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High performance solid-oxide fuel cell: Opening windows to low temperature application

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ABSTRACT

A key hindrance of operating solid oxide fuel cells (SOFCs) at low temperature is the relatively high cell resistance resulting in low power output density. In this work, we report an SOFC design based on an anode-supported cell (ASC) with thin film Yttria stabilized zirconia electrolyte (YSZ), capable of high power output densities of 1050 mW cm^{-2} using H_2 as fuel, at an operating temperature of 873 K. Such high cell performances have been realized by applying three optimization steps: (1) using $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$ -perovskite (LSC) as high performing cathode material at low temperature; (2) integration of an optimized $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ (CGO) inter-diffusion barrier layer and (3) optimization of the microstructure of the anode substrate by means of increasing the substrate porosity.

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Introduction

The Solid Oxide Fuel Cell (SOFC) is an attractive power generation device that directly and efficiently converts chemical energy from hydrogen or fossil fuels to electric power. Hence, this device combines the benefits of environmentally benign power generation with fuel flexibility [1,2]. However, the necessity to operate a conventional SOFC at high operating temperature ($>1073 \text{ K}$) results in high costs of applied materials, especially the metallic interconnect and balance-of-plant materials, and material compatibility challenges [3,4]. The reduction of the operating temperature of SOFCs

(823–923 K) is an effective approach for reducing the costs of applied materials and increasing lifetime of SOFCs [5,6].

Although it is well-known that the anode-supported cell (ASC) with thin film electrolyte is the most promising cell design for low temperature application [7,8], the performance of this type of cells at lower temperatures strongly declines due to rapidly increasing cathode polarisation losses [9,10]. The catalytic oxygen reduction activity of the currently used cathode materials like $(\text{La}, \text{Sr})_{(1-x)}\text{MnO}_{3-\delta}$ and $(\text{La}, \text{Sr})_{(1-x)}(\text{Fe}, \text{Co})\text{O}_{3-\delta}$ is rather low for operation below 973 K [11–13]. In the literature, $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$ perovskite (LSC) has been proposed as cathode material for low-temperature SOFCs. This compound with rhombohedrally distorted perovskite structure is a well-known mixed ionic and electronic conducting material

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(MIEC) with high catalytic activity for oxygen reduction at low temperatures [14–16]. Another advantage of LSC as cathode is its high tolerance towards CO_2 in the desired temperature regime. That aspect makes LSC a more favourable low temperature cathode material than the highly promising $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (BSCF), which has been demonstrated as good low temperature cathode but has low CO_2 -tolerance [17,18]. A drawback of the use of LSC is the reactivity with YSZ electrolyte resulting in the formation of SrZrO_3 , especially during the SOFC manufacturing procedure, involving sintering temperatures for the cathode as high as 1173–1373 K. SrZrO_3 has very poor oxygen ionic conductivity, leading to lower cell performance [12]. Therefore, a diffusion barrier layer between the YSZ electrolyte and LSC cathode is needed in order to prevent Sr diffusion from the cathode to the zirconia electrolyte. From the literature, $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ (CGO) has been found to be a more suitable blocking layer, compared to $\text{Ce}_{0.8}\text{Y}_{0.2}\text{O}_{1.9}$ (CYO), due to its high ionic conductivity and chemical compatibility with the LSC-cathode along with low reactivity with Sr-containing cathode [7,19]. This ceria inter-diffusion barrier layer needs to fulfil three requirements. First, this layer has to be thin resulting in the reduction of the ohmic contribution. Second, the ceria barrier layer has to be sintered at temperatures as low as possible in order to prevent inter-diffusion of cations between the ceria and zirconia layer, which creates an undesirable reaction zone with a lower ionic conductivity that results in enhanced ohmic losses [8,20]. Third, this layer must be dense in order to prevent any reaction between cathode and zirconia electrolyte. Two techniques have been explored for the optimization of applied CGO layers in order to achieve those requirements, namely the cost efficient screen-printing technique (SP) and physical vapour deposition technology (PVD). The ceria deposition procedure using PVD has been demonstrated already to be a suitable technique with respect to those requirements [21]. This technique has the advantage of lowering the deposition temperature of the CGO layer to 1073 K or even below, which prevents the interdiffusion between CGO and YSZ.

The anode substrates used in Anode Supported Cells (ASC) are usually fabricated by tape casting method. The investigation of Ni-YSZ cermet anode indicated that the anode substrate structure can significantly influence the performance of the fuel oxidation reaction. Increasing porosity and pore size will allow for high electrochemical activity and less hindered gas transport [22,23].

In the present work, the significant improved cell performance at 873 K has been achieved by the use of LSC as cathode and improvement of quality of CGO interdiffusion barrier layer. The final improvement of cell performance has been obtained by optimization of the anode substrate with respect to porosity and pore size distribution.

Experimental

Fabrication of anode-electrolyte support

NiO (MERK), 3 mol% YSZ (TOSOH) and pore-former powder obtained from commercial sources were mixed into a tape cast suspension, consisting of PVB binder dissolved in

ethanol-toluene mixture. After tape-casting and evaporation of the dispersion aid, the resulting green tape was cut in the appropriate dimension and the functional anode layer and electrolyte layer were applied by screen printing (200 mesh). The functional anode layer is prepared from a mixture of NiO (MERK) and 8 mol% YSZ ($\text{Zr}_{0.84}\text{Y}_{0.16}\text{O}_{1.92}$, TOSOH), powder from commercial sources. The electrolyte layer consists of 8 mol% YSZ. The screen print pastes were prepared by mixing these powders into a dispersant aid and binder system using a Dispermat milling device. The resulting green anode electrolyte support was sintered at 1673 K for 1 h. The sintered anode-electrolyte support consists of an approximately 550 μm thick anode substrate, an 8 μm thick electrochemical active anode functional layer and a 3–5 μm thick electrolyte layer. The state-of-the-art anode electrolyte support used as a reference to monitor the improvement in cell performance consists of an anode substrate supporting a bi-layer electrolyte of 8YSZ (4–5 μm) and $\text{Ce}_{0.8}\text{Y}_{0.2}\text{O}_{1.9}$, (CYO, 3–4 μm) that has been co-fired at 1673 K together with the anode substrate.

Preparation of ceria diffusion barrier layer

$\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ powder (CGO, from Rhodia) has been used for the ceria barrier layer development by means of screen printing technique (SP). Screen printing pastes have been prepared by mixing CGO powders into a dispersant aid and binder system using a Dispermat milling system. The pastes with additional sintering aid (cobalt nitrate salt, 0.6 mol dm^{-3}), aiming for dense and crack-free CGO layer after sintering, has been screen-printed onto the 5 × 5 cm^2 square-shaped anode-electrolyte support, followed by sintering at 1573 K. This sintering temperature has been found to be the most optimum one in our previous work. After the cell performance test, the microstructure and elemental composition of the CGO layer have been investigated by SEM (JEOL HSM-6330F Field Emissions Scanning Electron Microscope) equipped with an EDX spectrometer (Thermo Noran) on the cross section of the samples. Line-scans of the cross section of the tested samples were performed to determine the element distribution across the different layers. For the CGO layer prepared by PVD, the reactive sputtering technique was used. Therefore, a metallic alloy with a nominal composition of 80 at.% Ce and 20 at.% Gd has been sputtered in an argon/oxygen atmosphere with an oxygen partial pressure of 10^{-3} mbar.

Cathode manufacturing

The $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$ (LSC) screen printing pastes have been prepared by mixing the LSC powders (Praxair) into a dispersant aid and binder system using a Dispermat milling device. The resulting pastes have been screen-printed (SP) on top of the 5 × 5 cm^2 square shaped anode substrate support covered with either CGO or CYO layer. This LSC cathode has been first optimized for its electrochemical performance through microstructural modification by means of optimization of sintering step, aiming for sufficiently and uniformly small particles along with high catalytic oxygen reduction activity and well established particle-to-particle connectivity. The optimum sintering temperature has been determined to be

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