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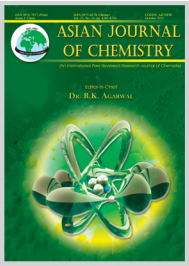

Liquid-Solid Reaction Method to Synthesize $La_{0.7}Sr_{0.3}MnO_3$ Powder as Cathode for Solid Oxide Fuel Cell†

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$La_{0.7}Sr_{0.3}MnO_3$ powders were prepared by liquid-solid reaction method using humidified (3 % H_2O) hydrogen as fuel and ambient air as oxidant. These powders were evaluated in terms of phase structure, morphology and fuel cell performances. An electrolyte-supported solid oxide fuel cell was prepared in a conventional manner. The results showed that the formation of $La_{0.7}Sr_{0.3}MnO_3$ perovskite phase was acquired at 1100 ºC for 5 h. The particles of the powders were uniform and were about 0.1-0.3 µm. At 750 ºC, the maximum power density of a single cell with $La_{0.7}Sr_{0.3}MnO_3$ powder were 135 mW cm^{-2}.

Key Words: Cathode, $La_{0.7}Sr_{0.3}MnO_3$, Solid oxide fuel cell.

INTRODUCTION

The solid oxide fuel cell offers several advantages compared with conventional power generation methods, such as: higher energy conversion efficiency (70 %), potential for cogeneration and a lower production of emission[1,2]. Recently, much attention is focused on lower temperature operation less than 800 ºC, because such operation enables to use low cost metallic interconnects. However, there are drawbacks, such as an increase of the ohmic loss at the solid-state electrolyte as well as the polarization loss at both electrodes under the lower temperature operation. In order to reduce the polarization loss, trial has been conducted to increase the electrochemical activity at both electrodes. Several perovskite compounds, containing La and Sr on A site and Co and/or Fe on B-site, have been employed as cathode materials for intermediate temperature operation solid oxide fuel cell, because they exhibit high electric and ionic conductivity, and high catalytic activity for oxygen[3]. Among the various cathode materials, (La, Sr) MnO_3 based perovskites, due to their high stability and high electrocatalytic activity for oxygen reduction, are extensively studied and investigated as cathodes[4]. In this study, $La_{0.7}Sr_{0.3}MnO_3$ (LSM) powders were prepared using liquid-solid reaction method were applied as cathode materials.

EXPERIMENTAL

In solid-liquid reaction method, stoichiometric amounts of the nitrate solution, Mn_2O_3 and small amounts of citric acid as complexing agent were mixed and milled to obtain a homogeneous suspension. The suspensions were sprayed by air spray and hot-air dried to form precursor particles. Subsequently, the precursor particles were calcined at 900-1250 ºC for 5 h to synthesize $La_{0.7}Sr_{0.3}MnO_3$ powders.

For the positive electrolyte negative (PEN), yttria stabilized zirconia (YSZ) dense electrolyte support with about 0.3 mm thickness and was prepared by tape casting method, then co-fired at 1400 ºC for 4 h. $La_{0.7}Sr_{0.3}MnO_3$/YSZ was used as cathode. The $La_{0.7}Sr_{0.3}MnO_3$ powder was characterized by an X-ray diffractometer (XRD) (D/max-rA, Rigaku). The samples were analyzed by scanning electron microscope (JEOL JSM-6400). The single cell was sealed on an alumina tube with a silver paste (DAD-87, China). The silver paste was coated on the each electrode as current collections. Humidified (3 % H_2O) hydrogen was used as fuel and ambient air as oxidant.

RESULTS AND DISCUSSION

Fig. 1 showed the XRD patterns for the $La_{0.7}Sr_{0.3}MnO_3$ powders prepared using solid-liquid reaction method. La_2O_3, LaO, MnO and Mn_3O_4 are detected after calcined at 900 ºC for 5 h. Formation of $La_{0.7}Sr_{0.3}MnO_3$ perovskite phase as a major phase is completed at 1100 ºC for 5 h. In comparison to solid phase reaction method, the calcination temperature was largely decreased[5]. Fig. 2 is the SEM image of $La_{0.7}Sr_{0.3}MnO_3$ powders after calcination at 1100 ºC for 5 h. The particles of the as-prepared powders are uniform and the size is about

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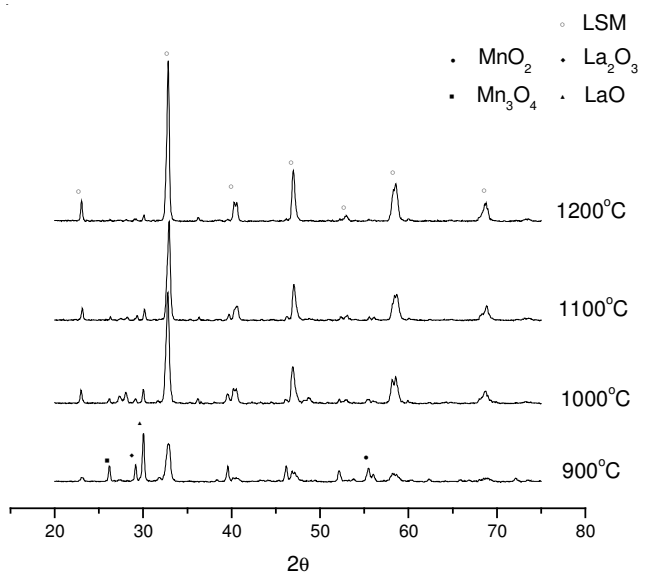

Fig. 1. XRD patterns of $La_{0.7}Sr_{0.3}MnO_3$ powders after calcination at various temperatures for 5 h in air

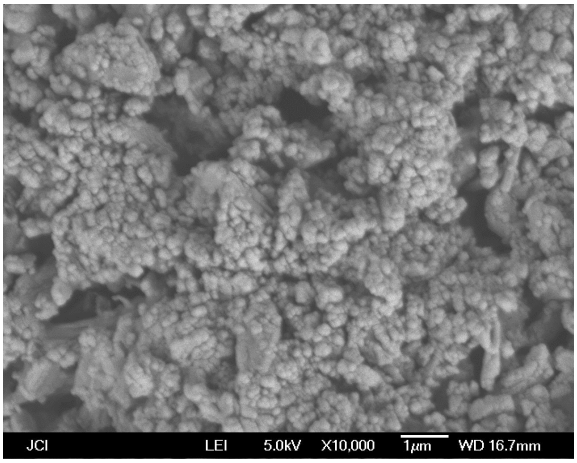

Fig. 2. SEM image of $La_{0.7}Sr_{0.3}MnO_3$ powders after calcination at 1100 ºC for 5 h

0.1-0.3 μm. In this experiment, the Mn_2O_3 was ball-milled with metallic nitrate solution and formed a suspension. After spraying the suspension, the droplets were formed, in which Mn_2O_3 particles were uniformly wrapped by La and Sr metallic nitrate complexes. Subsequently, the droplets were dried by hot air and the nitrate complexes decompose when the temperature is higher than 300 ºC. The nanosize La_2O_3, LaO, SrO particles were produced and were uniformly attached on the Mn_2O_3 surface. During the calcination, due to the uniformity of the mixing and the nanosizes of the reactant particles of La_2O_3, LaO, SrO, the perovskite phase can produce at relative low temperature and have fine reaction product. The scheme of the synthesis processing is shown in Fig. 3. Fig. 4 is the voltage-current (V-I) and power density curves of yttria stabilized zirconi-based cell with LSM/YSZ as cathode and Ni/YSZ as anode, using air as oxidant and humidified hydrogen as fuel. The maximum power density is 135 mW cm^{-2}.

Conclusion

By using the liquid-solid reaction method, the nanosize reactants and Mn_2O_3 were uniformly mixed. The calcination

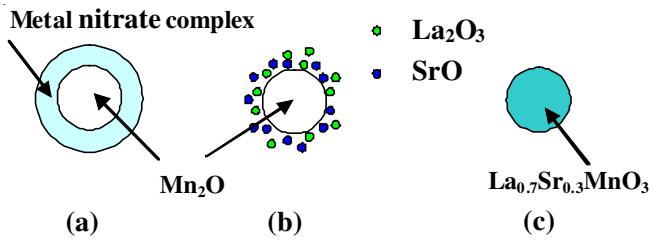

Fig. 3. Scheme of the powder synthesis processing

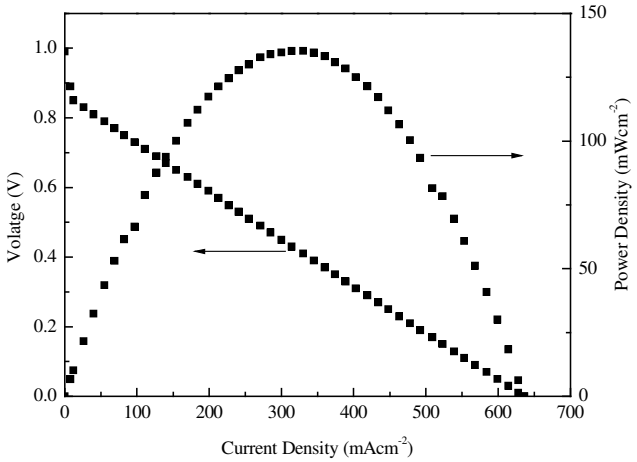

Fig. 4. Voltage-current (V-I) and performance curves of YSZ-based cells with LSM/YSZ as cathode and Ni/YSZ as anode, using air as oxidant and 3 % H_2O + H_2 as fuel at 750 ºC

temperature of the formation of the $La_{0.7}Sr_{0.3}MnO_3$ perovskite phase was largely decreased. The as-prepared $La_{0.7}Sr_{0.3}MnO_3$ powder is uniform. The maximum power density of an yttria stabilized zirconi electrolyte-supported cell is 135 mW cm^{-2} at 750 ºC by using humidified (3 % H_2O) hydrogen as fuel and ambient air as oxidant.

ACKNOWLEDGEMENTS

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