

Short communication

## Development of compact fuel processor for 2 kW class residential PEMFCs

Yu Taek Seo<sup>a</sup>, Dong Joo Seo<sup>a</sup>, Jin Hyeok Jeong<sup>b</sup>, Wang Lai Yoon<sup>a,\*</sup>

<sup>a</sup> Hydrogen System Research Center, Hydrogen and Fuel Cell Research Department, Korea Institute of Energy Research, 71-2, Jang-dong, Yuseong-gu, Daejeon 305-343, Republic of Korea

<sup>b</sup> Chemical Engineering Department, Kyungpook National University, Republic of Korea

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### Abstract

Korea Institute of Energy Research (KIER) has been developing a novel fuel processing system to provide hydrogen rich gas to residential polymer electrolyte membrane fuel cells (PEMFCs) cogeneration system. For the effective design of a compact hydrogen production system, the unit processes of steam reforming, high and low temperature water gas shift, steam generator and internal heat exchangers are thermally and physically integrated into a packaged hardware system. Several prototypes are under development and the prototype I fuel processor showed thermal efficiency of 73% as a HHV basis with methane conversion of 81%. Recently tested prototype II has been shown the improved performance of thermal efficiency of 76% with methane conversion of 83%. In both prototypes, two-stage PrOx reactors reduce CO concentration less than 10 ppm, which is the prerequisite CO limit condition of product gas for the PEMFCs stack. After confirming the initial performance of prototype I fuel processor, it is coupled with PEMFC single cell to test the durability and demonstrated that the fuel processor is operated for 3 days successfully without any failure of fuel cell voltage. Prototype II fuel processor also showed stable performance during the durability test.

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### 1. Introduction

The fuel cell-based power generation system is considered to be well qualified for a distributed cogeneration system for residential use, which is generally incorporating the polymer electrolyte membrane fuel cells (PEMFCs) stack with hydrogen generation system from fossil fuels or renewable sources. PEMFCs-based system has been widely developed because of its high efficiency, compactness and reduction of air pollutants emission such as CO<sub>2</sub> and NO<sub>x</sub> [1]. While introducing fuel cell systems into the market, small-scale hydrogen generation system from various hydrocarbon feedstocks is required due to limited existing hydrogen infrastructures [2]. Many types of fuel processing system are being developed to meet the objectives of producing hydrogen that is pure enough for supplying to

PEMFCs [3–5]. There exist well-developed natural gas infrastructures in Korea, in which natural gas pipeline connected to the most of families and provide the hot water for heating by gas-fired boilers. Accordingly natural gas fuel processor adopting steam reforming process should be suitable for the residential PEMFCs system that requires high hydrogen concentration in reformed gas. Although the autothermal reforming process need relatively short start up time, the produced reformed gas contains less than 50% of hydrogen due to high content of nitrogen for internal combustion. Steam reforming process shows hydrogen concentration of about 75% in dry gas basis [6]. In case of steam reforming process, external burner is used to transfer the endothermic heat of reforming reaction and could avoid the failure of catalytic combustion and difficulties in maintenance.

Recently many researchers have reported their developed fuel processor capable of producing hydrogen rich gas processed from natural gas. Viessmann is developing a PEMFCs system of 2 kW electrical and 3 kW thermal power output. The main

\* Corresponding author. Tel.: +82 42 860 3070; fax: +82 42 860 3309.  
E-mail address: [wlyoon@kier.re.kr](mailto:wlyoon@kier.re.kr) (W.L. Yoon).

advantages of its fuel processing system is to reduce CO concentration at the outlet of low temperature shift process to 0.3% (dry base) by maintaining outlet temperature of steam reforming process at about 600 °C, while remaining CH<sub>4</sub> concentration of 6% (dry base) [7]. It is also reported that Fraunhofer Institute had developed the natural gas steam reformer incorporated into the EDISON system and the fuel processor showed stable performance of delivering 2 N m<sup>3</sup> h<sup>-1</sup> for PEMFCs stack as reformer efficiency of 60% without recirculation of anode-off gas [8]. Other researchers keep the CH<sub>4</sub> concentration lower than 2% to produce more hydrogen in 1 kW class fuel processor and increase the outlet temperature of steam reforming process to 687 °C to obtain the CH<sub>4</sub> conversion of 96% in 5 kW class fuel processor [9,10]. Although their reforming capacity is different, its strategy should be worthwhile to develop our 2 kW class fuel processing system. Accordingly the Korea Institute of Energy Research (KIER) has decided to develop a compact, integrated fuel processing system to provide hydrogen rich gas of 2.0 N m<sup>3</sup> h<sup>-1</sup> by maintaining high reforming temperature and introducing reliable PrOx reactors. The main focus of the work is to integrate the unit processes of a natural gas steam reformer, water gas shift reactor, steam generator and internal heat exchangers into a single modular unit and to connect separate two-stage PrOx reactors for more convenient temperature control. In this project, the prototypes of fuel processor delivering 2.0 and 2.5 N m<sup>3</sup> h<sup>-1</sup> hydrogen were developed and tested to investigate the steady state behavior at nominal capacity.

In parallel with the system integration, the steam reforming and CO preferential oxidation (PrOx) catalysts have been developed to improve the system performance. Especially the PrOx process is important to avoid the poisoning of PEMFC anode catalyst and guarantee the durability of PEMFCs cogeneration system. The Pt-based catalysts have been used for CO removal process because of its high activity, in which CO was reduced to 100 ppm with the addition of excess air corresponding to [O<sub>2</sub>]/[CO]=2 and further reduced below 10 ppm with additive air corresponding to [O<sub>2</sub>]/[CO] ≥ 3, at which excess oxygen consumes hydrogen in reformed gas by oxidation and reduces the reforming efficiency [9]. Recently, it is reported that novel Ru catalyst has high CO removal performance in a single-stage PrOx reactor with additive air corresponding to [O<sub>2</sub>]/[CO]=1.5 by further reduction with gas mixture of hydrogen and nitrogen after aqueous reduction [11,12]. However, Ru catalyst also has undesired methanation activity for CO and CO<sub>2</sub>, which is very sensitive with reaction temperature causing complex hardware for temperature control. In order to maintain low additive air amount and avoid complex control hardware, two-stage PrOx reactors are adopted for the CO removal process. Pt-based catalyst is introduced to first stage with simple temperature control hardware and Ru-based catalyst is introduced to second stage without temperature control hardware. In this paper we report the characteristics of developed prototype fuel processors such as product gas composition and thermal efficiency. Also, it is presented that the performance of lab-prepared Ru-based catalyst adopted at second PrOx reactor and the effect of support structure in PrOx reactions.

## 2. Experiments

### 2.1. Preparation of Ru-based PrOx catalyst

The Ru-based catalyst was prepared by impregnation method. RuCl<sub>3</sub> (RuCl<sub>3</sub>·xH<sub>2</sub>O, Aldrich Chem. Co.) and 2–4 mm size of γ-Al<sub>2</sub>O<sub>3</sub> granule were used as metal precursors and support. They were agitated for at least 0.5 h and then the moisture was removed by rotary vacuum evaporator. The catalyst was further dried in oven at 110 °C overnight and calcinated at 300 °C for 4 h. The same procedure was used to prepare the Ru-based catalyst with α-Al<sub>2</sub>O<sub>3</sub> granule to verify the effect of the support structure. Catalytic activity was measured in a fixed bed flow reactor made of quartz tube under atmospheric pressure. Fifty milligrams of the catalyst was placed at the center of reactor. The test gas is provided by mass flow controllers at a flow rate of 100 ml min<sup>-1</sup> corresponding to GHSV of 120,000 h<sup>-1</sup>. The reactant gas mixture consists of 1 vol.% CO, 1 vol.% O<sub>2</sub> and 60 vol.% H<sub>2</sub> in N<sub>2</sub> balance. To measure the preferential oxidation activity, the temperature of the reactor was increased to the desired temperature under test gas flow. The gas composition at the inlet and the outlet of the reactor at steady state was analyzed by a gas chromatograph equipped with TCD, methanizer and FID.

### 2.2. Fuel processor assembly

The basic engineering design concepts are to produce cascade heat flow and to maximize heat transfer efficiency between the heat sources (enthalpy of natural gas combustion, enthalpy of reformed gas and exothermic heat from water gas shift reaction) and heat sinks (feeding water, natural gas and endothermic heat of steam reforming reaction) with minimization of heat loss to the ambient by integrating modularized unit process. The schematic design of fuel processor is explained in Fig. 1. The Korean town gas was used as a natural gas fuel for testing the fuel processor and the composition of the city gas was generally 90.2 vol.% CH<sub>4</sub>, 6.6 vol.% C<sub>2</sub>H<sub>6</sub>, 2.3 vol.% C<sub>3</sub>H<sub>8</sub> and 0.9 vol.% C<sub>4</sub>H<sub>10</sub> with 3.8 ppm of sulfur compounds as odorants. The THT and TBM are desulfurized in a bed of Mn-based adsorbent at room temperature. After preheating the water near boiling tem-

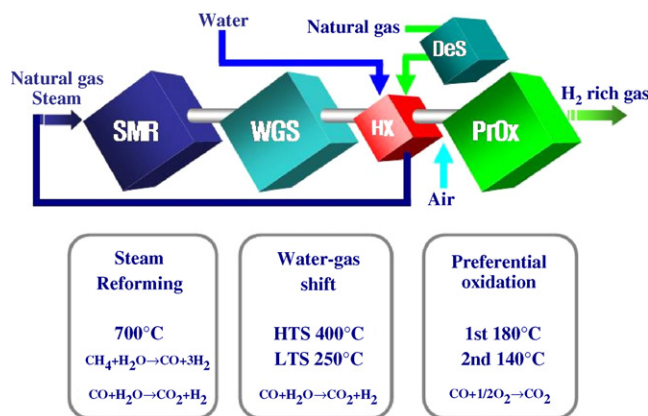


Fig. 1. Process flow diagram of fuel processor.

perature in steam generator using exothermic heat of water gas shift processes, the steam and desulfurized natural gas are mixed and introduced into the steam reforming process. The introduced hydrocarbons including  $\text{CH}_4$ ,  $\text{C}_2\text{H}_6$  and  $\text{C}_3\text{H}_8$  are converted to  $\text{H}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$ , residual  $\text{CH}_4$  and  $\text{H}_2\text{O}$  on a Ni-based catalyst at about  $680^\circ\text{C}$ . The endothermic heat of steam reforming reaction is mainly supplied by the combustion heat of natural gas and air by an external burner. The reformed gases from steam reforming process pass through two consecutive processes of high and low temperature water gas shift reaction at  $400$  and  $250^\circ\text{C}$ , respectively. The Fe- and Cu-based catalysts are used at each shift reaction processes. The CO concentration in the reformed gas is reduced to below 1.0 vol.% by exothermic shift reaction, in which more hydrogen is generated with  $\text{CO}_2$ .

The integrated fuel processor is composed of steam reformer, water gas shift reactors, steam generator, external burner and internal heat exchangers, while separate two-stage CO preferential oxidation is added to the fuel processor outside of the insulated system. Air is added to the outlet gas from shift reactor and the gas mixture is provided to the preferential oxidation process. The oxidation of CO by  $\text{O}_2$  reduces the CO concentration in reformed gas to below 10 ppm, which is the prerequisite to avoid the poisoning of anode catalyst in PEMFCs [9]. In two-stage preferential oxidation, the amount of air added to reactor can be reduced by optimizing the added air at each stage [11], and less exothermic heat is expected at each stage that result easier temperature control of catalyst beds. The CO preferential oxidation process using Pt-based catalyst is used at first stage and Ru-based catalyst is used at second stage because the higher exothermic heat in first stage might result in uncontrollable methanation reaction of  $\text{CO}_2$  on Ru-based catalyst.

The total volume of the system with thermal insulation is 381. The composition of reformed gas at each unit processed is analyzed by on-line gas chromatograph (HP 6890N system, Agilent Technologies Inc.) equipped with TCD, methanizer and FID using 8 vol.%  $\text{H}_2/\text{He}$  as carrier gas. Natural gas for burner and reforming process was provided with mass flow controllers (Alicat Scientific), while air for burner and preferential oxidation process was provided with blower and mass flow controllers (Alicat Scientific), respectively. The water flow rates were controlled by high speed metering pump (RHOCKC, Fluid Metering Inc.). All these components and fuel processor are housed as shown in Fig. 2.

### 3. Results and discussion

#### 3.1. Activity of the prepared $\text{Ru}/\text{Al}_2\text{O}_3$ catalysts

Fig. 3 shows the CO preferential oxidation activity of  $\gamma\text{-Al}_2\text{O}_3$  supported Ru catalysts at different Ru loadings of 0.5 and 1.0 wt% under the condition of  $[\text{O}_2]/[\text{CO}]=1.0$  and  $\text{GHSV}=120,000\text{ h}^{-1}$ . As the reaction temperature was increased to  $140^\circ\text{C}$ , CO conversion of 1.0 wt% Ru catalyst reaches only 50% while maintaining 50% of  $\text{O}_2$  consumption. However, when the reaction temperature rise to  $160^\circ\text{C}$ , CO conversion is greatly increased to 93% and reaches 99% at reaction temperature of  $180^\circ\text{C}$  with  $\text{O}_2$  consumption of 100%. In case

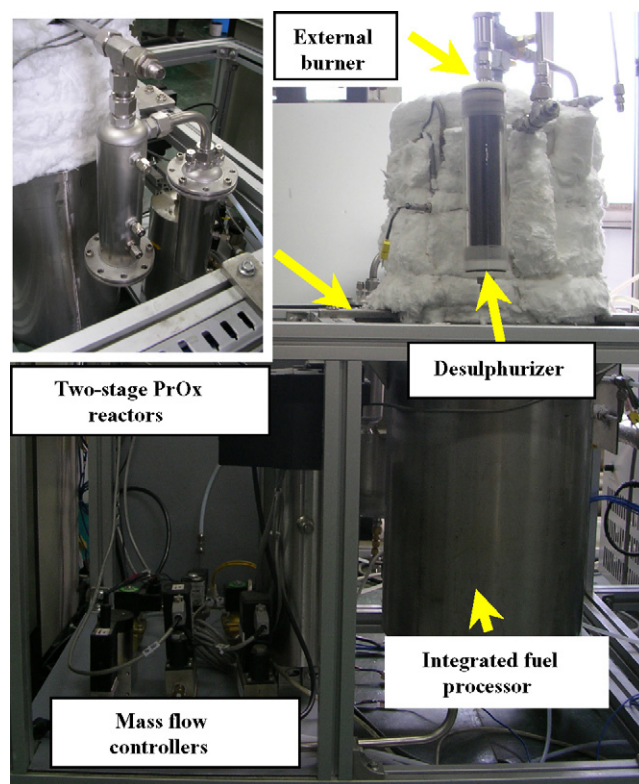


Fig. 2. Test system for 2.0 kW class natural gas fuel processor (inset: two-stage preferential oxidation reactor).

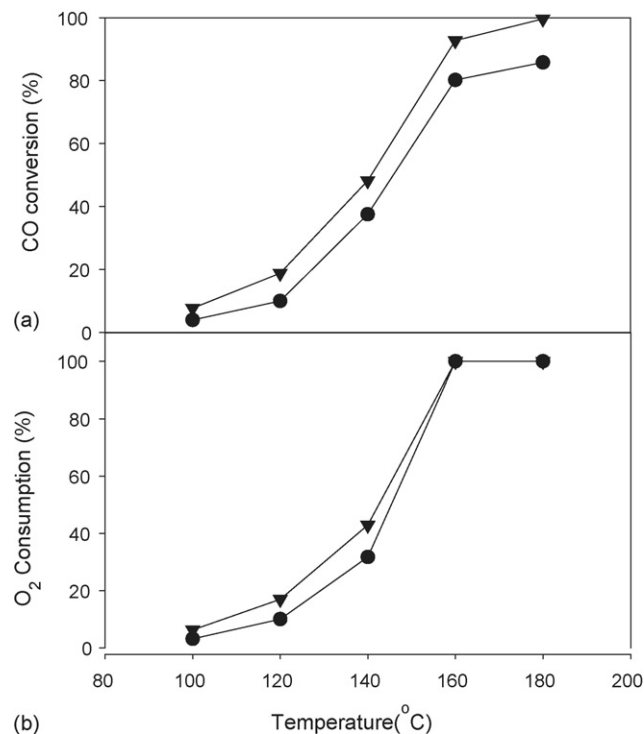


Fig. 3. The effect of Ru loading on CO preferential oxidation activity of  $\gamma\text{-Al}_2\text{O}_3$  supported Ru catalyst. (a) CO conversion, (b)  $\text{O}_2$  consumption. Reaction conditions: 1.0 vol.% CO, 1.0 vol.%  $\text{O}_2$ , 57.0 vol.%  $\text{H}_2$ ,  $\text{N}_2$  balance and  $\text{GHSV}=120,000\text{ h}^{-1}$  (dry base); (●) Ru loading=0.5 wt%, (▼) Ru loading=1.0 wt%.



of 0.5 wt% Ru catalyst, CO conversion remains 85% even at reaction temperature of 180 °C. It is noted that the added O<sub>2</sub> is completely consumed on both 0.5 and 1.0 wt% Ru catalyst at 160 °C, but the CO conversion is much higher on 1.0 wt% Ru catalyst, which means CO was oxidized more effectively in 1.0 wt% loading and unnecessary hydrogen consumption is avoided. Therefore high CO removal conversion is achieved with 1.0 wt% Ru catalyst at a temperature range of 160–180 °C, which is the suitable condition for second stage of PrOx reactors of prototype I because low exothermic reaction heat should be evolved after reducing CO concentration at first stage.

In addition with  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supported catalysts,  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> was used to verify the effect of support structure on the activity of 2.0 wt% Ru-based catalyst. The activities of CO preferential oxidation of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> supported Ru-based catalyst under the condition of [O<sub>2</sub>]/[CO] = 1.0 and GHSV = 120,000 h<sup>-1</sup> are shown and compared with  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supported catalyst in Fig. 4. The Ru-based commercial catalyst showed good CO conversion of 100% at around 120 °C, although the methanation of CO reduced the CO conversion above 140 °C. Lab-prepared Ru-based catalyst on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> showed 80% of CO conversion at 120 °C and reached 98% at 140 °C, while Ru-based catalyst on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> showed 20 and 50% of CO conversion at each temperature. In contrast to Ru-based commercial catalyst, lab-prepared Ru-catalysts suppress the methanation reaction above 140 °C and could maintain high CO conversion even at 180 °C. Moreover the added O<sub>2</sub> is completely consumed on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> supported catalyst above 120 °C, which indicate that the CO molecules are converted to CO<sub>2</sub> more effectively on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> supported catalyst under the investigated condition. The physical properties and microscopic characteristics of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> supported Ru-based catalyst are under investigation and will be reported in near future. However this basic activity study showed the possibility of introducing the catalyst to PrOx reactors of prototype II fuel processor.

### 3.2. Performance of fuel processors

The prototypes I and II fuel processors are designed to produce 2.0 and 2.5 N m<sup>3</sup> h<sup>-1</sup> hydrogen at nominal load operation,

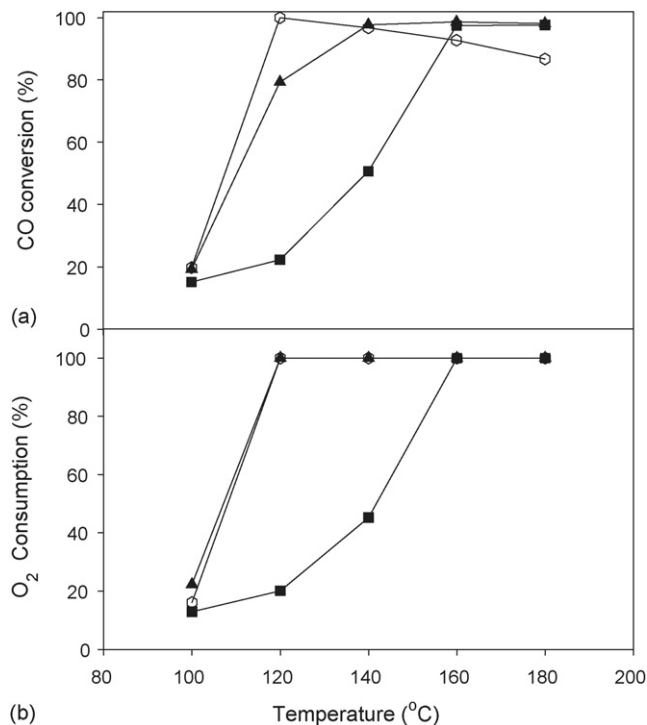


Fig. 4. The CO preferential oxidation activities of Ru catalysts on various supports. (a) CO conversion, (b) O<sub>2</sub> consumption. Reaction conditions: 1.0 vol.% CO, 1.0 vol.% O<sub>2</sub>, 57.0 vol.% H<sub>2</sub>, N<sub>2</sub> balance and GHSV = 120,000 h<sup>-1</sup> (dry base); (○) Ru-based commercial catalyst; (▲) 2.0 wt% Ru on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>; (■) 2.0 wt% Ru on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>.

respectively. The performance of developed fuel processors is shown in Table 1.

The operation of prototype I is carried out under the steam to carbon (S/C) ratio of 2.5 and the outlet temperature of steam reforming process of 680 °C. The composition of reformed gas from the low temperature shift process is 76.2 vol.% H<sub>2</sub>, 19.2 vol.% CO<sub>2</sub>, 0.3 vol.% CO and 4.3 vol.% CH<sub>4</sub> leading the methane conversion of 81%. The outlet CO concentration of steam reforming process is about 13 vol.%, and then reduced to 0.3 vol.% with employing high and low temperature shift processes at suitable operating temperature of 400 and 250 °C, respectively. As described above, two-stage reactors adopting Pt

Table 1  
Performances of fuel processor at nominal load operation

	2.0 kW	2.5 kW
Nominal capacity	2.0 kW	2.5 kW
Volume of fuel processor (including thermal insulation)	o.d. 265 mm × H700 mm, 38 l	o.d. 265 mm × H725 mm, 40 l
Flow rate of NG for process (l min <sup>-1</sup> )	11.1	13.0
Flow rate of water for process (g min <sup>-1</sup> )	25.2	35.6
S/C ratio	2.5	3.0
Composition of product gas (vol.%, dry basis)		
H <sub>2</sub>	71.4	71.3
CO <sub>2</sub>	19.2	18.4
CH <sub>4</sub>	4.3	3.7
CO	<10 ppm	<10 ppm
N <sub>2</sub>	5.1	6.6
Methane conversion (%)	81	83
Fuel processing efficiency (% HHV)	73	76

and Ru catalyst at each stage remove residual CO below down to 10 ppm, with adding of air for preferential oxidation reaction. Although, it has been reported that Ru-based catalyst has wider temperature window and high selectivity for CO oxidation, the side reaction named methanation could be occurred and resulted the conversion of CO and CO<sub>2</sub> to CH<sub>4</sub> with consumption of H<sub>2</sub> [11]. This unexpected reaction reduces H<sub>2</sub> concentration in reformed gas and highly exothermic reaction might cause problems of temperature control. Therefore, in two-staged cascade reactors, Pt-based catalyst reduce CO concentration below 400 ppm at 180 °C in first stage, and then Ru-based catalyst clean up residual CO below 10 ppm at 140 °C without any complex hardware for temperature control. The total flow rate of air added to two stages is 0.165 N m<sup>3</sup> h<sup>-1</sup>.

It has been reported that thermal efficiency of fuel processor should include the factor of the fuel utilization rate in anode of PEMFCs. However, in case of stand-alone fuel processing system without recirculation of the anode off gas, following equation, defining thermal efficiency as the heating value of the produced hydrogen referred to the heating value of the provided natural gas for reforming process and for external burner, is suitable to evaluate and characterize the overall reforming processes [13].

$$\eta = \frac{\Delta H_{\text{H}_2-\text{HHV}} \times n_{\text{H}_2}}{\Delta H_{\text{NG}-\text{HHV}}(n_{\text{NG}-\text{reforming}} + n_{\text{NG}-\text{burner}})}$$

The higher heating value is used to calculate the thermal efficiency. The produced hydrogen from steam reforming and water gas shift processes is slightly decreased during the preferential oxidation process because the added air is consumed to oxidise CO and also a small amount of hydrogen resulting a slight decrease in efficiency. The thermal efficiency of fuel processor at nominal capacity was 73% as a HHV basis. The fuel processing was operated with decreasing the S/C ratio from 3.0 to 2.5 and the thermal efficiency shows no abrupt change with maintaining CO concentration below 10 ppm. Therefore it is concluded that the S/C ratio of 2.5 is suitable for the operation of fuel processor.

A durability of prototype I was tested at nominal capacity operation with coupling to PEMFCs single cell. The composition of product gas and cell voltage of PEMFC single cell were measured after reaching the fuel processor steady state and shown in Fig. 5 under the condition of constant 400 mA cm<sup>-2</sup>, cell temperature of 70 °C, fuel utility of H<sub>2</sub>/Air = 70%/40%, RH of H<sub>2</sub>/Air = 100%/80% and operating pressure of H<sub>2</sub>/Air = 0 psig/0 psig. The CO concentration in the product gas was below 10 ppm during the three day operation and the H<sub>2</sub> concentration was stable as showing in no failure of producing electricity in PEMFC single cell. The performance and reliability of fuel processor prototype I has been confirmed with this operation.

The development of prototype I fuel processor provides much information to improve the design and operation of fuel processor. Recently, newly designed prototype II fuel processor had been built and tested. Prototype II is designed to produce 2.5 N m<sup>3</sup> h<sup>-1</sup> hydrogen for 2.5 kW class residential PEMFCs under the operating condition of S/C = 3.0 and [O<sub>2</sub>]/[CO] = 1.5. The improved performance is shown in Table 1 together with

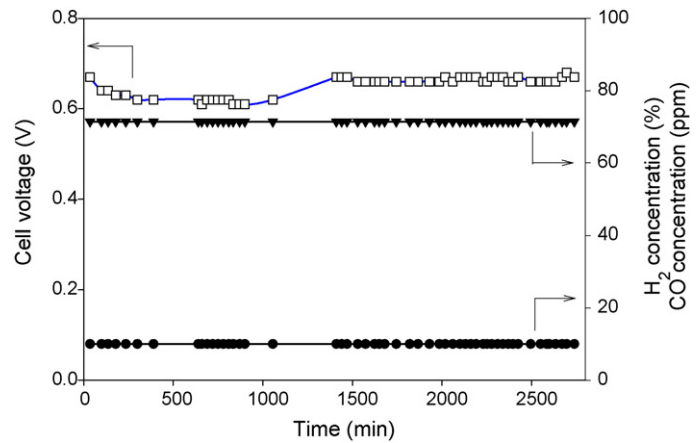


Fig. 5. Performance of fuel processor at nominal capacity operation. (▼) H<sub>2</sub> concentration; (●) CO concentration and (■) single cell voltage at constant 400 mA cm<sup>-2</sup>, cell temperature: 70 °C, fuel utility: H<sub>2</sub>/Air = 70%/40%, RH: H<sub>2</sub>/Air = 100%/80%, operating pressure: H<sub>2</sub>/Air = 0 psig/0 psig.

the performance of prototype I. The thermal efficiency and CH<sub>4</sub> conversion are both increased to 76 and 83%, respectively with optimizing the heat exchange rate between steam reforming process and steam generator. Two-stage PrOx reactors adopting Pt- and Ru-based catalyst at each stage reduce the CO concentration below 10 ppm with addition of air. In first stage, the temperature control unit is improved and the Pt-based catalyst reduces the CO concentration from ~1 mol% to 300 ppm at 180 °C. The Ru-based catalysts on α-Al<sub>2</sub>O<sub>3</sub> in second reactor clean up the CO below 10 ppm at 140 °C, in which exothermic heat is easily controlled by introducing internal heat exchanger inside the reactor and other coolant is not needed. The mixing unit is used for complete mixing of reformed gas and added air, resulting the optimization of additive air [O<sub>2</sub>]/[CO] = 1.5.

After confirming initial performance of prototype II fuel processor, durability test was carried out at nominal capacity. The composition of product gas and thermal efficiency were obtained after reaching the fuel processor steady state and shown in Fig. 6. It is noted that the composition is stable for more than 3 days

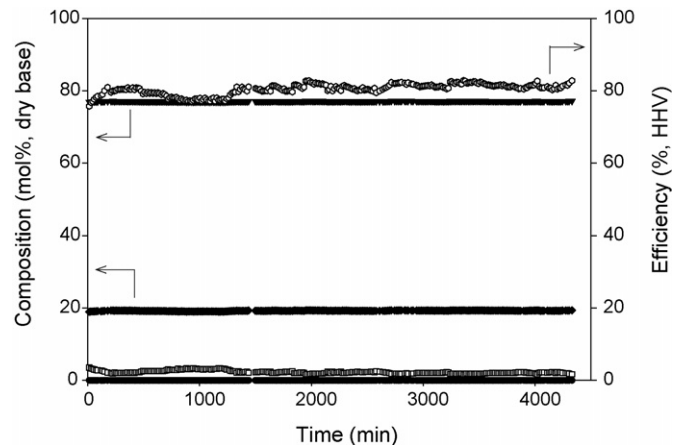


Fig. 6. Durability of prototype II at nominal capacity operation under the condition of S/C = 3.0, [O<sub>2</sub>]/[CO] = 1.5; (○) thermal efficiency; (▼) H<sub>2</sub> concentration; (◆) CO<sub>2</sub> concentration; (□) CH<sub>4</sub> concentration; (●) CO concentration.

and the CO concentration in the product gas kept below 10 ppm during the operation. The stability of fuel processor and the durability of two-stage PrOx reactors have been confirmed, therefore the coupling with PEMFCs stack will be started to complete the residential heat and power cogeneration system. The system integration would be completed during FY 2006 and the characteristics of the system will be investigated to obtain the information for further improved fuel processor, PEMFCs stack and integrated system.

#### 4. Conclusion

Prototype I fuel processor capable of delivering  $2.0 \text{ N m}^3 \text{ h}^{-1}$  hydrogen with dual pipe geometry has been developed, which features integrated structure of natural gas steam reforming, high and low temperature water gas shift reaction, steam generator and internal heat exchangers. The fuel processor shows the thermal efficiency of 73% at HHV basis and  $\text{CH}_4$  conversion of 81%. Two-staged cascade reactors employing Pt and Ru catalysts, respectively, were used to remove CO less than 10 ppm by the preferential oxidation. The developed fuel processor was connected to the PEMFC single cell to test the performance and reliability of the fuel processing system. CO concentration was remained below 10 ppm and produced  $\text{H}_2$  shows no failure of fuel cell voltage during 3 days operation. Newly designed prototype II fuel processor is under investigation and showing improved performance on thermal efficiency of 76% and  $\text{CH}_4$  conversion of 83%. The durability test of prototype II demon-

strated the stable performances and kept the CO concentration below 10 ppm. The integration of prototype II and PEMFCs stack will be performed and tested in near future.

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