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Journal of Power Sources 106 (2002) 68–75
JOURNAL OF POWER SOURCES
www.elsevier.com/locate/jpowsour

Development of a 50 kW PAFC power generation system

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Abstract

LG-Caltex Oil Corp. developed a 50 kW PAFC stack during 1994–1999. The aim of this study was the preparation of a 50 kW PAFC stack for the application of on-site power generation system. To manufacture the large-size electrodes (4225 cm^2), automated manufacturing facilities were used. These facilities provided uniformity and reproducibility of manufacturing of the electrodes. These electrodes showed high current density (400 mA/cm^2 at 0.65 V) in the unit cell measurement. Only 3% of degradation of cell voltage was observed in the unit cell test over the cumulative operation of 6000 h. The 50 kW PAFC stack was composed of 109 unit cells and 19 cooling plates. The 50 kW stack showed 85% of hydrogen utilization rate and 40% of electrical efficiency (LHV). Uniform voltage distribution among cells (within 5%) was observed during operation. LG-Caltex Oil Corp. is developing the prototype 50 kW PAFC power generation system during the years 2000–2002. This prototype will be composed of reformer, inverter, stack and control systems. The prototype will be tested for the commercial application of on-site power generation. The long run, reliability and economic feasibility of the prototype will be analyzed for commercial power generation.

Keywords: PAFC; Fuel cell; Stack; Fuel cell system; Reformer; Electrode

1. Introduction

Fuel cells are becoming widely accepted as a preferred means of generating electricity for distributed electrical power generation due to their high fuel conversion efficiency, environmental compatibility and flexibility of fuels [1,2]. During the last three decades, five types of fuel cells have been investigated such as alkaline fuel cells (AFC), phosphoric acid fuel cells (PAFC), molten carbonate fuel cells (MCFC), solid oxide fuel cells (SOFC) and proton exchange membrane (PEMFC). Among these fuel cells, PAFC is regarded as the most advanced technology in research and closest to commercialization. 50–200 kW of commercial PAFC power units have been developed and nearly 250 PAFC demonstration units are in operation in USA, Japan and Europe [3,4]. These PAFC power plants are demonstrated in office buildings and hotels for on-site power generation. Recently, PAFC power plants are also applied in the various application fields such as landfill gas utilization, cogeneration system, and uninterrupted power system (UPS) [5,6]. The PAFC power plant is used to remove the landfill gas, which must be flared or otherwise used for safety reason [5]. The 137 kW of output was obtained in the operation of PC-25C with landfill gas and 37.1% of efficiency at 120 kW was recorded in the operation [5]. The PAFC power plant was applied in the telecommunication equipment to supply the uninterrupted electric power, and the heat energy generated by fuel cell was used by absorption refrigerator to cool the utility [6]. Seventy-eight percent of total efficiency was estimated at the rated power of 200 kW.

PAFC technology has reached a level of maturity and the cell components and stacks are being manufactured at scale and in large quantity. On the other hand, to achieve market entry and economic competitiveness with conventional power generation systems, there is still need to increase the power density of the PAFC stack and to enhance the stability of system. Also, cost of the PAFC system should be further decreased by the development of low cost materials and improved manufacturing processes of the PAFC stack.

LG-Caltex Oil's business area includes oil refinery, petrochemical, LNG/LPG supplies and power generation. Experience in the energy business makes fuel cell development a natural extension of LG-Caltex Oil's energy portfolio. The fuel cell R&D activity at LG-Caltex Oil was initiated in 1989 under the governmental program of Korea. The 500 W, 2 kW, 15 kW and 40 kW PAFC stacks were developed during 1989 and 1994. Based on the R&D experience gained on the 500 W–40 kW stacks, a 50 kW PAFC stack was designed and manufactured from 1994 to 1999. During that period, large-size electrodes (4225 cm^2) were prepared with automated electrode manufacturing facilities. The 50 kW PAFC power generation system that consists of stack, reformer, inverter and heat recovery unit

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0378-7753/02/$ – see front matter
PII: S0378-7753(01)01052-7

has been developed with the financial aid of Ministry of Commerce, Industry and Energy, Korea (MOCIE) since 2000. We are also developing a 10 kW PAFC pilot system to obtain the technical data and experience for the integration of PAFC system. We are planning the demonstration of the 50 kW prototype to test the reliability and longevity for on-site application of the power generation system.

This article presents the technical results obtained from the development of the 50 kW PAFC stack in LG-Caltex Oil. The unit cell results and stack operation data of the 50 kW stack are discussed. The system development for 10 and 50 kW are also presented.

Table 1
Specification of 50 kW PAFC stack

Item	Specification
Number of cells	109
Area of electrode (cm^2)	4225
Operating pressure	Atmosphere
Operating temperature (°C)	185
Cooling type	Water cooling (pressurized water)
Number of cooling plate	19
Cell voltage (mV)	650
Current density (mA/cm^2)	216
Hydrogen utilization rate (%)	85
Electric efficiency (LHV) (%)	40
Weight (t)	2.5

2. Experimental

2.1. Preparation of electrode

To prepare the large-size electrode (4225 cm^2) with high current density and uniform electrochemical characteristics, automated manufacturing facilities such as spray, calendering, edge sealing machines were developed to prepare the electrodes. 10 wt.% of Pt/C catalyst was used in the anode, while 20 wt.% of alloy catalyst was used to prepare the cathode electrode. The metal loading of Pt catalysts were 0.6 mg Pt/cm^2. The alloy of the cathode catalyst consisted of Pt, Fe, and Co. The carbon paper was used as support of Pt catalyst. The carbon paper was impregnated with wet-proofing solution, which prevents the blockage of pores from the inflow of H_3PO_4 electrolyte and water. The carbon paper was dried at room temperature for 24 h and then calcined at

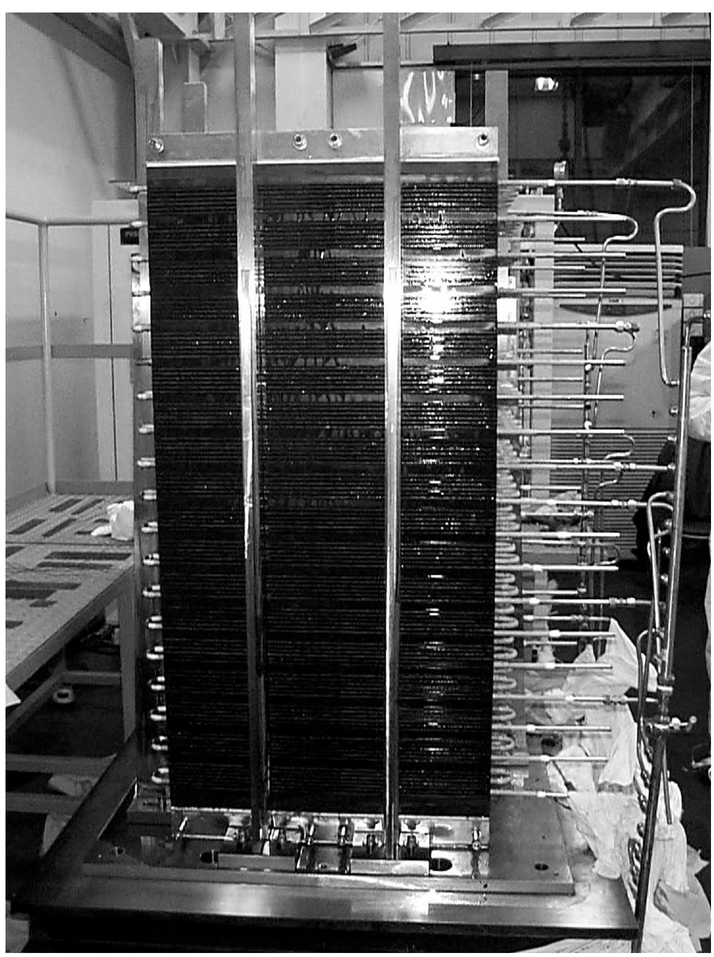

Fig. 1. Photograph of 50 kW PAFC stack.

360 °C for 2 h. The catalyst layer on the carbon paper was manufactured by spraying the catalyst slurry, which consisted of Pt/C catalyst, polyetra-fluoroethylene (PTFE) emulsion and iso-propyl alcohol. Then the catalyst layer was cold pressed, dried at 60 °C and sintered at 360 °C in inert atmosphere.

The matrix layer was prepared with a curtain coater. The curtain coater consisted of a coating system and conveyor belt. The matrix solution was prepared by mixing silicon carbide (SiC), polyethylene oxide (PEO) and PTFE. This matrix slurry was pumped from a tank into the coating head. From the coating head, the matrix slurry was a free falling film and returned back into the tank. The thickness of the matrix layer was optimized by controlling the speed of the conveyer belt and the thickness of solution curtain. The coated matrix was sintered at 360 °C for 1 h.

A single cell was fabricated by attaching the anode to cathode. Before testing the single cell, the cathode and anode were impregnated with phosphoric acid (105 wt.%) at 120 °C for 2 days. After placing the graphite plate and the current collector on the anode/cathode, the compression plates were attached at both sides. Then the single cell was compressed until a constant electric resistance was obtained by measuring the internal resistance of cell with milliohm meter. The effective electrode area of the single cell was 25 cm^2. Pure hydrogen was used as fuel on the anode side, air was used as an oxidant on the cathode. The flow rate of fuel and oxidant was adjusted with mass flow controller and a variable electronic load was connected to the current collector. The single cell tests were carried out at 185 °C.

2.2. 50 kW PAFC stack assembly and operation

Table 1 shows the specifications of 50 kW PAFC stack. The 50 kW PAFC stack consisted of 109 anodes and cathodes, respectively (Fig. 1). The anode and cathodes were 650 mm × 650 mm, and the height of stack was 1100 mm. The edges of electrodes were sealed with the mixture of SiC powder and PTFE solution. The electrodes were kept in 105% of H_3PO_4 for 48 h at 120 °C before stacking. The acid absorbed electrodes were then assembled in a stack with graphite bipolar plates. The water-cooling method was applied to control the stack temperature. The 19 cooling graphite plates were used to control the stack temperature during operation. Cooling plates have been inserted in every six cells in the stack. The temperature of the cooling water was controlled with a heat recovery system, which controls the stack temperature and supplies steam to the reformer. The 50 kW stack was operated at 160–185 °C using 85% of H_2 which supplied from the reformer process in the Yosu plant of LG-Caltex Oil Corp. Air was supplied to the cathode with an air blower. Stack data such as temperature of cooling plate, voltage of each cell were collected to the analog input of programmable logic controller (PLC).

3. Results and discussion

3.1. Unit cell performance and long-run test

Fig. 2 shows the polarization curves of unit cells for various cathode catalysts. The unit cell performance was measured at 185 °C and 10%Pt/C catalyst was used as anode. The open circuit voltage (OCV) was 0.9 V and 170 mA/cm^2 of current density was obtained in the 10% Pt/C catalyst at 0.65 V. The Pt loading was increased to 20 wt.%, and 250 mA/cm^2 was obtained in the unit cell as a result of the 20% Pt/C catalyst. When the 20% Pt-alloy (Pt-Fe-Co) catalyst was used as cathode, 400 mA/cm^2 of current density was measured at 0.65 V. The OCV was also

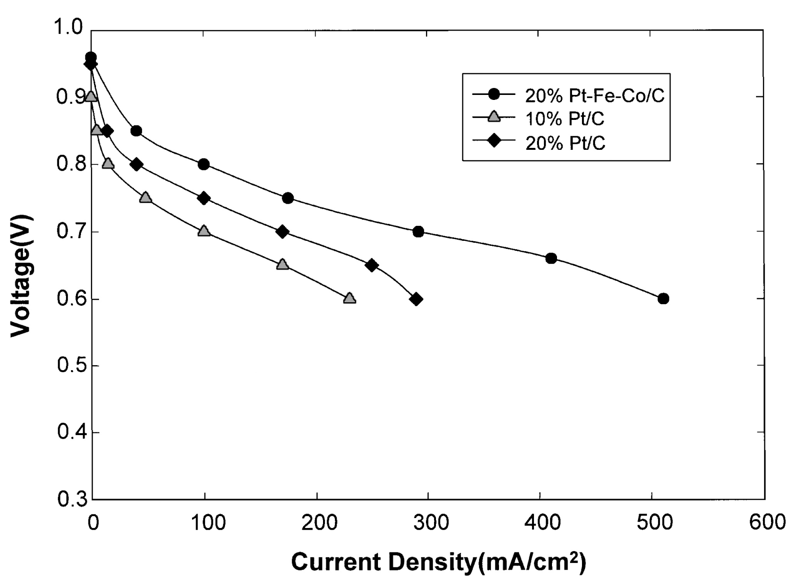

Fig. 2. Effect of cathode catalyst on the unit cell performance of PAFC (fuel, 100% H_2; oxidant, air; pressure, atmospheric pressure; temperature, 185 °C; anode, 10% Pt/C).

increased to 0.95 V. This shows that the high loading of Pt catalyst is effective to increase the current density in a PAFC. Also, alloy catalysts provide on increased current density than pure Pt catalyst. It has been reported that improved catalytic activity was observed in the Pt-alloy catalysts such as Pt-Cu-Fe [7], Pt-Cr [8], Pt-Co [9], Pt-Cr-Co [10], and Pt-Fe [11]. From our unit cell results, the performance of the 20% alloy catalyst (0.26 W/cm^2) was 60% higher than that of 20% Pt catalyst (0.16 W/cm^2). The improved activities of Pt-alloy catalysts were explained by the roughness effect [12] or ordered alloy structure [9]. The power density of 20% alloy catalyst is compared with other references, this result represents the highest performance of unit cell published to date [13].

When the manufacturing cost of 50 kW PAFC stack is evaluated, the catalyst cost is about 15–20% of total manufacturing cost of 50 kW PAFC stack. To compare the manufacturing cost of stack with the metal loading or the type of Pt catalysts, it is more cost effective (20–25%) to use the 20% alloy catalyst than other catalysts (10% Pt or 20% Pt catalysts). It comes from the increased current density of 20% alloy catalyst, which decreases the number of cells in the stacking of 50 kW stack. It also leads to a decrease in the cost of stack components such as bipolar plate (about 10%), carbon paper (2%) and other chemicals (6%). It shows that the high loading of Pt-alloy catalyst is cost effective to prepare the PAFC stack. Based on these results, we used the 20% Pt-alloy catalyst in the preparation of cathode electrode of 50 kW stack.

It has been reported that the alloy catalyst is more resistant to sintering in long run operation [14]. To test the performance degradation of PAFC electrode, the long-run operation of unit cell was tested. Fig. 3 shows the long-run result of unit cell measured at 185 °C for 6000 h. During the 6000 h of long-run test, the hydrogen was starved two times, which results in a 2–5 mV of voltage drop in the cell performance. A voltage drop of 2 mV was also observed when the outlet vent of cathode was blocked by accident. The long-run data shows a 5 mV voltage drop over 1000 h of operation with our 20% alloy catalyst. This shows that the performance of the electrode is constant over long-run operation. The lowest PAFC degradation rate publicly acknowledged is the 2 mV/1000 h at 200–250 mA/cm^2 in a short stack with 3600 cm^2 cells [13]. The voltage decay rate is rather higher than that of published data, the 5 mV/1000 h of degradation rate is within the range of tolerance. Our unit cell performance is measured on small 25 cm^2 cell and it may have some deviation with the performance of full-scale cell (4225 cm^2) in stack. To attenuate the problems occurring from the scale-up of electrode, such as reproducibility, degradation and uniform cell performance in large size cell, automated manufacturing facilities were used to prepare the 4225 cm^2 electrodes. This provides a more reliable and reproducible method in the production of the electrode than that of previous method. It is also effective to reduce the production period of 50 kW PAFC stack, which leads to a decrease in the cost of stack.

3.2. Performance of the 50 kW PAFC stack

Fig. 4 shows the performance of the 50 kW stack with an electrode area of 4225 cm^2 at an operating temperature of 185 °C. The 85% of hydrogen and air were used as fuel and oxidant, respectively. It is shown that 55 kW of DC power was obtained at the average cell voltage of 0.65 V. The current density of the 50 kW stack at 0.65 V was 216 mA/cm^2 (0.14 W/cm^2). The utilization rate of hydrogen was 85% and electric efficiency (LHV) was 40%. The average open circuit potential of the 50 kW stack was 1003.9 mV and the deviation of open circuit potential between each cell was

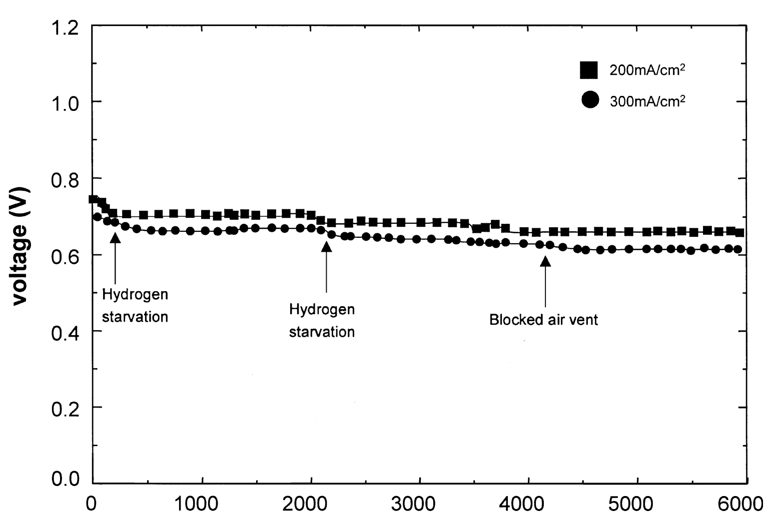

Fig. 3. Long-run data of unit cell performance with the usage of 20% alloy catalyst as cathode (fuel, 100% H_2; oxidant, air; temperature, 185 °C; anode, 10% Pt/C).

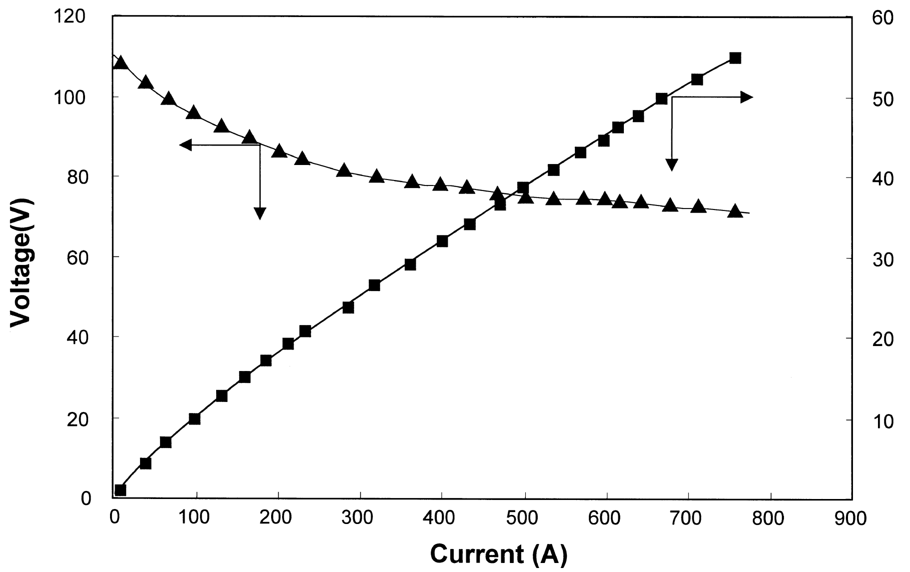

Fig. 4. I–V plot and power curves of operation of 50 kW PAFC stack (fuel, 85% H_2; oxidant, air; electrical efficiency (LHV), 40%; fuel utilization rate, 85%).

3.2 mV. Fig. 5 shows the voltage distribution of the cell group of the 50 kW stack at a constant load of 670 A. Each cell group is composed of 6 unit cells. The standard deviation of cell voltage was 7.4 mV, which denotes the uniform voltage distribution of the 50 kW stack. The uniform voltage distribution of the stack is necessary to prevent the decay of stack performance during long-run operation. To obtain the uniform distribution of voltage in stack, the fuel and air should be uniformly supplied in each cell. Also, the heat generated in each cell should be efficiently removed all over the stack. The voltage distribution data (Fig. 5) implies that fuel and air are uniformly distributed in the manifold and the cooling water removes the heat efficiently in the cooling plates.

The 50 kW PAFC stack was operated during the cumulative time of 650 h. The voltage decay or performance drop was not observed in the 650 h of operation. The 650 h of operation time is not long-time for the commercial operation of fuel cell power plant, but it shows the stable performance of the 50 kW stack was achieved and this stack can be used as a component of an on-site power generation system. We are planning to test the long-run operation after the installation of reformer during the current project (2001–2002).

To be applied as a commercial on-site system, there is need to improve the stability and reliability of stack for a commercial PAFC prototype. Namely, the flooding/drying of electrode, acid management and corrosion of carbon catalyst should be improved to enhance the stability of currently developed stack. This can be achieved by the modification of stack design, improvement of the H_3PO_4 supply method and stabilized Pt catalyst. Basic studies to improve the stability and reliability of stack are currently investigated in short-stack scale.

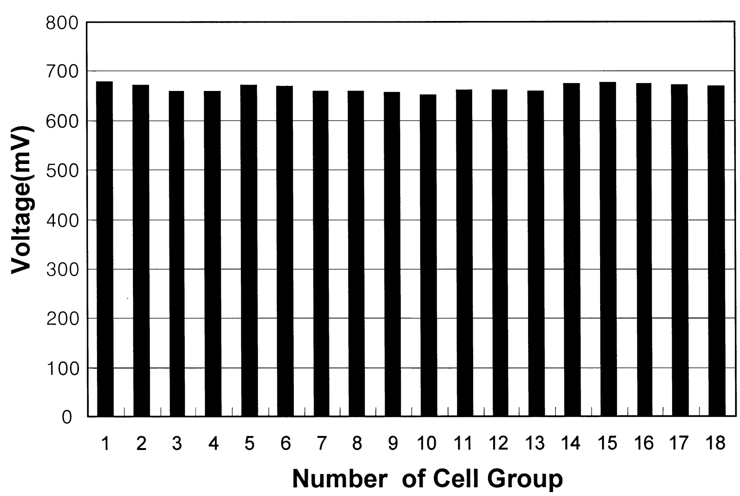

Fig. 5. Voltage distribution of 50 kW PAFC stack (I, 670 A; average voltage, 666 mV).

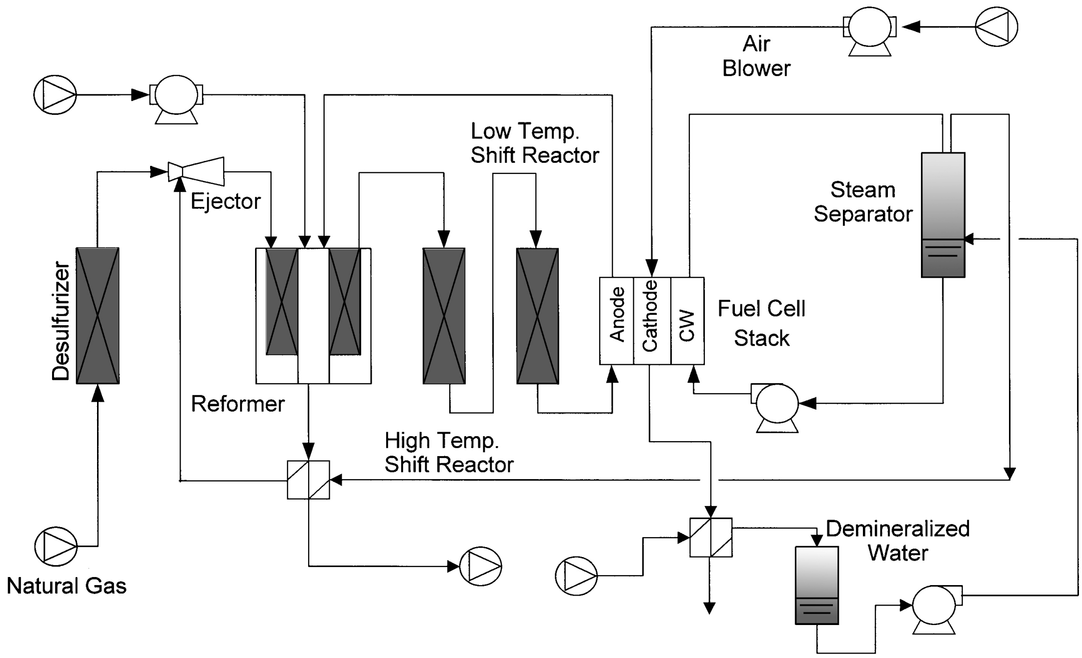

Fig. 6. Schematic process flow diagram of 10 kW PAFC pilot system.

3.3. Development of 10 kW pilot system

To obtain the technical data and experience of system integration, LG-Caltex Oil is developing the 10 kW PAFC pilot system. Fig. 6 shows the schematic process flow diagram (PFD) of 10 kW PAFC pilot system. It consists of three sections, namely fuel process system, fuel cell stack and heat recovery system.

The fuel processing system comprises three reactors: desulfurizer, steam reformer and CO shift reactor. Natural gas is used as fuel of 10 kW pilot system, and 20 ppm of odorants such as tetra hydro-thiophene (THT) and tertiary butyl mercaptan (TBM) are included in the natural gas of Korea. The THT and TBM are desulfurized in hydrodesulfurization (HDS) reactor. The commercial Co–Mo/Al_2O_3 catalyst (ICI 41-6) is used in HDS reactor, which converts the sulfur compounds into H_2S. Then the H_2S is removed by sulfur absorption bed, which is packed with ZnO (ICI 32-4) catalyst. Then natural gas and steam are injected into the steam reforming reactor through an ejector. The commercial reforming catalysts (ICI 25-4, 57–4) are packed in the tubular type reactor. The reformed gas contains high level of CO, the high temperature shift reactor (HTS) and low temperature shift reactor (LTS) are installed after the steam reforming reactor. The CO content in the reformed gas is reduced to 0.3–0.6% after passing through these shift reactors. The anode off gas is recycled into the burner of steam reforming reactor and used as fuel in the operation.

The 10 kW PAFC stack is composed of 18 unit cells. The electrode area is 4225 cm^2 and four cooling plates are inserted into the 10 kW stack. The heat recovery system is composed of a heat exchanger and a steam separator, which supplies the steam into the reformer and supplies the cooling water into the stack. The 10 kW stack was operated at 185 °C and atmosphere pressure. The 222 mA/cm^2 of current density at 700 mV (0.15 W/cm^2) was obtained in the operation of 10 kW PAFC stack. It shows the performance of 10 kW stack is similar to that of commercial PAFC power plants [13].

In the development of 10 kW pilot system, we tried to design the pilot system with compactness. The total size of 10 kW pilot system including reformer, heat recovery system, 10 kW stack and demi-water supplying system is the 1500 mm × 3000 mm × 2500 mm ($W \times L \times H$). The size of the system is a critical factor for the development of commercial power plant and should be reduced into a small package. Some modification of 10 kW pilot system after initial operation is currently under development. It includes modification of the heat exchanger of the reformer to increase the heat efficiency and the change of internal structure of steam reformer to fully utilize the heat generated

Table 2
Target specification of 50 kW PAFC power generation system

Item	Target specification
Fuel	Natural gas
Reforming type	Steam reforming
Reformer type	Plate type reformer
Stack cell size (cm^2)	4225
Number of cells in stack	108
Cooling type	Water cooling
Target electric efficiency (LHV) (%)	40
Inverter efficiency (%)	83–87
Total size	4000 mm × 2500 mm × 2300 mm

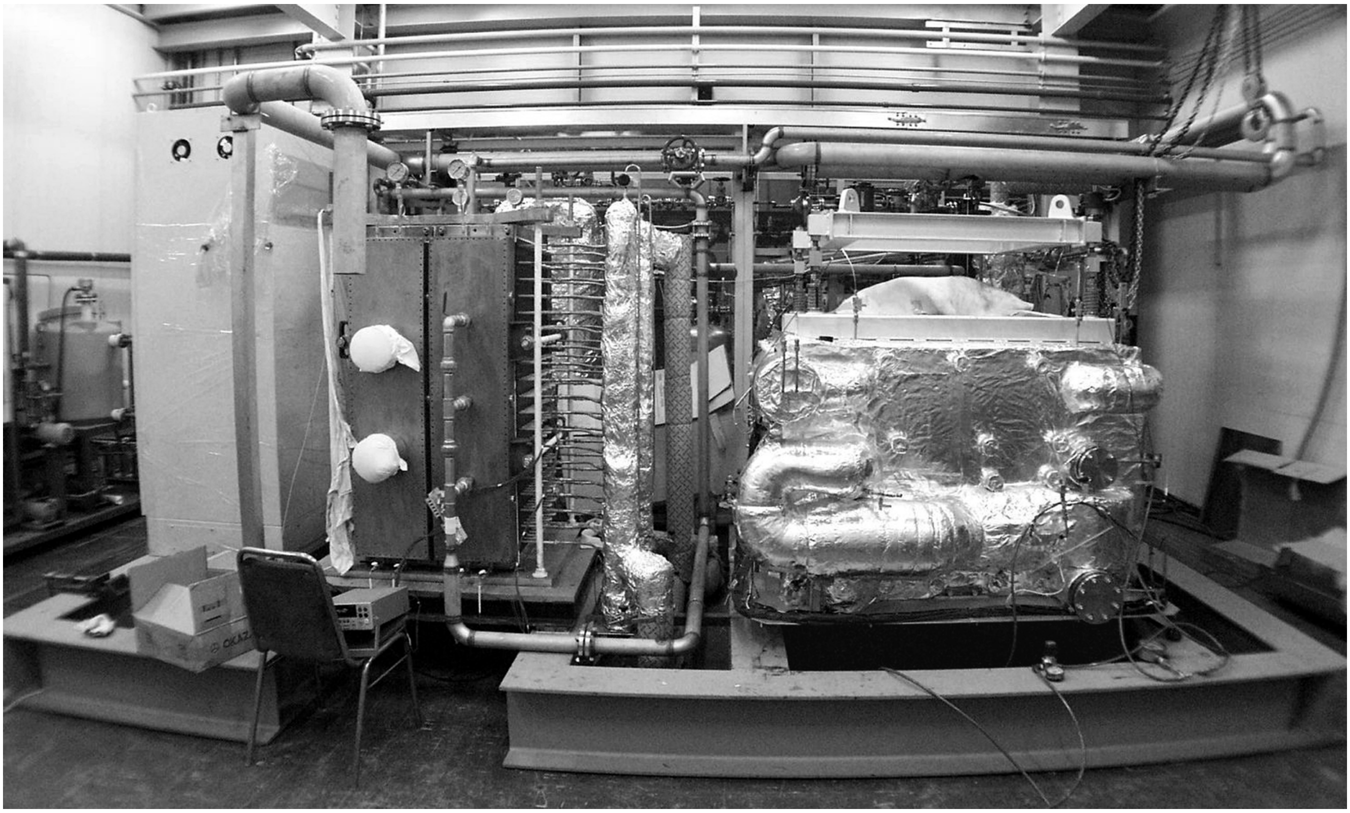

Fig. 7. The photograph of inner side of 50 kW PAFC power generation system.

by the burner. It will lead to a more efficient system compared with initial design and also gives the technical data for the integration of PAFC fuel cell system.

3.4. Development of 50 kW system

Table 2 shows the target specification of the 50 kW PAFC power generation system. The schematic design of the 50 kW power generation system is similar to the 10 kW pilot system (Fig. 6). It consists of reformer, stack, inverter and heat recovery system. Natural gas (13A) is used as fuel and the steam reforming reaction is applied in this system. In the 50 kW PAFC power generation system, the plate type reformer is applied. The plate type reformer has many advantages over the conventional tubular type reformer. It has a large heat transfer area and low pressure drop in the reforming reactor. It is also simple to control the temperature of the reformer due to the usage of catalytic combustor. The 50 kW stack is 108 unit cells with the cell size of 4225 cm^2. The inverter is installed to convert dc power generating from the stack to ac power. The efficiency of inverter is about 83–87% in the operation of 50 kW system.

Fig. 7 shows the photograph of the 50 kW PAFC power generation system. The total size of this system is the 4000 mm $\times$ 2500 mm $\times$ 2300 mm ($L \times W \times H$). In this size, all the components of 50 kW system are installed and assembled into a package. This system will be integrated in 2001 and long-run operation will be tested until the end of 2002. This system will be demonstrated in the R&D center of LG-Caltex Oil. The electric power generating from the 50 kW system will be supplied to the lighting of buildings and laboratories of our R&D center. This demonstration will give the stability and reliability data of the 50 kW system for commercial operation. The feasibility of the 50 kW system will also be analyzed for market introduction.

4. Conclusions

An overview of the 50 kW PAFC stack development and the 50 kW PAFC power generation system being developed by LG-Caltex Oil Corp was presented. The electrode prepared by automated manufacturing facilities gave high current density (400 mA/cm^2 at 0.65 V) and stability (3% degradation rate) over 6000 h of operation. The 50 kW PAFC stack showed 85% hydrogen utilization rate and 40% of electrical efficiency (LHV). The 10 kW pilot system and the 50 kW power generation system are being developed and will be demonstrated to evaluate the feasibility of these systems for commercial application.

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