Journal of Power Sources 231 (2013) 146–152

Contents lists available at SciVerse ScienceDirect

Journal of Power Sources

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

# Novel two-step processing route combining mechanical alloying and microwave hybrid sintering to fabricate dense $La_{9.33}Si_2Ge_4O_{26}$ for SOFCs

M. Santos [a], C. Alves [b], F.A.C. Oliveira [b], T. Marcelo [b], J. Mascarenhas [b], A. Cavaleiro [a], B. Trindade [a,*]

[a] CEMUC, Mechanical Engineering Department, University of Coimbra, Rua Luís Reis Santos, 3030-788 Coimbra, Portugal
[b] Laboratório Nacional de Energia e Geologia I.P., Estrada do Paço do Lumiar, 1649-038 Lisboa, Portugal

## HIGHLIGHTS

► A novel processing route was used to fabricate $La_{9.33}Si_2Ge_4O_{26}$ for SOFCs.
► High density materials were obtained by microwave hybrid sintering.
► The sintered materials exhibit an apatite structure.
► Volatilization of germanium was prevented during sintering.

ARTICLE INFO

*Article history:*
Received 11 October 2012
Received in revised form
14 December 2012
Accepted 27 December 2012
Available online 5 January 2013

*Keywords:*
SOFC
Lanthanum-based oxyapatite
Mechanical alloying
Microwave hybrid sintering
Mechanical properties

## ABSTRACT

In this work, microwave hybrid sintering at 1300 and 1350 °C was carried out for densification of $La_{9.33}Si_2Ge_4O_{26}$ mechanically alloyed powder with apatite structure. The pellets sintered at these two temperatures present the same structure (apatite) with relative densities of 92 and 96%, respectively. Mechanical analysis performed on sintered materials revealed the following results: hardness of 7.1 and 8.0 GPa, Young's modulus of 122 and 133 GPa, yield strength of 1807 and 2073 MPa and fracture toughness of 1.5 and 1.0 MPa $m^{1/2}$, respectively.



## 1. Introduction

Solid oxide fuel cells (SOFCs) are devices that allow direct conversion of chemical to electrical energy through an electrochemical reaction in a cleaner and more efficient way than conventional processes (e.g. gas turbines). The advantages of such cells include high-conversion efficiency, long-term stability, fuel flexibility and reduced emissions of polluting gases. However, it has proved difficult to develop this technology into commercially viable industrial products due to lack of appropriate electrolyte materials or manufacturing routes that would enable the cost of electricity per kW h to compete with existing technology. Yttria-stabilized zirconia (YSZ) has traditionally been used in electrolytes for SOFCs. This material exhibits high oxygen ion conductivity only at temperatures in the range of 850–1000 °C. Therefore, SOFCs using YSZ as an electrolyte must operate at similarly high temperatures, causing problems related to degradation of materials and mechanical and chemical compatibility in oxidizing and reducing atmospheres, cell sealing and life time. There is therefore a great deal of interest in lowering SOFC operation temperatures.

Recent research is being focused on the development of new materials for SOFC electrolytes with higher ionic conductivity at intermediate temperatures (500–800 °C) as alternative materials to YSZ. Rare earth silicates with an apatite-type structure, such as doped lanthanum oxides of general formula $La_{10}(MO_4)_6O_2$, where M = Ge, Co, Si, Al, or P, exhibit ionic conductivity higher than that of YSZ at intermediate temperatures, mainly due to the presence of oxide ion channels containing interstitial oxygen sites [1] combined with moderate thermal expansion coefficients and low electronic conductivities [2,3].

* Corresponding author. Tel.: +351 239790794.
*E-mail addresses:* bruno.trindade@dem.uc.pt, bruno.sacadura@gmail.com (B. Trindade).

0378-7753/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jpowsour.2012.12.101