

Development of kW Class SOFC Systems for Combined Heat and Power Units at KEPRI

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ABSTRACT

The Korea Electric Power Research Institute (KEPRI) has been developing planar solid oxide fuel cells (SOFCs) and power systems for combined heat and power (CHP) units. The R&D work includes solid oxide fuel cell (SOFC) materials investigation, design and fabrication of single cells and stacks, and kW class SOFC CHP system development. Anode supported cells composed of Ni-YSZ/FL/YSZ/LSCF were enlarged up to $15 \times 15 \text{ cm}^2$ and stacks were manufactured using $10 \times 10 \text{ cm}^2$ cells and metallic interconnects such as ferritic stainless steel. The first-generation system had a 37-cell stack and an autothermal reformer for use with city gas. The system showed maximum stack power of about $1.3 \text{ kW}_{\text{e,DC}}$ and was able to recover heat of $0.57 \sim 1.2 \text{ kW}_{\text{th}}$ depending on loaded current by making hot water. The second-generation system was composed of an improved 48-cell stack and a pre-reformer (or steam reformer). The thermal management subsystem design including heat exchangers and insulators was also improved. The second-generation system was successfully operated without any external heat source. Under self-sustainable operation conditions, the stack power was about $1.3 \text{ kW}_{\text{e,DC}}$ with hydrogen and $1.2 \text{ kW}_{\text{e,DC}}$ with city. The system also recuperated heat of about $1.1 \text{ kW}_{\text{th}}$ by making hot water. Recently KEPRI manufactured a 2 kW class SOFC stack and a system by scaling up the second-generation 1 kW system and will develop a 5 kW class CHP system by 2010.

Key words : SOFC, Combined heat and power units, Fuel reformer, Self-sustainable operation

1. Introduction

High temperature solid oxide fuel cells (SOFCs) offer high electricity conversion, superior environmental performance, combined heat and power (CHP), and fuel and size flexibility for transportation and stationary applications. In recent years, development of SOFCs has progressed remarkably and it is expected that SOFCs will replace conventional generators by providing highly efficient and environmentally friendly energy conversion. CHP applications for supplying residential energy needs have attracted particular interest. The market requirements for this application are increasing. Clearly, extension of the versatility of fuel usage and operation conditions (operation temperature and pressure) for SOFCs will be beneficial not only to the development of large scale systems but also for application to small systems.¹⁻³⁾

For a 1 kW-class SOFC system for CHP units, the Korea Electric Power Research Institute (KEPRI) has developed an anode-supported, planar type SOFC that can be easily mass-produced for commercialization by having cost-effective interconnects such as ferritic stainless steels. At higher temperatures, performance of SOFCs can be increased due to higher electrochemical activity of the electrodes and

lower ohmic losses, but the surfaces of the metallic interconnects at the cathode side are rapidly oxidized under such conditions. To improve the performance of anode-supported SOFCs, a thin film electrolyte fabrication was adopted and a functional layer (FL) is introduced on the Ni-YSZ anode substrate at the anode side. In addition, for efficient operation of the SOFC through higher electrode performance, alternative cathode materials consisting of LSCF instead of LSM were developed.

The developed 1 kW-class SOFC stack and CHP system showed $1.3 \text{ kW} \sim 1.6 \text{ kW}$ electric power. It also recovered heat of about 1 kW via generating hot water. As the second stage of this national project, the development of a 5 kW-class system for CHP is scheduled for completion by 2010. In the second stage, 5 kW-class SOFC modules and fuel (such as natural gas, LPG and diesel) reformers will be developed. The target for system efficiency is 35% and the target for total efficiency including heat recovery is 75% when using natural gas (NG).

2. Experimental Procedure

2.1. Cell Fabrication

For anode substrates, nickel oxide and yttria-stabilized zirconia powders ($\text{ZrO}_2 + 8\text{Y}_2\text{O}_3$, 8YSZ) were initially mixed and milled together at a weight ratio of 50:50. Then, 24 vol.% of graphite powder as a pore-former and organic binder was mixed into the initial powder mixture with ethyl alcohol, and the resulting mixture was dried in an oven. The

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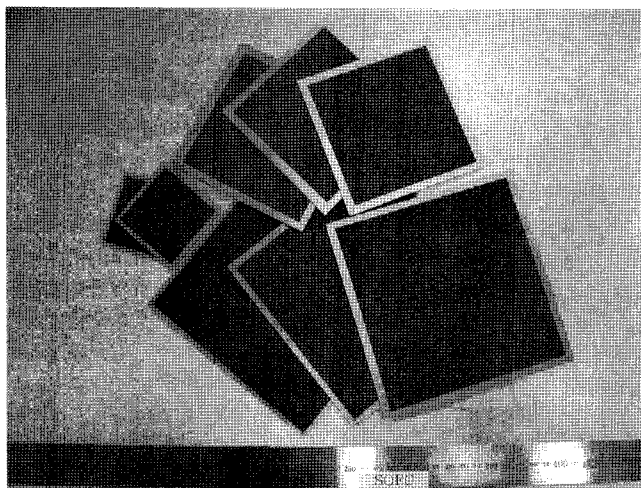


Fig. 1. Anode-supported cells at KEPRI (5×5 , 10×10 , 15×15 cm²).

NiO+YSZ powder mixture was pressed into a rectangular mold and pre-sintered at 1400°C for an hour to prepare a pre-sintered anode substrate. Thereafter, the YSZ was coated on the substrate by slurry coating and sintered at 1550°C for 2 h to form a dense electrolyte layer with a thickness of about 20 μ m.

The raw materials of the LSCF ($(\text{La}_{0.6}\text{Sr}_{0.4})(\text{Co}_{0.2}\text{Fe}_{0.8})\text{O}_3$) cathode were synthesized by the citrate method. The LSCF powder was calcined at 800°C for an hour and at 1000°C for 8 h. X-ray diffraction patterns were analyzed to confirm single perovskite phases in the LSCF. Each anode-supported single cell with composite cathode (LSCF+CSO(Sm-doped cerium oxide, $\text{Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{2.8}$)) was prepared on top of the fired YSZ thin electrolyte surface.

On the surface of the electrolyte (8YSZ) layer, the composite cathode mixtures of (LSCF+CSO) with a weight ratio of 50:50 were printed with a thickness of 40 μ m and heat-treated at 1100°C for 2 h.⁴⁾ Fig. 1 shows anode-supported cells.

2.2. Stack Preparation

Low temperature-melting borosilicate was used to seal the stack. Internal manifolds and channels were manufactured at the metallic interconnects. To prohibit undesired oxidation at the cathode side of the interconnects, LSM ($(\text{La}_{0.6}\text{Sr}_{0.4})\text{MnO}_3$) paste was coated on interconnect surfaces. Metallic (inconel and Ni) mesh was introduced for better electrical contact and current collection between interconnects and cells.

Stainless steel (STS 430) has several merits such as lower cost and easier manufacturing processes for interconnect material relative to other high temperature alloys. However, experience at KEPRI reveals that STS 430 shows low oxidation durability even at 650°C, especially for the cathode side. To overcome this problem, a composite paste of LSM was fabricated and coated on the cathode side of the STS 430 interconnects.

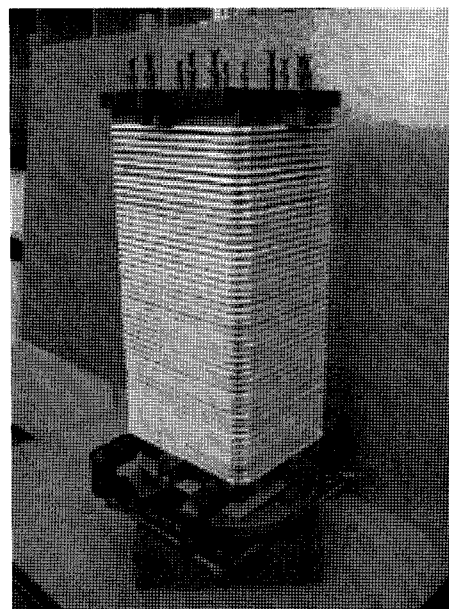


Fig. 2. 1 kW-class 48-cell SOFC stack.

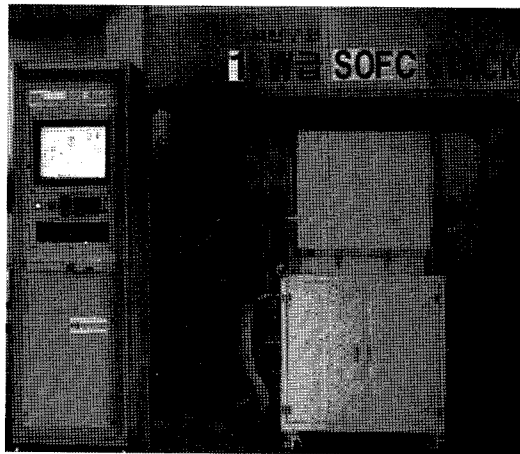
To make a SOFC stack module above 1 kW-class, a fabrication process to increase the cell area has been developed and a stack design has been studied. At present, a fabrication process for 15×15 cm² cells has been developed. However, in this work, 10×10 cm² cells were stacked with stainless steel interconnects.

2.3. System Integration & Operation

The 1 kW class SOFC system consisted of a stack, a stack room, a natural gas reformer, a desulfurizer, a catalytic combustor, a power conditioning system, heat exchangers, pumps, blowers and other equipment. KEPRI's first-generation 1 kW system contained a 37-cell stack but the second-generation system had a 48-cell stack of about $17 \times 14 \times 36$ cm³ that weighed 35 kg (see Fig. 2). The cell number in a stack was increased to enhance electric conversion efficiency by operating at a lower current. Increasing the cell number also offers advantages for the power conditioning process, which converts DC from the stack into AC and regulates voltage, due to higher stack voltage.

The SOFC stack was operated at high temperatures from 650°C to 800°C. A stack room was necessary to increase temperature during start-up and to thermally insulate the stack at high operating temperatures. The stack room was a cubic box made of ceramic board insulator and came down from the top of the stack. There was a heater (or burner) in the stack room to control the stack room temperature.

An external 1 kW-class city gas reformer that used porous Ni-Cr disc-type catalytic plate was developed in the first-generation SOFC system. In order to increase the electric efficiency of the second-generation system, a steam reformer was adopted. The second-generation system separated hot parts such as the stack, the reformer, the combustor and the heat exchangers from cold parts such as the blowers and



(a)



(b)

Fig. 3. 1 kW-class SOFC systems for demonstration, (a) the first generation, (b) the second generation.

pumps to aid system efficiency. Fig. 3 shows KEPRI's first and second-generation 1 kW-class SOFC systems.

The 1 kW-SOFC system was placed in a hood and proper pre-treatment was conducted before the test. City gas was used as fuel but its pressure was about 200 mm Aq. Therefore the system used the combination of a blower and a MFC (Mass Flow Controller) to overcome pressure drop and more accurately feed the fuel. The city gas was desulfurized and then converted into a hydrogen-rich gas in a reformer. Air and water were supplied to the reformer using a blower and a pump, respectively. The water was deionized and then evaporated through heat exchange with stack room gas. The reformat gas composition was analyzed by GC (Agilent 6890N) and then entered the SOFC stack. The air for the cathode was supplied using a blower.

Thermocouples were installed inside the stack room to measure and control stack temperature. Changing the loaded current to the stack with a DC loader, voltages of the

Table 1. Composition and Flow Rate of Reformat Gas, Water Free

Species	H ₂	N ₂	CO	CH ₄	CO ₂
Composition (mol %)	38.9	39.5	9.1	3.6	8.9
Flow rate (mol/min)	0.524	0.532	0.122	0.049	0.120

Table 2. Stack Performance under Self-sustainable Operation Conditions, Stack Current 35 A

Fuel	Voltage (V)	Power (kW)	Efficiency (%)
H ₂ 14.9 L/min	38.2	1.34	54
City gas 5.12 L/min	35.9	1.26	40

stack and each cell were measured. To calculate the recuperated heat, water temperatures at the inlet and outlet of the hot water maker and water flow rates were monitored. It was necessary to minimize parasitic electric power (such as the power electronics, pumps and blowers) to improve the system efficiency. Therefore, the total parasitic power consumed for system operation was monitored. The system was equipped with some thermocouples and pressure gauges to check operating status.

3. Results and Discussion

3.1. Performance of the first-generation system

The composition of reformat gas was analyzed before it entered the stack. To avoid water damage to the GC columns, water was removed from the reformat gas. Table 1 shows the results of the composition analysis. For the 37-cell stack, H₂ of 0.460 mol/min was necessary to generate 40 A and H₂ of 0.575 mol/min was necessary to generate 50 A. From the experimental result, the total amount of H₂ and CO in the reformat gas was 0.646 mol/min. The fuel utilization for the 40 A loaded condition was about 0.7 and about 0.9 for the 50 A condition. Besides H₂ and CO, there was CH₄ of 0.049 mol/min and it could also be used as a fuel via internal reforming inside an anode under that condition (sufficient water and temperature over 700°C). Considering those results, sufficient fuels were supplied to load a current up to 50 A.

When the system was operated with city gas, the stack performance was measured at different temperatures and currents. Fig. 4 shows the stack performance according to loaded current at 750°C. Stack voltage was about 28.3 V at 40 A and about 26.0 V at 50 A.

The 1 kW class SOFC system was developed as a CHP unit for higher efficiency residential applications. The unreacted anode off-gas was burned in a catalytic combustor with cathode off-air. Then the high temperature combustion gas exchanged heat with water to generate useful hot water. The amount of recuperated heat was 0.57 to 1.2 kW_{th} according to the loaded current.⁵⁾

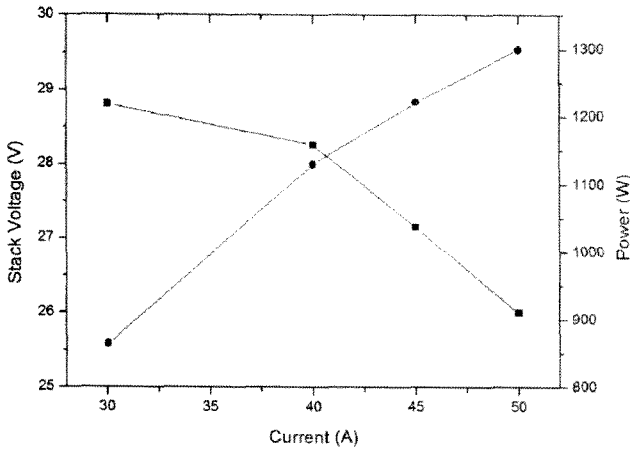


Fig. 4. I-V-P characteristic curve of 37-cell stack at 750°C, operated with city gas.

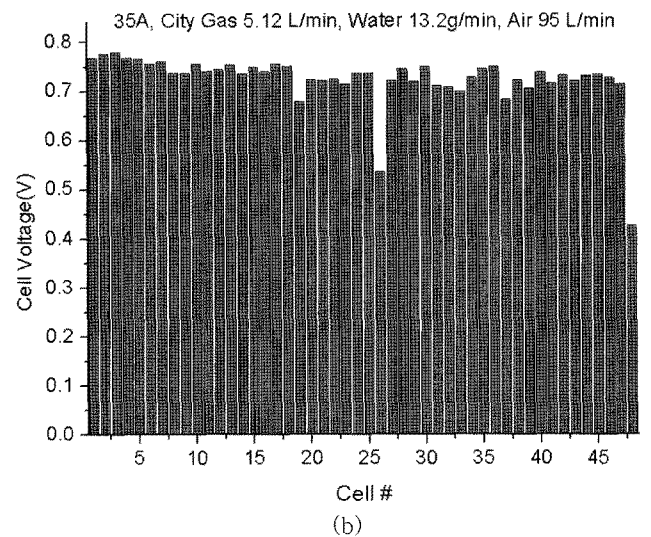
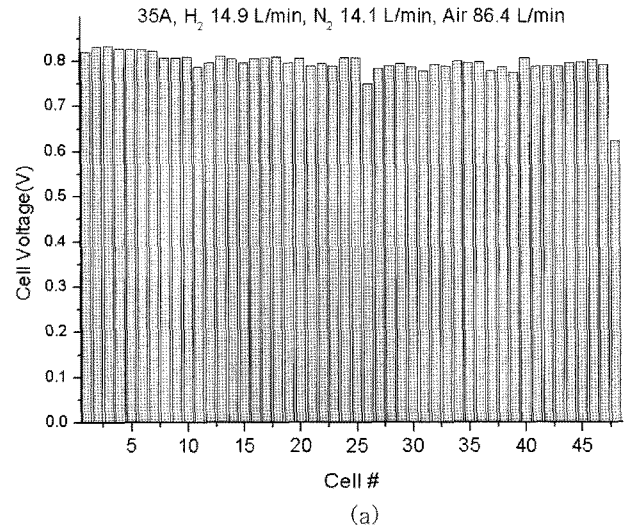


Fig. 6. Voltage distribution of each cell under self-sustainable operation condition, (a) with hydrogen, (b) with city gas.

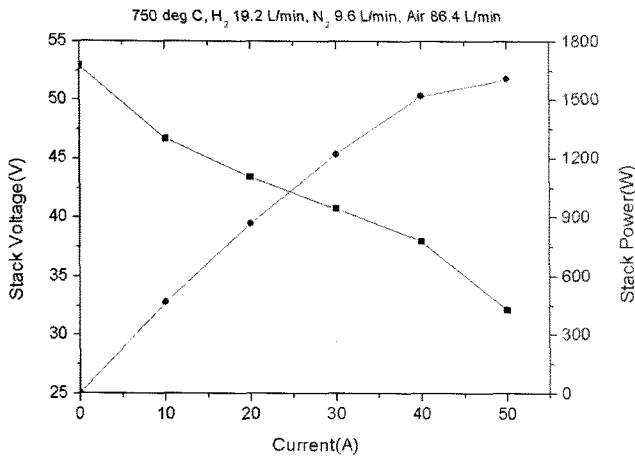


Fig. 5. I-V-P characteristic curve of 48-cell stack at 750°C, H₂ 19.2 l/min, N₂ 9.6 l/min, air 86.4 l/min.

3.2. Performance of the second-generation system

The second-generation 1 kW class SOFC system consisted of a hot box section, a cold balance of plant (BOP) section and a water reservoir. The hot box part contained a SOFC stack made up of 48 cells and ferritic stainless steel interconnectors, a fuel reformer, a catalytic combustor and heat exchangers. The thermal management and insulation system were especially designed for self-sustainable operation. The cold BOP section was composed of blowers, pumps, a water trap and system control units. When the 1 kW class SOFC system was operated at 750°C using hydrogen, the stack power was 1.2 kW at 30 A and 1.6 kW at 50 A (see Fig. 5). The SOFC system was then operated using hydrogen and city gas without any external heat source.

Under self-sustainable operation conditions, the stack power was about 1.3 kW_{DC} with hydrogen at 35 A. The fuel utilization for the 35A loaded condition was about 84% and the stack efficiency was about 52%~54%. For the self-sustainable operation condition using city gas, the stack power was about 1.2 kW_{DC} at 35 A and the stack efficiency was

about 40%. Fig. 6 shows each cell voltage in the stack for the self-sustainable operation conditions. The system also recuperated heat of about 1.1 kW_{th} by making hot water.

The dissipated power from the SOFC system equipped with a city gas (or natural gas) reformer was higher than anticipated because the BOPs were not optimized. This explains why the target power conversion efficiency of 35%(LHV) for the SOFC system was not approached. The SOFC system efficiency was low because of losses at the reformer and in the power conditioning system. In the next generation system, the balance of plant will be modified to improve thermal management. The target of the next system will be to minimize the parasitic power of the RPG system to blow 15% by improving BOPs and thermal management.

4. Conclusions

Anode-supported planar-type SOFCs have been studied and developed using high-efficiency cells and 1 kW-class stacks built with 48 cells and ferritic stainless steel (STS430). In addition, a 1 kW-class SOFC demonstration system for a CHP unit was operated at 650°C~800°C with hydrogen and city gas. The demonstration system showed 1.3 kW_{DC} electrical power at 35 A and 1.6 kW_{DC} at 50 A. To increase the efficiency of the SOFC stack and system, a 1 kW-class stack was built with a 48-cell stack and facilities for steam reforming. Fuel utilization for the 48-cell stack under the 35 A loaded condition was 75% and the electrical efficiency of stack was about 50% (LHV) with hydrogen fuel.

Further research will be conducted to achieve higher performance of KEPRI's SOFC system. The next stage of the development program, a national project for a 5 kW-class SOFC system to operate on fuels such as natural gas and diesel, will be undertaken in future research. The main target of the program is to achieve total efficiency, including heat recovery, of more than 75% (electric efficiency(AC) is more than 35%, LHV) using natural gas and obtain long-term durability of the system by 2010.

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