Preparation and Characterization of $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ Film by Electrophoretic Deposition Method

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Abstract. $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ (LSGM) precursor particles were successfully prepared by ultrasonic spray pyrolysis. LSGM thin films were prepared by the electrophoretic deposition (EPD) technique using ethanol containing iodine. The LSGM films were obtained by sintering at 1300 °C for 10 h. The X-ray diffraction patterns revealed that the diffraction peak of LSGM thin films was in agreement with the perovskite structure. The LSGM films attained a uniform thickness of 10 μm.

Introduction

Concerns about global warming due to the increased emission of greenhouse gases and the depletion of fossil fuels have caused a renewed interest in alternative energy sources. As a result, recently, there has been an increase in interest in solid oxide fuel cells (SOFCs) as a power generation system because of their high efficiency, low emission, fuel flexibility [1]. However, the high operating temperature (typically 1000 °C) of zirconia-based SOFCs precludes the use of metallic interconnect materials in SOFCs, which in turn has a significant impact on their cost. A reduction in the operating temperature to 600 °C would allow the use of high-temperature alloys for interconnects and would also increase the life and reliability of SOFC systems. So, the solid electrolyte materials at $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ (LSGM) is attracting a lot of attention. It exhibit a high ionic conductivity of 0.1 S/cm at 800 °C. The ionic conductivity of LSGM is around five times that of conventional yttria-stabilized zirconia [2,3]. However, it is difficult to prepare LSGM films owing to its complex composition. Electrophoretic deposition (EPD) is a colloidal process that can be conveniently used to prepare LSGM films. In this, particles are deposited directly from a stable colloid suspension by a dc electric field. It has the advantages of short formation time, few restrictions on the shape of the substrates, feasibility of use of simple deposition apparatus and suitability for mass production [4]. Another question that arises because of the reduction in operating temperature is how to reduce polarization losses at the electrodes. For intermediate temperature operation of SOFCs, Ni-samarium doped ceria (SDC) is a potential candidate for anode material [5]. In this study, LSGM precursor particles were prepared by ultrasonic spray pyrolysis. LSGM films were prepared by EPD in ethanol suspensions. Then, their particle characteristics were investigated.

Experiment

The LSGM particles were prepared by ultrasonic spray pyrolysis [6]. $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Sr}(\text{NO}_3)_2$, $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{C}_6\text{H}_5\text{GaO}_7$ were used as starting materials. The molar ratio of the metal component (La:Sr:Ga:Mg) was set at 0.8:0.2:0.8:0.2 in the starting solution. As-prepared precursor particles were calcined at 800 or 1200 °C for 2 h in air. The heating and cooling rates were 4 °C/min. The crystal phase of as-prepared particles and calcined particles was identified by X-ray diffraction (XRD, Shimadzu, XRD-6100) using $\text{CuK}\alpha$ radiation. Particle size and morphology were determined by scanning electron microscopy (SEM, JEOL, JSM-6390).

Stable suspensions for EPD were prepared by mixing LSGM particles, iodine, and ethanol. The concentrations of iodine and LSGM particles were 0.6 and 10 g/l, respectively. The LSGM particles had been calcined at 800 °C for 2 h in air. The solution was subjected to ultrasonic dispersion for 10 min and then stirred for 0 to 96 h. An aluminum foil was used as the substrate (cathode). A stainless mesh was used as the anode. The aluminum foil electrodes were mounted parallel to each other at a distance of 10 mm. Thereafter, the amount of LSGM particles deposited onto the cathode by EPD at a constant voltage of 100V for 1 min was measured at 12-h intervals.

$\text{Ni-Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{1.9}$ (Ni-SDC) added graphite (30 wt%) was used as the substrate (cathode) after pelletizing by cold isostatic pressing. Ni-SDC particles were prepared by ultrasonic spray pyrolysis.

$(\text{CH}_3\text{COO})_2\text{Ni} \cdot \text{H}_2\text{O}$, $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, and Sm_2O_3 were used as starting materials.

The molar ratio of the metal component (Ni:Ce:Sm) was set at 5.6:0.8:0.2 in the starting solution. An EPD- stable suspension was prepared by using ethanol with LSGM particles that had been calcined at 1200 °C for 2 h in air. The EPD was maintained at a constant voltage of 100 V for 5 min. The unburned LSGM films obtained were then dried in air for 1

Fig.1 XRD patterns of LSGM powders

Fig.2 SEM images of LSGM powders: (a) as-prepared, (b) sintered at 800 °C, and (c) sintered at 1200 °C

Fig.3 Amount of LSGM powder deposited by EPD at constant voltage of 100 V for 1 min, at 12-h intervals.

Fig.4 XRD patterns of LSGM films obtained by EPD at constant voltage of 100 V for 5 min, followed by sintering at 1300 °C for 10 h

Fig.5 SEM images of LSGM films obtained by EPD at constant voltage of 100V for 5 min, followed by sintering at 1300 °C for 10 h: (a) surface and (b) cross-section

day and sintered at 1300 °C for 10 h. There were few distortions when sintered, and a uniform film was formed from the LSGM particles calcined at 1200 °C for 2 h. The sintered LSGM films were then investigated by XRD and SEM.

Results and discussion

Figure 1 shows XRD patterns of as-prepared LSGM particles and those calcined at 800 and 1200 °C, respectively. Additionally, Figure 2 shows SEM images of the three types of particles. XRD diffraction patterns of calcined LSGM particles showed perovskite structure. The as-prepared particles were found to be amorphous. The crystal phase was improved by sintering at either 800 or 1200 °C for 2 h. However, an impurity phase peak was also observed. SEM showed the as-prepared particles and the LSGM particles calcined at 800 °C had spherical morphology and were

non-aggregated. On the other hand, SEM of the LSGM particles calcined at 1200 °C had more particles resulting in the collapse of spherical morphology. The average particle size ranged from 0.5 to 0.9 μm .

Figure 3 shows the influence of stirring time on the amount of LSGM particles deposited. The amount of deposition was determined by EPD at a constant voltage of 100 V for 1 min at 12-h intervals. The amount of deposition increased with increasing stirring time. However, the amount of deposition did not increase significantly at 36 h. The amount of sediment obtained was approximately 3 mg/cm^2 in 36 h. It is believed that the deposited LSGM particles were resistant to make the LSGM film. So, the electric field at 100 V was not sufficient to make the LSGM film. Hence, it is considered that a thicker LSGM film can be obtained by increasing the constant voltage level.

Figure 4 shows the XRD pattern of LSGM film obtained by EPD at a constant voltage of 100 V for 5 min. Figure 5 shows SEM images of the top view of the LSGM surface and the cross-sectional view of LSGM/Ni-SDC obtained by EPD at a constant voltage of 100 V for 5 min. XRD diffraction patterns of LSGM films showed a perovskite structure. The crystal phase was improved by sintering at 1300 °C for 10 h. However, a single phase could not be obtained. Further, a NiO-phase peak was observed. Ni migrated across the LSGM/Ni-SDC interface, as observed from the formation of a Ni-rich phase at the LSGM side. SEM showed no visible cracks on the LSGM films. The LSGM films that were obtained by EPD at a constant voltage of 100 V for 5 min and sintering at 1300 °C for 10 h had a uniform thickness of 10 μm .

Conclusion

LSGM precursor particles were successfully prepared by ultrasonic spray pyrolysis. LSGM films were prepared by EPD in ethanol using a small amount of iodine. The amount of deposition became constant at approximately 3 mg/cm^2 at 36 h. The LSGM films were obtained by EPD at a constant voltage of 100 V for 5 min and followed by sintering at 1300 °C for 10 h. The films obtained were of a uniform thickness of 10 μm . XRD diffraction patterns of LSGM films showed a perovskite structure. However, Ni will be oxidized to NiO, which is highly resistive, during the sintering of the films unless sintering is performed in a controlled atmosphere.

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