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### An active functional layer for carbon-tolerant anode of intermediate temperature solid oxide fuel cells

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#### ABSTRACT

A highly active anode functional layer NiTiO<sub>3</sub>-Sm<sub>0.2</sub>Ce<sub>0.8</sub>O<sub>1.9</sub> (NTO-SDC) is proposed to improve electrochemical performance of intermediate temperature solid oxide fuel cells (IT-SOFCs) with carbontolerant Ni-TiO<sub>2- $\delta$ </sub> anode. When exposing to the H<sub>2</sub> atmosphere, NTO in both anode support and anode functional layer can be totally reduced to Ni and TiO<sub>2- $\delta$ </sub>. Meanwhile, a small amount of Sm<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> pyrochlore structure phase owning acceptable oxygen-ion conduction can be observed after high temperature sintering at 1250 °C. Maximum power densities of anode-supported NTO/NTO-SDC/SDC/ La<sub>0.8</sub>Sr<sub>0.2</sub>Co<sub>0.2</sub>Fe<sub>0.8</sub>O<sub>3- $\delta$ </sub> (LSCF)-SDC single cells, are 338 mWcm<sup>-2</sup> and 223 mWcm<sup>-2</sup> with wet H<sub>2</sub> and methane fuels at 700 °C, respectively. Short-time stability test for anode-supported cells with NTO-SDC anode functional layer shows no obvious degradation with discharging at 0.5 V in wet methane fuel. Preliminary results have demonstrated that fabricating an anode functional layer of NTO-SDC is a facile and efficient strategy to improve electrochemical performance of IT-SOFCs with carbon-tolerant anode. © 2017 Elsevier B.V. All rights reserved.

#### 1. Introduction

Carbon-based fuels including biofuels and fossil fuels can be directly used for intermediate temperature solid oxide fuel cells (IT-SOFCs), which is one of most promising next-generation energy conversion devices [1]. Due to high catalytic activity Ni, countless carbon deposition on active sites of Ni-based anodes is observed for hydrocarbon fuels, resulting in deactivation and cell failure [2,3]. Recently, extensive achievements have been made in carbon-tolerant anodes: 1) exploring oxide anodes with low catalytic activity to hydrocarbon cracking reaction, such as La<sub>0.75</sub>-Sr<sub>0.25</sub>Cr<sub>0.5</sub>Mn<sub>0.5</sub>O<sub>3- $\delta$ </sub> [4,5], doped SrTiO<sub>3</sub> [6,7], Sr<sub>2</sub>Mg<sub>1-x</sub>Mn<sub>x</sub>MoO<sub>6- $\delta$ </sub> [8]; 2) applying an reforming layer to optimize the fuel composition, such as Ru-CeO<sub>2</sub>[9]. However, there are still challenges to develop low cost IT-SOFCs with carbon-tolerant anodes.

Now it seems that it is still important and promising to solve the problem of carbon deposition of Ni-based anodes, owing to its superior catalytic activity and mature technology. Xia et al. [10] used SDC impregnation to reduce the active sites of Ni framework and thus enhance the carbon tolerance of Ni-based anode. Liu et al. [11] employed nano-structured BaO to form numerous BaO/ Ni interfaces on the traditional Ni-YSZ anode, which resulted is high electrochemical performance and good cell stability in hydrocarbon fuels. Tietz et al. [12,13] reported a Ni/TiO<sub>2</sub> composite anode by the reduction of NiO/NiTiO<sub>3</sub> compacts, and achieved a power output of 0.09 W cm<sup>-2</sup> at 750 °C. Correspondingly, Rodriguez et al. [14] gained high performance using Au/TiO<sub>2- $\delta$ </sub> catalyst for the water-gas shift reactions, where water dissociated on oxygen vacancies in TiO<sub>2- $\delta$ </sub>, CO adsorbed on Au sites located nearby, and subsequent reaction steps occurred at the metal-oxide interfaces. Wang et al. [15] applied a NiTiO<sub>3</sub> (NTO) reforming layer on the Ni-YSZ anode to achieve significant carbon-tolerant improvements in humidified methane and propane fuels. Recently, Ling et al. [16] developed a robust Ni-TiO<sub>2- $\delta$ </sub> anode from the on-site reductive decomposition of NTO anode substrate, which can effectively suppress the carbon deposition by water-mediated carbonremoving reactions.

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Here, we proposed a highly active anode functional layer NiTiO<sub>3</sub>-Sm<sub>0.2</sub>Ce<sub>0.8</sub>O<sub>1.9</sub> (NTO-SDC) to improve electrochemical performance of IT-SOFCs with carbon-tolerant Ni-TiO<sub>2- $\delta$ </sub> anode, resulting from a better interface connection between the electrolyte and the anode as well as an increase in the three phase boundary (TPB) length [17].

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**Fig. 1.** XRD patterns of (a) NTO powders calcined at 800 °C for 2 h in air, (b) NTO powders reduced in H<sub>2</sub> at 700 °C for 5 h, (c) NTO-SDC mixtures (1:1 in weight ratio) co-fired at 1250 °C for 5 h, (d) co-fired NTO-SDC sample reduced in H<sub>2</sub> at 700 °C for 5 h.

#### 2. Experiment

NTO, SDC and LSCF powders were synthesized by an EDTAcitrate complexation process [18]. The anode-supported half cells of NTO/NTO-SDC/SDC were prepared by an one-step drypressing/co-firing process [19]. The cathode slurry of LSCF-SDC was then painted on SDC electrolyte, and sintered at 950 °C for 3 h in air to form single cells of NTO/NTO-SDC/SDC/LSCF-SDC.

Phase structure and chemical compatibility of NTO and SDC were performed by X-ray diffraction (XRD) testing with Cu-K<sub> $\alpha$ </sub> radiation at room temperature. Electrochemical performance of single cells was tested in a home developed cell-testing system, using humidified hydrogen (~3% H<sub>2</sub>O) and humidified methane as fuels, respectively. The flow rate of fuel gas was about 40 mL min<sup>-1</sup>. Fractured micro-structure of the cells tested with humidified CH<sub>4</sub> fuel was analyzed using scanning electron microscopy (SEM, JEOL JSM-6700F) with an energy dispersive X-ray (EDX) micro-analyzer.

#### 3. Result and discussion

Fig. 1(a) and (b) show XRD patterns of as-prepared NTO and reduced Ni-TiO<sub>2 $-\delta$ </sub> in wet H<sub>2</sub> at 700 °C for 5 h, respectively. The



Fig. 2. (a) SEM image and (b) BSE image of the cross-section of the after-testing single cell with of NTO/NTO-SDC/SDC (50 µm)/LSCF-SDC, the EDX element analyses of (c) Ti and (d) Ni.

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