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Sr- and Mg-doped $LaGaO_3$ powder synthesis by carbonate coprecipitation

Nam Soo Chae [a], Kil Soon Park [a], Young Soo Yoon [b], In Sang Yoo [a], Jong Sung Kim [a], Hyon Hee Yoon [a,*]

[a] *Department of Chemical Engineering, Kyungwon University, Sungnam 461-701, Republic of Korea*
[b] *Department of Advanced Technology Fusion, Konkuk University, 173-401 Seoul, Republic of Korea*

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Abstract

Sr- and Mg-doped $LaGaO_3$ (LSGM) and Co-doped LSGM (LSGMC) powders were synthesized by coprecipitation using ammonium carbonate or ammonium bicarbonate precipitant. Precursor, calcined particles, and sintered pellets of LSGM were characterized using TGA, XRD, and SEM. The calcined powders of LSGM showed uniform microstructures with nearly spherical morphology and average particle size of 100–200 nm. The ionic conductivity of the sintered LSGM and LSGMC pellets at 1400 °C was 4.5×10^{-2} S/cm and 1.13 S/cm at measuring temperature of 800 °C, respectively. The internal microstructure was observed by FIB-SEM and it was found that the internal pores affected adversely on the ionic conductivity of the sintered LSGM pellets. The ionic conductivity of the as-prepared powders would be improved by optimization of the preparation process. The results of this study indicated that ammonium carbonate coprecipitation could be used as a convenient and economic method to produce LSGM electrolyte for low temperature SOFCs.

Keywords: Solid oxide fuel cell; Coprecipiation; Electrolyte; LSGM; Ion conductivity

1. Introduction

Recent solid oxide fuel cell (SOFC) development has been focused on a lower temperature operation less than 800 °C [1]. One of the methods to reduce the operating temperature is the use of new electrolyte materials which has higher oxide ionic conductivity. Sr- and Mg-doped lanthanum gallate (LSGM) has been found to have higher oxide ionic conductivity over a wide range of oxygen partial pressures at a low temperature (600–800 °C) compared to commonly used electrolyte materials such as YSZ [2]. Further improvement of the ionic conductivity has been reported by doping cobalt into the LSGM. Many researchers had demonstrated high performance SOFCs using LSGM as the electrolyte.

LSGM powders are typically prepared by solid-state reactions. The conventional techniques require high temperatures for sintering. Several types of wet-chemical methods, such as sol–gel, hydrothermal treatment, and coprecipitation, have been reported for the synthesis of LSGM powders [3]. The wet chemistry-derived powders were reported to show better reactivity than those prepared by solid-state reaction. However, the required sintering temperature is still above 1400 °C [4]. Recently, a carbonate coprecipitation method was developed to prepare rare-earth-doped ceria [5]. It was reported that the oxide powders synthesized by carbonate coprecipitation method could be sintered to >99.5% of the theoretical density at temperatures of 1100–1250 °C. Lowering the sintering temperature of electrolyte materials will allow fine grain sizes as well as the saving of energy [6].

In this study, we applied the carbonate coprecipitation method for the synthesis of LSGM electrolytes. Cobalt-doped LSGM (LSGMC) electrolyte powder was also prepared using this method. The powders and sintered pellets obtained were characterized by XRD, scanning electron microscopy (SEM), and focused ion beam-scanning electron microscopy (FIB-SEM). An impedance analyzer was used to determine the ionic conductivity of the sintered pellets at various temperatures. The influence of precipitation reaction conditions was evaluated.

* Corresponding author. Tel.: +82 31 750 5356; fax: +82 31 750 5574.
E-mail address: hhyoon@kyungwon.ac.kr (H.H. Yoon).

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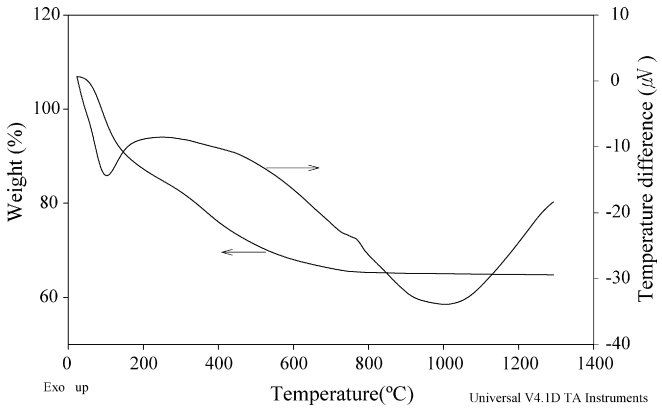

Fig. 1. TGA curves of the precursors of LSGM obtained by carbonate coprecipitation.

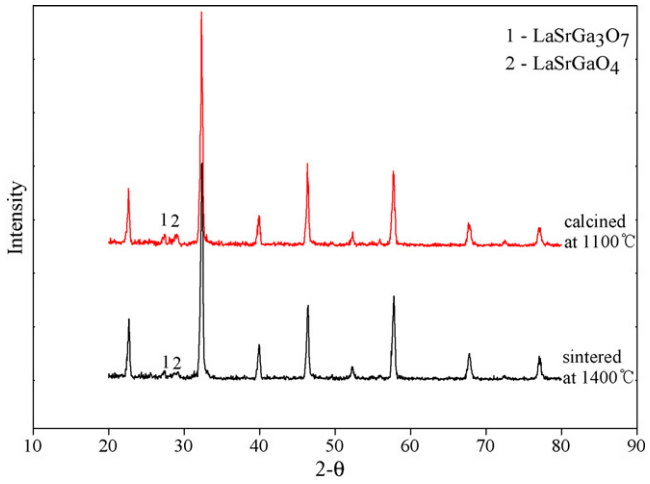

Fig. 2. XRD patterns of LSGM powder calcined at 1100 °C and LSGM pellet sintered at 1400 °C.

2. Experiment

LSGM and LSGMC powders were prepared by coprecipiation using ammonium carbonate $(NH_4)_2CO_3$ or ammonium bicarbonate $(NH_4)HCO_3$ as a precipitant. The starting salts are $La(NO_3)_3{\cdot}6H_2O$, $Sr(NO_3)_2$, $Ga(NO_3)_3{\cdot}xH_2O$, $Mg(NO_3)_2{\cdot}6H_2O$, and $Co(NO_3)_2{\cdot}6H_2O$. The stoichiometric amounts of each component of the final product $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{2.85}$ for LSGM and $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.1}Co_{0.1}O_{2.85}$ for LSGMC were dissolved in distilled water. The concentration of stock solution was 0.2 M for La^{3+}. An aqueous solution of ammonium carbonate or ammonium bicarbonate with a concentration of 1.5 M was used as the precipitant. The mixed salt solution of 200 mL was dripped at a speed of 5 mL/min into the precipitant solution in a beaker kept at 70 °C with mild stirring. The resulting suspension was aged at 70 °C for 2 h after completion of precipitation. The precipitate was filtered and dried at room temperature over 24 h. The dried powder was ground in an agate mortar for 10 min and calcined at various temperatures. The calcined powder was pressed into several disks of 15 mm diameter and 1 mm thickness at 30 MPa. The pellets were then sintered at various temperatures for 6 h in air [7].

The crystal structures of the sintered LSGM samples were analyzed via X-ray diffractometry. Differential thermal analysis/thermogravimetry (DTA/TG) of the precursor powder was performed using a TG–DTA analyzer. The morphology of the LSGM powders and sintered pellets were observed through a high resolution scanning electron microscopy (HRSEM) (Model S-4700, Hitachi). The internal microstructure of the sintered LSGM pellets was observed by a focused ion beam-scanning electron microscopy (FIB-SEM) (Model Nova-200, FEI Company). The oxide ionic conductivity of sintered samples was measured using a two-probe ac impedance method (Solatron 1280B) as a function of temperature from 773 K to 1173 K in air.

3. Results and discussion

The thermal behavior of precursors obtained using the ammonium coprecipitation method was investigated by DSC/TG analysis. Fig. 1 shows DSC/TG curves of LSGM precursors.

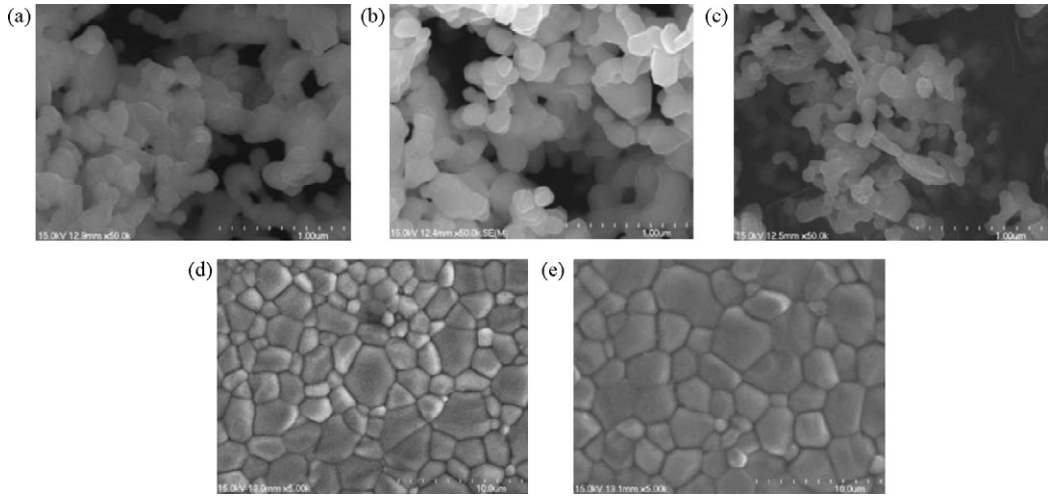

Fig. 3. SEM micrographs of LSGM powders calcined at different temperatures [(a) 900 °C, (b) 1000 °C, and (c) 1100 °C] and LSGM pellets sintered at different temperatures [(d) 1400 °C and (e) 1600 °C].