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Performance investigation of a micro-tubular flame-assisted fuel cell stack with 3,000 rapid thermal cycles

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HIGHLIGHTS

- A novel concept for micro-combined heat and power is proposed.
- Micro-tubular flame-assisted fuel cells are tested during 3000 thermal cycles.
- Maximum heating rate of 966 °C.min^{-1} and cooling rate of 353 °C.min^{-1} are achieved.
- Significant power density of ~257 mW cm^{-2} is achieved in combustion exhaust.
- A low voltage degradation rate is measured during the thermal cycling test.

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ABSTRACT

Solid oxide fuel cell research and development has faced challenges with slow startup, slow shutdown and a limited number of thermal cycles, which hinders the technology in areas like micro-combined heat and power. A novel micro combined heat and power system, based on a boiler/hot water heater with integrated micro-tubular flame assisted fuel cells (mT-FFCs), is proposed which requires rapid startup, shutdown and thousands of thermal cycles. A 9 cell mT-FFC stack is developed and operated in a two-stage combustor. Rapid startup and shutdown of the fuel cells is demonstrated. The first-stage combustor is ignited, turned off and re-ignited for a total of 3000 on/off, thermal cycles. A maximum heating rate of 966 °C.min^{-1} and a maximum cooling rate of 353 °C.min^{-1} is achieved while thermal cycling. Despite the presence of CO in the exhaust, the anode remains porous and crack free after ~150 h of thermal cycling testing. The mT-FFC stack continues to generate significant power, even after completing the cycling test, and a low voltage degradation rate is reported.

1. Introduction

Solid oxide fuel cells (SOFCs) are electrochemical power generation devices known, in general, for high efficiency, low emissions, high fuel flexibility, no moving parts and no noise [1–4]. While these advantages give SOFCs significant potential as a cleaner source of electricity, SOFCs are hindered by a few disadvantages. The U.S. Department of Energy has cited the main challenges as long startup time, limited number of shutdowns and corrosion and breakdown of the Balance of Plant (BoP) components at high temperature [5]. As an example of slow start-up time, a heating rate of 5 °C.min^{-1} or less has been cited in many cases for a planar SOFC [6–9] although higher rates around 50 °C.min^{-1} have been achieved with metal supported planar SOFCs [10,11]. Thermal cycling tests have been conducted to assess shutdown and startup potential, but less than 50 cycles is common in the literature for planar SOFCs and for commercial systems [12–17].

This challenge is particularly common for the dual chamber SOFC (DC-SOFC) in which the fuel and oxidant streams are physically separated [3,4]. While the DC-SOFC has advantages such as high power density and efficiency, its deployment is limited by slow startup and cycling time due to stringent sealing requirements [18]. At least three means of overcoming this challenge have been proposed including the single chamber SOFC (SC-SOFC) [19–21], the no chamber direct flame fuel cell (DFFC) [18,22–43] and the micro-tubular SOFC (mT-SOFC) [9,44–48]. The SC-SOFC, DFFC and mT-SOFC reduce the sealing constraint which allows for more rapid startup. DFFCs and mT-SOFCs have been investigated for rapid start-up, rapid shut-down and thermal cycling, but the maximum number of thermal cycles found in the literature is only 400 [44] for the papers reviewed in this study. A mT-SOFC has achieved a rapid startup of 361 °C.min^{-1}, but only 11 thermal cycles [44]. A planar DFFC has achieved a more rapid startup of 1160 °C.min^{-1}, but only 26 thermal cycles [34]. Recently, a dual

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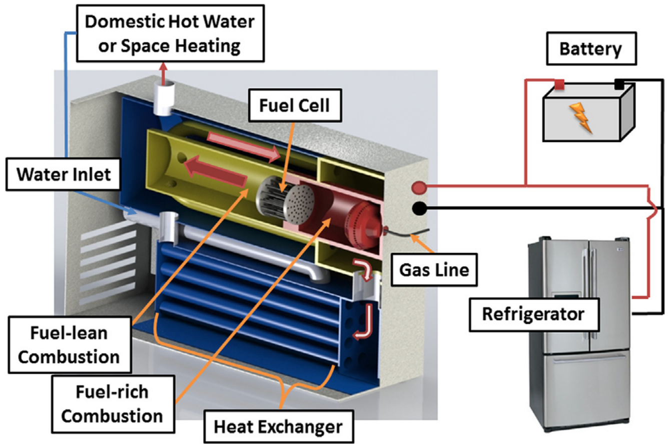

Fig. 1. One example of a FFC micro-CHP system showing a boiler with a mT-FFC stack integrated between fuel-rich and fuel-lean combustion zones.

chamber version of the original DFFC was proposed to achieve higher syngas utilization and is denoted the Flame-assisted Fuel Cell (FFC) [49–53]. To reduce the sealing constraints compared to planar and tubular SOFCs, a mT-SOFC was preferably proposed for use in the FFC setup. The combination of rapid startup capabilities while operating in combustion exhaust with a mT-SOFC is termed a micro-tubular FFC (mT-FFC) [49,50].

Combined Heat and Power (CHP) and micro-CHP systems have been proposed for the mT-FFC integrated with a residential furnace [49] and the DFFC integrated with similar systems [29,43]. Fig. 1 shows a FFC micro-CHP design integrated with a boiler. A similar integration is also possible in a furnace or hot water heater to enable a many different FFC micro-CHP systems. In this setup, a first-stage, fuel-rich combustion generates syngas and products of combustion which pass through a mT-FFC. The syngas is electrochemically converted to electricity and any remaining fuel is combusted in a second-stage, fuel-lean combustion. The heat is then recovered for hot water or space heating applications. While recent work has been conducted on fuel-rich combustion exhaust composition and FFC performance in a model fuel-rich combustion exhaust of methane and propane [49–53], a working system with actual exhaust needs further investigated. Furthermore, residential boilers startup and shutdown rapidly and can cycle over 3000 times each year [54]. A DFFC has been investigated for 26 thermal cycles only [34] while mT-FFCs have not even been investigated for thermal cycling yet. The rapid startup, shutdown and thermal cycling capabilities of a mT-FFC system need further investigation to realize the boiler shown in Fig. 1.

In this work, a mT-FFC system is developed and 9 fuel cells are connected in series to form a stack. The system is thermal cycled 3000 times and the startup and shutdown characteristics are investigated. Changes in the mT-FFC anode microstructure, stack degradation, open circuit voltage (OCV) and power density are assessed.

2. Experimental setup

2.1. Combustion chamber setup

A two-stage combustor was developed as an example of the micro-CHP system shown in Fig. 1. Fig. 2 shows the experimental setup. More details about the experimental setup are discussed in other work [55]. Methane and air (Air 2 in Fig. 2) are regulated and supplied to the fuel-rich combustion chamber via mass flow controllers. The methane flow rate is fixed at 2.4 L min^{-1} and the flow rate of air adjusts to achieve the proper equivalence ratio. The flow rates of fuel and air at different equivalence ratios are shown in Table 1. The fuel-rich combustion exhaust and mT-FFC exhaust composition was analyzed using a mass spectrometer (MS) and gas chromatograph (GC) at different equivalence ratios. The equivalence ratio (Ø) is defined in Eq. (1). Here, n_{fuel} and n_{air} are the molar flow rates of fuel and air, respectively. The expressions $n_{fuel}{}^S$ and $n_{air}{}^S$ are the molar flow rates of fuel and air needed for stoichiometric reaction, respectively.

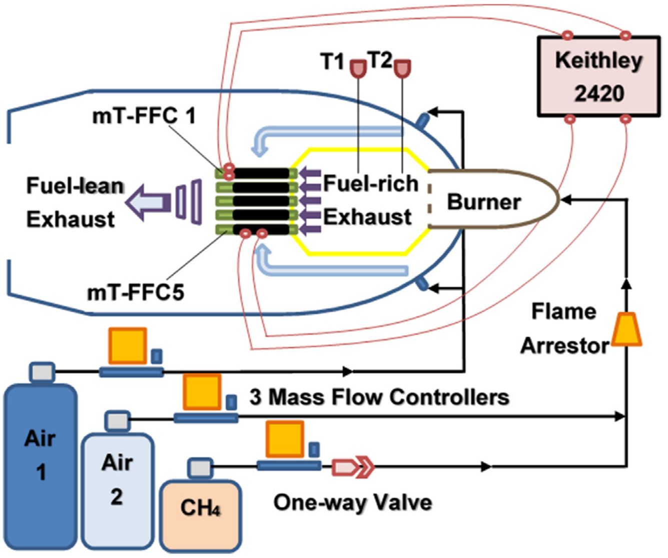

Fig. 2. Experimental setup of the mT-FFC two-stage combustion system.

Table 1
Fuel/air flow rates for the first-stage, fuel-rich combustion equivalence ratios (Ø) shown.

Equivalence ratio	Methane flow rate (mL.min^{-1})	Air flow rate (mL.min^{-1})
1.05	2400	21,760
1.10	2400	20,771
1.15	2400	19,868
1.20	2400	19,040
1.25	2400	18,278
1.30	2400	17,575
1.35	2400	16,924
1.40	2400	16,320
1.45	2400	15,757

$$\varnothing = \frac{n_{fuel}/n_{air}}{n^S_{fuel}/n^S_{air}} \tag{1}$$

A spark ignitor is used to ignite the fuel/air mixture inside the fuel-rich combustion chamber. A flame-arrestor prevents flashback after ignition. Four ports distributed evenly around the experimental apparatus allow air (Air 1 in Fig. 2) to flow over the outside of the fuel-rich combustion chamber for pre-heating. Some of the oxygen is then reduced at the mT-FFC cathode for electrochemical power generation. Fuel remaining at the mT-FFC outlet is oxidized with the remaining oxygen in a fuel-lean combustion. The temperature of the flame in the fuel-rich combustion chamber (T2 in Fig. 2) and 2 cm downstream (T1 in Fig. 2) are monitored with K-type thermocouples. The mT-FFCs are arranged in a circle with the air temperature around the fuel cell at the top of the stack (mT-FFC 1 in Fig. 2) and around the fuel cell at the bottom of the stack (mT-FFC 5 in Fig. 2) monitored with K-type thermocouples. Two additional thermocouples were placed inside mT-FFC 1 and mT-FFC 5 to monitor the anode temperature during startup and cool down. The thermal cycling test was conducted with the following steps. First, the methane and air flow rate were initiated at fuel-rich conditions with a LabVIEW program. After 5s, the ignitor sparked for 5s and the fuel/air mixture ignited. The mixture reacted for 75s and then the flow rates of methane and air were stopped. The stack was allowed to cool naturally for 95s. The cycle was then repeated. The total