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Plasma sprayed metal-supported solid oxide fuel cell and stack with nanostructured anodes and diffusion barrier layer



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ABSTRACT

Plasma sprayed LSGM (La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O₃ $_{-\delta}$) based metal-supported solid oxide fuel cells with nanostructured LDC (Ce_{0.55}La_{0.45}O₂ $_{-\delta}$)–Ni anode and LSCM (La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O₃ $_{-\delta}$) diffusion barrier layer are developed in our institute. The non-sealed single cell measurement configuration is applied to test the performance and the durability of a prepared 10 × 10 cm² cell. The tested cell can deliver the power densities of 543, 480, 352 and 214 mW cm⁻² at 0.6 V for the test temperatures of 800, 750, 700 and 650 °C respectively. Results obtained from long term durability test show that this cell with a porous Ni–Fe metal substrate, a porous LSCM diffusion barrier layer, a porous nano-structured LDC–Ni anode layer, a thin dense LDC isolation layer, a dense crack free LSGM electrolyte layer, a porous LSGM–LSCF (La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O₃ $_{-\delta}$) cathode interlayer and a porous LSCF cathode current collecting layer has a degradation rate of ~2.2% kh⁻¹ during the test period of 3700 h at conditions of 700 °C and 400 mA cm⁻² current density. The tested 18-cell stack in a sealed configuration shows that it can deliver a power of 606 W at 42.4% fuel utilization, 0.75 V and 800 °C. Each cell in the stack has an open circuit voltage larger than 1 V.

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1. Introduction

The SOFC (solid oxide fuel cell) is an electrochemical reactor for generating electricity and has some unique advantages over the traditional power generation technologies, including inherently high efficiency, low polluting gas emissions and fuel flexibility [1–3]. Typical SOFCs that use Ni–YSZ (yttria-stabilized zirconia) cermet as anodes, LSM (lanthanum strontium manganite)–YSZ composite as cathodes and YSZ as electrolytes are usually operated at high temperatures (\geq 800 °C). Over the past two decades, technical developments have focused on the development of both advanced SOFC materials and structures so that the reduction of cell operation temperature can be realized [4–8]. SOFCs with reduced operation temperature (600– 800 °C) provide numerous advantages such as wider choices of lowcost component materials, improved sealing and interfacial reaction prevention, and increased flexibility in the structure design [9–11].

Toward lowering operation temperatures, there is a tendency to shift ceramic-supported SOFCs to metal-supported SOFCs due to the potential benefits of low cost, high strength, better workability, good thermal conductivity and quicker start-up [12–14]. Fast start-up and thermal cycling are considered as the main causes of ceramic-supported SOFC breakage and stack failure [15,16]. Metal-supported

SOFCs are desired to be used in the auxiliary power unit of mobile application where high structural robustness and thermal shock resistance are required [17,18]. In addition, metallic substrates allow the use of conventional metal welding techniques for stack sealing and could significantly reduce the manufacturing costs of SOFC stacks.

SOFCs are typically manufactured using wet ceramic techniques such as tape casting combined with multi-steps of high temperature (up to 1400 °C) sintering to obtain dense electrolytes. It is difficult to incorporate metallic substrates into the wet ceramic manufacturing processes without oxidizing the metallic substrates or significantly changing metallic substrate properties. The metal substrate oxidation and the matching shrinkage between substrates and SOFC layers can be relatively easily solved in plasma spray processes, hence these processes are favorable for the sequential multi-layer depositions of metal-supported solid oxide fuel cells on porous metallic substrates [19,20]. Atmospheric plasma spraying (APS) is a well-established and fast deposition process used in widespread industrial applications. It allows reducing the interaction between layers of SOFC cells, for instance, the interaction between LSGM (La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O₃) electrolyte layer and Ni in the anode layer [18,21], as compared with the conventional high temperature sintering processes. In addition to high material deposition rates, APS processes can easily control the component composition and microstructure of a cell through variation of spray parameters, therefore, the atmospheric plasma spraying has thus appeared as a promising candidate for an inexpensive and fast cell production as well as has attracted much attention [18,19,22].



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The LSGM materials have been reported as a high ion conductivity electrolyte for intermediate temperature solid oxide fuel cells [23,24]. In our approach to establish the reduced temperature, high power density and durable SOFC technology, LSGM electrolyte, nanostructured LDC (Ce_{0.55}La_{0.45}O_{2 - δ})-Ni anode and LSCM (La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O_{3 - δ) diffusion barrier layer are adopted to fabricate the developed metal-supported SOFCs using atmospheric plasma spraying processes.}

2. Experimental

The atmospheric plasma spraying system mainly consisted of a modified Praxair SG-100 DC plasma torch to heat the injected powders from powder feeders and a planar electric furnace to preheat a porous Ni–Fe substrate. The modified Praxair SG-100 DC plasma torch with Mach I nozzle was operated at medium currents from 320 to 550 A and at high voltages from 88 to 105 V. A gas mixture from argon, hydrogen and helium was generated by a proprietary mixer and used as the plasma forming gas. More details on experimental apparatus and plasma spraying parameters can be found elsewhere [25,26].

Commercially available agglomerated powders of LDC, LDC-NiO-C (carbon), LSCF ($La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3 - \delta$)-C, LSGM and LSCM were applied to construct metal-supported solid oxide fuel cells using atmospheric plasma spraying processes. LDC, LDC-NiO-C and LSCF-C powders provided by Inframat Inc., USA were agglomerated powders that had average granule sizes ranging from 20 to 50 µm. The carbon black of 15 wt.% was added as a pore former. The carbon black was burnt out during the atmospheric plasma spraying process. The original particles of the LDC and LDC-NiO-C agglomerated powders were smaller than 100 nm and the sizes of original particles in the LSCF agglomerated powders were between 200 nm and 400 nm. The Ni content in LDC-NiO-C powders was approximately 50% by volume after hydrogen reduction. LSGM and LSCM were sintered and crushed powders provided by Marion Technologies, France. These powders were sieved into a smaller range of powder sizes before plasma spraying. Layers of LSCM, LDC-NiO, LDC, LSGM, LSGM-LSCF and LSCF were formed by APS in sequence on a 10×10 cm² porous Ni–Fe substrate to complete a Ni– Fe/LSCM/LDC-NiO/LDC/LSGM/LSGM-LSCF/LSCF cell. During plasma spraying, the Ni-Fe metal substrate was preheated at temperatures of 600-800 °C. The porous Ni-Fe substrate containing 8-10 wt.% iron had a thickness of ~1.2 mm and a permeability of ~1 darcy. A dense LDC isolation layer was used to inhibit the undesired reactions between the LSGM electrolyte and the LDC–NiO anode. A porous LSCF–LSGM composite cathode interlayer formed of LSGM and LSCF with a volumetric ratio of 50:50 was applied to match with the thermal expansion coefficients of the dense LSGM electrolyte and the porous LSCF current collector. The area of the LSCF current layer was 81 cm². After all layers of a cell were coated, it was heated at 875 °C for 3 h in air with an applied pressure ranging from 250 to 500 g cm⁻² to relax the stresses in the cell and to improve the adhesions between coated layers.

The electric performances of a 10×10 cm² single cell were measured by the SOFC measurement system located in our institute. Fig. 1 shows the schematic drawing of a cell test housing used in a nonsealed cell measurement configuration. In this non-sealed cell measurement configuration, a fuel mixture of hydrogen (800 $\text{cm}^3 \text{min}^{-1}$) and nitrogen (200 cm³ min⁻¹) was fed to the anode at 1000 cm³ min⁻¹ rate and the air oxidizer was fed to the cathode at 2000 cm³ min⁻¹ rate. Solartron 1255, Solartron 1287 and Prodigit 3356 DC electronic load were applied for the AC impedance and power measurement. In the AC impedance measurement, a 10 mV AC signal with frequencies varying from 0.01 Hz to 1 MHz was applied to the cell operated at the OCV (open circuit voltage) condition. Platinum grids and leads at the anode and cathode sides of the cell were used to measure cell current and voltage. Thermocouples close to the anode and cathode of a cell that was heated by an electrical furnace were applied to measure cell temperatures at different locations.

In the stack measurement, Crofer 22 interconnectors were applied to connect the prepared 18 cells with 10 × 10 cm² in size in serial and the home-made glass-ceramic sealant with the main compositions of BaO–Al₂O₃–B₂O₃–SiO₂ was applied for stack sealing. To reduce chromium poisoning, the surfaces of interconnectors at the cathode side were coated with LSM (La_{0.67}Sr_{0.33}MnO₃ – $_{\delta}$). The assembled 18-cell stack shown in Fig. 2 was located in an electric furnace with a temperature controller. The similar equipment was applied for the power measurement of the prepared stack. A dead weight of 30 kg was loaded on the stack for a better sealing and electrical contacts. A mixture of hydrogen fuel and nitrogen as well as an air oxidant were then fed into the stack. After reduction for several hours, the performance of the stack was tested at 800 °C. The SEM (scanning electron microscope) microstructures and



Fig. 1. Schematic drawing of the cell test housing used in a non-sealed cell measurement configuration.

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