



A novel microstructured metal-supported solid oxide fuel cell



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HIGHLIGHTS

- Novel metal-supported solid oxide fuel cell design for mobile applications.
- The proposed honeycomb microstructure allows saving about 65% of metallic material.
- The SOFC anode is inside of the interconnect layer, in honeycomb cells.
- This novel MSC exhibits performances of 300 W cm⁻² at 850 °C under pure H₂.

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ABSTRACT

An innovative design, alternative to the conventional metal supported fuel cells (MSC) is proposed. This new design of Solid Oxide Fuel Cell (SOFC), comprises a 200 μm layer of a honeycomb-metallic framework with hexagonal cells which supports a 250 μm layer of electrolyte. Each hexagonal cell is further functionalized with a thin 5–10 μm of Ni–YSZ anode. This new design allows a reduction of ~65% of the metallic supporting material, rendering performances over 300 mW cm⁻² under pure hydrogen at 850 °C, with an OCV of ~1.1 V.

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1. Introduction

Fuel cells are electrochemical devices able to produce electricity directly from the oxidation of a fuel and simultaneous reduction of an oxidant. The fuel cells provide many advantages over traditional energy conversion systems including: high efficiency to electrochemically generate electricity, reliability, modularity, fuel adaptability (hydrogen, CO, methane) and very low levels of SO_x and NO_x emissions [1–3].

The high temperature fuel cells are known as Solid Oxide Fuel Cells, and each single functional unit comprises at least three components: an anode, a cathode and a solid electrolyte. Another element is added when a stack is fabricated, i.e. the interconnect or bi-polar layer.

When the thickness of the electrolyte becomes too low (<150–300 μm) the mechanical stability of each cell and that of the whole stack can be seriously compromised and hence a supported configuration is typically used, the cathode-supported cells (CSC), or anode-supported cells (ASC) being the chosen configurations. They have demonstrated good results in commercial equipment, such as stationary power plants and/or combined heat and power (CHP) applications [2,4,5].

There is an increasing interest in mobile applications although the supported configurations with brittle ceramics or cermets may delay the implementation of these applications [6–8]. Metal supported fuel cells (MSC) provide benefits over traditional all-ceramic SOFCs due to: materials cost reduction (it is expensive to use ASC in commercial applications), short start-up times, and the possibility of improved sealing and stacking schemes [4,9]. These MSCs can be considered as potential electrical power supply for mobile applications due to the higher mechanical stability, electrical conductivity and better thermal and redox behavior of the used alloys,

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which are required for this task [10–13]. Regarding the metal alloys, ferritic FeCr stainless steels are the most widely used as metallic substrates due to their well-matched thermal expansion coefficients with YSZ electrolytes (YSZ $\sim 10\text{--}11 \times 10^{-6} \text{ K}^{-1}$ vs. FeCr-alloy $\sim 12\text{--}14 \times 10^{-6} \text{ K}^{-1}$) [4,14], low costs and adequate oxidation resistance at high temperature [15,16].

The main disadvantages of the MSC are related to electrode materials. Cathodes are typically sintered under air, and this atmosphere can oxidize the metal support. Hence the maximum sintering temperature should be below 900 °C in air or alternatively, the sintering of the cathode must be performed under reducing or vacuum atmosphere. However, these conditions are harsh for the typical cathode materials like lanthanum strontium manganite (LSM) or (LSCF), which tend to decompose during the sintering process [4,17]. Due to this, the common cathode material has been platinum [4], although very recently Zhou et al. have reported alternative electrodes such as $\text{La}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.9}\text{Sc}_{0.1}\text{O}_{3-\delta}$ or $\text{SrFe}_{0.75}\text{Mo}_{0.25}\text{O}_3$ [15,18–21]. As for the anode material, the common Ni–YSZ cermet is prone to Ni and Fe/Cr interdiffusion between the anode and metal layers when heated at high temperatures (1300–1400 °C), which leads to low power densities and severe degradation of the cell [16,22–24].

In a previous work [25] we have shown that the microstructure of any component of a solid oxide fuel cell is a key element to improve the performance of such devices. An interesting approach to control the materials microstructure can be the soft lithography allowing successful replications of micro- and nanostructures in the range of the 50 nm [26].

We have shown [27] that a supporting electrolyte with honeycomb patterning provides high mechanical strength to the whole structure, supporting a 100 μm layer of YSZ and rendering high volumetric power densities. The reported procedure to control the honeycomb microstructure involved the use of NOMEX mesh (DUPONT™) as a molding tool of a ceramic-based material (e.g. YSZ), creating a backbone with hexagonal cells in a honeycomb structure. The original NOMEX mesh and the sintering conditions controlled the final details of the microstructure.

In this work, one of main aim will the fabrication and testing of microstructured and multi-layered samples comprising a 200 μm metal-supporting layer of ferritic FeCr stainless steels with honeycomb patterning which acts as support of a $\sim 250 \mu\text{m}$ electrolyte layer of YSZ. The hexagonal holes in the crofer microstructure will be filled with a thin (5 μm) active layer of Ni–YSZ anode, Fig. 1.

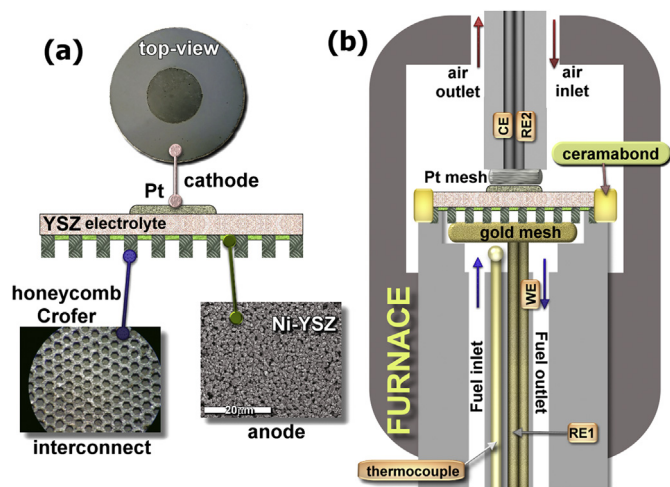


Fig. 1. (a) Schematic representation of the assembled metal supported fuel cell. (b) 2-Point setup for fuel cell tests (WE: working electrode, CE: counter electrode and RE_x: reference electrodes).

This novel design may have several important advantages over the traditional *state-of-the-art* configuration:

- (1) It allows the production of robust MSC with a 200 μm layer of microstructured metal, in fact, in this way any type of patterning can be produced.
- (2) It offers the possibility to fabricate at mass-scale in a cost-effective way.
- (3) It saves up to $\sim 65\%$ of the supporting material, which will eventually lead to a decrease of the cost of any MSC assembled in this configuration. This opens up the possibility of the fabrication of SOFC devices with an elevated ratio of kW per cm^3 and/or kW per kg [27].
- (4) The anode is situated just inside each hexagonal cavity, in direct contact with YSZ. Hence any type of reactivity between anode and interconnect will be restricted to the inner walls of these hexagonal cells and hence the performance should not be affected by undesired formation of secondary phases [16,22–24].
- (5) The use of interconnect material on one side of each SOFCs offers the possibility of cold sealing through an electrical resistance welding process.
- (6) The use of a metal honeycomb microstructure allows the use of such structure as a current collector and hence no platinum is used in the anode compartment as current collector.

2. Experimental procedure

2.1. Materials

Ytria (8 mol%) stabilized zirconia (YSZ) was supplied by Pi-KEM Ltd. (Staffordshire, UK), NiO (99,99%, Sigma–Aldrich, St. Louis, MO, USA) and the metal alloy Crofer® 22APU were provided by IKER-LAN. Methyl-ethyl-ketone (Sigma–Aldrich, St. Louis, MO, USA). Ethanol (Scharlau, Barcelona, Spain). Triton-Q (Dow Chemical, Midland, MI, USA). dibutyl phthalate (Scharlau, Barcelona, Spain). Butvar B98 polyvinyl butyral (Sigma–Aldrich, St. Louis, MO, USA).

A master and the corresponding negative microstructured mold was prepared using standard micro- and nanofabrication processes [26,28] and the honeycomb microstructures from this master were transferred to poly(dimethylsiloxane) (PDMS) stamps by soft lithography, Fig. 2(a) and (b).

2.2. Electrolyte, anode and interconnect slurries

A modified procedure [27] for tape-casting was used for the preparation of three organic-based slurries for each component of the honeycomb-MSC.

The slurries were obtained by mixing the following materials: 10 g of starting powders (i.e. 10 g of YSZ in one case, 10 g of crofer in another case and finally, 6 g of NiO + 4 g of YSZ), 10 g of a mixture of methyl-ethyl-ketone and ethanol (3:2, w/w) solvents; 0.5 g of Triton-Q (dispersant); 2 g of dibutyl phthalate (plasticizer) and 1 g of Butvar B98 polyvinyl butyral (binder). In all cases, the components were ball-milled for 2 h at 200 rpm in a zirconia vessel with zirconia balls.

2.3. Fabrication of green and sintered multilayered samples

The slurries were hand casted over a PDMS rubber with the honeycomb patterning and dried for 24 h after each deposition. First, three layers of metal alloy were deposited and then another four layers of electrolyte. After 48 h the multilayered sample of 78.5 cm^2 of area, Fig. 2(c)–(e) is easily peeled off from the PDMS

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