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# Performance and degradation of $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.85}\text{Mg}_{0.15}\text{O}_{3-\delta}$ electrolyte-supported cells in single-chamber configuration

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## ABSTRACT

Electrolyte-supported cells were made of a  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.85}\text{Mg}_{0.15}\text{O}_{3-\delta}$  (LSGM2015) electrolyte (200  $\mu\text{m}$  thickness) prepared by ethylene glycol complex solution synthesis, isostatic pressing and sintered at 1400 °C, a Ni-SDC anode, a  $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{3-\delta}$  (SDC) buffer-layer between anode and electrolyte, and a  $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$ -SDC cathode. The cells were tested in single-chamber configuration using methane–air mixtures. The results of X-ray diffraction and SEM-EDS showed a single-phase in the electrolyte and conductivities ( $\sim 0.01 \text{ S cm}^{-1}$  at 650 °C) close to the typical values. Good cell power densities of 215 and 102  $\text{mW cm}^{-2}$  were achieved under  $\text{CH}_4/\text{O}_2 = 1.4$  of at 800 and 650 °C, respectively. However, the cell stability tests indicated that the operating temperature strongly influenced on the cell performance after 100 h. While no significant change in the power density was observed working at 650 °C, a clear performance degradation was evidenced at 800 °C. SEM-EDS revealed an appreciable degradation of the electrolyte and both the electrodes.

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## 1. Introduction

Last decades, great efforts have been made by many researchers to find new electrolyte materials in order to reduce the operating temperature of SOFCs [1,2], thus improving the chemical stability at long-term, the cost-effective materials for interconnectors and the balance of plant [3]. Sr- and Mg-doped lanthanum gallates ( $\text{LaGaO}_3$ ), commonly well-known as LSGM, are considered as an alternative to yttria-stabilized

zirconia (YSZ) electrolyte, especially at intermediate temperatures (600–800 °C). The doping of the  $\text{LaGaO}_3$  perovskite with Sr (in La-sites) and Mg (in Ga-sites) introduces a quantity of oxygen vacancies that results in a high conductivity ( $>0.10 \text{ S cm}^{-1}$  at 800 °C) [4]. Moreover, it exhibits a negligible electronic conductivity at temperatures lower than 1000 °C over a broad range of oxygen partial pressures, from pure oxygen ( $P(\text{O}_2) = 1 \text{ atm}$ ) to moistened hydrogen ( $P(\text{O}_2) = 10^{-22} \text{ atm}$ ), and a stable performance over long operating times [4,5], resulting in a larger performance of the cell

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[6]. The highest oxide-ion conductivities are found for the  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.85}\text{Mg}_{0.15}\text{O}_{3-\delta}$  composition at 800 °C (0.166 S cm<sup>-1</sup>) and 700 °C (0.079 S cm<sup>-1</sup>), respectively [4,7]. However, LSGM ceramics often present problems related to their chemical stability by the formation of resistive phases, such as  $\text{LaSrGa}_3\text{O}_7$ ,  $\text{LaSrGaO}_4$  or  $\text{LaGaO}_4$  [8]. In their manufacturing process, the LSGM purity strongly depends on several factors, including the purity of starting materials, methods of powder synthesis and shaping, temperature sintering, etc. Conventional synthesis method by solid-state reaction (SSR) involves high sintering temperatures, which facilitate the presence of compositional inhomogeneities, due mainly to the Ga evaporation, thus easily leading to the formation of secondary phases [9]. Alternative synthesis routes, which are based on wet chemical synthesis, such as sol–gel, co-precipitation, hydrothermal synthesis, glycine-nitrate [10–13], have been proposed to reduce the final sintering temperature. The chelation of complex cations, leading to the polyesterification in the presence of a polyhydroxy alcohol, such as ethylene glycol, to form a polymeric resin is a cost effective and simple alternative with excellent results for both the Sr- and Mg-doped  $\text{LaCrO}_3$  and  $\text{LaGaO}_3$  ceramics [14,15]. Both the phase purity and the microstructure strongly depend on the synthesis method, the composition and the sintering temperature, thus affecting both the electrical and mechanical properties [16].

Despite several LSGM compositions present good ionic conductivities to be used as electrolytes in SOFCs at intermediate temperatures [17,18], these ceramics have been not widely implemented, as these may present important chemical reactions with the electrodes during their sintering. The interfacial reaction between the Ni-based anode and the LSGM electrolyte has been reported in several works [19,20]. It induces the formation of secondary phases at the anode-electrolyte interface, thus causing a strong drop in the cell performance [21]. However, it can be avoided using a buffer layer based on doped ceria between the anode and the electrolyte, which can suppress the migration of Ni, La and Ga [19,22]. Owing to these problems, only a few studies about LSGM cells have been reported and much less literature is available about the single-chamber SOFCs (SC-SOFCs) using LSGM as electrolyte [23–25]. SC-SOFCs operate on a uniform mixture of fuel and oxidant by ensuring the selective catalytic and electrochemical reactions on each electrode [26]. While the anode should present a higher electrocatalytic activity for the oxidation of the fuel, the cathode should exhibit a higher catalytic activity for the reduction of oxygen. So, the cell performance strongly depends on the selectivity of the electrodes and the operating conditions, such as the temperature, the flow rate and the fuel-oxidant ratio [27]. The main advantage of SC-SOFCs, with respect to the conventional dual-chamber SOFCs, is to simplify the device design and operate in mixtures of hydrocarbon (natural gas, methane, propane...) and air, without the separation between the fuel and the oxidant, as their operating temperatures are optimal for the hydrocarbon reforming in the anode, which is well-known as internal reforming [28]. Therefore, SOFCs under these conditions become thermally and mechanically more resistant than conventional fuel cells. In addition, these devices generate quite heat that can be used for warm water supply

and heating, increasing the (combined) efficiency even further. Thus, it is expected that these advantages could provide a fast commercialization of the SC-SOFCs during the transition phase from fossil fuels to renewable energy sources.

Unfortunately, the stability of the used materials with respect to both the microstructural degradation and the material decomposition is still preventing a broad implementation of SOFCs. As the degradation of the cells based on LSGM electrolytes is a key factor to be studied, which has been much less investigated in single-chamber configurations than in the conventional dual chamber devices, the aim of this work is the study of the performance and degradation of  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.85}\text{Mg}_{0.15}\text{O}_{3-\delta}$  (LSGM2015) electrolyte-supported cells in single-chamber configuration. We are thus continuing our investigation on single-chamber Ni-SDC/SDC/LSGM/LSC-SDC fuel cells fed by mixtures of methane and air gas [25]. Then, the cell stability tests at short-term indicated a performance degradation around 10–15% after 55 h operating at 750 °C and 6 thermal cycles (between the room temperature and 750 °C). Now, we are extending our previous results on the cell performance degradation operating for 100 h at different temperatures, under optimal single-chamber conditions. In this case, an electrolyte with the LSGM2015 composition has been used due to its high electrical conductivity ( $\sim 0.15$  S cm<sup>-1</sup> at 800 °C) previously reported by several authors [6]. After the stability tests, the microstructures of the cells have been analysed by scanning emission microscopy (SEM) and emission dispersive spectroscopy (EDS) in order to identify the causes of the performance degradation. In addition, both the purity and the microstructure of the electrolytes sintered at different temperatures have been preliminary characterized by X-ray diffraction, SEM-EDS and impedance spectroscopy.

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## 2. Experimental

### 2.1. Synthesis and characterization of the materials

For preparing LSGM2015 powders, stoichiometric amounts of  $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  (Merck, 99.0%),  $\text{Sr}(\text{NO}_3)_2$  (Fluka, 99.0%),  $\text{Ga}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (Alfa Aesar, 99.999%),  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (Merck, 99.0%) were dissolved in nitric acid (65%) and ethylene glycol (1:8 v/v). The as-obtained solutions were thermally treated in three steps: 80 °C for 2 h, 120 °C for 3 h to obtain a black resin embedding all cations, and finally 180 °C up to combustion of the polymeric gel, which converted the resin into an expanded porous solid. After milling in an Agate mortar, the resulting powder was calcined at 900 °C for 5 h and attrition milled for 2 h in ethanol with zirconia balls. Afterwards, 1 g powder was uniaxially pressed into a pellet with 12 mm diameter at 100 MPa, isostatically pressed at 200 MPa, and finally sintered in air for 12 h at different temperatures (1400 and 1450 °C). After attrition milling, BET surface area of the powders was determined using a Quantachrome Accusorb instrument. The density of the sintered pellets was measured by the Archimedes method with distilled water. The theoretical densities were calculated from the division of the molecular weight by the cell volume as proposed by Datta et al. [29], giving a value of 6.68 g cm<sup>-3</sup> for LSGM2015. The materials were characterized

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