4 Ω cm for the same pellets sintered at 1200 °C for 2 h. Furthermore, the bulk resistances of nano-sized YSZ sintered at 1400 and 1200 °C are close to that of micron-sized YSZ sintered at 1400 °C for 2 h. This indicates that the bulk conductivity of YSZ is not affected significantly by the grain size.

There is, however, a significant difference in the grain boundary resistance (the low frequency arc) of the YSZ specimens. The grain boundary resistance of micron-sized YSZ sintered at 1400 °C is 7.24×10^3 Ω cm, i.e. much smaller than 5.46×10^4 Ω cm for nano-sized YSZ sintered at 1400 °C and 1.56×10^5 Ω cm for nano-sized YSZ sintered at 1200 °C. The grain boundary resistance of YSZ pellets is strongly dependent on the impurity level, porosity and grain size [25–27]. Steil et al. [25] found the porosity has some effect on the grain boundary resistance and little effect on the bulk resistance of Tosoh YSZ pellets sintered at temperatures from 1300 to 1650 °C. Badwal and Drennan [26] studied the ionic conductivity of samples made of 10 mol% Y_2O_3/ZrO_2 powders sintered at different temperatures between 1300 and 1900 °C and also found that porosity affects the grain boundary resistance while the bulk resistance remains unchanged.

From sintered density measurements and SEM observations, the porosity of the pellets made of nano-sized YSZ powders is similar to that of micron-sized YSZ powders after sintering at 1400 °C. In addition, the grain size of micron-sized YSZ pellets is slightly smaller than that of nano-sized YSZ pellets. Therefore, the difference in the grain boundary resistance of YSZ pellets is not due to a difference in particle size and porosity. Traces of impurities, such as silica, can significantly deteriorate the ionic conductivity of the YSZ electrolyte [27]. Thus, the high grain boundary resistance associated with nano-sized YSZ pellets sintered at 1400 °C may be due to the existence of an impurity. The high grain

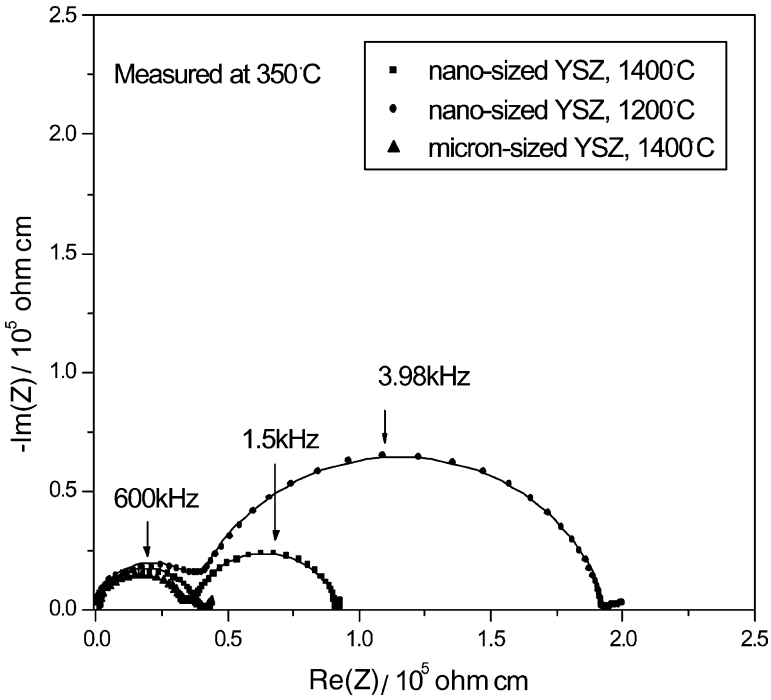

Fig. 3. Impedance spectra of curves of nano-sized YSZ and micron-sized YSZ pellets measured at 350 °C in air. Solid symbols are experimental data and solid lines are fitting results.

boundary resistance of nano-sized YSZ sintered at 1200 °C is most likely due to its high porosity level (∼8%) and small average particle size.

3.2. Sintering behaviour of YSZ thin films on anode substrates

The performance and impedance curves of anode-supported thin YSZ electrolyte cells made of micron-sized and nano-sized YSZ powders at 800 °C are presented in Fig. 4. The impedance curves were measured at open-circuit. The sealing of the cell was checked before the test. The open-circuit potential of the anode-supported cell with micron-sized YSZ thin electrolyte is 1.06 V, while for the nano-sized YSZ thin film it is only ∼0.60 V. This indicates that there may be some cross-over of fuels in the cell made of nano-sized YSZ powders. Using platinum as a cathode, the cell with thin micron-sized YSZ electrolyte achieved maximum power density of 0.25 W cm^{-2} at 800 °C, i.e. seven times higher than the 0.036 W cm^{-2} of the cell with thin nano-sized YSZ electrolyte. From the impedance curves measured at 800 °C (Fig. 4b), it can be readily seen that the ohmic resistance and polarization resistance of the cell with a thin-film electrolyte made of nano-sized YSZ is significantly higher than that of the cell with micron-sized YSZ thin-film electrolyte. The performance of the thin electrolyte cell prepared from nano-sized YSZ powder is inferior to that of the cell made from micron-sized YSZ powders. It should be pointed out that the performance of a SOFC with a thin micron-sized YSZ electrolyte using a Pt cathode is still very poor, since the Pt cathode contributes a high polarization resistance (∼1.8 Ω cm^2 at open-circuit) at 800 °C. Replacing the Pt cathode with a LSM/YSZ composite cathode, an anode-supported SOFC with a thin micron-sized YSZ electrolyte can achieve a maximum power density of 0.98 W cm^{-2} and a total cell resistance of 0.57 Ω cm^2 at 800 °C, as shown in Fig. 5.

The sintering behaviour of a YSZ electrolyte film on Ni/YSZ substrates was also studied by means of scanning electron microscopy. Micrographs of the cross-sections of anode-supported SOFCs and of the surface of the electrolyte thin films made of nano-sized and micron-sized YSZ powders sintered at 1400 °C for 2 h in air are shown in Fig. 6. Micrographs of anode/electrolyte bilayers with electrolyte thin films made of nano-sized and micron-sized YSZ powders fired at 500 °C for 2 h in air are presented in Fig. 7. At a sintering temperature of 500 °C, organic additives in the electrolyte film are decomposed and the micrographs indicate the microstructure status of the green YSZ film. The thickness of green films made of micron-sized and nano-sized YSZ powders is ∼22 and ∼56 μm, respectively. The green film of the micron-sized YSZ layer appears to be denser than that of the nano-sized YSZ layer. After sintering at 1400 °C, the thickness of the micron-sized YSZ film is ∼13 μm while that of the nano-sized YSZ film is ∼33 μm (Fig. 6a and b). The linear shrinkage is ∼40% for both YSZ films. The YSZ film made from micron-sized powders is quite dense with a few closed pinholes (Fig. 6a and c). On the other hand, the YSZ thin film prepared from nano-sized YSZ powders is very porous with many open

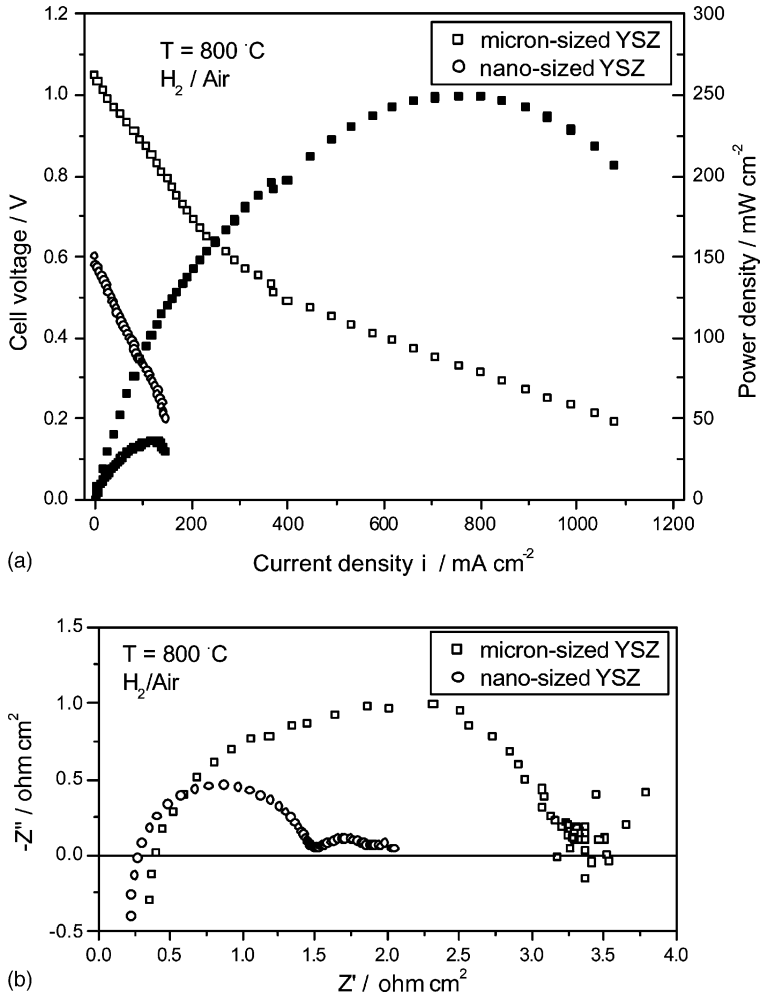

Fig. 4. (a) Cell voltage and power density vs. current density and (b) impedance curves at open-circuit voltage tested at 800 °C for anode-supported solid oxide fuel cells with thin-film YSZ electrolyte made of micron-sized YSZ and nano-sized YSZ powders and with Pt cathode.

pores through the coating (Fig. 6b and d). This indicates that there will be insufficient densification taking place in the YSZ electrolyte film made of the nano-sized YSZ powders despite the high sintering temperature of 1400 °C. The sintering behaviour of the YSZ film on Ni/YSZ substrates is clearly very different from that of the pellets, and this is particularly true for nano-sized YSZ powders. A porous electrolyte leads not only to a sharp decrease in the open-circuit voltage, but also to a significant increase in both ohmic and polarization resistance. This explains the poor cell performance of the thin electrolyte cell prepared from nano-sized YSZ powders.

The green density of the deposited YSZ film was estimated by measuring the green density of YSZ films prepared by tape casting. A YSZ film of ∼100 μm in thickness was prepared by tape casting using the same formula as that for the YSZ slurry in spray coating except for the amount of solvents (see Table 1). The green density of the tape-cast film made of nano-sized YSZ powder is very low, viz. only ∼25%, which is 13% lower than that using micron-sized YSZ powder (∼38%). According to Shi [28], if the green density of ceramics is lower than a critical value, namely, ∼33%, it is difficult to reach full densification under pressureless solid-state sintering conditions due to the existence of thermodynamically stable large pores. The low green density of the YSZ film made of nano-sized YSZ powders would make the densification of the YSZ electrolyte film difficult as large pores in particular have great effect on the densification process. The low green density of nano-sized YSZ films can be related to the existence of high agglomeration as the powders in the suspension tend to agglomerate due to the high surface energy of the nanoscale particles. In the case of pellets made of nano-sized YSZ powders, the green density is 39%, which is high enough to achieve a relatively high sintered density even at a sintering temperature of 1200 °C. In addition, it is found that the shrinkage of the anode-support (NiO/YSZ = 56/44) is ∼18% after sintering at 1400 °C for 2 h. As mentioned above, the shrinkage of pellets made of nano-sized YSZ powder is 27% at a sintering temperature of 1400 °C, which is significantly higher than that of anode support. This mismatch in shrinkage may play a role in the densification of the thin-film

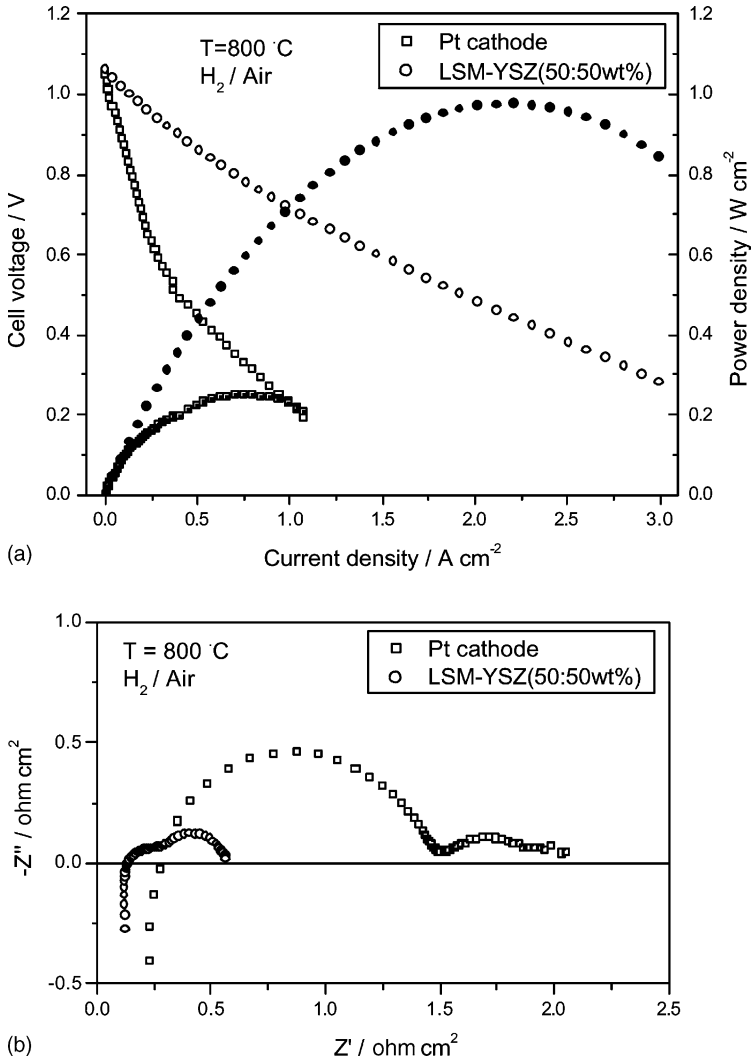

Fig. 5. (a) Cell voltage and power density vs. current density and (b) impedance curves at open-circuit voltage tested at 800 °C for anode-supported solid oxide fuel cells with thin-film YSZ electrolyte made of micron-sized YSZ and with LSM/YSZ (50:50 wt.%).

electrolyte on the anode support. It is noticed that the shrinkage of the pellets made of the micron-size YSZ powders is ∼19% at sintering temperature of 1400 °C, i.e. very close to ∼18% of the Ni/YSZ anode at the same sintering temperature. Thus, in case of sintering of a thin-film electrolyte on an anode support, the film is under compression rather than tension while sintering [29]. The relative sintered density, ρ_T, of ceramic pellets/thin films (herein thin-film YSZ electrolyte on anode support) can be calculated based on the following equation:

$$\rho_T = \rho_0 \frac{L_0}{L_T} \left(\frac{\Phi_0}{\Phi_T} \right)^2 \quad (1)$$

where ρ_0 is relative green density of the thin-film YSZ electrolyte; L_0 and L_T are the thickness of the green and the sintered thin-film YSZ electrolyte, respectively; Φ_0 and Φ_T are the diameter of the green and the sintered thin-film YSZ electrolyte, respectively. The values of these parameters are listed in Table 2. The calculated values of ρ_T of sintered electrolyte thin films made of micron-sized YSZ and nano-sized YSZ powders are ∼95 and ∼62%, respectively. It is clearly seen that, due to the differences in the shrinkage and sintering behaviour between the nano-sized thin-film electrolyte and the anode support, the nano-sized YSZ thin-film electrolyte cannot fully shrink like the pellets, and thus results in a porous structure after sintering at 1400 °C for 2 h. In order to obtain a very dense thin-film electrolyte made of nano-sized YSZ powders, two approaches could be used, one is to increase the green density through reducing the agglomeration of nano-sized YSZ particles, the other is to control carefully the shrinkage of the anode substrate to match to the shrinkage profile of the YSZ thin film. More research will be carried out to co-sinter the nano-sized YSZ thin film/anode-support bilayer at reduced temperatures.

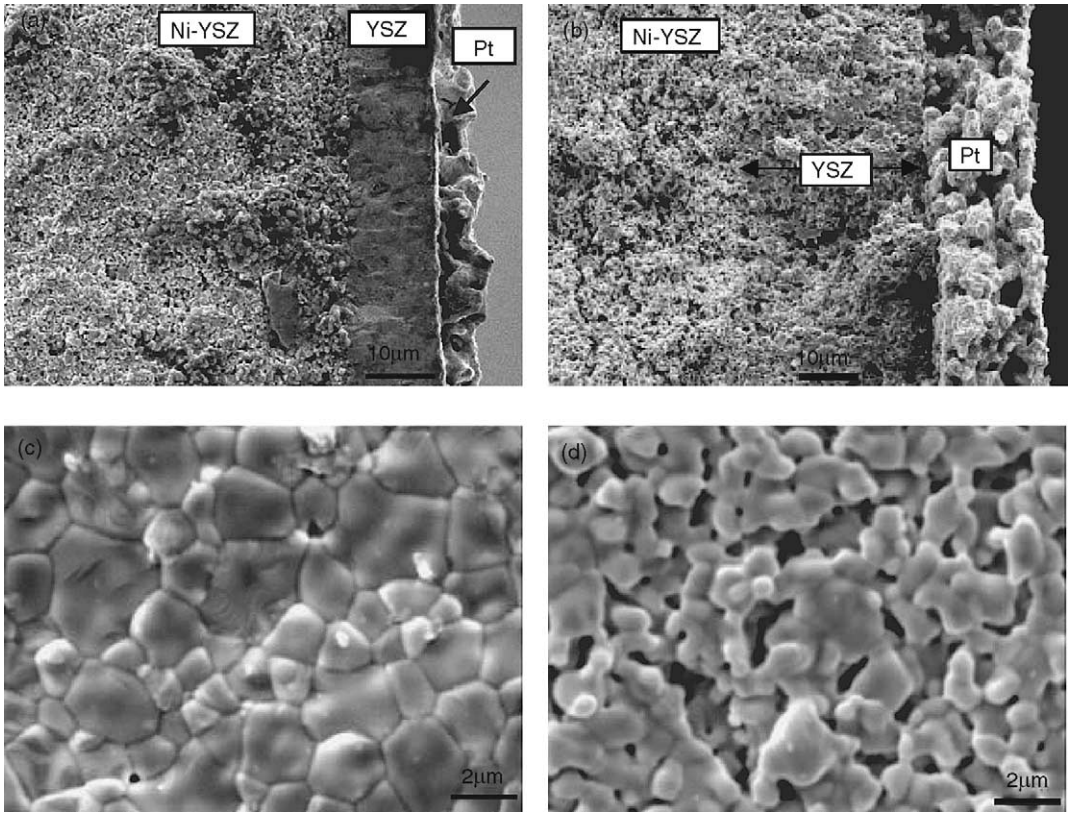

Fig. 6. Scanning electron micrographs of cross-sections of anode-supported thin-film YSZ electrolyte cell made from: (a) micron-sized YSZ powder sintered at 1400 °C; (b) nano-sized YSZ powder sintered at 1400 °C; and of surface of YSZ electrolyte made from: (c) micron-sized YSZ powder sintered at 1400 °C; (d) nano-sized YSZ powder sintered at 1400 °C.

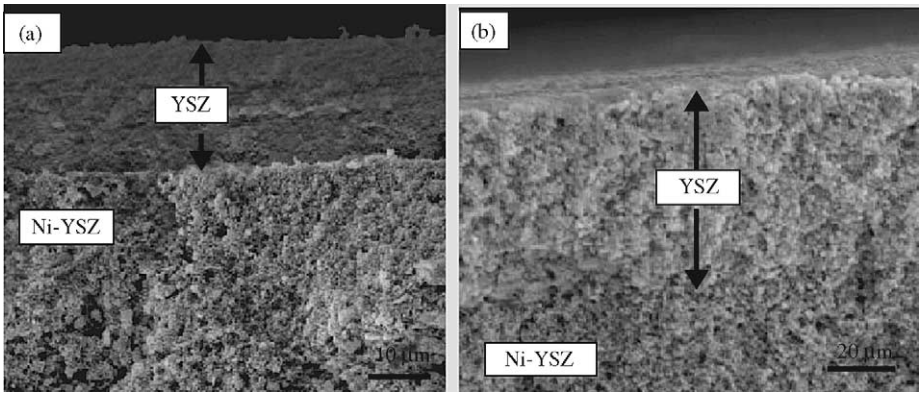

Fig. 7. Scanning electron micrographs of cross-sections of thin-film YSZ electrolyte fired at 500 °C made from: (a) micron-sized YSZ powder; (b) nano-sized YSZ powder on anode-support.

Table 2
Relative green and sintered density of thin-film YSZ electrolyte on anode support

YSZ powder	Green film fired at 500 °C for 2 h			Thin film sintered at 1400 °C for 2 h		
	Diameter (Φ_0) (mm)	Thickness[a] (L_0) (μm)	Green density[b] (ρ_0) (%)	Diameter (Φ_T) (mm)	Thickness[a] (L_T) (μm)	Sintered density (ρ_T) (%)
Micron-sized	24.0	22	∼38	19.8	13	∼94.5
Nano-sized	24.0	56	∼25	19.8	33	∼62.3

[a] Thickness of green and sintered thin-film YSZ electrolyte is measured from electron micrographs.
[b] Green density is estimated from value of green density of tape-cast film.

4. Conclusions

The characteristics of YSZ powders have a significant effect on the formation of dense and thin YSZ films on anode support/substrates for the development of SOFCs which operate at intermediate temperatures. Pellets made of nano-sized YSZ powders achieve a relative density of ∼96 and ∼92% at a sintering temperature of 1400 and 1200 °C, respectively. When the nano-sized YSZ powders are coated on to anode-support substrates, however, thin nano-sized YSZ films are unable to sinter and densify even at 1400 °C but form a porous structure. The reasons are most likely related to the low green density of the nano-sized YSZ film and the significant difference in the sintering kinetics of the nano-sized YSZ layer and the Ni/YSZ cermet substrate. On the other hand, with micron-sized YSZ powder, a dense and almost pore-free YSZ thin film can be achieved on the Ni/YSZ cermet substrate after sintering at 1400 °C for 2 h. The results indicate the importance of the sintering behaviour and the nature of the YSZ powder in the development of an anode-supported thin YSZ electrolyte SOFC for operation at intermediate temperatures.

References

[1] N.Q. Minh, J. Am. Ceram. Soc. 76 (1993) 563.
[2] S.P.S. Badwal, Solid State Ionics 143 (2001) 39.
[3] R. Vaβen, D. Simwonis, D. Stöver, J. Mater. Sci. 36 (2001) 147.
[4] T. Iwara, J. Electrochem. Soc. 143 (1996) 1521.
[5] A. Khandkar, S. Elangovan, M. Liu, Solid State Ionics. 52 (1992) 57.
[6] B.C.H. Steele, A. Heinzel, Nature 414 (2001) 345.
[7] R. Maric, S. Ohara, T. Fukui, H. Yoshida, M. Nishimura, T. Inagaki, K. Miura, J. Electrochem. Soc. 146 (1999) 2006.
[8] T. Ishihara, T. Shibayama, M. Honda, H. Nishiguchi, Y. Takita, J. Electrochem. Soc. 147 (2000) 1332.
[9] K. Wang, J.H. Wan, J.B. Goodenough, J. Electrochem. Soc. 148 (2001) A788.
[10] R. Doshi, V.L. Richards, J.D. Carter, X. Wang, M. Krumpelt, J. Electrochem. Soc. 146 (1999) 1273.
[11] C. Xia, M. Liu, Solid State Ionics 144 (2001) 249.
[12] C. Xia, F. Chen, M. Liu, Electrochem. Solid-State Lett. 4 (2001) A52.
[13] S. de Souza, S.J. Visco, L.C. de Jonghe, Solid State Ionics 98 (1997) 57.
[14] J.W. Kim, A.V. Virkar, K.Z. Fung, K. Mehta, S.C. Singhal, J. Electrochem. Soc. 146 (1999) 69.
[15] C. Wang, W.L. Worrell, S. Park, J.M. Vohs, R.J. Gorte, J. Electrochem. Soc. 148 (2001) A864.
[16] T. Tsai, E. Perry, S. Barnett, J. Electrochem. Soc. 144 (1997) L130.
[17] P.K. Srivastava, T. Quach, Y.Y. Duan, R. Donelson, S.P. Jiang, F.T. Ciacchi, S.P.S. Badwal, Solid State Ionics 99 (1997) 311.
[18] X.J. Chen, K.A. Khor, S.H. Chan, L.G. Yu, Mater. Sci. Eng. A 335 (2002) 246.
[19] T. Matsushima, H. Ohrui, T. Hirai, Solid State Ionics 111 (1998) 315.
[20] S.K. Tadokoro, E.N.S. Muccillo, J. Alloys Compd., in press.
[21] K. Hishinuma, M. Abe, K. Hasegawa, Z. Nakai, T. Akiba, S. Sōmiya, in: S.P.S. Badwal, M.J. Bannister, R.H.J. Hannink (Eds.), Science and Technology of Zirconia V, Technomic Publishing, Lancaster, 1993, p. 207.
[22] D.D. Upadhyaya, T.R.G. Kutty, C. Ganguly, in: S.P.S. Badwal, M.J. Bannister, R.H.J. Hannink (Eds.), Science and Technology of Zirconia V, Technomic Publishing, Lancaster, 1993, p. 310.
[23] A.E. Edelstein, R.C. Cammarato (Eds.), Nanoparticles: synthesis, properties and applications, Institute of Physical Publishing, Philadelphia, 1996, p. 170.
[24] J.B. Bauerle, J. Phys. Chem. Solids 30 (1969) 2657.
[25] M.C. Steil, F. Thevenot, M. Kleitz, J. Electrochem. Soc. 144 (1997) 390.
[26] S.P.S. Badwal, J. Drennan, J. Mater. Sci. 22 (1987) 3231.
[27] C.C. Appel, N. Bonanos, J. Euro Ceram. Soc. 19 (1999) 847.
[28] J.L. Shi, J. Mater. Res. 14 (1999) 1398.
[29] S.J. Visco, L.S. Wang, S. de Souza, L.C. De Jounghe, Mater. Res. Soc. Symp. Proc. 369 (1995) 638.