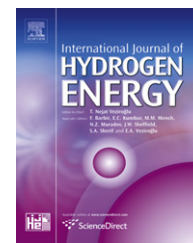


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Effect of hydrogen sulfide inclusion in syngas feed on the electrocatalytic activity of LST-YDC composite anodes for high temperature SOFC applications

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ARTICLE INFO

Article history:

Received 14 December 2011

Received in revised form

14 January 2012

Accepted 24 January 2012

Available online 3 March 2012

Keywords:

SOFC

Syngas

H₂S

Lanthanum strontium titanate

Yttrium cerium oxide

Anode catalyst

ABSTRACT

The performance of La_{0.4}Sr_{0.6}TiO_{3±δ}-Y_{0.2}Ce_{0.8}O_{2-δ} (LST-YDC) composite anodes in solid oxide fuel cells significantly improved when 0.5% H₂S was present in syngas (40% H₂, 60% CO) or hydrogen. However, electrochemical impedance spectroscopy measurements at OCV showed that polarization resistance of the cell increased when the concentration of H₂S exceeded 0.5%. Gas chromatographic and mass spectrometric analyses revealed that the rate of electrochemical oxidation of all fuel components improved when H₂S was present in the fuel. Electrochemical stability tests performed under potentiostatic conditions showed that there was no power degradation caused by the presence of H₂S in different feeds, and that there was power enhancement when 0.5% H₂S was present.

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1. Introduction

Solid oxide fuel cells (SOFCs) have been developed as alternatives to conventional energy systems with the objectives of decreasing energy production costs, more efficient utilization of energy resources and minimization of environmental pollution. Through the selection of appropriate anode catalysts SOFCs are able to use a wide range of fuels with the best known conversion efficiency at high operating temperature, and produce less environmental pollution than generators such as conventional combustion systems. These specific advantages make SOFCs attractive options as systems for energy production [1,2].

Hydrogen (H₂) has been extensively used as the fuel for SOFCs, partly in order to have a clean energy production system. Ni-YSZ based catalysts have been found to be among

the most active anode materials for electro-oxidation of H₂ when compared to alternative anode candidates including ceramic based materials. High electronic and catalytic activities [3], comparable thermal expansion coefficient (TEC) with YSZ [4], in addition to low cost, make this material a viable and economical anode for SOFC applications. However, using Ni as a catalyst has some drawbacks including: short lifetime because of particle coarsening [5]; low redox stability [6]; and, above all, its instability when using hydrocarbons [7,8] and sulfur-containing fuels [9–11].

Replacing H₂ with other fuel types including syngas (H₂ + CO) and hydrocarbons would provide a major advance toward applications of SOFCs as economical and efficient candidates for future energy conversion systems. Thus, to enable this advance it is necessary to replace Ni with another material in order to widen options for SOFC's feeds. As

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a result, a challenging but promising issue in SOFC development is replacing Ni-based anode catalysts with other types of materials with no or minimal loss of performance.

Many approaches have been investigated for increasing the stability and resistance of the anode when H₂S-containing hydrocarbons are used as anode feed. Among these, an attractive option uses anodes comprising strontium titanate (SrTiO₃) based materials with cubic perovskite structure. Appropriate electronic conductivity, TEC compatibility with yttria stabilized zirconia (YSZ, typically 8% Y₂O₃), stability toward redox reactions, and its high stability towards sulfidation when there is less than 10 wt% H₂S in the feed [12] make this material one of the most promising candidates for anode material when using sour fuels [13]. Unfortunately, the catalytic activity of this material is not as high as that of Ni and LSCM anode materials [14]. Nevertheless, SrTiO₃-based anode materials remain a potentially valuable and economically viable choice for use with high concentration H₂S-containing feeds as its stability toward H₂S could compensate for its lower catalytic activity. Marina et al. [15] tried to improve the catalytic activity of a lanthanum-substituted SrTiO₃, La_{0.4}Sr_{0.6}TiO₃ (LST), by partly substituting the B-sites with Fe, Ni, Cu, Co and Ce in order to decrease the polarization resistance of the anode. Addition of cerium ions significantly decreased the impedance of the anode and enhanced the performance of the cell when humidified H₂ was used as the feed. They also suggested that Ce ions could partially replace Ti⁴⁺ ions; however, CeO₂ itself was detected as a secondary phase. They also examined the stability of this composite material in H₂ containing H₂S, and showed it was stable for over 100 h when 100 ppm H₂S was present in H₂ under humidified conditions. Partially replacing Ti⁴⁺ with Mn³⁺ and Ga³⁺ was shown to be another effective approach for improving electrochemical activity of LST anode materials [16,17]. However there are no data on the tolerance of these materials to sour feeds. An yttrium strontium titanate (YST)-YSZ composite impregnated with CeO₂ and Ru was used as the anode material for H₂S-containing feed [18]. This anode also offered stability in low concentrations of H₂S. Cerium oxide based materials are effective catalysts for the water gas shift reaction [19,20] and oxidation and reforming of hydrocarbons [21,22]. Also, CeO₂ is an effective support for noble metal catalysts as it improves their stability [20].

It was found that the presence of H₂S increased the rate of conversion of methane as SOFC fuel using a composite LST and YSZ anode [23], an effect of participation by H₂S in the reaction mechanism without the H₂S itself being converted. However, it was not known whether this effect was peculiar specifically to the LST-YSZ anode and methane as fuel or whether similar effects would occur over other anode catalysts or when using other anode feeds. Herein we describe the effect of H₂S inclusion in the anode feed on the catalytic activity and the chemical and electrochemical stability of composite LST and Y_{0.2}Ce_{0.8}O_{2-δ} (YDC) anodes in SOFC fueled with syngas comprising 40% H₂ and 60% CO.

2. Experimental section

LST and YDC both were synthesized using a citrate-nitrate gel combustion method [24]. Lanthanum(III) nitrate

hexahydrate (La(NO₃)₃.6H₂O; Aldrich, 99.999%), strontium nitrate (Sr(NO₃)₂; Aldrich, puriss), yttrium(III) nitrate hexahydrate (Y(NO₃)₃.6H₂O; Acros, 99.9%), cerium(III) nitrate hexahydrate (Ce(NO₃)₃.6H₂O; Aldrich, 99.99%), titanium(IV) propoxide (Ti(OC₃H₇)₄; TP; Aldrich, 98%), citric acid (CA; Acros, 99%) and triethanolamine (TEA; MP Biomedicals) were used as starting precursors.

For LST preparation, CA was initially added to stirred water preheated at 60 °C. After dissolving CA, La(NO₃)₃.6H₂O and Sr(NO₃)₂ were added to prepare a homogenous solution. TP was stabilized in TEA at 60 °C for 1 h until its color changed to pale green. Then the citrate-nitrate solution was slowly added over 1 h into the latter solution while it was vigorously stirred at room temperature. The final molar ratio was 2:2:1 for CA: TEA: total metal ions.

YDC solution was prepared by adding stoichiometric amounts of Y(NO₃)₃.6H₂O and Ce(NO₃)₃.6H₂O into the citrate solution with 2:1 M ratio of CA: total metal ions and stirred continuously to form a homogeneous solution. The resulting gels were formed after continuously stirring solutions for 24 h at 80 °C. The LST and YDC gels were combusted at 550 °C and 350 °C respectively for 2 h to obtain crystalline powders. After combustion each of the LST and YDC powders was ball milled for 2 h and calcined at 900 °C for 4 h to burn off carbon residues. To increase the crystallinity of LST it was calcined at 1300 °C for a further 4 h.

Membrane electrode assemblies (MEA) were prepared having the following configuration LST:YDC 50:50 | YSZ (0.3 mm) | LSM:YSZ 50:50 | LSM. The electrodes were prepared using spin coating of electrode materials suspensions onto YSZ disks (D = 2.5 cm, t = 0.3 mm, Fuel Cell Materials) using a spin coater (Cookson Electronics, Model No. P6204), drying, then sintering at 1200 °C for 2 h.

A SOFC setup with coaxial alumina tubes (inlet and outlet) was used for feeding fuel and air to the anode and cathode sides of the MEA, respectively, and removing effluent, as described previously [25]. Each MEA was sealed between the outer anode and cathode chamber tubes using glass sealant. Gold wire and gold mesh welded together were used as current collectors. A Pt wire connected to Pt formed from

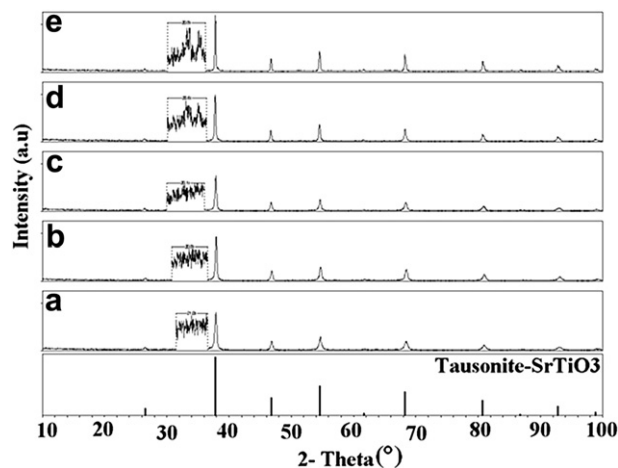


Fig. 1 – XRD patterns of LST powder calcined at: (a) 900 °C; (b) 1200 °C; (c) 1300 °C; (d) 1400 °C; (e) 1600 °C for 4 h in air.

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