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Performance of a natural gas fuel processor for residential PEFC system using a novel CO preferential oxidation catalyst

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Abstract

Natural gas fuel processors have been developed for 500 W and 1 kW class residential polymer electrolyte fuel cell (PEFC) systems. These fuel processors contain all the elements—desulfurizers, steam reformers, CO shift converters, CO preferential oxidation (PROX) reactors, steam generators, burners and heat exchangers—in one package. For the PROX reactor, a single-stage PROX process using a novel PROX catalyst was adopted. In the 1 kW class fuel processor, thermal efficiency of 83% at HHV was achieved at nominal output assuming a H_2 utilization rate in the cell stack of 76%. CO concentration below 1 ppm in the product gas was achieved even under the condition of $[O_2]/[CO] = 1.5$ at the PROX reactor. The long-term durability of the fuel processor was demonstrated with almost no deterioration in thermal efficiency and CO concentration for 10,000 h, 1000 times start and stop cycles, 25,000 cycles of load change.

Keywords: Fuel processor; Natural gas; CO preferential oxidation; Polymer electrolyte fuel cell; Residential use

1. Introduction

Polymer electrolyte fuel cell (PEFC) systems have been increasingly developed for transportation and stationary power sources applications, especially for vehicle and residential use. From a feasibility study for residential energy demand in Japan, 0.5–1 kW class PEFC cogeneration systems have been developed for residential use. An AC power generation efficiency of not less than 31.5% (HHV) is required at nominal output for these systems in order to enable significant energy saving. Moreover, a durability of over 90,000 h is required of these systems to operate for 10 years, the minimum life of household appliances to be economical.

The hydrogen-rich reformate from hydrocarbons like natural gas is a convenient fuel for stationary PEFC systems, especially for residential use, in terms of infrastructure. Natural gas fuel processors adopting an autothermal reforming process or a steam reforming process have been mainly developed for those PEFC systems [1–5]. In autothermal reforming, the endothermic reaction heat of steam reforming and the exothermic reaction heat of partial oxidation are balanced. Autothermal reforming has the advantages of quick start-up and load change compared to steam reforming. As for efficiency, it is supposed that the power generation efficiency of a residential PEFC system adopting autothermal reforming is lower than that of a steam reforming system in the case of a cell stack operated at a H_2 utilization rate below 80%. That is because the heat of an anode-off gas from the cell stack cannot be effectively recovered for the reforming process in autothermal reforming but can in steam reforming. It seems difficult to operate the reformate-fueled PEFC stack at a high H_2 utilization rate for the long term, in contrast to the H_2-fueled PEFC stack. As for life, the long-term durability of the autothermal reforming process has not been confirmed. In contrast, more than 40,000 h durability of the natural gas steam reforming process, comprising a desulfurizer, a steam reformer and a CO shift converter, has already been confirmed in the commercial phosphoric acid fuel cell (PAFC) systems [6]. Therefore, it can be thought that a natural gas fuel processor adopting the steam reforming process is more suitable for the residential PEFC system which requires both high power generation efficiency and long durability.

On the other hand, the reformate after the CO shift converter contains about 0.5 vol.% CO. This CO concentration is acceptable for the anode catalyst of the PAFC operated at ca. 473 K. However, the anode catalyst of the PEFC operated at ca. 353 K is poisoned by only 10 ppm of CO [7–9]. Although CO-tolerant anode catalysts containing Ru in ad-

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dition to Pt have been developed for the PEFC [10–12], the acceptable CO concentration of the CO tolerant anode catalyst appears about to be 10 ppm for long-term durability. Therefore, for a PEFC system using natural gas, a CO removal process is required in addition to the reforming process established for the PAFC.

Generally, a CO preferential oxidation (PROX) process using Pt-based catalysts has been used for the CO removal process because of its high activity. It was reported that CO was reduced to ca. 100 ppm on a Pt/Al_2O_3 catalyst with the addition of excess air corresponding to $[O_2]/[CO] = 2$ [7]. It has also been reported that additive air corresponding to $[O_2]/[CO] \geq 3$ was needed to remove CO completely on the conventional Pt/Al_2O_3 catalyst [13]. The excess O_2 consumes H_2 in the reformed gas by combustion, which causes a decrease in the power generation efficiency of the PEFC system. Since power generation efficiency is the most important requirement for the residential PEFC system, a low $[O_2]/[CO]$ operation is necessary for the CO PROX process. Recently, multi-stage PROX reactors have been mainly adopted for the CO removal process [3,14–18]. In the multi-stage PROX, the total amount of additive air can be reduced by optimizing the additive air at each stage. It has been reported that CO was reduced to less than 10 ppm by a two-stage PROX reactor at a total additive air corresponding to $[O_2]/[CO] = 1.5$ [3]. However, complex hardware is required to control temperatures and oxidant injections along the multi-stage catalytic layer.

On the other hand, although several kinds of the Pt-based catalysts have been studied to improve the CO PROX activity [13,19–26], their performance in an actual reactor under practical conditions has not been confirmed. On the other hand, there were several reports that Ru/Al_2O_3 catalysts showed higher CO removal performance than conventional Pt/Al_2O_3 catalysts [10,12,27]. Generally, a Ru catalyst also possesses considerable methanation activity [28]. Because temperature control is quite difficult in actual reformate due to self-catalytic endothermic CO_2 methanation, a Ru catalyst is seldom used solely but sometimes used in the second-stage reaction.

In a previous work, we reported that the novel Ru catalyst, which is further reduced by H_2/N_2 after aqueous reduction, has extremely high practical CO removal performance even in a single-stage PROX reactor for residential PEFC applications. On the catalyst, CO can be reduced from 0.5 vol.% to below 10 ppm between 358 K and 443 K and below 1 ppm between 373 K and 393 K at $[O_2]/[CO] = 1.5$ [29]. Also, we reported on natural gas fuel processors adopting the steam reforming process and the single-stage CO PROX process for residential PEFC cogeneration systems [30]. In this paper, performances of the natural gas fuel processors for 500 W class and 1 kW class residential PEFC systems including the partial load operations and longer durability test results are reported. Moreover, the performance after 90,000 h of operation is estimated.

2. Experimental

The usual Japanese city gas (13A) was used as a natural gas fuel for testing the fuel processors. The composition of the city gas was mainly 88.0 vol.% CH_4, 6.0 vol.% C_2H_6, 3.0 vol.% C_3H_8 and 3.0 vol.% C_4H_{10}. The city gas contained 5 ppm of sulfur compounds as odorants. For the anode-off gas from the cell stack, a simulated anode-off gas provided by a cylinder gas was used. The typical composition of the simulated anode off gas was 49 vol.% H_2, 3 vol.% CH_4, 42 vol.% CO_2 and 6 vol.% N_2. This composition corresponds to an anode-off gas when 75% of H_2 in the reformate from the fuel processor operated at the design conditions ($S/C = 2.5$ and $[O_2]/[CO] = 1.5$) is utilized at the cell stack.

The tests of fuel processors with load fluctuations or with frequent starts and stops were carried out by automatic control. The flow rates of the natural gas, simulated anode-off gas, air and water were controlled by mass flow controllers. The temperatures of the fuel processors were controlled by temperature controllers having PID controls with thermocouples. The flow rates of the natural gas, simulated anode-off gas, and air were measured by dry gas meters. The flow rate of the product gas was measured by a wet gas meter. The flow rate of water was measured by change in weight. The CO and CH_4 concentrations in the product gas and the CO, CH_4 and O_2 concentrations in the exhaust gas were always measured by infrared gas analyzers. The gas composition of the product gas at a stationary state was analyzed by a gas chromatograph equipped with TCD and FID. The results are shown in dry base. The detection limit of both CO and CH_4 were 0.5 ppm, while the detection limit of O_2 was 20 ppm. The concentration below the detection limit was plotted as 0 ppm in the figures.

The thermal efficiency of the fuel processor was defined as (heating value of H_2 consumed at the cell stack)/(heating value of total natural gas supplied to the fuel processor). The total natural gas means the sum of (natural gas supplied to the reforming process) and (natural gas supplied to the burner of the fuel processor). In actual measurements for the thermal efficiency of the fuel processor without a cell stack, the numerator was calculated as the difference between (heating value of the product gas of the fuel processor) and (heating value of the simulated anode-off gas supplied to the burner of the fuel processor). The numerator means the product of (heating value of H_2 provided to the cell stack from the fuel processor) and (H_2 utilization rate of the cell stack). Therefore, the H_2 utilization rate can be calculated as ((heating value of the product gas of the fuel processor) − (heating value of the simulated anode-off gas supplied to the burner of the fuel processor))/(heating value of the H_2 provided to the cell stack from the fuel processor). All thermal efficiencies are shown as the higher heating value. On the other hand, the CH_4 conversion of the fuel processor was defined as ($CO + CO_2$ in the product gas)/($CH_4 + CO + CO_2$ in the product gas).

3. Process flow in the natural gas fuel processors

Natural gas fuel processors were developed for 500 W class and 1 kW class residential PEFC systems. The appearance of the 1 kW class natural gas fuel processor without an outer thermal insulator is shown in Fig. 1. The sizes of the fuel processors are 280 (W) × 440 (L) × 395 (H) for the 500 W class and 280 (W) × 440 (L) × 395 (H) for the 1 kW class, including thermal insulation. Those fuel processors contain desulfurizers, steam reformers, CO shift converters, CO PROX reactors, steam generators, burners and heat exchangers in one package. The natural gas is provided to the desulfurizer. Sulfur compounds contained in the raw fuel are removed in the desulfurizer using a high performance desulfurization agent [31,32]. Meanwhile, water is provided to the steam generator and steam is generated in it. The desulfurized natural gas and the steam are mixed and the mixed gas is provided to the steam reformer. The mixed gas is converted to H_2, CO and CO_2 by the steam reforming reaction in the steam reformer. The designed steam/carbon (S/C) ratio and CH_4 conversion at the steam reformer are 2.5 and 92%, respectively. The outlet gas of the steam reformer is provided to the CO shift converter using a heat resistant Cu–Zn catalyst [31,32]. CO concentration in the reformed gas is reduced to ca. 0.5 vol.% by the water–gas shift reaction in the CO shift converter. Air for CO PROX is added to the outlet gas of the CO shift converter, and the mixed gas is provided to the PROX reactor. In the PROX reactor, CO is removed to below 10 ppm by single-stage CO PROX on the novel Ru catalyst [29]. The designed $[O_2]/[CO]$ ratio at the PROX reactor is 1.5. These fuel processors use desulfurization, steam reforming, and CO shift catalysts, which have shown more than 40,000 h of durability in commercial PAFC systems [6]. They are designed for 90,000 h of operation without exchanging any catalyst. For burner fuel, both anode off gas and natural gas are provided to the burner in order to maintain the heat balance in the fuel processor.

4. Results and discussion

4.1. Initial performance

Table 1 shows the initial performances of the 500 W class and the 1 kW class fuel processors at nominal load operations. These fuel processors were operated under the conditions of $S/C = 2.5$ and $[O_2]/[CO] = 1.5$ assuming that the CO concentration at the outlet of CO shift converter is 0.5 vol.%. The thermal efficiency of the 1 kW class fuel processors was 83% at HHV (78% at LHV) assuming a fuel utilization rate in the cell stack of 76%. The thermal efficiency of the 500 W class fuel processors was 80% at HHV assuming a fuel utilization rate in the cell stack of 74%. Both the low S/C and the low $[O_2]/[CO]$ ratio operations contributed to achieving high thermal efficiencies in these fuel processors. These values satisfy our target thermal efficiencies of fuel processors for residential PEFC systems [30]. It has been reported that the thermal efficiency of a natural gas fuel processor without a CO removal reactor adopting autothermal reforming was calculated to be 78–84% when it was defined as the lower heating value of H_2 in the product gas divided by the lower heating value of the fuel fed to the fuel processor [4]. It has also been reported that the thermal efficiency of a natural gas fuel processor adopting autothermal reforming was 78.4% at HHV [1]. In these thermal efficiencies, the factor of the fuel utilization rate was

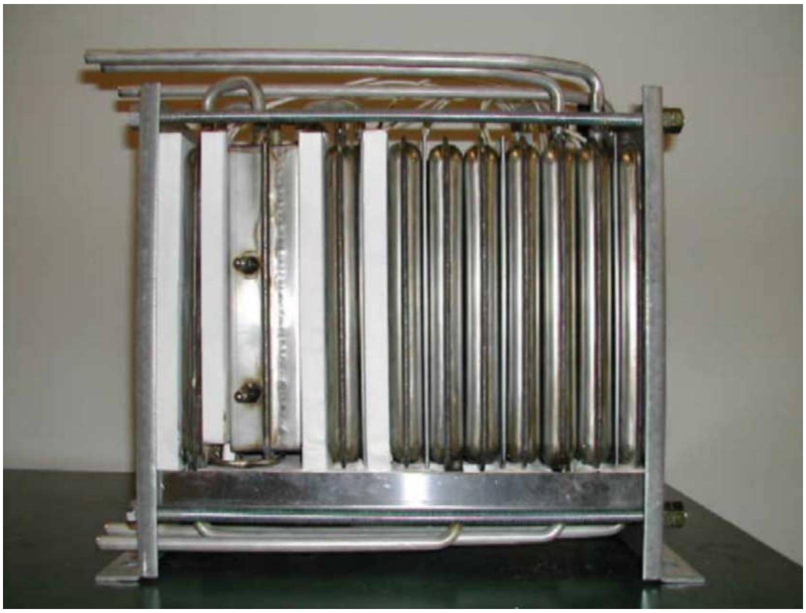

Fig. 1. Appearance of the natural gas fuel processor for 1 kW class residential PEFC system (without outer thermal insulation).

Table 1
Initial performances of fuel processors at nominal output operation

	Class	
	1 kW	500 W
Flow rate of natural gas for process (l/min (normal))	4.2[a]	2.1[b]
Flow rate of water for process (g/min)	9.8	5.0
S/C ratio at steam reformer	2.5	2.5
Flow rate of air for CO PROX (l/min (normal))	0.80	0.40
$[O_2]/(CO)$ ratio at CO PROX	1.5	1.5
Flow rate of natural gas for burner (l/min (normal))	0.18[a]	0.14[b]
Flow rate of simulated anode off gas for burner (l/min (normal))	9.3[c]	5.1[d]
Flow rate of product gas (l/min (normal))	22	12
Composition of product gas		
H_2 (vol.%)	75.4	75.4
N_2 (vol.%)	2.8	2.8
CH_4 (vol.%)	1.8	1.8
CO (vol. ppm)	0.5	<0.5
CO_2 (vol.%)	20.0	20.2
CH_4 conversion (%)	92	93
Calculated H_2 utilization rate (%)	76	74
Thermal efficiency (%, HHV)	83	80

[a] Composition of natural gas: 88.9 vol.% CH_4, 6.8 vol.% C_2H_6, 3.1 vol.% C_3H_8, and 1.2 vol.% C_4H_{10}.

[b] Composition of natural gas: 88.0 vol.% CH_4, 6.0 vol.% C_2H_6, 3.0 vol.% C_3H_8, and 3.0 vol.% C_4H_{10}.

[c] Composition of simulated anode off gas: 49.5 vol.% H_2, 2.7 vol.% CH_4, 41.9 vol.% CO_2, and 5.9 vol.% N_2.

[d] Composition of simulated anode off gas: 48.5 vol.% H_2, 2.8 vol.% CH_4, 42.7 vol.% CO_2, and 6.0 vol.% N_2.

not included. Therefore, the thermal efficiencies of the fuel processors adopting steam reforming seem not to be low compared to those of fuel processors adopting autothermal reforming in the PEFC system operated at a fuel utilization rate of ca. 75%. As for outlet CO level, the CO concentrations in the product gases were less than 1 ppm in both fuel processors adopting the single-stage PROX reactor in spite of the low O_2/CO operating condition. Such high CO removal performance was achieved by using the novel PROX catalyst.

Table 2 shows the initial performances of the 1 kW class fuel processor at partial load operations. At 75, 50 and 30% load operation conditions, the flow rates of natural gas for the process were set to 3.2 l/min (normal), 2.1 l/min (normal) and 1.3 l/min (normal), respectively. At 75% load operation, the thermal efficiency was almost the same as that at nominal (100%) load operation shown in Table 1. The thermal efficiency at half load operation was kept at 80%. The decrease in thermal efficiency from 100% load operation to 30% load operation was within 10%. As for CO removal performance, CO in the product gas was less than 1 ppm at all partial load operation conditions. Thus, it was confirmed that CO was sufficiently removed under widely varying operating conditions by the single-stage PROX reactor using the PROX catalyst.

Table 2
Initial performances of 1 kW class fuel processor at partial load operations

	Load (%)		
	75	50	30
Flow rate of natural gas for process (l/min (normal))	3.2	2.1	1.3
S/C ratio at steam reformer	2.5	2.5	2.5
Outlet CO concentration (ppm)	<0.5	<0.5	<0.5
CH_4 conversion (%)	92	92	93
Thermal efficiency (%, HHV)	83	80	77
Calculated H_2 utilization rate (%)	76	75	75

4.2. *Long-term durability*

A long-term durability of 10 years is required for residential PEFC systems. Therefore, it is very important to establish the long-term durability of fuel processors. The durability test of a 1 kW class fuel processor has been carried out at nominal output operation. Fig. 2 shows the result. The product gas composition and the thermal efficiency have been stable for more than 10,000 h. The CO concentration in the product gas after 10,000 h of operation was below 10 ppm. Thus, durability of the fuel processor for more than 10,000 h has been demonstrated. The durability of the PROX catalyst in an actual reactor has been also confirmed for more than 10,000 h.

Also, the performance of a fuel processor using deactivated catalysts corresponding to 90,000 h of operation, which were prepared by accelerated degradation methods, was tested. The accelerated degradation methods for steam reforming and CO shift catalysts were established on the basis of abundant experiences in commercial PAFC systems operated for more than 40,000 h. The accelerated degradation method for the PROX catalyst was established on the base of a large number of test results including long-term durability tests for more than 16,000 h in a micro reactor and for more than 8,000 h in an actual reactor [33]. Table 3

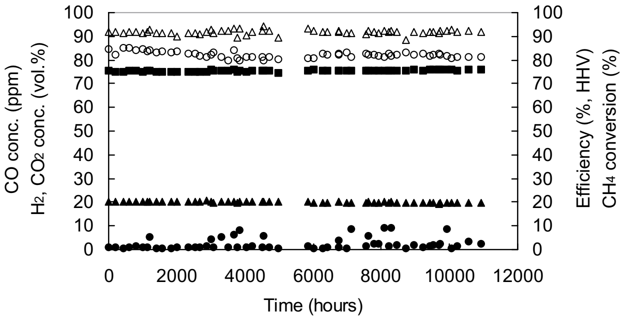

Fig. 2. Long-term durability of the fuel processor at nominal output operation. Fuel processor: 1 kW class; operation conditions: S/C = 2.5, $[O_2]/[CO] = 1.5$; (○) thermal efficiency; (△) CH_4 conversion; (●) outlet CO concentration; (▲) outlet CO_2 concentration; and (■) outlet H_2 concentration.

Table 3
Performances of 1 kW class fuel processor containing accelerated degradation catalysts

	Load (%)			
	100	75	50	30
Flow rate of natural gas for process (l/min (normal))	4.2	3.2	2.1	1.3
S/C ratio at steam reformer	2.5	2.5	2.5	2.5
Outlet CO concentration (ppm)	2.9	1.8	1.8	3.5
CH_4 conversion (%)	92	92	92	92
Thermal efficiency (%, HHV)	78	77	78	75
Calculated H_2 utilization rate (%)	75	74	74	75

shows the performance of the 1 kW class fuel processor containing these deactivated catalysts corresponding to 90,000 h of operation. There was not so large a decrease in the thermal efficiency of the fuel processor containing accelerated deactivated catalysts compared to initial efficiency shown in Tables 1 and 2. As for CO removal, the CO concentrations in the product gases were below 10 ppm in all partial load operations. It can be expected that there is no need to exchange any catalysts in the fuel processor during 90,000 h of operation.

4.3. *Influence of load fluctuation*

Continuous operation with load fluctuations is presumed to be the operation pattern for an early commercial PEFC system for residential use. Accordingly, the durability of the fuel processor against load fluctuation was tested. A 1 kW class fuel processor was used for the test. A nominal output operation for 15 min and a half load operation for 15 min or 7 min were repeated alternately for more than 6800 h. The procedure for decreasing load from 100 to 50% was carried out instantaneously, while the time for increasing load from 50 to 100% was set to 8 min. While the loads fluctuated, the flow rates of natural gas, water and air for PROX were controlled to keep both the S/C and the PROX air/Reformate ratios constant. Fig. 3 shows the change in the product gas composition at the nominal output operation. More than 25,000 load changes were carried out over 6,800 h of operation. After 25,000 load changes, the product gas composition was stable and the outlet CO concentration was below 10 ppm. Fig. 4 shows the changes in outlet CO concentrations of the CO shift converter and the CO PROX reactor at the load changes. Although the outlet CO concentration of the shift converter fluctuated according to the load change, the outlet CO concentration of the PROX reactor was consistently less than 10 ppm during the load change. In this test, it happened that the outlet CO concentration of the shift converter at nominal output operation reached to ca. 0.75 vol.%, exceeding the design value of 0.5 vol.%. However, the outlet CO concentration of the PROX reactor was below 10 ppm because CO can be reduced to below 10 ppm even at $[O_2]/[CO] = 1.0$ on the novel PROX catalyst [29]. Thus, the stable performance of the fuel processor and the wide operating range of the PROX catalyst were confirmed for continuous operation with control over load fluctuations.

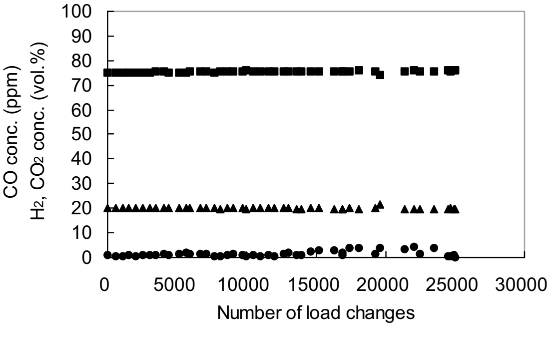

Fig. 3. Durability of the fuel processor for load changes. Fuel processor: 1 kW class; operation condition: S/C = 2.5; (●) outlet CO concentration; (▲) outlet CO_2 concentration; and (■) outlet H_2 concentration.

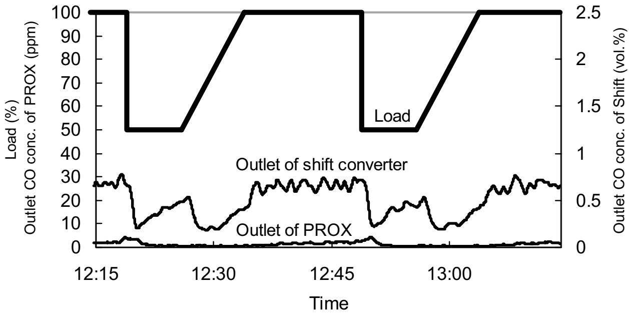

Fig. 4. Changes of outlet CO concentrations of the CO shift converter and the CO PROX reactor during load change. Fuel processor: 1 kW class; and operation condition: S/C = 2.5.

4.4. *The influence of starts and stops*

Although the continuous operation with control over load fluctuations is presumed for an early commercial PEFC system for residential use, several hundred starts and stops are unavoidable over 10 years of operation. Thus, a start and stop cycle test for the fuel processor was carried out. A 500 W class fuel processor was used for the test. The fuel processor was operated at the nominal load for 2 h every day. It took ca. 1 h for the start-up. Such a long time for start-up is a disadvantage of a fuel processor adopting the steam reforming process. Fig. 5 shows the change in the product gas composition at nominal output operation. In this test, from the beginning to 90 start–stop cycles, a N_2 was used to purge at both the start and stop processes. From the 91st to the 230th start–stop cycle, N_2 was used to purge only at the stop. After 231st start–stop, N_2 was not used as a purge at either the start or the stop process. That is because it is not desirable for a residential PEFC system to have a N_2 cylinder for these procedures. Even after 350 start–stop cycles, the product gas composition was stable and the outlet CO

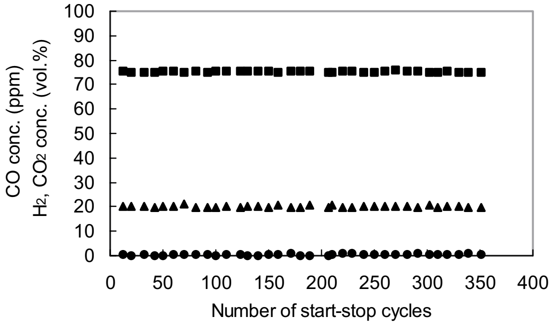

Fig. 5. Durability of the fuel processor for start–stop cycles. Fuel processor: 500 W class; operation conditions: S/C = 2.5, $[O_2]/[CO] = 1.5$; (●) outlet CO concentration; (▲) outlet CO_2 concentration; and (■) outlet H_2 concentration.

concentration was less than 10 ppm. Moreover, there was little difference in the outlet gas compositions regardless of whether N_2 was used to purge at the start and stop processes or not. The start and stop cycle test for a 500 W class fuel processor has also been carried out without any N_2 purge at all in the start and stop processes. The fuel processor was started up every 4 h and operated for 1 h at the nominal load. Fig. 6 shows the change in the product gas composition at the nominal output operation. After 1000 start–stop cycles, the product gas composition was stable and the outlet CO concentration was below 10 ppm. Thus, it was confirmed that not only the fuel processor but also all catalysts used in the fuel processor were durable for more than 1000 start and stop cycles without a N_2 purge.

4.5. Results of practical use

Our natural gas fuel processors have already been adopted in several manufacturers' residential PEFC cogeneration systems. In actual PEFC systems, it has been demonstrated that the fuel processors can show high thermal efficiencies and extremely low CO concentrations, similar to stand-alone performances without cell stacks. The total accumulated operation time of 51 fuel processors, installed in actual PEFC systems or under stand-alone durability tests, has reached more than 94,000 h. In these fuel processors, no troubles in the PROX catalyst have ever occurred. Thus, the stable performance and reliability of the novel PROX catalyst were confirmed even in actual residential PEFC systems.

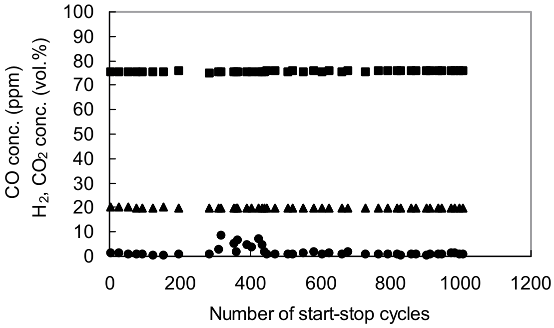

Fig. 6. Durability of the fuel processor for start–stop cycles without N_2 purge. Fuel processor: 500 W class; operation conditions: S/C = 2.5, $[O_2]/[CO] = 1.5$; (●) outlet CO concentration; (▲) outlet CO_2 concentration; and (■) outlet H_2 concentration.

5. Conclusion

The performance of natural gas fuel processors containing all elements—desulfurizer, steam reformer, CO shift converter, single-stage CO PROX reactor containing novel PROX catalyst—were studied. In initial performance, a thermal efficiency of 83% at HHV was achieved in the 1 kW class fuel processor at nominal load on assuming a H_2 utilization rate in the cell stack of 76%, while thermal efficiency of 80% at HHV was achieved in the 500 W class fuel processor at nominal load on assuming a H_2 utilization rate in the cell stack of 74%. Those high thermal efficiencies can be achieved at operating conditions of low S/C ratio of 2.5 and low $[O_2]/[CO]$ ratio of 1.5 assuming 0.5 vol.% of CO concentration at the outlet of CO shift converter. In spite of the low $[O_2]/[CO]$ ratio operation, CO concentrations below 1 ppm in the product gases were achieved under all partial load operations by using the novel PROX catalyst. As for durability, stable performance of the fuel processor was demonstrated for more than 10,000 h. Stable performances were also demonstrated in both tests of more than 1000 start–stop cycles and more than 25,000 load changes. It was also confirmed that the PROX catalyst has a wide operation range and long-term durability in actual reactors for residential PEFC systems.

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