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### A natural-gas fuel processor for a residential fuel cell system

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#### ABSTRACT

A system model was used to develop an autothermal reforming fuel processor to meet the targets of 80% efficiency (higher heating value) and start-up energy consumption of less than 500 kJ when operated as part of a 1-kWe natural-gas fueled fuel cell system for cogeneration of heat and power. The key catalytic reactors of the fuel processor – namely the autothermal reformer, a two-stage water gas shift reactor and a preferential oxidation reactor – were configured and tested in a breadboard apparatus. Experimental results demonstrated a reformate containing ~48% hydrogen (on a dry basis and with pure methane as fuel) and less than 5 ppm CO. The effects of steam-to-carbon and part load operations were explored. © 2008 Elsevier B.V. All rights reserved.

#### 1. Introduction

Fuel cell-based cogeneration systems are attractive for households because of the balanced heat and power output achievable from these environmentally clean and efficient devices. Fuel cell power systems are being deployed in Japan to meet or supplement the heat and electric power needs of private residences [1,2]. The ability to operate fuel cell systems with an infrastructure fuel such as natural gas, liquefied petroleum gases, or kerosene provides a tremendous advantage for distributed power generation. These stand-alone fuel cell systems typically convert the fuel in a fuel processor that delivers the fuel cell quality hydrogen to the fuel cell. The Japan Institute of Energy has established a correlation between the volume of units deployed and their unit cost, shown in Table 1, where such systems are economically attractive and can compete with existing sources of heat and power, namely electric power from the grid or fuel burners for heat. The correlation indicates that technological advancements are needed to meet the long-term (year 2016) cost target of  $4000 \, \text{kWe}^{-1}$ .

The fuel cell systems currently deployed in the Japanese residential demonstration programs use steam reformers to convert the

fuels [1,2]. This approach yields high concentrations of hydrogen for the fuel cell stack, and these systems can operate efficiently at steady state at the design point. However, the duty cycle of the residential cogeneration systems consist of daily start-stop cycling, and load that varies between 30 and 100% of capacity. Steam reformers with their large thermal mass, require considerable time and energy (fuel) to warm up to operating temperatures, and are slow to respond during load transitions. It is anticipated that, compared to a steam-reformer, an autothermal reformer (ATR)-based system can significantly reduce the thermal mass of the fuel processor, since the ATR operates at much higher (1–2 orders of magnitude) space velocities. Furthermore, ATRs have been demonstrated to have fast start-up performance [3] and can be more responsive to load changes. With less fuel energy wasted at start-up, these systems are expected to be more energy efficient over their lifecycle - an appropriate measure of a key performance metric driving the development of these distributed heat and power applications.

The purpose of this paper is to discuss the development of a natural-gas fueled ATR-based fuel processing system (FPS) as a part of a 1-kWe fuel-cell cogeneration system (FCS), for residential applications. Some key requirements of this system are [1–3]

- The FPS is to be fueled by natural gas with an average composition of 88% CH<sub>4</sub>, 5.8% C<sub>2</sub>H<sub>6</sub>, 4.5% C<sub>3</sub>H<sub>8</sub> and 1.7% C<sub>4</sub>H<sub>10</sub>.
- The FPS must maintain greater than 80% efficiency (based on the fuel's higher heating value (HHV)) over its operating range of 30–100% of capacity.

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#### Table 1

Deployed volume and costs of residential fuel cell systems in Japan.

	Fiscal year			
	2008	2009	2012	2016
Anticipated volume of deployed units (×10 <sup>3</sup> )	1.1	5	70	>300
Required unit cost (×10 <sup>3</sup> ) By using existing technology By using advanced technology	>\$30	\$21	\$12 \$8	\$4

- The fuel cell system should generate the electric power at greater than 31% electric efficiency (HHV).
- The start-up energy consumed by the FCS must be less than 3.6 MJ (1 kWh) per start.

#### 2. The fuel cell system configurations

GCTool, a fuel cell system design and analysis code [4], was used to identify two FPS configurations that can meet the efficiency and start-up energy targets and are self-sufficient in terms of water used. The first configuration, termed C1, relies on passive nonphase-change devices to recover process water and supply steam to the ATR. It offers higher thermal efficiency than the current stateof-the-art practice as the condenser that produces low-grade heat, which is difficult to utilize because of a pinch-point temperature limitation, has been eliminated. The operating steam-to-carbon (S/C) ratio in the ATR, however, is constrained by the fuel-cell stack temperature and by the pressure drops in the FPS (34 kPa or 5 psi) and in the downstream components. The second configuration, termed C2, overcomes this limitation by raising additional steam using the waste heat in the burner effluent. The S/C in C2 is higher, resulting in improved FPS efficiency but the system is more complex than C1.

Fig. 1 shows the thermal integration of the fuel processor with the downstream components in the configuration C1. At reference conditions, the reformate leaving the ATR is first used to preheat the process air to  $600 \,^{\circ}$ C in a high-temperature recuperator and then is quenched with steam to the inlet temperature of the first-stage water gas shift (WGS) reactor. The steam quench increases the overall S/C in the fuel processor to 4.9. The important parameters for the reference system are summarized below.

ATR: 750 °C (at exit), 34 kPa, 3% heat loss Air feed at 550 °C, 77 °C  $T_{\text{dewpoint}}$ ,  $\Delta P = 34 \text{ kPa} (5 \text{ psi})$ Fuel feed at 80 °C, 77 °C  $T_{dewpoint}$ ,  $\Delta P = 34$  kPa (5 psi) WGS reactor, two stages, with inlet temperatures of 375  $^\circ\text{C}$  and 300  $^\circ\text{C}$ PrOx reactor, with inlet temperature of 100 °C, 0.2 ppm CO at exit Air management Anode air blower: 34 kPa head, 60% efficiency Cathode air blower: 13.7 kPa head, 60% efficiency Water management Enthalpy wheel humidifier (EWH) for air humidification [5] Membrane humidifier (MH) for fuel humidification [5] Stack condensate is recovered Fuel cell stack (FCS) module 80% fuel utilization (FU), 50% air utilization (OU) Air preheated to 80 °C, 77 °C T<sub>dewpoint</sub> Reformate at 80 °C, >77 °C T<sub>dewpoint</sub> 0.75 V cell voltage at rated power 80°C stack temperature 13.7 kPa (2 psi) total pressure drop in stack and downstream components Parasitic losses 90% DC/AC inverter efficiency 95% DC/DC converter efficiency 95% blower motor efficiency 60% blower efficiency

#### Table 2

Performance of FPS in C1 and C2 configurations.

	C1	C2	C2*
ATR temperature	750 °C	750 °C	750 °C
S/C (ATR)	1.65	2.21	6.20
S/C (FPS)	4.9	5.5	6.2
%CO (ATR)	10.2	8.8	7.5
%CH <sub>4</sub> (ATR)	0.05	0.03	0.01
Efficiency (HHV)			
FPS (%)	88.4	88.5	88.5
FCS (%)	31.0	31.1	31.1
Total (%)	82.6	82.6	82.6
Heat recovery			
Stack radiator (W)	1300	1300	1300
Anode cooler (W)	70	160	270
Burner HX (W)	200	100	0
Desuperheater (W)	90	90	90
Condenser (W)	0	0	0
Total (W)	1660	1660	1660

Table 2 compares the thermal efficiencies of the FPS in configurations C1 and C2, the electric efficiencies of FCS, the combined thermal and electric efficiencies, and the heat loads on the heat exchangers. Also included in Table 2 is the performance of C2\* in the limit of maximum S/C that is possible in ATR.

Fig. 2 shows the temperatures and the concentrations of CO and H<sub>2</sub> exiting the components in the FPS. Humidified natural gas and process air enter the ATR at a mixing-cup temperature of 506°C. The reformate exits the ATR at 750 °C, is cooled to 519 °C in the high-temperature recuperator (HRC), and is then further cooled to 375 °C by steam injection. The WGS reaction is mildly exothermic, so that the reformate temperature rises to 429 °C after WGS1. The reformate is cooled to 300 °C in the heat exchanger HW2. There is a 5 °C temperature rise in WGS2. The heat exchanger HP1 cools the reformate to 120 °C - the target inlet temperature for the PrOx reactors. The reformate temperature rises in each of the PrOx reactors and is cooled back down in the heat exchangers (HP2 and HAC) as needed for the next component. The dew point temperature of the reformate leaving PrOx is >77 °C so that no further humidification is required to meet the >90% RH target. The reformate is cooled to 80°C in the heat exchanger HP1.

The heat exchangers HW2 and HP1 are cooled using the process water that is converted to steam to quench the reformate to 375  $^\circ C$  at the inlet to WGS1.

The CO concentration in the reformate is 10.4% at the exit of the ATR (on a dry basis). The WGS reactors reduce the CO concentration to 1.1% in the first stage and to 0.3% in the second stage. The PrOx reactors further reduce the CO concentration to 2 ppm.

The H<sub>2</sub> concentration in the reformate is 39.8% at the exit of the ATR (on a dry basis). The WGS reactors raise the H<sub>2</sub> concentration to 45.0% in the first stage and to 45.9% in the second stage. The H<sub>2</sub> concentration decreases in the preferential oxidation reactors (PrOx) to 44.9%.

The two-stage PrOx reactor in the reference system has an overall CO selectivity of 55%, operates at a stoichiometry of 1.1 (i.e., O-to-CO ratio) and achieves a CO selectivity of 63% (i.e., 37% of added  $O_2$  goes into oxidizing H<sub>2</sub> rather than CO). The second stage operates at a stoichiometry of 2.2 and achieves a CO selectivity of 44%. The overall CO stoichiometry in the two-stage PrOx is 1.8 [6].

We have estimated the start-up energy for the FPS. At 100% heat transfer effectiveness, the amount of fuel energy needed for FPS start-up is 430 kJ, with 280 kJ required to heat the catalytic reactors, 40 kJ for the heat exchangers, and 110 kJ to heat 500 g of process water to the stack temperature of 80 °C. Fig. 3 shows

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