INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2017) 1–8

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/he

Effect of anode thickness and Cu content on consolidation and performance of planar copper-based anode-supported SOFC

Vincenzo De Marco [a,*], *Alessandro Iannaci* [b], *Saqib Rashid* [a], *Vincenzo M. Sglavo* [a,b]

[a] *Department of Industrial Engineering, University of Trento, Via Sommarive 9, 38123, Trento, Italy*
[b] *INSTM, Trento Research Unit, Via G. Giusti 9, 50121, Florence, Italy*

ARTICLE INFO

Article history:
Received 25 January 2017
Received in revised form
21 March 2017
Accepted 30 March 2017
Available online xxx

Keywords:
Planar intermediate temperature –SOFC
Copper-based anode
Electrochemical impedance spectroscopy
Co-sintering

ABSTRACT

Planar, Cu-containing Gadolinia-doped ceria anode-supported solid oxide fuel cells to be used at intermediate temperature (500–750 °C) were produced in the present work. The Intermediate temperature solid oxide fuel cells were fabricated using Li_2O as sintering aid for Gadolinia-doped ceria, varying the anode-to-electrolyte thickness ratio (r) from 2 to 10 and the CuO content in the anode from 45 vol% to 55 vol%. Co-sintering of the thermo-pressed green cells was carried out at 900 °C for 3 h. The electrolyte densification was favoured by increasing the r value, this being accounted for the enhanced compressive stresses induced by the supporting anode on the electrolyte upon sintering. Larger CuO content positively influences the overall cell performance, due to the improved electronic conductivity of the anode. Nevertheless, CuO concentration cannot exceed 50 vol% because of the tensile stresses (and corresponding flaws) generated in the electrolyte for larger amount. IT-SOFC containing 50 vol% CuO was characterized by an Open Circuit Voltage ≈0.82 V and a maximum power density of 200 mW cm^{-2} at 700 °C.

Introduction

Wide fuel flexibility [1–4], high efficiency and potential coupling within hybrid systems [5–7] are some of the fundamental reasons that make Solid Oxide Fuel Cells (SOFC) among the most promising devices for environmental-friendly energy conversion. The research is currently leading towards the reduction of the operating temperature, within the so called "intermediate temperature" (IT) range (500–750 °C), because of the improved stability and durability of the system, associated to lower overall costs [8,9]. The first approaches carried out to reduce the operating temperature tended to increase the electrolyte conductivity. This suggested the substitution of traditional yttria-stabilized zirconia (YSZ) with Gadolinia-doped ceria (GDC) [10]. The main issue associated with the use of this latter ceramic is its limited densification unless very high sintering temperatures are used (>1500 °C) [11]. In some cases, sintering aids are used although they can determine variable effects on conductivity [12–15]. Recent works have shown that the use of Li_2O allows to decrease the sintering temperature of GDC down to 950 °C [16].

* *Corresponding author*. Fax: +39 0461281977.
E-mail addresses: vincenzodemarco.86@gmail.com (V. De Marco), alessandro.iannaci@uniroma2.it (A. Iannaci), saqib.rashid@studenti.unitn.it (S. Rashid), vincenzo.sglavo@unitn.it (V.M. Sglavo).
http://dx.doi.org/10.1016/j.ijhydene.2017.03.221
0360-3199/

GDC electrolyte densification which can be carried out at 900 °C with the use of Li_2O sintering aid. This allows to increase CuO content in the anode and keep the tensile, thermal stress in the electrolyte below the critical level. Dense Li-doped GDC electrolyte can be obtained at 900 °C using ten times thicker 45/50 vol% CuO-containing anode. IT-SOFC containing 50 vol% CuO reaches power density of 200 mW cm^{-2} at 700 °C.

Acknowledgments

This work was partially realized with the financial support by Caritro foundation (Cassa di Risparmio di Trento e Rovereto, Italy) by the project “Bando per progetti di ricerca scientifica nell'ambito della gestione e salvaguardia dell'ambiente, territorio e sviluppo sostenibile (2016)”.

REFERENCES

[1] Hu B, Keane M, Patil K, Mahapatra MK, Pasaogullari U, Singh P. Direct methanol utilization in intermediate temperature liquid-tin anode solid oxide fuel cells. Appl Energy 2014;134:342–8.

[2] Papurello D, Borchiellini R, Bareschino P, Chiodo V, Freni S, Lanzini A, et al. Performance of a solid oxide fuel cell short-stack with biogas feeding. Appl Energy 2014;125:254–63.

[3] Minh NQ. Ceramic fuel cells. J Am Ceram Soc 1993;76:563–88.

[4] Steele BCH. Materials for fuel cells. Nature 2001;414:345–52.

[5] Buonomano A, Calise F, Dentice D'Accadia M, Palombo A, Vicidomini M. Hybrid solid oxide fuel cells–gas turbine systems for combined heat and power: a review. Appl Energy 2015;156:32–85.

[6] Ferrari ML. Advanced control approach for hybrid systems based on solid oxide fuel cells. Appl Energy 2015;38:364–73.

[7] De Marco V, Florio G, Fragiacomo P. Optimal operation conditions for a methane fuelled SOFC and microturbine hybrid system. J Renew Energy 2015;2015:13. Article ID 508138.

[8] Brett DJL, Atkinson A, Brandon NP, Skinner S. Intermediate temperature solid oxide fuel cells. Chem Soc Rev 2008;37:1568–78.

[9] Aguadero A, Fawcett L, Taub S, Woolley R, Wu KT, Xu N, et al. Materials development for intermediate-temperature solid oxide electrochemical devices. J Mater Sci 2012;47:3925–48.

[10] Inaba H, Tagawa H. Solid State Ionics 1996;83:1–5.

[11] Inaba H, Nakajima T, Tagawa H. Sintering behaviors of ceria and gadolinia-doped ceria. Solid State Ionics 1998;106:263–8.

[12] Esposito V, Zunic M, Traversa E. Improved total conductivity of nanometric Samaria-doped ceria powders sintered with Molten $LiNO_3$ additive. Solid State Ionics 2009;180:1069–75.

[13] Mori M, Suda E, Pacaud B, Murai K, Moriga T. Effect of components in electrodes on sintering characteristics of Ce0.95Gd0.1O1.95 electrolyte in intermediate-temperature solid oxide fuel cells during fabrication. J Power Sources 2006;157:688–94.

[14] Nicholas JD, De Jonghe LC. Prediction and evaluation of sintering aids for cerium Gadolinium oxide. Solid State Ionics 2007;178:1187–94.

[15] De Marco V, Sglavo VM. Effect of Bismuth oxide as sintering aid for Gadolinia-doped ceria at 1050°°C. ECS Trans 2015;68:413–20.

[16] De Marco V, Grazioli A, Sglavo VM. Production of planar copper-based anode supported intermediate temperature solid oxide fuel cells cosintered at 950°°C. J Power Sources 2016;328:235–40.

[17] Gong M, Liu X, Trembly J, Johnson C. Sulfur-tolerant anode materials for solid oxide fuel cell application. J Power Sources 2007;168:289–98.

[18] Flytzani-Stephanopoulos M, Sakbodin M, Wang Z. Regenerative adsorption and removal of H_2S from hot fuel gas streams by rare earth oxides. Science 2006;312:508–10.

[19] Papurello D, Lanzini A, Fiorilli S, Smeacetto F, Singh R, Santarelli M. Sulfur poisoning in Ni-anode solid oxide fuel cells (SOFCs): deactivation in single cells and stack. Chem Eng J 2016;283:1224–33.

[20] Konar R, Mukhopadhyay J, Sharma AD, Basu RN. Synthesis of Cu-YSZ and Ni-Cu-YSZ cermets by a novel electroless technique for use as solid oxide fuel cell anode: application potentiality towards fuel flexibility in biogas atmosphere. Int J Hydrogen Energy 2016;41(2):1151–60.

[21] Di Bartolomeo E, Basoli F, Luisetto I, Tuti S, Zurlo F, Salehi Z, et al. Ni and Ni-Co La0.8Sr0.2Ga0.8Mg0.2$O_{3-\delta}$ infiltrated cells in H_2 and CH_4/CO_2 mixture. Appl Catal B Environ 2016;191:1–7.

[22] Mahato N, Banerjee A, Gupta A, Omar S, Balani K. Progress in material selection for solid oxide fuel cell technology: a review. Prog Mater Sci 2015;72:141–337.

[23] De Marco V, Grazioli A, Sglavo VM. Production and co-sintering at 950°C of planar half-cells with CuO-GDC cermet supporting anode and Li_2O-doped GDC electrolyte. Ceram Eng Sci Proc 2016;37:31–8.

[24] Azzolini A, Sglavo VM, Downs JA. Fabrication and co-sintering of thin tubular IT-SOFC with Cu_2O-GDC cermet supporting anode and Li_2O-doped GDC electrolyte. J Eur Ceram Soc 2015;35:2119–27.

[25] Azzolini A, Sglavo VM, Downs JA. Production and performance of copper based anode supported SOFC. ECS Trans 2015;68(1):2583–96.

[26] Atkinson A, Selcuk A. Residual stress and fracture of laminated ceramic membranes. Acta Mater 1999;47:867–74.

[27] Mori M, Yamamoto T, Itoh H, Inaba H, Tagawa H. Thermal expansion of nickel-zirconia anodes in solid oxide fuel cells during fabrication and operation. J Electrochem Soc 1998;145:1374–81.

[28] Cologna M, Contino AR, Montinaro D, Sglavo VM. Effect of Al and Ce doping on the deformation upon sintering in sequential tape cast layers for solid oxide fuel cells. J Power Sources 2009;193(1):80–5.

[29] Cologna M, Contino AR, Sglavo VM, Modena S, Ceschini S, Bertoldi M. Curvature evolution and control in anode supported solid oxide fuel cells. Ceram Eng Sci Proc 2010:95.

[30] Cologna M, Sglavo VM, Bertoldi M. Sintering and deformation of solid oxide fuel cells produced by sequential tape casting. Int J Appl Ceram Technol 2010;7(6):803–13.

[31] Montinaro D, Sglavo VM, Bertoldi M, Zandonella T, Aricò A, Lo Faro M, et al. Tape casting fabrication and co-sintering of solid oxide “half cells” with a cathode-electrolyte porous interface. Solid State Ionics 2006;177:2093–7. no. 19–25 SPEC. ISS.

[32] Mukhopadhyay M, Mukhopadhyay J, Sharma AD, Basu RN. Engineered anode structure for enhanced electrochemical performance of anode-supported planar solid oxide fuel cell. Int J Hydrogen Energy 2012;37(3):2524–34.

[33] Cai PZ, Green DJ, Messing GL. Constrained densification of alumina/zirconia hybrid laminates, II: viscoelastic stress computation. J Am Ceram Soc 1997;80:1940–8.

[34] Green DJ, Guillon O, Juergen Roedel J. Constrained sintering: a delicate balance of scales. J Eur Ceram Soc 2008;28:1451–66.