



35-We polymer electrolyte membrane fuel cell system for notebook computer using a compact fuel processor

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ABSTRACT

A polymer electrolyte membrane fuel cell (PEMFC) system is developed to power a notebook computer. The system consists of a compact methanol-reforming system with a CO preferential oxidation unit, a 16-cell PEMFC stack, and a control unit for the management of the system with a d.c.–d.c. converter. The compact fuel-processor system (260 cm³) generates about 1.2 L min⁻¹ of reformat, which corresponds to 35 We, with a low CO concentration (<30 ppm, typically 0 ppm), and is thus proven to be capable of being targeted at notebook computers.

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1. Introduction

Fast-developing technologies related to electronic devices, such as cellular phones, computers, camcorders, and cordless tools, have led to an increase in the demand for high-performance energy sources. Currently, rechargeable batteries are the major players in the market for powering hand-held devices. Nevertheless, as energy demands increase with the popularization of broadband-mobile computing and an ever-increasing number of functions, existing battery technology has come up against the need for new energy-providing devices such as mobile fuel cells.

Some of the promising fuel cell technologies for portable applications are liquid-fed direct methanol-fuel cells (DMFCs) and polymer electrolyte membrane fuel cells (PEMFCs), which have a low operating temperature and fast start-up, consequently targeting high-performance notebook computer systems that usually require higher specific energy. The DMFCs have lower power den-

sities than PEMFCs and suffer the problems of methanol crossover and slow methanol electrocatalysis [1]. Thus, PEMFCs are preferred more than DMFCs for the portable applications.

Polymer electrolyte membrane fuel cells provide an attractive alternative to conventional batteries over a wide range of power and energy capacities. The advantages of using PEMFCs are high electrical efficiency, flexibility with respect to power and capacity, long lifetime, and good ecological impact [2,3].

Hydrogen is the best fuel for fuel cells. For notebook computer applications, the hydrogen-supply units should be compact with high specific energy. Metal hydrides, chemical hydrides and fuel reformers are candidates for hydrogen-supply. A compact micro-fuel reformer is particularly attractive because of its high specific energy and capability for instant recharging of fuel [4]. Nevertheless, fuel-reformer technology has become a bottleneck for the practical use of fuel cells.

Hydrogen for PEMFCs can be produced in an on-board fuel processor from a liquid fuel such as methanol or gasoline. Although the term 'reformer' is often used for the whole system, the production of hydrogen actually occurs via three processes, as follows.

(1) the *autothermal reforming* of a hydrocarbon, i.e.,



wherein partial oxidation and steam reforming (SR) take place in the absence of water and oxygen, respectively;

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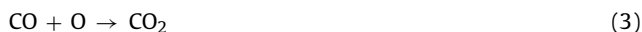
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(2) the water–gas shift reaction, which eliminates most of the CO and produces more hydrogen, i.e.,



(3) preferential oxidation (PROX) to decrease any remaining CO to ppm level [3].



The PROX reaction is a selective catalytic oxidation of CO in the H_2 -rich reformat using O_2 as an oxidant. Many auxiliary processes, such as fuel vapourization, sulfur removal, heat integration and effluent-gas combustion, can make PEMFC a very complicated device. To apply the fuel cell system to a notebook computer, the fuel system should be miniaturized and integrated within the system.

For powering a notebook-computer system, Samsung SDI Co. is in the process of developing a 50-We PEMFC for its continuous use (more than 10 h) on a single fuel cartridge using methanol as a fuel source (targets: $>150 \text{ mW cm}^{-2}$ power density; $>600 \text{ Wh L}^{-1}$ energy density; $<500 \text{ g}$ weight; $<930 \text{ cm}^3$ volume).

As a milestone, a 35-We PEMFC has been contracted and a continuous durability test lasting 24 h is reported here. Hydrogen with a low CO level ($<30 \text{ ppm}$, typically 0 ppm) is supplied using a compact methanol-reforming fuel processor, which includes a catalytic combustor, a methanol steam reformer, and a CO preferential oxidation reactor.

This study addresses the characteristics of the fuel cell–power system in terms of the design and performance of the fuel processor, which powers, the 35-We PEMFC that possesses the capability for carrying out various operations in a commercial notebook computer. The results from the durability test assure the reliable performance of the system in terms of the power output for within-the-range gas compositions.

2. System design

The 35-We PEMFC system for powering a notebook computer consisted of three parts, namely: fuel processor, stack and process controller. A schematic representation of the system for a notebook computer is shown in Fig. 1 and its three-dimensional graphic (3500 cm^3) representation is presented in Fig. 2.

The fuel cell power system operates at ambient temperature. The methanol-reforming system supplies hydrogen to the stack after reducing the CO content with a PROX system. The electrical power generated by the stack from the hydrogen and air fed from a

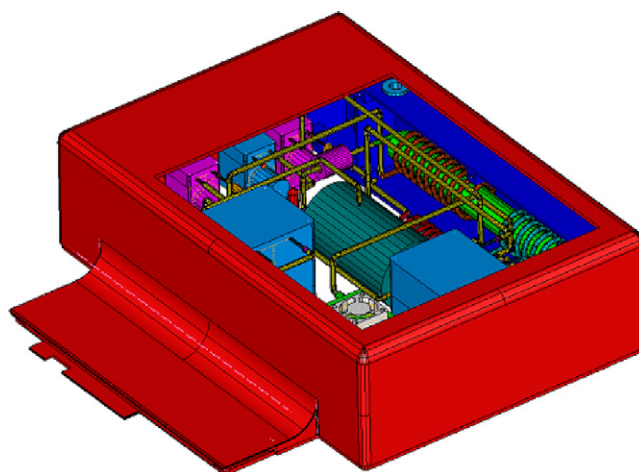


Fig. 2. Three-dimensional graphic presentation of 35-We PEM fuel cell system for notebook computer.

pump is supplied to a notebook computer. A control unit regulates the air-flow rate and the methanol-fuel pressure during the entire operation and conducts a controlled start-up and shut-down of the fuel cell and power converter.

2.1. Fuel-process system

A compact methanol-fuel process for a 35-We PEMFC for a notebook computer consists of a reformer with a catalytic combustor and a carbon monoxide catalytic reducer. The reformer, in turn, consists of an inner combustion-reaction unit (3/8 in. outside diameter, 3/8 in. stainless steel (SUS) 304), an outer methanol SR unit (1 in. outer diameter, SUS 304), and an outermost evaporation/heat-exchanger unit. The inner combustion-reaction unit supplies a major portion of the heat to the reforming unit by the total oxidation of methanol in air. Its flue gas subsequently exchanges the remaining heat with the incoming reforming-feed stream. The reforming-feed stream consists of methanol and water – both of which are evaporated by the heat exchange with the flue gas – is fed to the reforming unit and therein undergoes an endothermic SR reaction.

The reformatted gas stream is cooled while passing through a separator located in the fuel tank, and the unreacted methanol and water are condensed back to their liquid forms. The reformatted gas stream (reformat hereinafter) is composed of 74% H_2 , 24% CO_2 , and about 1% CO. A stainless-steel thermos unit insulates the reformer that has a total volume of 260 cm^3 . The percentage of CO is further reduced by PROX of CO with air.

2.2. Stack

Single-cell module: Independent of the investigated flow field, each fuel cell was constructed with an Umicore® membrane-electrode assembly (Series R300E, catalyst loading of anode and cathode: $0.8 \text{ mg Pt cm}^{-2}$; PtRu anode+Pt cathode) sandwiched between TORAY® carbon paper (TGPH-090, Japan).

The set-up was placed between the flow-field plates, which were made of SGL SIGRACET® material (R8710). Stainless-steel plates were placed on either side to provide external electric circuit connections and to compress the stack. The application of a high clamping pressure (120 N cm^{-2}) of the cell reduced the contact resistance and affected the fuel cell performance positively. The total active area of the single-cell module was about 90 cm^2 .

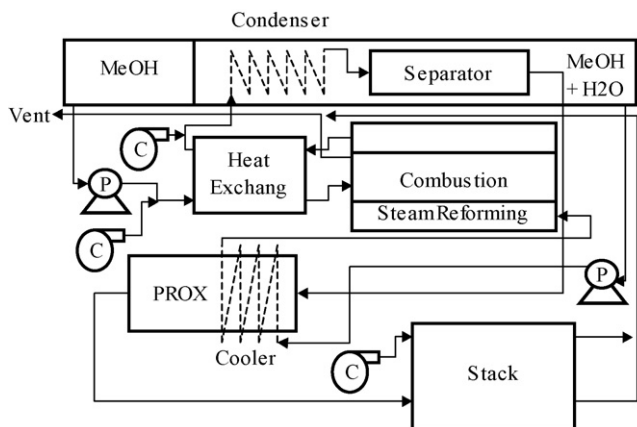


Fig. 1. Schematic diagram of 35-We PEM fuel cell system for notebook computer.

Sixteen-cell stack: The 16 cells of the stack were assembled similar to the single-cell module. The main difference is that the 16-cell system needs no additional plates to provide electric connections or compression force. The active area of the whole stack was approximately 144 cm².

An air pump (69 mm × 63 mm × 85.5 mm, Schwarzer Precision, Germany) was used for supplying air (4.5 L min⁻¹) to the cathode. Around 1.2 L min⁻¹ of reformat was fed to the anode. The stack test was performed at ambient temperatures. The temperature of the stack was maintained below 333 K by four fans situated on top of the stack. All physical parameters, such as the electric current and voltage of the fuel cell, were recorded with a data-acquisition system (34970A, Agilent) and an on-line computer.

2.3. Control unit

A control unit regulated the air-flow rate and methanol-fuel pressure during testing and achieved a controlled start-up and shut-down of the fuel cell d.c.–d.c. power converter. Due to confidentiality issues, detailed information is not provided in this article.

3. Experiments

The reactors designed for the reforming process and the preferential oxidation of CO (PROX) were fabricated by the WON Engineering Co. (Korea). For methanol combustion, 0.5 g of 1 wt.% Pt/γ-Al₂O₃ (pellet type, 3.2-mm o.d., Aldrich) and 1.5 g of Ru/γ-Al₂O₃ (sphere type, 3.0-mm o.d., Sud-Chemie Inc.) were used. For methanol steam reforming, 6 g of Cu–Zn–Al₂O₃ (pellet type, 3.2-mm o.d., Sud-Chemie Inc.) was used.

Three PROX catalysts were evaluated, namely: 0.5 wt.% Pt/γ-Al₂O₃ (Aldrich), Ru on γ-Al₂O₃ (RUA, Sud-Chemie Inc.), and water-pretreated 5 wt.% Pt/γ-Al₂O₃ (Poasen Co. Korea).

The water-pretreated 5 wt.% Pt/γ-Al₂O₃, prepared using a new water-pretreatment method, was supplied by the Poasen Co., Korea. A water-pretreatment method has been previously reported [5–9] for the alumina-supported platinum catalysts used in the PROX reaction. The sphere type of catalyst was 5 wt.% Pt/γ-Al₂O₃ (WT5Pt/Al–S), prepared by the incipient wetness method, typical of those reported in literature [10].

Conventional pretreatment consists of calcination in oxygen and reduction in hydrogen, both for 1 h at 773 K. By contrast, the new pretreatment consists of interrupting the reduction step, cooling the catalyst to room temperature, and adding liquid water to incipient wetness while retaining the catalyst under a hydrogen atmosphere. The catalyst is then slowly heated back to 773 K and the reduction continues. For obtaining a honeycomb-type catalyst (WT5Pt/Al–H), the powdered WT5Pt/Al–S (<10 μm, ϕ) is coated on to a honeycomb monolith (400 cpi, Corning).

The flow rate of the simulated reformat (74.9% H₂, 22.1% CO₂, 1% CO, balance N₂; Dukyang energen Co., Korea) was controlled by a ball flow meter during the PROX-activity evaluation over the three kinds of catalysts. An air pump (34 mm × 35 mm × 34 mm, Schwarzer Precision, Germany) was used for adding air to the simulated mixture of gases at a flow rate up to 500 ml min⁻¹. The total flow rate of the reformat was measured by a wet gas meter. For high-temperature activity measurements, a heater was used.

A gas chromatograph (6890N, Agilent) with a packed molecular sieve and thermal conductivity detector was used to determine the concentrations of H₂, O₂, CH₄, and CO₂. An on-line CO analyzer (California Analytical Instrument Inc., 601C, USA), sensitive at ppm levels, was employed for accurately determining CO concentrations. The conversions of CO and O₂ and the selectivity to CO₂

were calculated using the following formulae:

$$\text{CO conversion (\%)} = \frac{[\text{CO}]_{\text{out}}}{[\text{CO}]_{\text{in}}} \times 100 \quad (4)$$

$$\text{O}_2 \text{ conversion (\%)} = \frac{[\text{O}_2]_{\text{in}} - [\text{O}_2]_{\text{out}}}{[\text{O}_2]_{\text{in}}} \times 100 \quad (5)$$

$$\text{CO selectivity (\%)} = \frac{0.5 \times [\text{CO}_2]_{\text{out}}}{[\text{O}_2]_{\text{in}} - [\text{O}_2]_{\text{out}}} \times 100 \quad (6)$$

The thermal or reforming efficiency, $\eta(\text{H}_2)$ is defined as the lower heating value (LHV) of the hydrogen produced, if it includes a purification step for complete fuel-processing units [11–12]. The LHV of the fuel can be calculated using the following equation:

$$\eta(\text{H}_2) = \frac{[n(\text{H}_2)\text{LHV}(\text{H}_2)]}{[n(\text{fuel})\text{LHV}(\text{fuel})]} \quad (7)$$

4. Results and discussion

4.1. Methanol steam reforming

Typical catalysts for methanol SR are based on Cu–Zn–Al₂O₃. Attempts to improve this catalyst have continued so as to yield better activity, CO₂ selectivity and durability. Methanol SR can theoretically yield 75% hydrogen at 100% CO₂ selectivity [12]. The catalytic SR of methanol as a hydrogen source is hence attracting interest for portable-power fuel cell applications. i.e.,



The reaction is only moderately endothermic, and it requires a relatively low process temperature (473–673 K) with small steam-to-carbon ratios to produce a reformat with a high H₂ concentration. Its low operating temperature makes it an attractive candidate for portable-power systems.

A cylindrical reformer consists of an inner combustion reactor and an outer SR reactor. The inner combustion reactor supplies heat to the SR reactor. Conversion through SR is controlled by the combustion.

To obtain a sufficient amount of reformat with low CO concentration, the SR reaction temperatures are controlled by the feeding rate of the mixture of methanol and water (steam-to-carbon ratio is 1.2) for SR, and by the feeding rate of fuel (0.18–0.24 ml min⁻¹) with an air-flow rate of 2 L min⁻¹ for the combustion reaction.

For power above 35 We, the flow rate of reformat should be more than 1.2 L min⁻¹ with around 1% CO, under which conditions the fuel cell efficiency for electrical power from H₂ reaches about 50%. To satisfy these conditions, the concentration of CO in the reformat is measured as a function of the temperature of SR (Table 1). The reactor temperatures reported have been measured at the outlet of the reactor surface. The maximum temperature difference is almost 20–50 K between the catalysts and the thermocouple.

With increase in the SR temperature, the rate of reformat production is higher with increasing CO concentration. At 364–374 K, the reformat is composed of 74% H₂, 24% CO₂, and less than 1% CO

Table 1
Reformat production rate and CO concentration for SR reformat as function of temperatures

Steam reforming temperature (K)	Reformat (L min ⁻¹)	CO (%)
364	1.17	0.9
370	1.20	1.18
382	1.21	1.47
408	1.29	1.71
488	1.30	3.56

Table 2
Optimum conditions for reformer for 35 We fuel cell system

Methanol combustion	
Volume of combustion reactor	36 cm ²
Fuel feed rate for combustion	0.18–0.24 ml min ⁻¹
Flow rate of air for combustion	2 L min ⁻¹
Methanol steam reforming	
Volume of steam reforming reactor	38 cm ⁻²
S/C ratio at steam reformer	1.2
Fuel rate for steam reformer	1.2–1.22 ml min ⁻¹
Average surface temperature of steam reformer	364 K
Flow rate of reformat	1.17–1.23 L min ⁻¹
Thermal efficiency (LHV)	82%
Conversion of steam reforming	62%
Volume of insulated reformer	222.5 cm ² (4.5 cm outer diameter × 14 cm)
Composition of reformat	74% H ₂ , 24% CO ₂ , and 1% CO or so

o.d. = outer diameter

using an airflow of 1.17–1.23 L min⁻¹. Table 2 shows the optimum operating conditions for the reformat.

The durability of the reforming process for 200 h has been confirmed by monitoring the reformat obtained. The temperature is maintained at 364–374 K with a stable SR activity. The thermal efficacy of this reformer is 82%, as calculated using Eq. (7).

4.2. PROX of CO

The CO concentration from a reformer/water–gas shift unit is typically about 1 mol%, which is determined by the thermodynamic equilibrium of the water–gas shift reaction. The PEMFC anode is a Pt catalyst that is very sensitive to CO poisoning at low temperatures. According to the Partnership for a New Generation of Vehicles (a broad partnership between the US government, industry, universities, and national labs), the target CO concentration for a fuel processor is 10 ppm [13]. Several approaches are currently applied to achieve this target: CO PROX, catalytic methanation, and Pd-membrane separation. Among these, PROX is the preferred method as it involves the lowest cost for reducing CO levels to the desired value without excessive hydrogen consumption [12].

To achieve this low CO concentration, the PROX reactor is placed between the shift reactor and the fuel cell anode. The Los Alamos National Laboratory (the leading laboratory for PROX systems under the Department Of Energy) has developed what many consider to be the state-of-the-art PROX catalyst and reaction system. It can achieve low concentrations of CO (10–20 ppm) in a multistage reactor with a Pt/Al₂O₃ or Ru/Al₂O₃ catalyst. The reaction chemistry is complicated and involves not only the catalytic oxidation of both CO and H₂ but also methanation and water–gas shift.

As for many catalytic processes, the challenges in this process include activity and selectivity. The conversion of CO has

to be 99.9% to achieve a concentration of 10 ppm. At the same time, the reformat contains mostly hydrogen and its oxidation obviously decreases the overall fuel efficiency. Because the PROX reactor is placed between the methanol-SR reactor (~523 K, typically Cu–Zn–Al₂O₃) and the PEMFC (~353 K) in our 35-We PEM fuel cell system, the PROX reactor should be able to operate between these temperatures.

Operation of PROX at low temperatures is also very important for start-up of a fuel cell system. Therefore, the PROX reactor must operate over a wide temperature range to be practical. In practice, unreacted methanol and a higher CO concentration can be more difficult to remove or transformed by the low SR activity and selectivity.

Supported noble-metal catalysts, such as Pt, Ru and Au, have been effective for the enhancement of the PROX reaction [3–12,14,15,10,16,17]. At low temperatures, highly dispersed gold on an oxide support exhibits high activity and selectivity (maximum at 553 K) [13]. The CO conversion and selectivity are lower at higher temperatures (423–473 K) because H₂ oxidation and selectivity decrease at low CO concentrations. Supported Pt has a high conversion of CO at high temperatures (423–473 K) but the activity decreases at low temperatures. In addition, methanation does not occur even at high reaction temperatures (~573 K) [15]. Therefore, supported Pt could be used as a practical PROX catalyst if the low-temperature activity (at 298–373 K) could be improved.

For its application in a notebook computer fuel cell system, the PROX catalyst should have a very high CO oxidation activity, with selectivity over a wide temperature range and a fast start-up. The following tests were carried out to find an ideal PROX catalyst that satisfies these conditions.

4.2.1. Evaluation of PROX catalysts

The conversion and selectivity of CO as functions of the ratios of O₂ to CO (λ) and temperatures are shown in Table 3 for three catalysts, namely: WT5Pt/Al–S, 0.5Pt/A, and Ru/Al. Table 4 shows the notations, types and amounts consumed for the three catalysts. The excess oxygen present with respect to the amount of oxygen required for the oxidation of CO to CO₂ is characterized by the process parameter, (λ), i.e.,

$$\lambda = 2c_{\text{O}_2} c_{\text{CO}}^{-1} = 2p_{\text{O}_2} p_{\text{CO}}^{-1} \quad (9)$$

where $\lambda = 1$ is sufficient to effect for the complete oxidation of CO to CO₂ in the absence of any oxygen-consuming side-reaction such as H₂ oxidation.

The test conditions were 1 atm., a flow rate of 1.2 L min⁻¹ (STP), temperatures of 298, 393 and 423 K, and 1.4 or 1.8 λ . Because of the high loading (wt.%) of the noble metal, Pt, 2 g of WT5Pt/Al–S catalyst were used whereas 20 g of 0.5Pt/Al and Ru/Al were used.

The loading (wt.%) of Ru and preparation of Ru/Al are proprietary under Sud-Chemie Inc. The reaction was started at room

Table 3
CO conversion and CO selectivity over catalysts with different ratios of O₂ to CO (Lambda) and temperatures (74.9% H₂, 22.1% CO₂, 1% CO, and balance N₂; GHSV: 2400 for 0.5Pt/Al and Ru/Al and 40 000 for WT5Pt/Al–S)

Temperature (K)	Lambda (λ)	CO conversion and CO selectivity (%)					
		WT5Pt/Al–S		0.5Pt/Al		Ru/Al	
		CO conversion	Selectivity	CO conversion	Selectivity	CO conversion	Selectivity
298	1.4	75.5	85.4	12.1	95.5	2.6	99.5
298	1.8	82.5	75.7	7.5	98.5	5.3	99.2
393	1.4	95.2	77.3	92.5	66.1	77.9	55.6
393	1.8	90.1	50.1	78.7	43.7	92.0	51.1
423	1.4	99.8	71.3	79.3	56.6	59.4	42.4
423	1.8	80.9	44.9	99.1	55.1	76.6	42.5

Table 4

Notation, type and amount of catalysts used for PROX-catalyst evaluation

Notation	Type	Used catalyst amount (g or cm ³)	Catalyst
WT5Pt/Al-S	Sphere (0.5 mm outer diameter)	2 g	Water-pretreated 5wt.% Pt/ γ -Al ₂ O ₃ (Poasen)
WT5Pt/Al-H	Honeycomb (400 cpsi)	12 cm ³ (8 mm outer diameter \times 60 mm)	
0.5Pt/Al	Pellet (3.2 mm outer diameter)	20 g	0.5 wt.% Pt/ γ -Al ₂ O ₃ (Aldrich)
Ru/Al	Sphere (3 mm outer diameter)	20 g	Ru/ γ -Al ₂ O ₃ (Aldrich)

o.d. = outer diameter

temperature and was gradually increased at the desired temperature. Measurements at each temperature were taken after 30 min to allow the reaction to stabilize. The simulated reformat (74.9% H₂, 22.1% CO₂, 1% CO, balance N₂, Dukyang energen Co.) was used as the reactant. The reactor was a stainless-steel tube type (50 mm outer diameter \times 100 mm length). The catalyst bed was held in place with glass-wool plugs. The reactor temperatures reported were measured at the outlet of the reactor surface. The maximum temperature difference was almost 30 K between the catalyst and the thermocouple, and the maximum bed temperature might be even higher [6] because of the highly exothermic reactions.

Most studies on the PROX reaction failed to report the temperature gradient. The experimental conditions are similar to the operating conditions of a PROX reactor. In all experiments, the CO conversion increases with increase in O₂ concentration, whereas the CO selectivity decreases. In addition, above 393 K, the O₂ conversion is almost 100%. The decrease in CO conversion at high temperature is caused by hydrogen oxidation: increasing gas chromatography (GC) water peaks are observed without other peaks such as those for CH₄. Therefore, selectivity is improved at low O₂ concentrations and lower reaction temperatures because hydrogen oxidation is suppressed.

The conversion and selectivity for CO while using WT5Pt/Al-S are much greater over a larger temperature range, especially at lower temperatures. At lower ratios of O₂ to CO above 393 K, the conversion and selectivity are higher due to the limited H₂ oxidation, a side-reaction. The superior CO conversion and selectivity compared with the 0.5Pt/Al are not caused by the higher gas hourly space velocity (GHSV): 2400 and 40 000 for 0.5Pt/Al and WT5Pt/Al-S, respectively. Korotkikh and Farrauto [15] indicated that lower space velocity is positive for CO conversion, but negative for selectivity. Therefore, the conclusion is that the water-pretreated Pt catalyst have higher activity and selectivity.

The Ru/Al displays very low activity and selectivity. Ruthenium catalysts are not practically useful in the PROX system due to their higher operating temperature (413–473 K) compared with that of the PEMFC (about 253 K) and the active methanation process for both CO and CO₂, which is a side-reaction that occurs above 473 K.

Our previous studies [3,5–9] found that the activity and selectivity of the CO oxidation in H₂ are improved over a very large temperature range (255–473 K) by the water-pretreatment method for a 5 wt.% Pt/ γ -Al₂O₃ catalyst. The improvement is attributed to a reduction in the size of the metallic Pt nanoparticles and an increase in their chemical interaction with the alumina support.

The WT5Pt/Al-S offers some advantages over the standard pretreated catalyst. It is more active and selective over a very broad range of temperatures. For the typical operating temperatures of the PROX reactor (273–423 K), the conversion and selectivity are substantially higher. In addition, at low temperatures that could be encountered during the start-up of mobile notebook-computer systems, the catalyst gives a very high conversion and selectivity. Thus, the autothermal PROX system can be preferably designed using WT5Pt/Al catalysts.

4.2.2. Autothermal PROX system

An autothermal PROX system can be developed using the WT5Pt/Al catalyst because, at low ignition temperatures, it shows high activity and selectivity over a large temperature range. It has higher efficiency because it does not require a heating system (no electrical consumption by the remaining side-reaction processes) or the cycling of heat. A coiled-tube heat-exchanger, surrounding a honeycomb-type PROX catalyst, can maintain the active temperatures of the reaction that can remove CO to below 50 ppm from about 1% CO in the real reformat, thus avoiding CO poisoning of the anode catalysts in the stack.

4.2.2.1. Honeycomb-type PROX catalyst. For mobile applications, the use of particulate, spherical, pelleted or powder-type of PROX catalysts is unsuitable because of lack of mechanical stability against attrition and pressure drop. In the PROX reaction, heat from the CO and/or H₂-oxidation reactions should be released for selectivity and to avoid side-reactions, the reversible water-gas shift reaction, and methanation. The heat may be passively cooled through radiative heat loss or actively cooled using a heat-exchanger placed within the bed. When using a particulate type of catalysts, the cooling systems are ineffective because of the large geometric surface area.

For mobile applications, minimization of the size and the weight of the PROX reactor are important and can be accomplished through the direct application of the PROX catalyst on to a suitable compact heat-transfer structure [18]. Therefore, the heat-transfer technology should satisfy the following conditions: (i) high surface area to volume ratio; (ii) suitability for catalyst application; (iii) light weight.

Using a ceramic monolith is advantageous for a large geometric surface area, with little backpressure, and for the adiabatic PROX operation. There is a positive temperature gradient moving down the catalysts as the reaction moves towards completion. In addition, the reduced thickness of the catalytic layer on the walls of the monolith can minimize the intraparticle (internal) transport resistance.

The active WT5Pt/Al-S was wash-coated on to a ceramic honeycomb (WT5Pt/Al-H, straight-channel monoliths containing 400 cpsi (62 cells cm⁻²) supplied by the Poasen Co.). It has a lower space velocity (3600 GHSV, 20 cm³ catalyst).

The activity and surface temperatures of the outlet reactor of WT5Pt/Al-H with different ratios of O₂ to CO (λ) in the reformat are shown in Fig. 3. The reaction started at room temperature. The temperature was increased by the desired ratios of O₂ to CO. Measurements at each ratio were taken after 30 min to allow the reaction to stabilize. The reformat from the SR reformer was used as the reactant. The reactor was a stainless-steel tube type (34 cm³, 25-mm outer diameter \times 70 mm). The honeycomb-type catalyst was held in place with glass-wool plugs.

At 1.4 (λ), the activity of WT5Pt/Al-H is much improved, but with lower selectivity, compared with the WT5Pt/Al-S. The decrease in CO conversion is caused by hydrogen oxidation: the absence of an O₂ peak is observed with increasing GC water peaks and without other peaks such as those for CH₄. The CO conversion generally

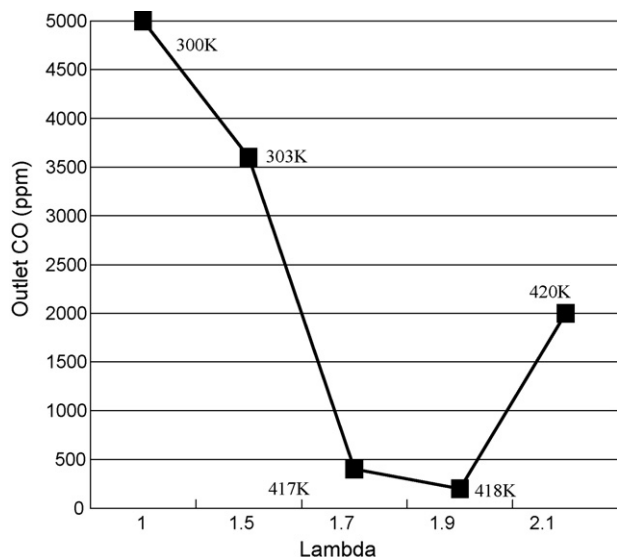


Fig. 3. Activity and surface temperature of outlet reactor of WT5Pt/Al-H with different ratios of O₂ to CO (74.9% H₂, 22.1% CO₂, and 0.8% CO; GHSV: 6300).

increases with increasing ratios of O₂ to CO and peaks at 1.9 (λ). The temperature increases with the ratio of O₂ to CO due to the highly exothermic reactions of the H₂ and CO oxidations. Thus, the operation temperature and activity of an autothermal PROX system can be controlled by the ratio of O₂ to CO.

At 1.9 (λ), the initial conversion (typically 0 ppm for 20 min) is higher than reported and decreases gradually with increasing reaction temperatures. The raised temperatures are caused by the accumulation of heat because the generated heat is more than that released. To maintain higher activity, a cooling system for the reactor should be provided.

4.2.2.2. Cooling system for PROX reactor. Thermal management of the exothermic PROX reaction is important for higher activity. Hot-spot formation appears to be prevalent, with large temperature differentials between the front and the back of the honeycomb structure (a maximum difference of 40 K). Inclusion of the heat-transfer elements within the honeycomb structure is not practically effective in maintaining the optimum reaction temperature for PROX under H₂ excess. Thus, an additional cooling system should be necessarily applied.

A coiled, stainless-steel tube (1/8 in. outer diameter) was used in the PROX reactor as a heat-exchanger (Fig. 4). For coolant, a SR mixture was fed and circulated through the coiled tube. Subsequently, the heated coolant was fed to the SR reactor for thermal efficiency of the reformer. This cooling system could keep the PROX temperature below 420 K.

For stable reaction temperatures, the WT5Pt/Al-H was cooled by the heat-exchanger tube at 1.8–1.9 (λ). It can remove CO to below 30 ppm (typically 0 ppm for 5 h) in a simulated reformat mixture (74.9% H₂, 22.1% CO₂, 1% CO, and balance N₂) for 10 h. This satisfies one of the required conditions (no data in this article).

4.3. Evaluation of fuel process

The designed fuel process, a reformer system that included a PROX reactor, was integrated and evaluated. The total size of the reformer system is below 260 cm³. The reformer consists of the outer methanol SR with an inner methanol combustion reactor. It was operated under optimum conditions (see Table 2) to generate reformat (74% H₂, 24% CO₂, and 1% CO). The rate of reformat gen-



Fig. 4. Photograph of PROX reactor with coiled heat-exchanger (34 cm³, 25 mm outer diameter \times 70 mm length.).

eration is stable with varying temperatures, to yield a stable \sim 1% CO concentration, which is removed by the designed 'autothermal' PROX system using a WT5Pt/Al-H catalyst at 1.8 (λ). Activity data from the processing of fuel for about 2 h are shown in Fig. 5. The CO is removed to less than 50 ppm, typically 0 ppm. After PROX reformer processing, the composition of the reformat is 72% H₂, 24.3% CO₂, and 3.67% N₂ (\pm 5% error range). Thus, this compact fuel processor can successfully generate reformat for a 35-We PEMFC system for application in a notebook computer.

4.4. Evaluation of 35-We fuel cell system with a notebook computer

4.4.1. Durability of stack in 50 ppm CO

To define the characteristics of CO tolerance of the anode in the designed stack (see Section 2.2 for details), the stack tests were carried out with pure H₂ and 50 ppm CO-balanced H₂.

The output power from the stack was monitored as a function of time and is shown in Fig. 6. With pure H₂, high stable power (about 43 We) is delivered. As expected, the power declines with time at a rate of 5 We h⁻¹, when 50 ppm CO is present in the fuel. The power is lower than 35 We – a typical target – after 2 h 20 min with the CO-containing fuel. After the fuel is switched back to pure H₂, the power recovers quickly and stabilizes after about 10 min (open triangle symbol). But when the power is recorded, the stack performance is not the same as its initial one, which indicates that there is some adsorbed CO left behind on the anode.

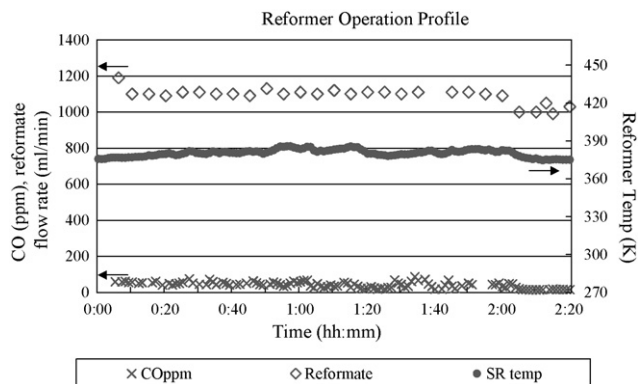


Fig. 5. CO concentration, rate of generated reformat, and reformer temperature plotted against time for integrated reformer system.

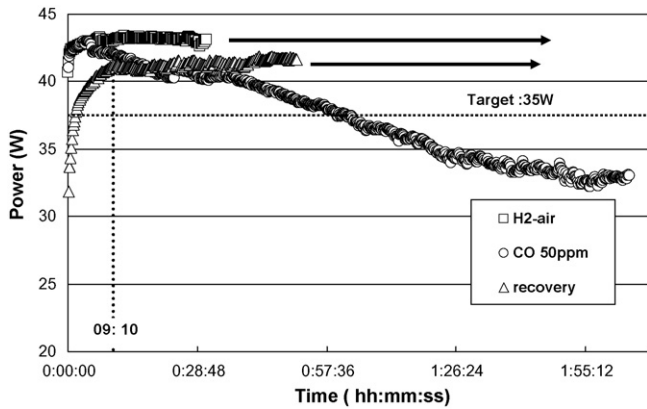


Fig. 6. Stack performance for anode fuel of pure H₂ and 50 ppm CO, with balance made up by H₂ (88 A (current), 144 cm² (active area), 0.6 A cm⁻² (current density), 735 ml min⁻¹ fuel for anode, 4.6 L min⁻¹ air for cathode).

Therefore, a constant concentration of 50 ppm CO in the fuel can poison the anode and yield a lower output of power. The designed reformer system should be able to remove CO below 50 ppm with sufficient reformate for a 35-We PEM fuel cell system to be used in a notebook computer.

4.4.2. Evaluation of 35-We fuel cell system with notebook computer for 24 h

The 35-We fuel cell system was installed in a notebook computer (Q10, Samsung Electronics) and monitored continuously for 24 h. The CO concentration, the rate of reformate generation, and the SR temperature for a 24-h study are presented in Fig. 7. Variations in the output power from the stack and the power consumed by the notebook computer are observed, as shown in Fig. 8. The results are summarized in Table 5.

It should be noted that the start-up power for operation of the fuel cell system and the notebook computer is supplied by a Li-ion battery, and later the power is provided by the fuel cell stack after 30 min in the steady-state.

Two peaks of increased CO concentration are observed, as shown in Fig. 7 at about 12 and 20 h. The former is due to caused by the overflow of highly condensed methanol from the condenser to the PROX unit (the condenser is located between the reformer and the PROX unit in the fuel cartridge). It causes the oxidation of methanol instead of CO. The CO level is brought back to below 20 ppm after

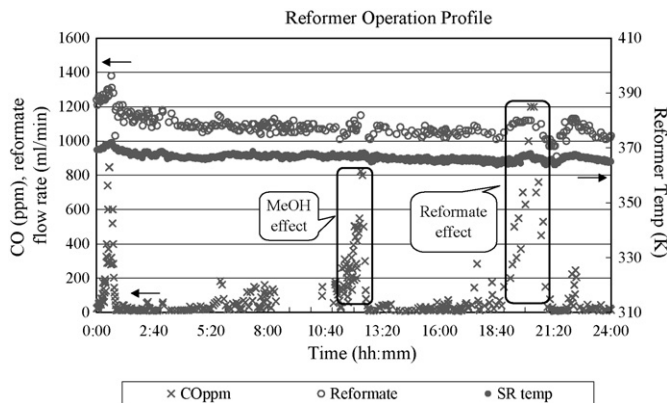


Fig. 7. CO concentration, rate of generated reformate, and reformer temperature plotted against time for 35 We fuel cell system for notebook computer (Q10, Samsung Electronics) for 24-h demonstration.

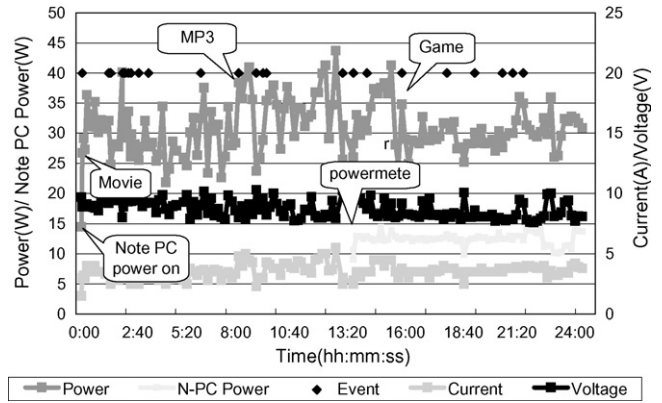


Fig. 8. Powers, currents, and voltages plotted against time for 35 We fuel cell system installed in notebook computer (Q10, Samsung Electronics) for 24-h demonstration.

emptying the condenser. The latter results from the high concentration of CO from the reformer due to the higher reformer temperature. It also accounts for the low CO concentration after reducing the reformer temperature. Operation conditions should be optimized in future work.

The notebook computer was operated for 24 h using various programs such as playing mp3 songs, videos, and games. It gave very stable power and fully responded to the loads of computer operations (see Fig. 8). The voltage and current remained stable. Although twice the concentration of CO, much higher than 50 ppm, was present in the reformate, the output power was not affected. Thus, varying the CO concentrations may help in the regeneration of CO-poisoned anode catalysts.

No catalytic deactivation is observed for the reformer using both PROX and electrode catalysts after this demonstration.

Thus, the compact fuel process developed in this study has been successfully adopted in the 35-We fuel cell system for a notebook computer. In real PEMFC systems, it has been shown that these fuel processors have high thermal efficiencies and low CO concentrations with adequate rates of reformate generation. The fuel cell system successfully provides power for the operation of a notebook computer.

The topics of current research are:

- (i) increasing the output power of the stack (stack 60 W = balance-of-process 10 W + system 50 W) for high performance
- (ii) minimization of fuel cell system size (<1200 cm³, typically <100 cm³ reformer)
- (iii) faster start-up (<5 min, now 15 min (max.))

Table 5

Summary of results for the 35 We fuel cell system installed in notebook computer; () implies used amount of fuel and water without recycling

Used methanol for 24 h	983 (.452) cm ³
Used water for 24 h	344 (555) cm ³
The rate of used methanol to water	60 cm ³ /23 cm ³ h ⁻¹
Average surface temperature of steam reformer	364 K
Flow rate of reformate	1–1.2 L min ⁻¹
Surface temperature of reformer	364 K
Conversion of steam reforming	62%
Average power for stack	35 We
Average power for system	13 We
Variation of voltage	11–8 V
Variation of current	5.5–2.5 A
Consumed power for balance of process	10 We
Total volume of 35-We fuel cell system	3500 cm ³
Fuel tank included in the system	400 cm ³

- (iv) optimization and control of operation of the fuel process
- (v) better power density for the stack ($>500 \text{ mW cm}^{-1}$)
- (vi) less weight of the total system.

5. Conclusion

A compact methanol-fuel processor for a 35-We fuel cell system for a notebook computer has been developed with high thermal efficiency. The volume of the fuel-processing unit is 260 cm^3 , and it generates about 1.2 L min^{-1} reformat with a low CO concentration ($<30 \text{ ppm}$, typically 0 ppm). For preferential oxidation (PROX) of CO, tests for catalyst evaluation have been conducted, and the most promising candidate is a wash-coated, honeycomb-type, water-pretreated, Pt-on-alumina catalyst.

A stainless-steel coiled-tube PROX is used as a reactor for the heat-exchanger. A SR mixer is furnished to feed and circulate the coolant through the coiled tube. Then, the heated coolant is fed to the SR reactor to increase the thermal efficiency of the reformer. This cooling system can sustain or optimum PROX temperature to enable high activity and selectivity of CO oxidation.

The compact fuel process has been installed in a 35-We fuel cell system for a notebook computer. In actual PEMFC systems, it has been used for 24 h operation and the fuel processor has proved to have high thermal efficiency and low CO concentration with adequate reformat generation. The fuel cell system successfully generates power for operation of the notebook computer with high stability and durability.

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