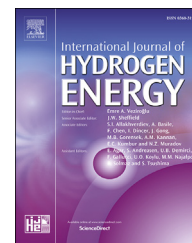




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# Role of electrocatalytic properties of infiltrated nanoparticles in the activity of cathodes of solid oxide fuel cells – A case study of infiltrated $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_x\text{Mn}_{1-x}\text{O}_3$ ( $x=0, 0.5, \text{ and } 1$ ) on Pt electrode

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## ABSTRACT

In this paper, critical role of infiltrated nanoparticles (NPs) on the electrocatalytic activity of cathodes of solid oxide fuel cells (SOFCs) for  $\text{O}_2$  reduction reaction is studied on well-defined and microstructurally clean Pt electrodes using infiltrated  $\text{La}_{0.8}\text{Sr}_{0.2}\text{CoO}_3$  (LSC),  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_3$  (LSCM), and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  (LSM) NPs. The promotion factor,  $f_p$ , as defined as the ratio of electrode polarization resistance,  $R_E$  of the electrode prepared by the conventional method to that of the infiltrated electrode measured under identical conditions, is close to unit for the  $0.2 \text{ mg cm}^{-2}$  NPs infiltrated Pt electrodes, indicating that the promotion effect of infiltrated perovskite NPs on the electrocatalytic activity of Pt electrodes is very small or negligible under open circuit conditions. However, under *dc* bias of 100 mV,  $f_p$  is 35, 18 and 7 for the infiltrated LSC-Pt, LSCM-Pt and LSM-Pt electrodes, respectively. The electrode activity is significantly promoted by the presence of the NPs under cathodic polarization conditions. The electrochemical activity of Pt electrodes with infiltrated NPs depends strongly on the nature of the infiltrated materials with the order of  $\text{LSC} > \text{LSCM} > \text{LSM}$ . The higher oxygen surface exchange rate as well as faster oxygen transportation kinetics of the B-site Co-rich perovskite oxides is responsible to the better catalytic activity of the infiltrated Pt cathodes.

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## Introduction

Operating solid oxide fuel cells (SOFCs) at intermediate temperature range (600–800 °C) has significant advantages over high temperature SOFCs operated at conventional temperature of 1000 °C, including the significantly increased selection of electrode and electrolyte materials, applicability of lost cost interconnect materials, significantly enhanced microstructural stability and reduced manufacturing cost [1]. Reduction in the thickness of the supported electrolyte films and manipulation of electrode microstructure as well as developing alternative electrode materials have also significantly enhanced fuel cell performance at reduced temperatures [2–8]. Wet impregnation or infiltration is an effective means to increase the electrocatalytic activity of SOFC electrodes at intermediate temperatures by combination of the structural stability of rigid skeleton and high catalytic activity of infiltrated nanoparticles (NPs) [9–11]. However, the catalytic role of the infiltrated NPs is not well studied and there are considerable discrepancies in the literature [12–16]. For example, Søggaard et al. [17] reported that CeO<sub>2</sub> and Sm-doped CeO<sub>2</sub> (SDC) nanoparticles have a similar promotion effect on the (La,Sr)CoO<sub>3</sub>-Gd-doped ceria (LSC-GDC) and (La,Sr)MnO<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub>-ZrO<sub>2</sub> (LSM-YSZ) composite cathodes. On the contrary, Wang et al. [18] reported that compared to ceria, the infiltration of SDC NPs resulted in much greater enhancement in the oxygen surface exchange coefficient (*k*) of LSM.

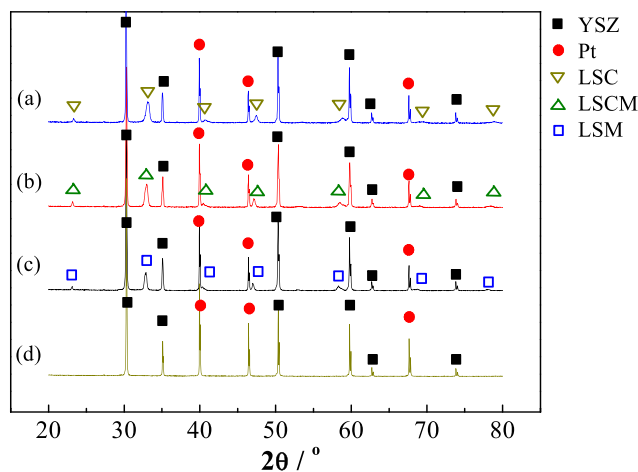
We also studied the effect of the infiltrated CeO<sub>2</sub> and Gd-doped CeO<sub>2</sub> (GDC) NPs on Pt electrode and showed that the electrochemical performance of the infiltrated Pt cathodes is closely dependent on the catalytic properties of the infiltrated NPs [19]. The advantage of using Pt as electrode skeleton is that it is chemically inert and can thus avoid the potential interferences of the changes of physical and chemical properties of the conventional perovskite oxide electrodes during the infiltration process [20–22]. Herein, we expanded the scope of the study to the infiltrated La<sub>0.8</sub>Sr<sub>0.2</sub>CoO<sub>3</sub> (LSC), La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3</sub> (LSM), and La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.5</sub>Mn<sub>0.5</sub>O<sub>3</sub> (LSCM) NPs on Pt electrodes. LSM is a state-of-the-art cathode material for high temperature SOFC, but the low ionic conductivity and high activation energy for O<sub>2</sub> reduction reaction lead to a significant increase of electrode polarization resistance at reduced temperatures [23–25]. LSC is a mixed ionic and electronic conductor (MIEC) with extraordinary electrical conductivity, exhibiting very good electrode activity at reduced temperatures [26–28]. The combination of LSM and LSC to form LSCM compromises the electrode activity and thermal and chemical stability of both LSM and LSC [29–31]. The present results clearly demonstrate that the electrode activity of the infiltrated Pt cathodes is closely associated with the catalytic activity of the infiltrated NPs and the electrode performance of infiltrated Pt increases in the order of infiltrated LSC > LSCM > LSM.

## Experimental

Electrolyte pellets were fabricated from 8 mol% Y<sub>2</sub>O<sub>3</sub>-stabilized ZrO<sub>2</sub> (YSZ, Tosoh, Japan) using die pressing and sintering

at 1500 °C for 5 h. The pellets were about 0.8 mm thick and 18 mm in diameter. Platinum paste (Pt ink 6082, Metalor) was diluted by ink vehicle (Fuel Cell Materials, USA). The diluted Pt paste was applied on the YSZ pellets by slurry coating and sintered at 1100 °C for 3 h to form thin and porous Pt skeleton electrode. The electrode geometric area was 0.5 cm<sup>2</sup>. La<sub>0.8</sub>Sr<sub>0.2</sub>CoO<sub>3-δ</sub> (LSC), La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.5</sub>Mn<sub>0.5</sub>O<sub>3-δ</sub> (LSCM) and La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3-δ</sub> (LSM) precursor solutions with concentration of 0.5 mol L<sup>-1</sup> were prepared by dissolving stoichiometric amounts of La(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O (99.9%, Alfa Aesar), Sr(NO<sub>3</sub>)<sub>2</sub> (99.0%, Sigma Aldrich), Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (98.0–102.0%, Alfa Aesar), Mn(NO<sub>3</sub>)<sub>2</sub> (50 wt% aqueous solution, Alfa Aesar), citric acid (99.5%, Sigma-Aldrich), and deionized water. The molar ratio of metal ions to citric acid was 1:1.5. The solution was infiltrated into as-prepared Pt electrodes and heat-treated at 800 °C in air for 1 h. The loading of infiltrated LSC, LSCM and LSM in the Pt electrodes was 0.2 mg cm<sup>-2</sup>. Platinum paste was applied to the other side of YSZ pellets as the counter and reference electrodes. Pt mesh was placed on the surface of the Pt working electrodes as the current collector.

A three-electrode arrangement was used for the electrochemical measurement. The electrochemical activity of the electrodes was measured by a Gamry Potentiostat Reference 3000. A current interrupt technique was used to subtract the IR contribution from the measured polarization curves. Impedance responses were measured under open-circuit and under dc bias (i.e., overpotentials) in a frequency range of 0.1 Hz–100 kHz. Electrode polarization resistance (*R<sub>p</sub>*) was measured by the difference between the low and high frequency intercepts and electrode ohmic resistance (*R<sub>o</sub>*) was obtained from the high frequency intercept on the impedance curves. The impedance behavior for the O<sub>2</sub> reduction was also studied as a function of cathodic current passage at 200 mA cm<sup>-2</sup> and 800 °C. Before the test, the electrodes were stabilized under open circuit conditions at 800 °C for 4 h. The current passage was interrupted from time to time to measure the impedance curves at open circuit. The phase of infiltrated oxides was characterized by an X-ray diffractometer (XRD, Bruker D8 Advance Diffractometer). The microstructure of the electrodes before and after the polarization treatments was



**Fig. 1** – XRD patterns of infiltrated (a) LSC-Pt, (b) LSCM-Pt, (c) LSM-Pt and (d) pristine Pt cathodes on YSZ electrolytes.

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