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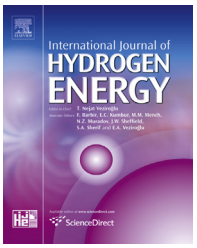

Electrochemical studies of perovskite cathode material for direct natural gas fuel cell

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ABSTRACT

Natural gas is the most promising renewable energy source and its widespread availability ensured its importance for early applications in stationary fuel cells as a reliable and low cost fuel. Therefore it is very important to efficiently utilization of natural gas in low temperature fuel cells. Herein, we demonstrate the synthesis of perovskite material of Yttrium doped $Sr_{0.92}Fe_xTi_{1-x}O_{3-\delta}$ (x = 0.25, 0.30) (YSFT) by solid state reaction method and further investigated as a new cathode material for a low temperature solid oxide fuel cell fueled by natural gas. The YSFT is characterized by X-ray powder diffraction, Brunauer–Emmett–Teller and scanning electron microscopy. The perovskite structure is achieved at relatively low temperature (850 °C). The average crystalline size is found 28 nm and 36 nm for x = 0.25 and 0.30 respectively. TGA results showed the lattice oxygen loss of YSFT is about 0.206% in its original weight in the temperature range of 25–1000 °C. The maximum electronic conductivities of 2.3 Scm^{-1} and 2.07 Scm^{-1} are achieved for x = 0.25 and x = 0.30 at 550 °C in air atmosphere respectively. It is observed that the oxygen reduction is enhanced due to the perovskite crystal structure and oxygen vacancies play an important role in the redox reaction to improve the performance of fuel cell. The YSFT perovskite cathode material based fuel cell with natural gas have achieved the power density of 250 $mWcm^{-2}$ for x = 0.25 at 550 °C. The fuel cell device has demonstrated very stable results by running continuously for 5 h with domestic available natural gas.

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Introduction

Fuel cells have got more attention because of higher efficiencies as compared to conventional energy conversion systems (combustion engines) [1–3]. Among all types of fuel cell, solid oxide fuel cell (SOFC) based on oxide ion conducting solid electrolyte operates at high temperature have number of advantages including high efficiency, low pollutant emission and fuel flexibility. The conventional SOFC are operated at high temperatures up to 1000 °C [4,5]. Such a high operating temperature also causes several problems like degradation, interfacial reaction, poisoning and corrosions and also a big challenge to commercialize. Hydrogen is a basic fuel for fuel cells but SOFC have potential to operate with other fuels than hydrogen like hydrocarbons, natural gas (methane), biogas, methanol, ethanol, ammonia, urea etc. [6,7]. Hydrogen has some critical issues like; highly flame-able, very difficult to handling, storage and transportation. Therefore, researchers try to use natural gas instead of hydrogen [8]. Among all hydrocarbons, natural gas has much importance because of its widespread and easy availability. Natural gas is the most important fuel for applications of stationary SOFC. A much higher efficiency of SOFC conversion of natural gas would be achieved by dramatic reduction in production of CO_2 in the same time when it is used as the fuel. The cracking of natural gas at anode can produce the layer of carbon that might be blocked the active sites of active material and reduced the fuel cell performance. The carbon deposition can be reduced if the sufficient amount of fuel can directly oxidized with controlled supply of oxygen transportation from cathode side [9]. The number of improvements in the utilization of hydrocarbons with new electrodes in intermediate and high temperature SOFC has been reviewed in literature [10].

The low temperature SOFC operating on natural gas requires the cathode materials which enhance the oxygen reduction reaction (ORR). To achieve the good electrochemical performance of fuel cell at low temperature the cathode polarization resistance needs to be kept as low as possible.

Perovskite metal oxide shows good conductivity (mixed electronic/ionic conductors (MIECs)), chemical stability, catalytic activity, high surface area and fine particle size. Owing to excellent structural stability of perovskite type materials are much attractive for SOFC electrodes [11–13]. For SOFC cathode, the active electrochemical sites for quick reduction of oxygen can be enhanced by using the MIECs. The number of studies are reported by different researchers related to perovskite cathode materials for SOFC such as perovskite strontium doped lanthanum magnetite (LSM) has been used as cathode material in high temperature SOFC due to their high thermal compatibility, mechanical stability and high electronic conductivity. However LSM has poor ionic conductivity with low catalytic activity at low temperature [14]. Barium strontium cobalt iron (BSCF) perovskite materials were frequently used for cathode materials for LT-SOFC. As it has high rate of oxygen diffusion, low polarization resistance and low cost. The oxygen reduction occurs at both triple phase boundary and surface of electrode which enhance the performance of SOFC. BSCF is compatible with ceria based electrolytes at intermediate to low temperature range with hydrogen fuel [15]. Lanthanum strontium cobalt iron (LSCF) perovskite has great potential as cathode material for SOFC with high conductivity and low polarization losses at low temperature 700 °C. LSCF show good performance with ceria based electrolytes at low temperature with hydrogen fuel [16]. It is also shows mixed ionic and electronic conduction, high electrochemical activity and long term stability. In perovskite LSCF, A-site is comprised of La and Sr while B-site is comprised the mixture of Fe and Co. The development of new perovskite cathode materials with low polarization resistance is therefore highly desired for the efficient LT-SOFC operating at natural gas for future energy demands at low cost [17].

In the present study, Yttrium doped $Sr_{0.92}$ Fe_xTi_{1-x} $O_{3-\delta}$ ($x = 0.25, 0.30$) (YSFT) perovskite oxide materials were synthesized by a solid state reaction method and perovskite phase has been achieved at low temperature (850 °C). The crystalline structure was determined by XRD and microstructure was analyzed by SEM. EIS results revealed low internal resistance of the fuel cell and maximum power density of 220 mW cm^{-2} has been achieved at 550 °C with natural gas operation.

Experimental

Synthesis of cathode material

The perovskite $Y_{0.08}Sr_{0.92}Fe_xTi_{1-x}O_{3-\delta}$ (YSFT) with $x = 0.25$ and 0.30 were synthesized by using conventional simple and cost effective solid state reaction method. The stoichiometric amounts of yttrium nitrate hexahydrate ($Y(NO_3)_3{\cdot}6H_2O$, Sigma–Aldrich 99.99%), strontium nitrate hexahydrate ($Sr(NO_3)_2{\cdot}6H_2O$, Sigma–Aldrich 99.99%)iron nitrate nonahydrate $Fe(NO_3)_3{\cdot}9H_2O$, Sigma–Aldrich 99.99%) and titanium dioxide (TiO_2, Sigma–Aldrich 99.99%) were mixed and grinded in a mortar and pestle. The above mixture of materials was put in to preheated furnace at 850 °C for 4 h to remove the nitrates in air. After cooled down to room temperature, the black powders were grinded again with mortar and pestle for further use as a cathode material for LT-SOFC.

Fuel cell preparation

Button size fuel cell of 13 mm diameter and 0.8 mm thickness was prepared at 300 Mpa pressure using co-pressing process. The cell composed of anode, cathode and electrolyte. The electrolyte is SDC-carbonate, anode is LiNiCuZnO nanocomposite and cathode is our prepared YSFT. The SDC-Carbonate for electrolyte and LiNiCuZnO as anode was prepared as reported in our earlier report [18]. The mass ratio of Anode, electrolyte and cathode was 0.4 g:0.3 g:0.3 g were put in to stainless (LiNiCuZn/SDC-carbonate/YSFT) steel die of 13 mm diameter and press under pressure of 300Mpa using hydraulic press. The fabricated fuel cells were sintered at 600 °C in a furnace for 30 min to make them hard. To investigate the fuel cell performance the anode and cathode surfaces of fuel cell were painted by silver paste for better current contacts. The active area of fuel cell was 0.64 cm^2. The fuel cell was

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