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Ionic transport mechanism of $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{2.85}\text{-(Li/Na)}_2\text{CO}_3$ composite electrolyte for low temperature SOFCs

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ABSTRACT

$\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{2.85}$ (LSGM)-(52 mol% Li_2CO_3 : 48 mol% Na_2CO_3) composite was investigated as a novel electrolyte for low temperature solid oxide fuel cells (LT-SOFCs) operated at 400–600 °C. In this study, the stability of conductivity of LSGM-30 wt% $(\text{Li/Na})_2\text{CO}_3$ composite electrolyte was been investigated by AC impedance measurements. The composite sample showed a relatively stable conductivity ($0.098\text{--}0.12\text{ S cm}^{-1}$) over 120 h measurement in air at 600 °C. As the experiment time elapsed, the conductivity had a slight decline (0.085 S cm^{-1}). Further, it was also the first time to discuss the conduction behaviours of LSGM-30 wt% $(\text{Li/Na})_2\text{CO}_3$ composite electrolyte in various dry and wet atmospheres (nitrogen, hydrogen, oxygen, carbon dioxide and air) by electrochemical impedance spectroscopy. It was found that wet atmosphere could lower transition temperature at low temperature environment (450 °C), carbon dioxide was not inert gas for composite conduction but could improve the ionic conductivity. The value of O^{2-} conductivity in oxygen and H^+ conductivity in hydrogen was obtained by DC polarization method. Based upon the performances, the possible conduction mechanism of the composite electrolytes was discussed. The results indicated that the mixed $\text{O}^{2-}/\text{H}^+/\text{CO}_3^{2-}$ conduction happened in LSGM- $(\text{Li/Na})_2\text{CO}_3$ composite electrolyte with the increasing dominance of CO_3^{2-} conduction at high temperatures (600 °C).

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Introduction

Global warming stemming from the consumption of large amounts of fossil fuels, has become one of the most serious global environmental problems. In recent years, it has become

a hot topic to reduce the exhaust emissions by replacing the conventional power-generation systems with innovative technology [1]. Solid oxide fuel cells (SOFCs) have been regarded as an attractive electrochemical device, which directly convert the chemical energy of fuel into electrical energy [2]. SOFCs have gained increasing attention as one of the

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promising ways for green energy utilizations of fossil fuels with high efficiency, low emission, and fuel-flexible power generation systems that can be adapted for both small power units and large scale power [3,4]. In recent years, an enormous number of research efforts have been invested in the development of low temperature SOFCs capable of operating at temperatures range from 400 °C to 600 °C. However, conventional SOFCs have to be operated at high temperature (800–1000 °C) to obtain adequate ionic conductivity in the yttria stabilized zirconia (YSZ) electrolyte. They suffer from lots of disadvantages in the long-term stability, sealing problems and high manufacturing cost, further restricting the development of their commercialization [5]. Therefore, decreasing the operating temperature from the current regime of 800–1000 °C to 400–600 °C has been a longstanding goal in the SOFCs study. Such low temperature (400–600 °C) operation can allow for the use of inexpensive alloy interconnects, simplify the gas sealing challenge, reduce the materials, system cost, and enhance the fuel cell durability [6,7].

Carbonate/doped ceria composite electrolyte shows enhanced properties compared to pure doped ceria [8], such as high ionic conductivity, improved stability and low electron conductivity at low temperatures (400–600 °C), which has been considered promising electrolyte for using in low temperature solid oxide fuel cells (LT-SOFCs), with ionic conductivities as high as 0.01–0.1 S cm⁻¹ achieved at 400–600 °C [9,10]. Such LT-SOFCs could be achieved a power density of 900 mW cm⁻² at 550 °C with hydrogen and air as fuel and oxidant, respectively [11]. In the process of single cell test, water appears in both sides the anode and the cathode clearly, so it is generally believed that the conduction of O²⁻ and H⁺ exists in the composites. However, up to now, there is no unified understanding about the kind of dominant ion conduction, the exact conduction mechanism for the enhanced ionic conductivity has remained unclear [12].

Carbonate/doped ceria composite electrolyte has more advantages as they have higher conductivity than YSZ at low temperature (400–600 °C). However, beyond 600 °C and in a reducing atmosphere, part reduction of Ce⁴⁺–Ce³⁺ of rare earth doped cerium oxide (DCO) occurs inside the composites becoming mixed electron/ion conductors [13]. On the other hand, perovskite oxide of strontium- and magnesium-doped LaGaO₃ (LSGM) with high oxide ionic conductivity (0.02 S cm⁻¹ at 600 °C), and negligible electronic conduction over a wide oxygen partial pressure (P_{O2}) range, good chemical stability and mechanical strength [14,15]. LSGM appears to be one of the optimal electrolytes when solid oxide fuel cells (SOFCs) are operated in the range of 600–800 °C. With the compounding of carbonate, the LSGM-carbonate composite electrolytes obtain a higher conductivity (0.12 S cm⁻¹), ideal thermal stability and notable fuel cell performance of 617 mW cm⁻² at 600 °C [16].

Nonetheless, carbonate composite electrolyte has been widely studied. But it remains unclear how the conduction mechanism influences the ionic conductivity of the novel nano-composite electrolyte. In this work, the stability of the conductivity of the LSGM-30 wt% (Li/Na)₂CO₃ composite was investigated by AC impedance measurements, the conduction behaviours of composite electrolyte in various dry and wet atmospheres (nitrogen, hydrogen, oxygen, carbon dioxide and air) were measured by electrochemical impedance

spectroscopy, the proton and oxygen ion conductivities of LSGM-30 wt% (Li/Na)₂CO₃ composite electrolyte were independently measured by the DC polarization method under different gas atmospheres (hydrogen and oxygen) in the temperature range of 450–600 °C, and the possible ionic conduction mechanism of composite electrolyte was discussed.

Experimental

Preparation of LSGM-30 wt% (Li/Na)₂CO₃ composite

LSGM powders were firstly prepared using the polyacrylamide gel combustion process [16]. Then, the LSGM powders were mixed with binary carbonates (52 mol% Li₂CO₃;48 mol% Na₂CO₃) with the content of 30 wt%. The mixtures was thoroughly ground in mortar, heat-treated at 680 °C in air for 40 min at a heat rate of 10 °C min⁻¹, and then taken out directly from the furnace to room temperature for fast cooling, followed by grinding again for use.

Electrochemical performance

For conductivity measurement, LSGM-30 wt% (Li/Na)₂CO₃ composite electrolyte was cold pressed at 300 MPa into cylindrical pellets with 13 mm diameter and about 1 mm thickness in a fixed mould machine. Then the green pellets were sintered at 600 °C for 1 h with a heating rate of 5 °C min⁻¹. Silver paste was painted onto both sides of the electrolyte pellets as the current collector, and then heated at 600 °C for 40 min. The DC conductivity measurements of the pellets were carried out in different dry and wet atmospheres (nitrogen hydrogen oxygen carbon dioxide) by using the electrochemical workstation (IM6EX, ZAHNER-Elektrik GmbH & Co. KG, Germany) at 400–650 °C.

Two types of single cells were fabricated with LSGM-30 wt% (Li/Na)₂CO₃ electrolytes. Both NiO and lithiate NiO were mixed with SDC-30 wt% (Li/Na)₂CO₃ in an equal volume for composite electrode. The composite electrode and electrolyte were co-pressed into a pellet at a pressure of 300 MPa and then sintered at 600 °C for 30 min in air. After sintering, the cells had diameter of 13 mm and thickness of 1.5 mm, silver paste was painted on the electrode and sintered at 600 °C for 40 min. Electrical conductivity of symmetrical cell A (Ag paste/mixed anode/electrolyte/mixed anode/Ag paste) was characterized in reducing atmosphere by using ZY6951 Programmable DC power (ZYTEST-CHINA) to measure the cell voltage value with the change of time under a constant current (0.1A). Cell B was operated (Ag paste/composite cathode/electrolyte/composite cathode/Ag paste) in oxidation atmosphere. In this method, stainless steel was employed as testing holder for polarization measurement, the schematic diagram as shown in Fig. 1.

Results and discussion

Stability of composites

The conductive stability of LSGM-30 wt% (Li/Na)₂CO₃ composite electrolyte was investigated by AC impedance

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