

Current collectors for a novel tubular design of solid oxide fuel cell

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Abstract

Current collection/interconnection of novel tubular solid oxide fuel cells (SOFCs) is achieved by means of alloy metal interconnects based on Ducrolloy ($\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$) coated with various electronically conductive coatings. Coatings applied to the metal interconnect improve cell performance. For example, at 900°C in air, a $\text{Ag-La}_{0.8}\text{Sr}_{0.2}\text{CrO}_3$ (Ag-LSC) coated interconnect is used in conjunction with an 8 mol% yttria stabilised zirconia (YSZ) tubular electrolyte based SOFC. At 0.7 V a cell performance of 40 mA/cm² is achieved for the coated sample, compared with a lower performance of 16 mA/cm² attained with an uncoated interconnect. The thermal shock resistant properties of the tubes are not compromised by use of the metal interconnect. © 1999 Elsevier Science S.A. All rights reserved.

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1. Introduction

Solid oxide fuel cells (SOFCs) are highly energy efficient devices used to generate electricity from a variety of fuels. The cells may be fabricated in a variety of designs that include planar, tubular, segmented in-series and monolithic. To obtain higher power outputs, SOFCs are connected electrically in series or parallel by means of an electronically conducting interconnect [1]. The literature shows there to be many forms of the interconnect/current collector as determined by the cell design. These vary from metal wires connected with metal gas separator plates [2,3], Ni felts used in combination with metal plates [4], Ni/Cr wires [5], doped LaCrO_3 [6] and chromia forming metal plates [7]. Of these, doped LaCrO_3 and chromia forming alloys (or a combination) are the most widely employed [6].

The design and material components of the interconnect are highly dependent on its application with respect to the type of SOFC. For example, interconnects have been designed with reduced thermal stresses which overcome problems associated with thermal cycling, mismatches in thermal expansion coefficient (TCE) and improved electrical

contact. Yamazaki et al. [8] have reduced the thermal stress (where thermal stresses arise from differences in thermal expansion between the metal and ceramic components) in the planar SOFC stacks by implementation of a sliding mechanism between the separator and electrolyte, and by the introduction of flexible wire interconnectors between the electrodes and the separators. Flexible wires have also been used in conjunction with tubular cells, which are detailed in a previous work by the authors [9]. For the planar SOFC, Brückner et al. [10] coated a metal interconnect with a conductive perovskite to improve electrical contact; the metal alloy plate used was Ducrolloy ($\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$), a metal designed for interconnect applications to have a conductive oxide coating and a TCE closely matching other cell components. As described by Brückner et al. [10], electrical contact between interconnect and cathode is reduced due to the unevenness of their surfaces. Application of a cathode coating on the interconnect can increase the electrical contact between the cathode and interconnect, by compensating for the corrugations present on their respective surfaces. Under operational temperature and slight mechanical pressure, the coating can adjust to the uneven surfaces, and thus enlarge the electrical contact area considerably. Coatings can also be used to reduce the chromium evaporation from the metal interconnect to the cathode, which is detrimental to cell

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performance. Therefore, tailoring the coating to have better electronic conductivity and/or reducing the chromium evaporation would be a plausible way to improve cell performance.

The use of silver cermets as cathodes in SOFCs has been shown to improve the cell performance by lowering the resistivity of the electrode and increasing the contact area between the electrode and electrolyte [11]. Using a similar philosophy, silver cermets could be fabricated and applied as a coating to the surface of the metal alloy interconnect, reducing the resistance between the interconnect and cathode. In utilising the highly shock resistant properties of the tubular SOFCs with a metal interconnect of similar TCE and highly conductive coatings, it was envisaged that a thermally shock resistant method of interconnection could be achieved.

In this study, tubular cells are combined in a novel way with a planar type $\text{Cr5Fe1Y}_2\text{O}_3$ metal alloy interconnect. Various conductive perovskite and silver cermet coatings are applied to the $\text{Cr5Fe1Y}_2\text{O}_3$ metal plate and are studied and compared as potential cathode current-collector/interconnector systems for both medium (600–850°C) and high temperature (900–1000°C) based tubular electrolyte SOFCs.

2. Experimental

The tubular solid oxide fuel cells used in this study consisted of extruded electrolyte tubes, either YSZ or $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ (CGO), with the cathodes (air electrodes) on the outside of the tubes and the anodes (fuel electrodes) on the inside. Current collection for the cathode was achieved using silver wire or perovskite/cermet coated metal alloy plates, while current collection for the anode was performed using nickel wires forced down the inside of the tube; details of the cathodes, anodes and current collectors used in conjunction with the YSZ and CGO electrolytes are given in Table 1.

The proposed novel, highly thermal shock resistant, stacking combination of tubular cells with inter-

connector/current-collector plates is shown schematically in Fig. 1. The YSZ electrolyte consisted of an extruded YSZ tube, sintered at 1450°C for 1 h, with an outside diameter of approximately 2.4 mm and a thickness of 150 μm (extrudate supplied by Viking; extrusion and sintering performed by SAPCO). The CGO electrolyte was fabricated by extruding CGO extrudate through an in-house designed, high strength, steel piston extruder to form a tube. The extrudate was made by combining $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ (CGO) (NexTech Materials, Worthington, OH), heat-treated at 900°C for 4 h, with commercial binders specific to extrusion (Rohm and Haas, Formulation Chemicals); details of the chemicals are given in Table 2. The resulting green body tube was dried in air, then sintered at 1600°C for 2 h. Full details of extrudate preparation, extrusion and firing can be found elsewhere [12]. The final sintered tube had an outside diameter and wall thickness of approximately 3.14 mm and 400 μm , respectively.

The $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ and $\text{La}_{0.8}\text{Sr}_{0.2}\text{CrO}_3$ perovskite powders were prepared using the glycine nitrate process [13]. Stoichiometric mixtures of aqueous nitrate solutions containing $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Sr}(\text{NO}_3)_2$ and $\text{Cr}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ or $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (Rectapur, Univar and Acros, respectively, $\geq 99.9\%$ purity) with glycine, were heated in a beaker until ignition. The resulting powders were calcined in air at 900°C for 12 h.

The cathode for the YSZ cell comprised two layers; a contacting electrochemical layer in contact with the YSZ tube and a thicker cathode layer over the contacting layer. The contacting electrochemical layer, a 2:1 ratio of $\text{La}_{0.82}\text{Sr}_{0.18}\text{MnO}_3$ (LSM) commercial powder (Merck, LSM82X96010-2) to YSZ (TOSOH 8YSZ, 99.9% pure) ink, was painted on to the YSZ tube using a brush, and then dried in an oven at 140°C for 0.5 h. The second layer of LSM was then applied by brush over the first layer and again dried in an oven. The cathode for the CGO electrolyte was fabricated in a similar fashion, however, the cathode comprised three layers; a contacting layer of 2:1 ratio of $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ to CGO, an intermediate layer of 3:1 ratio of $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ to CGO and a third layer applied over the first two layers consisting of

Table 1

Cathode current-collectors, cathode layers, anodes and anode current-collectors used in conjunction with 8 mol% Y_2O_3 stabilized ZrO_2 (YSZ) and $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ (CGO) cells

	YSZ tubular cell	CGO tubular cell
Cathode current collectors	Ag wire Ag– $\text{La}_{0.8}\text{Sr}_{0.2}\text{CrO}_3/\text{Cr5Fe1Y}_2\text{O}_3$ $\text{La}_{0.8}\text{Sr}_{0.2}\text{CrO}_3/\text{Cr5Fe1Y}_2\text{O}_3$ $\text{Cr5Fe1Y}_2\text{O}_3$	Ag wire Ag– $\text{La}_{0.8}\text{Sr}_{0.2}\text{CrO}_3/\text{Cr5Fe1Y}_2\text{O}_3$ Ag– $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3/\text{Cr5Fe1Y}_2\text{O}_3$ $\text{Cr5Fe1Y}_2\text{O}_3$
Layer 1 cathode	$\text{La}_{0.82}\text{Sr}_{0.18}\text{MnO}_3$	$\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$
Layer 2 cathode	$\text{La}_{0.82}\text{Sr}_{0.18}\text{MnO}_3$:2:1YSZ	$\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$:3:1CGO
Layer 3 cathode	–	$\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$:2:1CGO
Electrolyte	YSZ	$\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$
Anode	Ni–YSZ	Ni–YSZ
Anode current collect	Ni wire	Ni wire

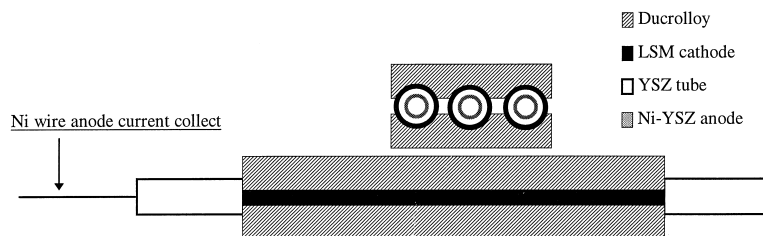


Fig. 1. Illustration of novel, small diameter, SOFC tubes combined with Ducrolloy metal plate current-collectors/interconnectors ($\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$).

$\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$. Each layer was dried and fired (1300°C for 1 h) before the next layer was applied. Before the inks were prepared, the ceramic powders were placed in 250 ml Ngalene bottles with ethanol and zirconia milling media and milled for 24 h. The mixture was then poured into evaporating dishes and the solvent removed by placing the dishes in an oven. To prepare the inks, the powders were mixed using a glass stirring rod with decal medium (Johnson Matthey Colour, Ref. no. 7340/s) and mineral spirits (Aldrich); the mineral spirits were used to modify the viscosity of the inks. The outermost cathode layer consisted of 20 g ceramic powder mixed with 7.3 g decal medium and 1.5 g of mineral spirits. The intermediate and contacting layers consisted of 20 g powder mixed with 20 g of decal medium and 5.6 g of mineral spirits.

The anode was deposited on to the entire inside length of the tubular cells by draining anode ink through the cell. Preparation of the anode ink involved various steps. Five grams of 8 mol% YSZ (TOSOH 8YSZ, 99.9% pure) was calcined in a furnace (Carbolite RHF 17/3) and heated at 10 K/min to 1500°C , holding for 10 min, and then cooled down to room temperature. The calcined YSZ was then sieved through a $180\ \mu\text{m}$ sieve. About 10.5 g of NiO (Alfa Chemicals), 2.9 g 1,1,1-trichloroethene, 2.1 g of methanol, 0.1 g dibutyl phthalate (Aldrich 99%) and 0.1 g of KD1 dispersant (Zeneca) were added, with zirconia milling media, to a plastic container, and milled for 2 h. The calcined YSZ was then added to the container and milled for a further 1 h. On completion of the milling, 0.1 g of PVB (polyvinyl butyral, Aldrich) was added to the mixture and milled for a final 2 min. The anode ink was used immediately. A PVC tube was used to connect a syringe to

the ceramic tubular electrolyte, ink was then poured into the syringe and allowed to drain through the cell. The cell was initially left to drain and then placed in an oven at 140°C to dry. A second coating was applied in a similar fashion. A total anode coating of approximately $30\ \mu\text{m}$ was achieved. After application of electrodes, the cells were fired using a ramp rate of 1 K/min to 500°C , 5 K/min to 1300°C , 1300°C (dwell 1 h), and then 10 K/min to room temperature.

After firing the electrodes on to the YSZ tube, the current-collectors were added. For relative comparison of cell current–voltage (I – V) curves with different methods of current collection, a single YSZ cell was used. Similarly, a single CGO cell was used to test the effect which different current collection methods had on the CGO cell performance. The cathode current-collector systems used in this work were silver wires wound around the cathodes and coated (perovskite and cermet coated) and uncoated $\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$. The silver wires were used as a standard to compare the effect the alternative $\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$ metal plate based current-collector/interconnect systems had on fuel cell performance. As has already been described, Fig. 1 shows the proposed tubular cell and flat plate current-collect system; the system employed in this work, however, used either a single YSZ or CGO cell wedged between the current-collect plates. Before the $\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$ was used as a current-collector, the various perovskite and cermet coatings were applied. Inks were prepared in a similar manner as those used for the $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$ contacting layers described previously. In order to collect current from the metal plates, silver wires were wound around the metal plates and attached to the $\text{Cr}_5\text{Fe}_1\text{Y}_2\text{O}_3$ using silver paste. This was then dried in an oven for 15 min at 140°C and then fired in-situ up to the operating temperature of the cells during tests.

Anode current collection was achieved by means of two 0.5 mm nickel wires (99.98%, Advent Research Materials) placed down the inside of the tubes, such that two nickel wires remained in contact with the anode by the mechanical force exerted by the wires on the inside of the tubes.

A purpose-built furnace was used for cell tests. A gas line connected to a dual supply of hydrogen and nitrogen was then attached to the end of the cell, while the current-collecting wires for the electrodes were brought outside the furnace. Before testing, the cells were heated in the fur-

Table 2

Amounts (wt.%) of organic additives relative to the weight of the ceramic CGO powder

PEG 1:10 H_2O (wt.%)	AMP-95™ (wt.%)	Binder 1 (wt.%)	Binder 2 (wt.%)	H_2O (wt.%)
6.9	1.8	8.2	4.9	12.1

PEG: Poly(ethylene glycol)-400, (Union Carbide, Danbury, CT).

H_2O : de-ionized water.

AMP-95™: pH control agent (Angus Chemical, Buffalo Grove, IL).

Binder 1: B1-1051 Duramax™.

Binder 2: B1-1052 Duramax™.

nance to their respective operating temperatures, 900°C for the YSZ electrolyte and 600°C for the CGO electrolyte, while nitrogen was passed through the tube. Upon reaching operating temperature, the nitrogen supply was slowly turned off as hydrogen was introduced. Anode reduction was achieved by passing pure hydrogen, at a rate of 25 cm³/min, bubbled through water at 25°C, through the cell for 15 min. The *I*–*V* characteristics of the cells were then measured using a potentiostat (PGP201 Potentiostat/Galvanostat, Radio Copenhagen) in air at the operating temperatures of the cells, and a constant flow rate of 25 cm³/min H₂.

3. Results and discussion

As previously described [10], the electrical contact area between the metallic interconnect and cathode can be greatly increased by coating the interconnect with a perovskite, and thus can improve cell performance. The results given in Fig. 2 clearly show this behaviour. When the YSZ cell is used in conjunction with the coated Cr5Fe1Y₂O₃, the current densities achieved are far higher than those obtained when using the uncoated Cr5Fe1Y₂O₃. The use of a Ag–La_{0.8}Sr_{0.2}CrO₃ coating yields a cell performance that almost equals the standard silver wire current collect. The *I*–*V* characteristics of the La_{0.8}Sr_{0.2}CrO₃ coating shown in Fig. 2, although less than that achieved using the Ag–La_{0.8}Sr_{0.2}CrO₃, show an overall gain in cell performance over the uncoated interconnect. Furthermore, the performance of the La_{0.8}Sr_{0.2}CrO₃ coated interconnect is not overly inferior to the silver wire interconnect, and thus is an appropriate method of interconnection. The difference in performance between the two coatings is attributed mainly to the lower resistivity of the Ag–La_{0.8}Sr_{0.2}CrO₃ cermet compared with the

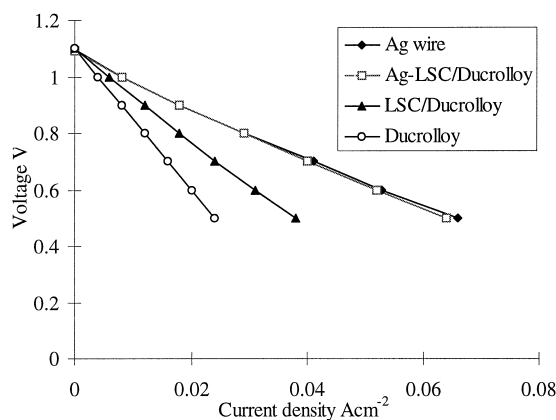


Fig. 2. *I*–*V* characteristics of YSZ electrolyte cells using different methods of cathode current-collection; where Ag–LSC is Ag–La_{0.8}Sr_{0.2}CrO₃, and Ducrolloy is Cr5Fe1Y₂O₃. The cell is tested at 900°C using a constant flow rate of saturated hydrogen bubbled through water at 25 cm³/min.

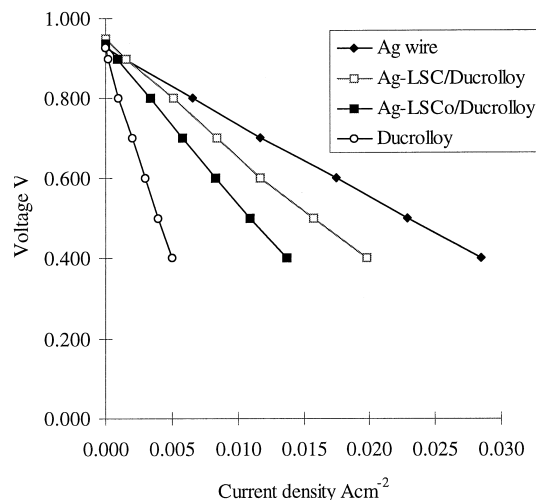


Fig. 3. *I*–*V* characteristics of CGO electrolyte cells using different methods of cathode current collection; where Ag–LSC is Ag–La_{0.8}Sr_{0.2}CrO₃, Ag–LSCo is Ag–La_{0.6}Sr_{0.4}CoO₃ and Ducrolloy is Cr5Fe1Y₂O₃. The cell was tested at 600°C using a constant flow rate of saturated hydrogen bubbled through water at 25 cm³/min.

La_{0.8}Sr_{0.2}CrO₃ ceramic. Both coatings weakly adhered to the surface of the interconnect, but can be easily wiped off using a paper towel. The high thermal shock resistant properties of the tubular design are not compromised, as the assembly can be brought up to temperature within minutes, even though the tubular cell is wedged between the two interconnect plates. This design allows for rapid start-up when compared with other tubular and planar cell designs that can take hours to reach operating temperature [9].

The results obtained for the CGO-based SOFC system at 600°C are given in Fig. 3. The *I*–*V* characteristics in show the Ag–La_{0.8}Sr_{0.2}CrO₃ coating to yield a better performance than the Ag–La_{0.6}Sr_{0.4}CoO₃ coating even though the ceramic component of the latter coating is reported to have a much higher conductivity of 1000 S/cm [16] compared with 36.6 S/cm for the La_{0.8}Sr_{0.2}CrO₃ component of Ag–La_{0.8}Sr_{0.2}CrO₃ [17]. On removal of the Ag–La_{0.6}Sr_{0.4}CoO₃ coated interconnect from the cell, it is found that the coating has bonded together and de-laminated from the surface of the interconnect. We believe that this de-lamination induces a higher resistance and results in poorer cell performance. It is assumed that the thermal expansion coefficient difference between the coating and the interconnect is too large. For example, the TCE of La_{0.6}Sr_{0.4}CoO₃ and silver have reported values of 18.8 [14] and 25.2 × 10⁻⁶ K⁻¹, respectively, compared with 11.3 × 10⁻⁶ K⁻¹ for Cr5Fe1Y₂O₃ [15].

In order to maintain a closer match of TCE between the coating and Cr5Fe1Y₂O₃, Ag–La_{0.8}Sr_{0.2}CrO₃ is used (TCE of La_{0.84}Sr_{0.16}CrO₃ is 9.6 × 10⁻⁶ K⁻¹); as the Ag–La_{0.8}Sr_{0.2}CrO₃ coating is found to adhere to the surface of the interconnect when testing the YSZ electrolyte cell.

When Ag–La_{0.8}Sr_{0.2}CrO₃ coated Cr₅Fe₁Y₂O₃ and silver wire current collectors are used alternately on the same YSZ cell, the performance is approximately equal. By contrast, when the same current-collectors are utilized in a similar manner on the CGO electrolyte cell, the performances achieved are lower for the Ag–La_{0.8}Sr_{0.2}CrO₃ coated Cr₅Fe₁Y₂O₃ than for the silver wire. The drop in performance relative to the Ag wire current-collect is attributed to the use of a lower operating temperature of 600°C. The conductivity of La_{0.8}Sr_{0.2}CrO₃ is temperature dependent, and at lower temperatures is vastly reduced. This increases the resistivity of the coating (or increases the contact resistance between the interconnect and the cathode), and thus reduces cell performance. At 900°C, using the YSZ electrolyte, La_{0.8}Sr_{0.2}CrO₃ is expected to show a higher conductivity, and thereby reduces the resistance between the interconnect and the cathode. The change in temperature affects the conductivity of the coating, which in turn affects the performance of the cell; this is also illustrated in the work by Kimura et al. [5].

4. Conclusions

Coating of the metal interconnect (current-collector) with a variety of perovskites and cermets is shown to improve cell performances. On comparison with a silver wire-current collect, the Ag–La_{0.8}Sr_{0.2}CrO₃ and alternate La_{0.8}Sr_{0.2}CrO₃ coated metal interconnects used in conjunction with the YSZ electrolyte cell (operating temperature of 900°C) yield favourable performances. Similar results are demonstrated for the CGO electrolyte cell. On the other hand a reduction in the operating temperature, by use of the CGO electrolyte, results in a drop in cell performance. This effect is attributed to increased resistivity of the ceramic component of the coating at lower temperature. Furthermore, for the CGO cell, a high mismatch in TCE between the Ag–La_{0.6}Sr_{0.4}CoO₃ and the interconnect results in a reduced cell performance. It is also demonstrated that the high thermal shock resistant properties of the tubular cells wedged between the interconnects are not compromised. Future work will involve investigation of chemical bonding between the interconnect and cathode using various perovskite and cermet based materials; it is

envisaged that improved bonding will allow better cell performance, although this may compromise the thermal shock resistant properties of the cell.

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