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Effect of Al and Ce doping on the deformation upon sintering in sequential tape cast layers for solid oxide fuel cells

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ABSTRACT

Water-based tape casting is an attractive production route for planar solid oxide fuel cells (SOFCs) due to its high productivity and reduced environmental issues. In this work planar anode supported SOFCs with thin electrolyte were prepared by water-based sequential tape casting and co-sintering. An in situ high temperature monitoring apparatus was assembled to allow the determination of free sintering shrinkage of thin green tape cast layers and to follow the curvature developed in multilayers during the entire sintering process.

The instantaneous curvature developed upon co-sintering was studied as a function of the firing schedule and layer composition. It was found that by tailoring the electrode composition it is possible to reduce the shrinking rate difference between anode and electrolyte thus obtaining defect-free electrolyte, minimising the residual curvature of the half-cell and improving the electrochemical performances of the cell.

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1. Introduction

Planar anode supported solid oxide fuel cells are ceramic devices constituted of stacked layers of different materials with dissimilar physical and mechanical properties. The fabrication of such devices typically requires the multi-step deposition and successive firing of the different layers. The cost of such high temperature treatments represents a significant fraction of the overall production expenses. In order to maintain the SOFC costs to an acceptable level, the production of the anode and electrolyte precursor layers by relatively cheap powder technologies, like tape casting or screen printing and subsequent one-step co-sintering, is the preferred route.

In general, during co-sintering of multilayered ceramics, stresses are generated due to the mismatch in thermal expansion coefficient and different sintering rate [1–4]. The stresses generated by dissimilar sintering rates are the primary cause of the deformation of the cell, specifically a curvature, which is developed upon co-firing; and if such stresses overcome the intrinsic strength of the layers, they can lead to flaws and defects formation. Co-sintering is a particularly delicate issue in SOFC processing: the anode needs to sinter in order to acquire enough mechanical strength to support the cell but needs to preserve sufficient porosity for fuel flow; conversely, the thin electrolyte acting as the gas separator between fuel and oxygen needs to densify at least to a point where it retains non-interconnected porosity only. As a matter of fact, SOFC are not tolerant to the presence of defects and even a small crack in the electrolyte leads to leakage and hot spots associated to direct combustion that decreases electrochemical performances and accelerates degradation. Moreover, any defect is detrimental for SOFC mechanical properties, which are of utmost importance in the real operation of a stack even for stationary applications, where the cells can be subjected to severe stresses deriving from external applied load or from thermal or RedOx cycles. Many efforts are being currently done in order to reduce the curvature development and flaws generation, by varying the starting powder grain size [5,6], through selective coarsening of NiO or YSZ powder [7,8] or by optimising the electrodes sintering [9] or co-sintering temperature [10].

Yttria (8 mol%) stabilised zirconia (YSZ) for the electrolyte and a mixture of YSZ and Ni are by far the most studied and employed compositions in SOFC production [7,11–13]. Although such well-known materials are the mayor constituents of anode supported half cells, a variety of doping elements are usually added in small quantities in both the anode and the electrolyte in order to enhance mechanical properties, electrochemical performances, long-term stability and resistance to RedOx cycles. For example, the addition of small quantities of Al_2O_3 (in the order of 1%) to cubic YSZ has been reported to act as sintering aid [14,15] which enhances hardness and fracture toughness, reduces grain growth in YSZ electrolyte [16]

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Table 1
Composition of electrolyte and anode slurry (vol.%)[a].

Components	Electrolyte	Anode
Powder	28.4	21.3
Dispersant	1.5	2.8
Water	58.2	53.0
Binder	11.9	22.9

[a] Binder and dispersant are reported on dry basis.

and significantly increases flexural strength of NiO/YSZ anode [17]. Oxides like Cr_2O_3, TiO_2, Al_2O_3, and Sc_2O_3 can improve anode RedOx stability [18]. Addition of small quantities of Mo [19], precious metals like Ru and Pt [20] or CeO_2 [21] has been observed to reduce the carbon deposition on Ni/YSZ anode during methane reforming. The use of CeO_2 has also been shown to improve the electrochemical performance of Ni/YSZ anodes [22].

Despite the importance and the widespread use of reactive elements and sintering aids and the delicate issue of co-firing for the successful production of reliable cells, no specific studies have been carried out addressing the effect of doping elements on the anode sintering rate and, consequently, on the transient stresses developed upon co-sintering with the electrolyte, which are the primary cause for cell curvature and defect formation. The aim of the present work is to study the effect of selected doping substances (like Al_2O_3 or CeO_2) on the anode sintering kinetics and, consequently, on the cell curvature and curvature rate; in addition it is aimed to determine whether the addition of doping elements can be successfully employed to reduce defects development upon sintering and residual curvature, with the ultimate goal of obtaining flat and higher quality cells.

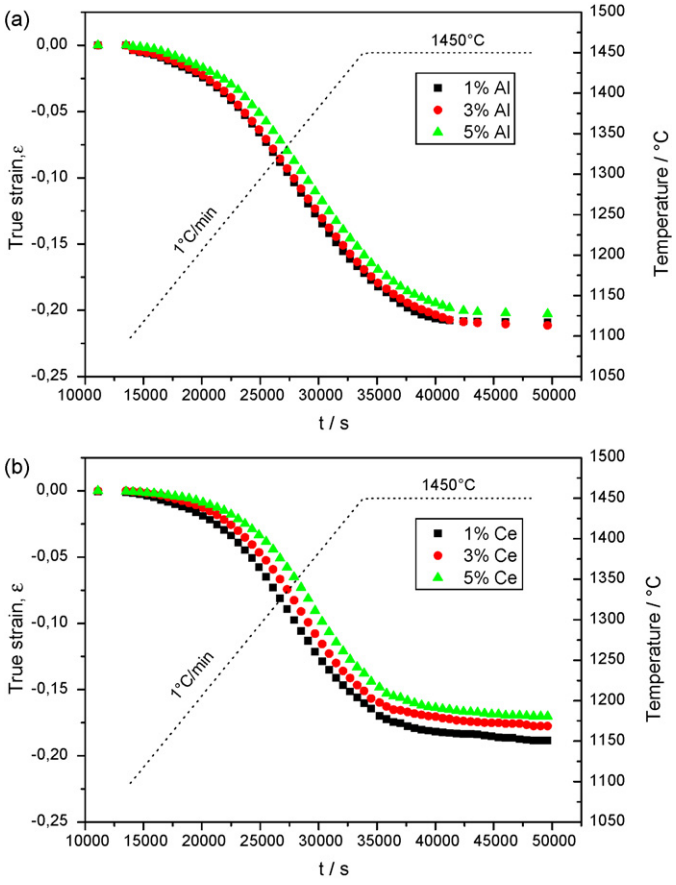

Fig. 1. True strain as a function of time for (a) Al- and (b) Ce-doped pressed pellets.

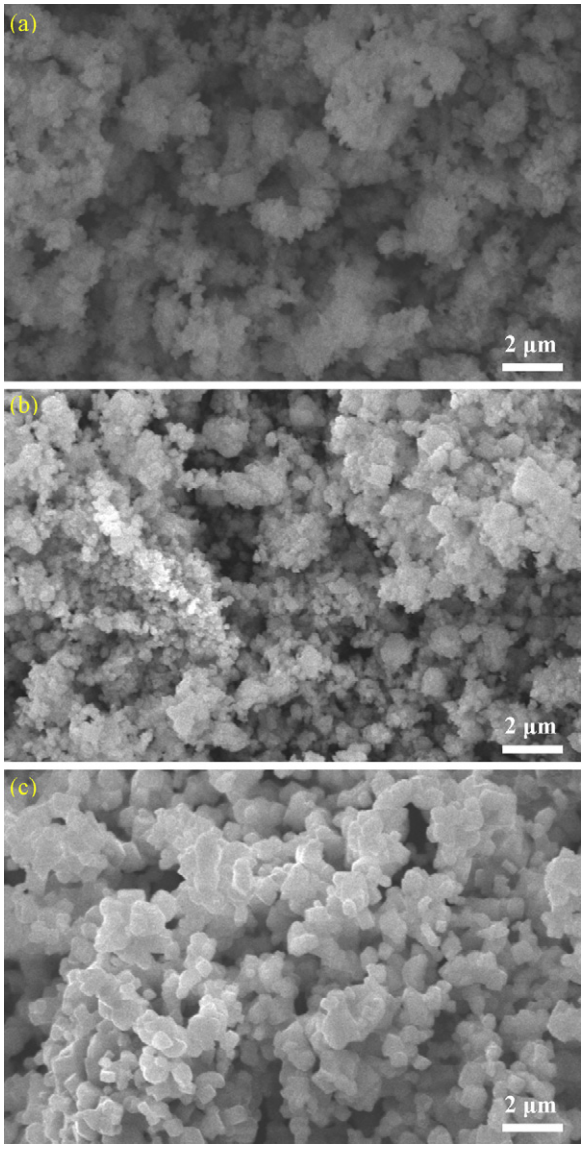

Fig. 2. Micrographs of the modified NiO powders. 1 mol% Al-doped (a), 5 mol% Ce-doped (b) and simply calcined (c).

2. Materials and methods

Due to their wide use as doping elements for typical SOFC anode, Al and Ce were selected in the present work. NiO powder with 1, 3 and 5 mol% Al or Ce doping agent was produced by adding $Al(NO_3)_3{\cdot}9H_2O$ (Riedel-De Haen, Germany) or $Ce(NO_3)_3{\cdot}6H_2O$ (Alfa Aesar, Germany) to starting NiO powder (J.T. Baker, USA) in a plastic jar containing ethanol and zirconia balls (Inframat Advanced Materials, USA); the mixture was milled for 18 h. The powders were then dried and calcined for 10 h at 900 °C. Pure NiO powder was also calcined for 10 h at 900 °C for comparison. Specific surface area (SSA) was determined by nitrogen adsorption (BET) method (ASAP 2010, Micromeritics, USA). Doped NiO powder was mixed with 8 mol% yttria stabilised zirconia (YSZ) powder (TZ-8YS, SSA 6 $m^2\ g^{-1}$, Tosoh, Japan) in a ratio 58 wt% NiO and 42 wt% YSZ; binder (B1000, Duramax, Rohm and Haas, France) and distilled water were then added and the blend was mixed for 5 h in a rotating plastic drum containing zirconia balls. The powders were then dried and manually ground in a mortar. A portion of the obtained powder was then pressed at 125 MPa for 120 s into 20 mm diameter/1 mm thickness pellets that were used for preliminary sintering analyses.

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