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Development of 1 kW SOFC power package for dual-fuel operation

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ABSTRACT

A compact SOFC power generation system was developed by integrating a 1 kW SOFC stack and balance-of-plant. The system was designed for dual-fuel operation using both natural gas (NG) and liquefied petroleum gas (LPG). An adiabatic pre-reformer was employed in a fuel processing system to convert C₂₊ hydrocarbons in the fuel into CH₄-rich gas which was further processed in a main reformer to produce H₂-rich gas for the SOFC stack. The SOFC system was operated for 350 h under thermally self-sustaining condition, and on-load fuel switching from NG to LPG was carried out during the operation. The system performance was not significantly affected by NG/LPG composition ratios and the performance was stable during continuous operation in NG or LPG.

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1. Introduction

Fuel cell system offers cogeneration capability with potential reduction of greenhouse gas emissions which is essential for the future residential power generation. Several residential PEMFC systems have been already demonstrated with an electrical efficiency over 30% and a total efficiency over 70% while higher electrical efficiency is expected in SOFC systems [1–5]. Natural gas (NG) is a convenient fuel for residential application, but liquefied petroleum gas (LPG) can be utilized in remote areas where NG is not available. LPG can be also used as a back-up to NG in the case of emergency. Under the program initiated by National RD&D (Research, Development and Demonstration) Organization for Hydrogen and Fuel Cell in Korea, we have been developing fuel-flexible SOFC system

for residential application. The program is being led by Korea Electric Power Corporation and the final target is to develop 1 kW power package with an electrical efficiency higher than 35% using dual gaseous fuels such as NG and LPG.

Although internal fuel reforming is possible in SOFC, direct introduction of NG or LPG to the SOFC is not desirable since higher hydrocarbons other than methane contained in the fuel (C₂₊ hydrocarbons) are prone to cause carbon deposition in the SOFC anode. Actually C₂₊ hydrocarbons are suspected as carbon deposition precursor on Ni-based catalyst during steam reforming reaction [6]. Therefore, it is necessary to remove C₂₊ hydrocarbons to ensure long-term stability of the steam reformer or the anode of SOFC stack. Pre-reforming of the fuel has been considered as a means to convert C₂₊ hydrocarbons into methane-rich gas for high-temperature fuel cells [7–11].

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We developed a compact SOFC system employing a pre-reformer for dual-fuel operation, with emphasis on thermal integration of the system to cope with the possible problems which can arise from the dual-fuel operation such as thermal management issues and carbon formation. Previous study on fuel flexibility in the SOFC system revealed that significant challenges on thermal management may result from the use of different types of fuels in the same integrated fuel cell reformer, and the SOFC system may require significant changes in operating condition and/or system design in order to operate well on a variety of fuels [12]. Although operation of SOFC using NG and LPG was studied before, the experiment was conducted not in the fully integrated system, but in a temperature-controlled testing station [8]. Moreover, previous literatures related to the dual-fuel operation of the high-temperature fuel cell did not deal with thermal integration of the system in detail [10,11]. In this study the heat duties of heat exchangers necessary for dual-fuel operation were estimated by process simulation and the calculated results were utilized in the development of a SOFC system. Performances of the SOFC system using both NG and LPG were investigated for 350 h. In addition, the effect of fuel switching from NG to LPG on performance of the system was closely studied.

2. System design

2.1. System configuration

A photograph of the SOFC power package is shown in Fig. 1. The power package has overall dimension of $60 \times 60 \times 130 \text{ cm}^3$ which is more integrated than the previous system [13,14]. The main power package divides into two parts: lower cold balance-of-plant (BOP) and upper hot box.

2.1.1. Cold BOP

The cold BOP systems developed by Hyosung Corporation are comprised of both mechanical and electrical BOP. The

mechanical BOP includes an air blower, an NG compressor, a water pump, mass flow controllers for hydrogen and LPG, a desulfurizer, and a water condenser. The electrical BOP consists of a data acquisition/control system and a power conditioning system which connects the SOFC power output to the electric grid.

2.1.2. Hot box

In the center of the hot box, a SOFC stack is installed. Fuel processing system and thermal management system are located around and below the stack as shown in Fig. 2. The hot box is covered by thermal insulating plates with heating elements embedded on them for initial heating of the system.

2.1.3. SOFC stack

Fig. 3 shows the stack comprised of 46 planar SOFCs (LSCF/YSZ/Ni-YSZ). The cells with an effective electrode area of $10 \times 10 \text{ cm}^2$ per cell were stacked by using glass sealants and interconnects made of ferritic stainless steel. The details of the cell and stack fabrication were reported in [13].

2.1.4. Fuel processing system

The fuel processing system is designed for the dual-fuel operation to utilize both NG and LPG by employing a pre-reformer. The fuel is first introduced to a desulfurizer charged with an adsorbent (TOSPIX™ 94, Tokyo Gas) for removal of organic sulphur compounds in the fuel. Then the fuel is mixed with steam and delivered to a pre-reformer packed with commercial pre-reforming catalyst (C11-PR, Sud-Chemie) to produce CH_4 -rich stream. The pre-reformed gas is further heated and processed in a main reformer filled with commercial steam reforming catalyst (FCR-4, Sud-Chemie) to generate H_2 -rich gas for SOFC.

2.1.5. Pre-reformer

Fig. 4 shows a schematic of the pre-reformer. Inlet, middle and outlet temperatures of the pre-reformer are monitored by thermocouples. Total volume and weight of the catalyst are 500 mL and 700 g, respectively. Three baffles are placed inside of the reactor to maximize utilization of the catalyst. The pre-reformer is placed below the SOFC stack and covered with ceramic insulating material (Ceramblanket 1260, Thermal Ceramics) for adiabatic operation.

2.1.6. Main reformer

The main reformer is in the form of a counter-flow heat exchanger, receiving heat needed for the endothermic steam reforming reaction from cathode off-gas. Additional heat can be supplied through recycling of the anode off-gas. Total volume and weight of the catalyst used are 160 mL and 200 g, respectively. Internal reforming of the methane-rich gas in the stack is usually adopted in place of external reforming to prevent overheating of the SOFC stack. However, previous operating results of a 1 kW SOFC stack indicated that thermally self-sustaining operation could be achieved using hydrogen or NG as a fuel without employing internal reforming [13,14]. Therefore, in this study, stack was operated only in external reforming mode.

2.1.7. Fuel

LNG supplied from Korea Gas Company was evaporated and supplied to the system at a pressure of 0.2 atm. The



Fig. 1 – photograph of a 1 kW power package.

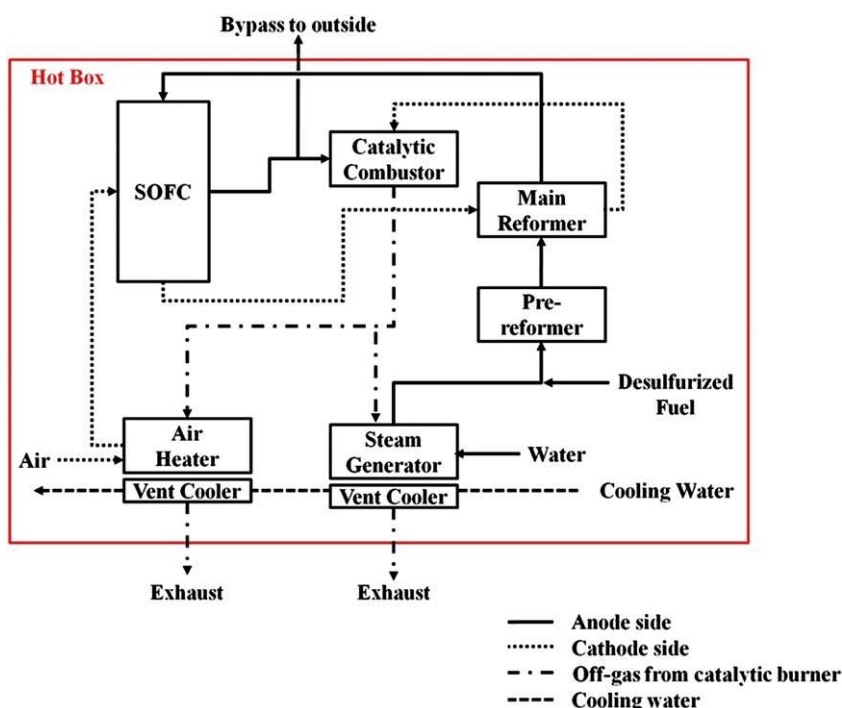


Fig. 2 – Schematic of the hot box in 1 kW SOFC package.

composition of NG measured by a gas chromatograph (GC) was 97.6% CH_4 , 1.7% C_2H_6 , 0.6% C_3H_8 , and 0.1% $\text{n-C}_4\text{H}_{10}$. LPG obtained from a local supplier consisted of 99.8% C_3H_8 and 0.2% $\text{n-C}_4\text{H}_{10}$. LPG was introduced to the system at a pressure of 3 atm. Fuel energy flows of NG and LPG based on the lower heating values (LHVs) of the fuels are 0.612 and 1.522 kW per

standard L/min (slm), respectively. All fuel energy flows expressed in this paper are calculated based on LHVs of fuels.

2.1.8. Thermal management system

The well-designed thermal management system is needed to sustain operating temperatures of the stack, reformer and other BOP in desired ranges simultaneously with increasing the system efficiency. Therefore, it is necessary to utilize heat generated from the stack and heat of combustion of the anode off-gas to supply heat for a steam generator, air pre-heater and fuel reformer where endothermic reforming reaction occurs. The thermal management system of the power package was developed as shown in Fig. 2. The off-gas from the cathode side of the stack is introduced to the main reformer to supply heat for endothermic methane steam reforming, and then mixed with anode off-gas in the catalytic



Fig. 3 – Photograph of a 1 kW SOFC stack.

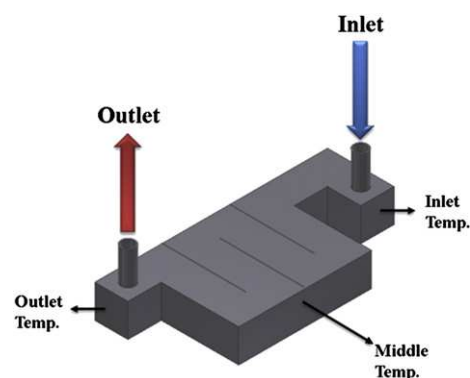


Fig. 4 – Schematic of a pre-reformer.

burner to generate hot gas which is divided into two streams for the air heating and the steam generation. The flow rate of the streams can be controlled by valves located in the cold box after the streams are cooled by vent coolers using cold water. The inlet temperature of the pre-reformer is regulated mainly by temperature of the generated steam. Counter-flow heat exchangers are employed for air heating and steam production. The residual heat of the hot gas is recovered by cooling water in two vent coolers and cogeneration is possible accordingly. If the system is overheated due to excessive heat generation of the catalytic burner, a part of the anode off-gas can be bypassed to outside. Part of the bypassed anode off-gas can be recycled by an additional blower to the main reformer to supply heat for steam reforming of methane.

2.2. Process simulation

Process simulation was conducted using the commercial software, Aspen Plus V7.1 to calculate heat duties of heat exchangers to meet the desired temperature of major components of the system such as the pre-reformer, the reformer and the stack when NG or LPG containing a fuel energy flow of 2.7 kW is supplied.

The pre-reformer should be operated within the reaction condition bound by thermal decomposition of C_{2+} hydrocarbons and carbon deposition [15]. In general, the pre-reformer is operated at 300–400 °C with a steam to carbon ratio higher than 2 to prevent carbon formation. The temperature of the stack should be sustained at 700–750 °C to maximize the power generation and keep stability of the sealant at the same time. Therefore, the desired temperatures of pre-reformer inlet, main reformer inlet and outlet, anode inlet and cathode inlet are fixed at 400, 650, 700, 700 and 700 °C, respectively (Fig. 5). The simulation condition refers to the actual experimental condition employed in the continuous operation. Fuel energy flow of 2.7 kW is supplied to the system to produce electricity of 1 kW. Water is mixed with the fuel at a steam-to-carbon ratio of 2.75. When NG is supplied to the fuel processor, the molar flow rates of NG and steam are 0.197 and 0.542 mol/min, respectively. When LPG is supplied, the molar flow rates of LPG and steam are 0.078 and 0.645 mol/min, respectively. Nitrogen supply (0.178 mol/min) to the fuel mixture as

a reference gas for GC analysis is considered in the calculation. Air is supplied to the cathode at the rate of 3.386 mol/min. Inlet temperatures of fuel, water and air are set to be 25 °C with exhaust gas temperature of 60 °C. The pre-reformer is assumed to be an adiabatic Gibbs reactor while the main reformer is assumed to be a Gibbs reactor.

According to the calculation, the total heat generation from the stack and the combustion of the anode off-gas is 4.29 kW for NG supply and 3.99 kW for LPG supply. Part of the heat (2.74 kW for both NG and LPG supply) among the total generated heat is utilized in heat exchangers and the heat duties of heat exchanges are represented in Fig. 6. The residual heat is defined as the total generated heat minus the used heat, and it can be recovered by cooling water, lost through the surface of the hot box, and lost in the form of bypassed anode off-gas. It is concluded that, if capability of heat exchangers can cover the calculated heat duty and heat balance is well established in a steady state (residual heat = 0), the auto-thermal operation of 1 kW system using NG and LPG is possible. Consequently, the heat exchangers with proper heat exchanging capacity were selected according to the calculated heat duties.

3. System operation

3.1. Initial operation

The system was first heated up to 750 °C according to the designated procedure to stabilize the sealants in the stack by the electric furnace. Then, the system was operated in an auto-thermal condition using internal heat generation without external heat from the heating elements. The hot box temperature was sustained near 750 °C by controlling the generation rate of the hot gas in the off-gas burner. The initial performance of the system was first obtained under the flow of diluted hydrogen, then fuel was switched to NG followed by LPG while keeping the fuel energy flow ca. 3.0 kW. Composition of the fuel supplied to the system is summarized in Table 1. Initial operation under NG or LPG flow was carried out with constant supply of water at a rate of 16.4 g/min. The flow rate of nitrogen added to either NG or LPG as a reference for GC analysis was 4.0 slm. The air flow rate of the cathode was kept constant at 75.9 slm.

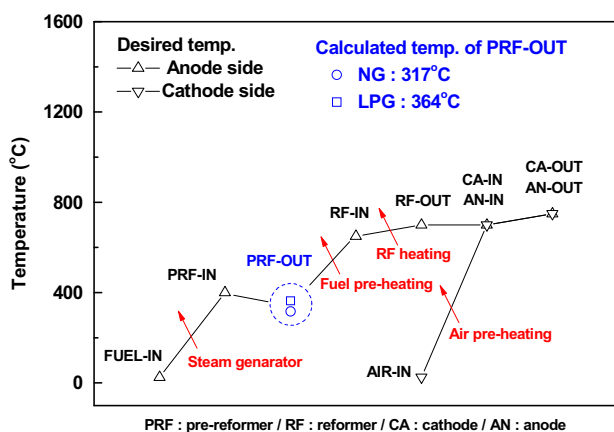


Fig. 5 – Desired temperature profile of the system.

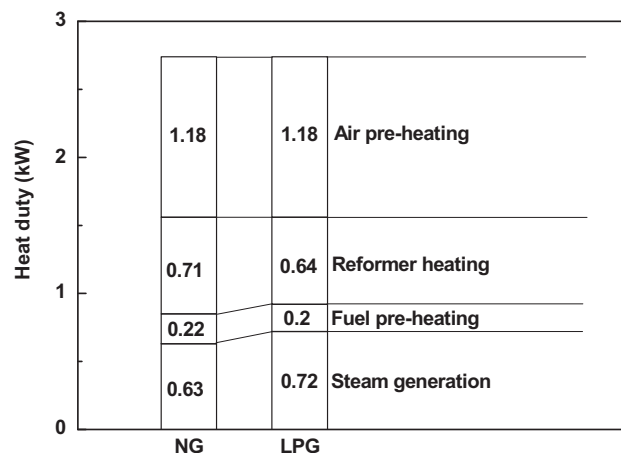


Fig. 6 – Calculated heat duties of heat exchangers.

Table 1 – Fuels supplied to the SOFC system.

Operating condition		Flow rate					S/C	Fuel energy flow (kW)
		H ₂ (slm)	NG (slm)	LPG (slm)	H ₂ O (g/min)	N ₂ (slm)		
Initial operation	H ₂	16.9				8.4		3.04
	NG		4.84		16.4	4.0	4.09	2.96
	LPG			2.21	16.4	4.0	3.08	3.36
Continuous operation	NG		4.42		10.1	4.0	2.75	2.70
	LPG			1.82	12.0	4.0	2.75	2.77

The effect of NG/LPG composition ratios in the fuel on the stack performance and gas composition of the pre-reformed and reformed gases was also investigated to confirm the capability of the system for dual-operation. Performance of the stack was analyzed by a DC electric load (American reliance, PLA 1.5k-120-200). Gas composition was analyzed by a gas chromatograph (Agilent 7890). The power consumed by BOP such as an air blower, a water pump and an NG compressor was measured to calculate electrical efficiency of stack and system.

3.2. Continuous operation

The operation of the system was conducted for 350 h by NG and LPG alternatively using the fuel composition as shown in Table 1. Temperatures of the stack, the pre-reformer and main reformer were measured by thermocouples. Condensed water from exhaust gas was recycled to the steam generator after refined in a filter filled with ion exchange resin. During the continuous operation, fuel switching from NG to LPG was carried out.

4. Results and discussion

4.1. Initial performance

Initial performance of the stack is presented in Fig. 7. The stack showed power output of 1 kW at current of 30 A irrespective of the fuel type, although the performance was higher under the operation by hydrogen at lower current.

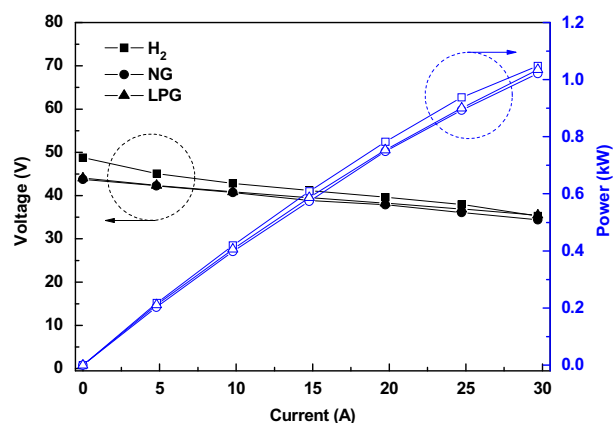


Fig. 7 – Initial performance of the stack operating with H₂, NG and LPG.

Stack performance using NG is quite similar with that using LPG. GC analysis of the anode inlet gas showed H₂ level of 70–75% (N₂-free, dry basis) and CO₂ levels of 14–15% with about 9–11% CO and 3–4% CH₄ under the operation with NG and LPG. The fuel utilization at 30 A calculated based on gas composition of anode inlet gas was 0.60 and 0.66 for NG and LPG operation, respectively. The air flow rate was 75.9 slm which corresponds to oxygen utilization of 0.3 at 30 A. The only BOP working under operation with H₂ was an air blower which consumed power of 150 W at air flow rate of 75.9 slm. However, 20 W was consumed by the water pump during NG or LPG operation, and additional 30 W was consumed by an NG compressor during NG operation. Therefore, the powers consumed by BOP systems were 150, 200 and 170 W for H₂, NG and LPG operations, respectively. Considering the parasitic loss caused by BOP, DC electrical efficiencies of the system under hydrogen, NG and LPG operation became 29.5, 27.7 and 25.9%, respectively at 30 A based on LHV of fuels.

The operating current was limited to 30 A in this study to control the hot box temperature within the desired temperature range (<800 °C). The stack temperature during the test was generally higher than expected because of the gas leakage which led to unintended combustion of fuel and air in the stack. It was found that the stack temperature became higher than 800 °C due to severe heat generation if the current increased higher than 30 A. If the operating current can be increased higher than 30 A by solving the leakage problem and introducing internal reforming to keep the stack temperature within the desired range, then the electrical efficiency of the system would be enhanced over 30%. The system was originally designed for grid connected operation, but additional

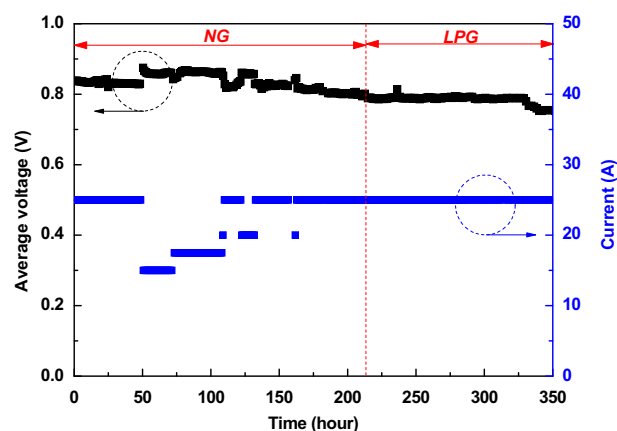


Fig. 8 – Evolution of the average cell voltage during the continuous operation.

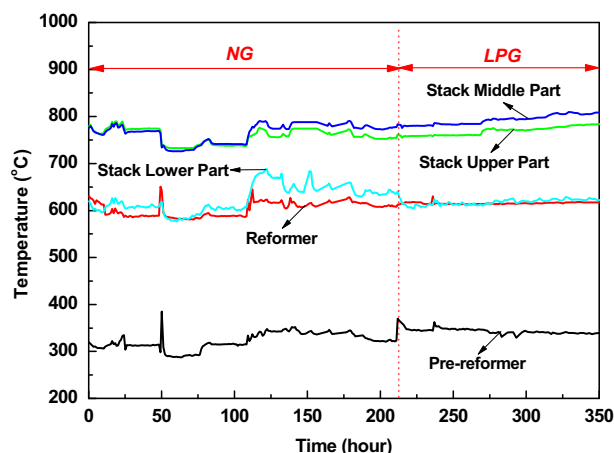


Fig. 9 – Evolution of the temperature of system components during the continuous operation.

loss induced by the power conditioning system reduced the efficiency further down to 24–25% since the power conversion efficiency was 91%.

4.2. Continuous operation with NG and LPG

The performance of the power package under NG and LPG was investigated for 350 h until the water pump was stopped due to sludge formation. Fig. 8 shows evolution of the average cell voltage during continuous operation. NG was first supplied to the stack for 213 h and then switched to LPG for 137 h. The operating current was mainly 25 A except for some period when NG was supplied. The stack showed relatively stable performance during the operation with NG or LPG although there was degradation in performance. The degradation was caused by some cells placed in the upper and middle parts of the stack due to gas leakage in the cells. The gas leakage was confirmed from evidence such as the constant increase of the stack temperature (Fig. 9) and excessively oxidized interconnects due to combustion of fuel and air which were found when the stack was taken apart after the experiment.

As shown in Fig. 9, temperatures of pre-reformer and main reformer were changed according to the operating current, but were stable after the operating current was kept constant

at 25 A. During the NG and LPG operation, compositions of pre-reformed gas and reformed gas were measured by the GC. It was found that all C_{2+} hydrocarbons included in NG or LPG were converted into methane-rich gas in the pre-reformer, and methane is almost converted to hydrogen-rich gas in the main reformer. Methane contents in the pre-reformed gas and the reformed gas were 54–58% and 3–4% (N_2 -free, dry basis), respectively.

4.3. Thermal management of the system

The temperature of the hot box was controlled primarily by hot gas generation rate in the off-gas burner. The temperature was also influenced by heat generation from the stack which, however, seemed to be nearly constant during the continuous operation of the system. The hot gas generation rate could be controlled by regulating the fuel utilization. But, in this study, the rate of fuel supply was kept constant and we regulated the amount of the bypassed anode off-gas to control the hot gas generation rate. Part of the bypassed anode off-gas was supplied to the main reformer but some amount of the bypassed gas was oxidized in the reactor placed out of the system. The hot gas generated in the off-gas burner was divided into two streams for steam generation and air heating at a ratio of 1:2 for NG operation and 1:1.16 for LPG operation.

Measured temperatures of the major components during NG and LPG operations are compared with the desired ones in Table 2. It was found that the operating temperature of the pre-reformer was relatively well controlled within the desired temperature ranges by the steam generator working properly in the designed range. The temperatures of the reformer, anode gas and cathode gas were lower than the desired temperatures to keep the hot box temperature lower than 800 °C. As was mentioned previously, the stack temperature increased during the operation due to gas leakage in the cell placed at upper part of the stack. In addition, the stack was cooled mainly by gas convection which caused more than 100 °C difference in cathode gas temperature between gas inlet and outlet as shown in Table 2. Consequently, the temperature gradient inside of the stack may be great which leads to increase the internal stress and affect the stack stability adversely. This phenomenon becomes serious when the power output of the stack becomes larger. Therefore, advanced thermal management strategy of the stack such as

Table 2 – Desired and measured temperatures of the system components during continuous operation.

		NG operation		LPG operation	
		Desired temp. (°C)	Measured temp. (°C)	Desired temp. (°C)	Measured temp. (°C)
Anode side	Pre-reformer inlet	400	360	400	343
	Pre-reformer outlet	319	340	362	350
	Reformer inlet	650	592	650	582
	Reformer outlet	700	646	700	612
	Anode inlet	700	615	700	603
	Anode outlet	750	641	750	655
Cathode side	Cathode inlet	700	558	700	558
	Cathode outlet	750	696	750	696

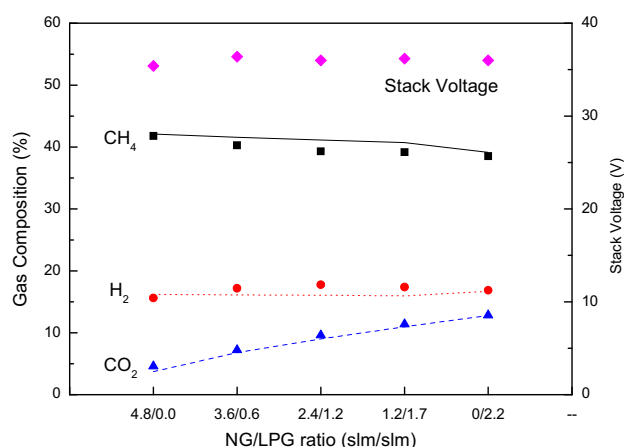


Fig. 10 – Stack voltage and gas concentration at the pre-reformer outlet according to various NG/LPG ratios.

direct utilization of methane-rich gas from the pre-reformer to induce endothermic internal reforming should be introduced to resolve the overheating and thermal unbalance of the stack.

4.4. Effect of on-load fuel switching

Switching of the fuel from NG to LPG at constant current of 25 A did not cause significant change in stack performance or gas composition. As shown in Fig. 10, under a number of NG/LPG ratios during the fuel switching, the stack and the pre-reformer showed stable performance. Assuming that the pre-reformer was operated in an adiabatic condition, equilibrium gas composition was calculated (lines in Fig. 10) and compared with the measured one. As shown in Fig. 10, equilibrium gas composition is well matched to the experimental data, indicating that operation of the pre-reformer can be regarded as adiabatic. Fig. 11 shows evolution of pre-reformer temperature before and after the on-load gas switching from NG to LPG at operating time of 241 h. The pre-reformer temperature was first increased and then decreased during the fuel switching, and it reached to the steady state after 12 h. The differences

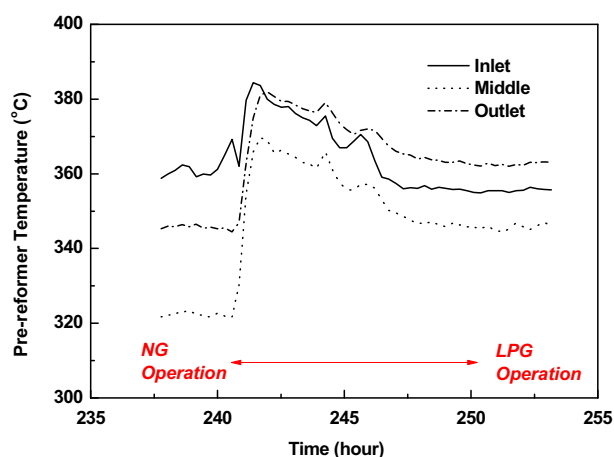


Fig. 11 – Evolution of pre-reformer temperature before and after gas switching from NG to LPG.

between the inlet and middle temperatures were 38 °C for NG and 12 °C for LPG. According to the calculation of adiabatic pre-reforming temperature keeping the same enthalpy and pressure of the gas mixture, the differences should be 61 °C for NG and 19 °C for LPG. There is a possibility that pre-reforming reaction is already finished before the gas reached to the middle part of the pre-reformer. Actually, less amount of catalyst may be needed for both NG and LPG pre-reforming and optimization on catalyst loading should be done in the future.

5. Conclusion

A compact 1 kW SOFC system powered by both NG and LPG was developed by the introduction of the pre-reformer. The power output of the stack was 1 kW at 30 A, regardless of the fuel type. DC electrical efficiencies of the system under hydrogen, NG and LPG operation became 29.5, 27.7 and 25.9%, respectively at 30 A based on LHVs of fuels. It was found that the pre-reformer converts all C_{2+} hydrocarbons into methane-rich gas which was processed in the main reformer to generate hydrogen-rich gas enough to generate power of 1 kW. The evolution of the system performance using NG and LPG alternatively was also investigated for 350 h under thermally self-sustaining condition and on-load fuel switching from NG to LPG was carried out during the operation. The performance of the system was not significantly affected by NG/LPG composition ratios. There was the degradation of the stack performance due to the gas leakage in the stack. To increase the efficiency and to ensure the long-term stability of the system, the stack needs further improvement in reliability. In addition, advanced thermal management strategy such as internal reforming should be introduced.

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