



Highly thermal integrated heat pipe-solid oxide fuel cell

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HIGHLIGHTS

- A novel HP-SOFC by introducing high temperature heat pipe.
- Temperature gradient decreased from 31 K/cm to 13 K/cm.
- Maximum power of a single HP-SOFC reached 1.78 W at 0.6 V for flame operation.

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ABSTRACT

Temperature gradient is a significant problem for the practical application of solid oxide fuel cells (SOFCs), which may lead to low power density and the degradation of SOFCs. In order to equalize the temperature distribution and improve the electrochemical performance, the concept of a heat pipe with liquid sodium metal is introduced into the design of SOFCs. A highly thermal integrated heat pipe-solid oxide fuel cell (HP-SOFC) was fabricated and investigated. The HP-SOFC consists of a heat functional layer, a current-collecting layer, an anode layer, an electrolyte layer, and a cathode layer. For an extreme flame operation, the temperature gradient along the axis of the tubular SOFC decreases from 31 to 13 K/cm due to the high heat-transfer rate of the heat functional layer. For a single fuel cell, the power output is significantly improved by 65%, increasing from 73 to 120 mW/cm² at 0.6 V with a methane-rich flame at an equivalence ratio of 1.7. In addition, prospects for other possible applications of the HP-SOFC are discussed.

1. Introduction

Solid oxide fuel cells (SOFCs) are one of the most promising types of fuel cells and have attracted great attention for their high efficiency, flexible fuel selection, low emissions [1–3], and vibration-free operation [4]. Without the limitation of the Carnot cycle, a net electrical efficiency of 40–60% can be achieved by commercially available SOFCs [5]. Working at 600–1000 °C, SOFCs have great potential for combined heat and power (CHP) applications [6–8] or trigeneration power plants [5].

During the operation or start-up process of SOFC systems, temperature non-uniformity is a significant problem for their practical application, which has been verified in a number of studies [9–12]. For example, the temperature difference along the axis direction was found to be approximately 200 K for a typical 1.5-m-long tubular SOFC by Siemens-Westinghouse [13]. Another case is a tubular SOFC stack system design by Kattke et al., in which the temperature difference along the axis of a single tubular SOFC with a length of 125 mm was greater than 100 K [14]. Moreover, this problem is especially serious in

the study of flame fuel cells (FFCs) [15,16], which are advantageous for their fuel flexibility, simple setup, and quick start-up [17–24]. Wang investigated an FFC unit as a black-start-up device for micro co-generation systems, and the temperature difference along the axis of the 100-mm-long micro-tubular SOFC was greater than 300 K in experiments [25].

The temperature non-uniformity can result in severe damage to SOFCs. At some local hotspots, an operating temperature significantly greater than the design value may cause electrode sintering and undesirable reactions between electrolytes and electrodes [26,27]. The sintering of the electrode pore structure can reduce the reactive sites and consequently lower SOFC performance [28]. Moreover, due to the thermal stress caused by the temperature gradient and sintering, delamination and thermal cracking problems appear easily [29]. Meanwhile, the local high temperature and temperature gradient may also lead to sealing failure in some specific situations.

In order to ensure sufficient electrochemical activity without excessive degradation, the temperature range for SOFCs must be maintained within specific bounds [30]. Some methods have been proposed

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to equalize the temperature distribution of SOFCs in previous studies. The most common method is to adjust the operational parameters [6], primarily pumping excess air into the cathode to reduce temperature gradients [31–33]. Vijay et al. [34] designed a secondary air channel, with flow in the counter direction to the cathode air channel, for the purpose of minimizing the thermal gradients in SOFCs. Simulation results showed that the new design effectively eliminated the thermal gradient with satisfactory fuel utilization. However, the increased bulk air flow indicates that a larger air blower is required, which may consume approximately 15% of the total electrical generation of the SOFCs [30,35]. To reduce the extra energy consumption, Nagata used an endothermic reaction in the reformation of raw fuel to cool the cell stack [27], with highly demanding requirements for catalysts and complex construction.

To reduce extra energy consumption and the requirements of catalysts, it is of great significance to explore a novel and feasible method of equalizing the temperature distribution in SOFCs. In previous studies, Dillig et al. used planar high-temperature heat pipes in the solid oxide cell (SOC) systems to enhance the heat management mechanism [36,37].

Heat pipes are common heat-transfer devices that have been widely used in the fields of spaceflight, electronic devices [38], and communication base stations [39]. Heat pipes have usually been utilized for the purposes of cooling or waste-heat recovery [40,41]. High-temperature heat pipes usually use liquid alkali metals (Na, K or NaK alloys) as heat-transfer fluids [42], which provide very high heat-transfer rates due to the evaporation-transport-condensation process. The working fluid in the evaporator section absorbs heat from the outside and evaporates, and then the saturated vapor propagates to the condenser section carrying a large amount of heat. When facing a cold source in the condenser section, the vapor condenses and releases heat to the outside. After condensation, the working fluid is transferred to the evaporator section to implement the next cycle. Consequently, the excess heat in the devices can be removed due to the high heat-transfer rate of the heat pipe, and the temperature gradient between the evaporation and condensation zones is significantly reduced. In this way, the energy provided for SOFCs can be utilized efficiently, extra energy consumption for reducing thermal gradients can be eliminated, and costly high-performance catalysts are not required.

In this study, by introducing the concept of a high-temperature heat pipe into the design of SOFCs, a highly thermal integrated heat pipe-solid oxide fuel cell (HP-SOFC) was proposed and fabricated in order to obtain a flatter temperature distribution. An extreme example of flame operation was selected and tested in the experiment.

2. Experiment

2.1. HP-SOFC structure and fabrication

The HP-SOFC consists of five functional layers, namely cathode, electrolyte, anode, current collecting, and heat functional layers, from inside to outside, as shown in Fig. 1(a). Specifically, the cathode layer consists of a LSCF/CGO cathode layer (10 μm) and a CGO barrier layer (10 μm). The electrolyte is made up of ScSZ with a thickness of 40 μm . The anode layer consists of a Ni/YSZ anode-support layer (750 μm) and a Ni/ScSZ anode-active layer (20 μm). The current collecting layer is made up of a 30-PPI nickel foam, which also functions as a fuel-flow channel for the anode. The heat functional layer is an annular high-temperature heat-pipe structure, consisting of an envelope, a heat-pipe wick, and the working fluid inside. The envelope of the heat pipe is comprised of 1-mm-thick 310S stainless steel. The heat-pipe wick is a special wire-mesh structure also comprised of 310S stainless steel. The thickness of the wick material is 0.16 mm, with a pore radius of 38 μm . According to the operational temperature of SOFCs, 600–1000 $^{\circ}\text{C}$, liquid sodium was chosen as the working fluid, which can produce phase change and provide good stability in this temperature range. The

boiling point of sodium is approximately 883 $^{\circ}\text{C}$, and the latent heat of vaporization for sodium can reach 3874 kJ/kg, which makes sodium more suitable as the working fluid than other alkali metals. The use of liquid sodium metal as the heat-transfer medium provides a high heat-transfer rate that reduces the temperature gradient.

First, the anode-supported NiO-YSZ layer was fabricated by cast-forming a given tubular structure. The NiO-YSZ tube was pre-sintered at 1000 $^{\circ}\text{C}$ for 2 h in air, then dip-coated in NiO-ScSZ and ScSZ slurries separately to prepare the anode-active and electrolyte layers. After drying, the tube was co-sintered at 1400 $^{\circ}\text{C}$ for 4 h in air. Next, the tube was dip-coated sequentially in the CGO slurry and LSCF-CGO slurry, dried, and co-sintered at 1200 $^{\circ}\text{C}$ for 3 h in air. Details of the slurry composition and fabrication process can be found in the literature [43,44]. Next, the tube was wrapped in a 30-PPI nickel foam to prepare the current collecting layer. Finally, the tube was surrounded by a heat-pipe layer with liquid sodium metal as the heat-transfer medium. The tubular HP-SOFC with a length of 100 mm was then obtained, and the active area of the SOFC was 14.9 cm^2 .

2.2. Experimental setup

The schematic of the experimental setup is shown in Fig. 1(b). Methane and air were premixed in the mixing chamber, and then flowed through the flashback protection chamber and into a two-layer porous media burner. The porous media burner consists of two layers of alumina pellets of different diameters. The upstream layer is a 20-mm-thick bed packed with 2–3-mm-diam Al_2O_3 pellets, and the downstream layer is a 60-mm-thick bed packed with 7.5-mm-diam Al_2O_3 pellets. The fuel-rich flame, which acts as both fuel reformer and heater for the SOFC, can be stabilized at the interface of the upstream and downstream layers. The details of the experimental setup and the scale of the reactor can be found in Ref. [25]. The HP-SOFC was positioned on top of the downstream layer of the porous media burner. A version of an ordinary SOFC (a cell without the heat functional layer) was simultaneously tested for comparison.

There are 13S-type thermocouples along the axis of the reactor used to measure the temperature profiles. The temperature distribution of the porous media burner was measured by thermocouples T1–T7 at 10-mm intervals. The temperature along the axis of the tubular SOFC was measured by thermocouples T8–T13 at 20-mm intervals. An electrochemical workstation (Gamry, USA) was used to measure the electrochemical performance of the SOFC in methane flame operation.

2.3. Experimental procedure

A premixed methane/air mixture was ignited at the burner exit. The ignition mixture was at a slightly leaner equivalence ratio than the stoichiometric mixture in order to allow the flame to propagate into the porous media. In this study, the equivalence ratio is defined as

$$\Phi = \frac{n_{\text{methane}}/n_{\text{air}}}{n_{\text{methane}}^S/n_{\text{air}}^S},$$

where n_{methane} and n_{air} are the molar flow rates of methane and air, respectively, n_{methane}^S and n_{air}^S are the molar flow rates of methane and air in stoichiometric conditions, respectively.

After ignition, the flow rates of methane and air were changed to a desired fuel-rich equivalence ratio, so that the inlet methane could be reformed into H_2 and CO, which were the effective fuels for the SOFC anode. After fuel-rich conditions were obtained, the SOFC was inserted into the combustion chamber, located on the top of the downstream porous media layer. When all the thermocouple readings varied by less than 10 K in 20 min, an indication that the system had reached thermal equilibrium, the temperature distribution was recorded. The combustion products samples, i.e., the fuel source for the SOFC, were captured in a gas sample bag and measured by gas chromatography (GC). The polarization curves and electrical impedance spectra (EIS) of the SOFC

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