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Development of disc-type intermediate-temperature solid oxide fuel cell

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Abstract

The Kansai Electric Power Co., Inc. (KEPCO) and Mitsubishi Materials Corporation (MMC) have been jointly developing disc-type intermediate-temperature solid oxide fuel cells (IT-SOFCs) since 2001. Operation temperatures between 600 and 800 °C have been set as the target. These temperatures enable SOFCs to use inexpensive metallic separators for cell-stacking and are sufficiently high for steam reforming of the hydrocarbon fuel. A 1 kW-class demonstration power generation system with a module that had a steam-reforming function was developed in 2003. The third generation module and the fourth generation module were developed in 2004. The third generation module was integrated into the system for various tests including long-term stability. The fourth generation module was examined for performance evaluation. The system included an SOFC module, a dc–ac inverter, a desulfurizer, a heat-recovery unit, and an automatic control device. The long-term stability test for over 1000 h gave no decrease in stack voltage. In parallel, the long-term stability test of a single cell stack unit was continued for over 10,000 h. In addition, hot water at 90 °C was obtained by the heat-recovery unit using high temperatures off-gas from the SOFC module. The newly developed fourth generation module gave an electrical efficiency of 60% (dc/lower heating value) using town gas as a fuel at an average stack temperatures of about 750 °C. A 10 kW-class SOFC co-generation system has been under development in a New Energy and Industrial Technology Development Organization project since 2004.

Keywords: Lanihanum gallate; Ni-SDC; Intermediate-temperature solid oxide fuel cell; Power-generation module; System

1. Introduction

The solid fuel cell (SOFC) is expected to give a higher efficiency than any other types of fuel cell. In addition, the SOFC has the advantages of low pollution and fuel flexibility. Since a SOFCs are usually operated at high temperature near 1000 °C. They have some weak points such as the limited choice of materials for the stack components, materials stability and high manufacturing cost. Recently, reduction of the operation temperature of SOFC has been drawing a great deal of attention because it gives several advantages, namely, a wider choice of low-cost and high-performance component materials, higher stability, reduced degradation and increased freedom for structural design. The Kansai Electric Power Co., Inc. (KEPCO) and Mitsubishi Materials Corporation (MMC) have been jointly developing disc-type intermediate-temperature solid oxide fuel cells (IT-SOFCs) since 2001. An operation temperature range between 600 and 800 °C has been set as the target because it enables the SOFC to use stainless-steel separators and to reform town gas internally. There are two approaches to the development of IT-SOFCs. One is to use a very thin YSZ membrane to renders the ohmic loss due to the electrolyte as small as possible [1–2]. Another is to use new electrolyte materials that show high oxide-ion conductivity at temperatures below 800 °C [3–4], comparable to with that of YSZ at 1000 °C. It has been reported that Co-doped lanthanum gallate compounds $La(Sr)Ga(Mg,Co)O_{3-\delta}$ possess excellent oxide-ion conductivity at intermediate temperatures [5–7]. KEPCO and MMC have developed a high-performance cell which uses this electrolyte. Previously, there have been three generations of module developed. The first-generation was a 1 kW-class module with 25 cells of 154 mm in diameter and an electrical efficiency of 43% (dc/lower heating value (LHV)) was obtained using

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hydrogen [8]. A second-generation 1 kW-class module with 41 cells of 120 mm in diameter was fabricated, and thermal self-sustainability was confirmed using methane as the fuel [9]. A third-generation 1 kW-class module with 46 cells (120 mm in diameter) was constructed, from which the 1 kW-class demonstration system was developed, as described later. The demonstration system was comprised of a module, controller, inverter, and heat-recovery unit. An electrical efficiency of 48% (ac/LHV, auxiliaries loss excluded) was obtained using town gas [10].

Recent work by KEPCO and MMC is described in this paper. After evaluation of the initial performance of the demonstration system, a long-term stability test was conducted for over 2000 h. A heat-recovery evaluation was carried out during the stability test. In parallel with the stability test, a long-term test of a single cell stack unit was performed for over 10,000 h.

Further improvement in the fundamental design of the module gave rise to fourth-generation module, and the module attained the electrical efficiency of 60% [dc/LHV].

Development of a 10 kW-class SOFC system started in the New Energy and Industrial Technology Development Organization (NEDO) project, 'Development of Solid Oxide Fuel Cell System Technology', in 2004. A scaled-down test module has been constructed and attained the electric power output of 6.25 kW.

2. Cells, modules and demonstration system

2.1. High performance cell

A photograph of the electrolyte-supported disc-type planar cells is shown in Fig. 1. The planar cells are fabricated using a highly-conductive lanthanum gallate-based electrolyte, $La(Sr)Ga(Mg,Co)O_{3-\delta}$ a $Ni\text{-}Ce(Sm)O_{2-\delta}$ cermet anode, and a $Sm(Sr)CoO_{3-\delta}$ cathode. The thickness of the electrolyte is 200 μm and the thickness of both electrodes range between 30 and 50 μm. The diameter of cell is 120 mm.

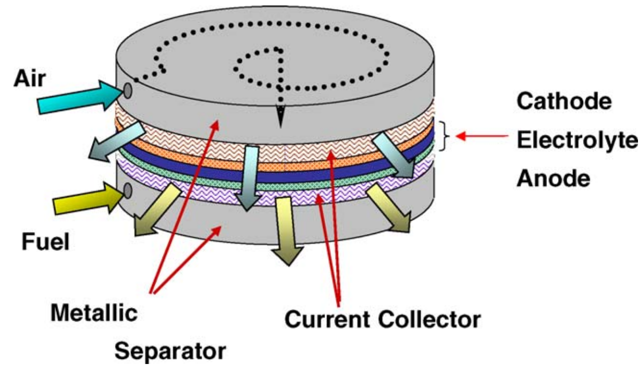

Fig. 2. Schematic diagram of single-cell stack unit.

2.2. Single-cell, stack unit

A schematic diagram of a single cell stack unit is given in Fig. 2. The cell is sandwiched between metallic separators and has a seal-less structure which is a notable feature. Fuel and air are supplied to the cell from individual central holes through a spiral channel within the stainless-steel separator. Gas distribution over each electrode is maintained uniformly via structure-controlled current-collectors that are placed between the cell and the separator. The remaining fuel after the fuel cell reaction is burned around the stack to supply additional heat to maintain the cell stack temperature and for the steam reforming of town gas.

2.3. 1 kW-class demonstration co-generation system

The components of the 1 kW-class demonstration co-generation system, are shown in Fig. 3, and a diagram of

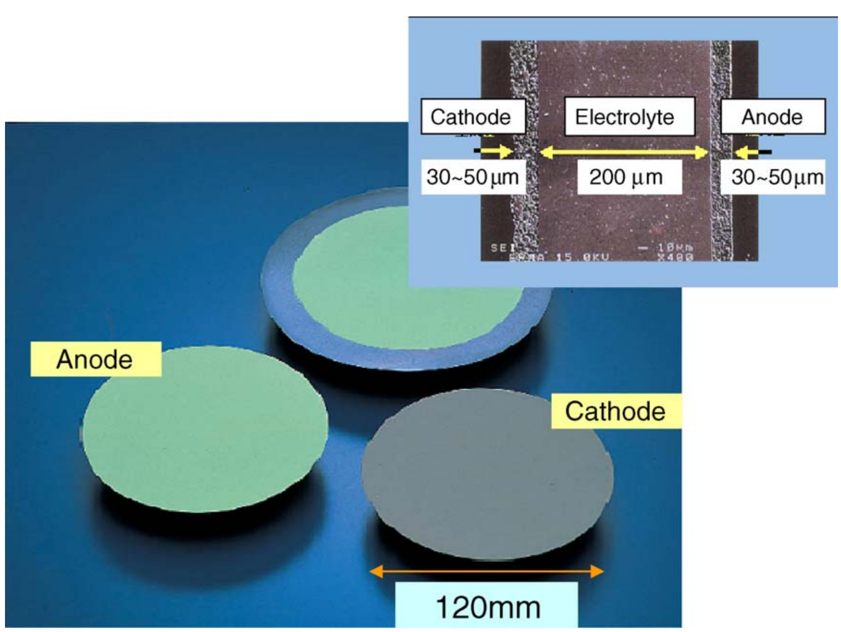

Fig. 1. Photograph of electrolyte-supported disc-type planar cells.

Fig. 3. Features of 1 kW-class demonstration co-generation system.

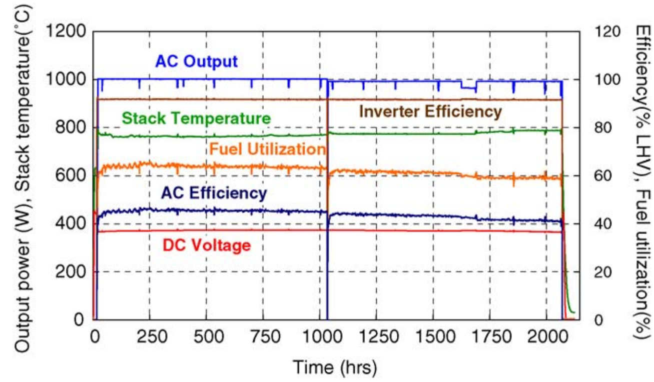

Fig. 5. Change of characteristics during the 2000 h of operation.

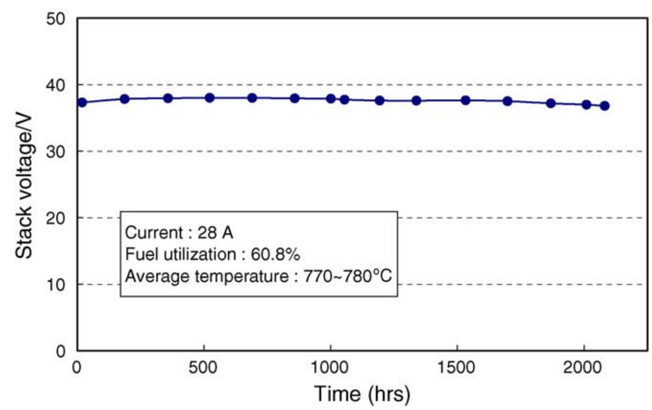

Fig. 6. Stack voltage during endurance testing.

the system is given in Fig. 4. The system contains the third-generation module that has a stack of 46 cells of 120 mm diameter, a controller unit with a function for automatic operation (i.e., start-up mode, power-generation mode, hot-standby mode, shut-down mode, and emergency stop), a power-conditioner unit with both an ac–dc inverter and a dc–ac converter, and a heat-recovery unit which makes hot water between 60 and 90 °C from the exhaust gas of the SOFC module. Air, town gas and water are fed to the module. Town gas is supplied through a desulfurizer and water is supplied through a purifier. A steam generator and a pre-reformer are contained within the module in order to generate hydrogen from town gas by using the joule heat of the stack and the combustion heat of the remaining fuel.

3. Long-term stability test

3.1. Long-term stability of 1 kW-class system

Long-term operation of a 1 kW-class system was carried out from November 2004 to March 2005. The change in fuel cell characteristics during 2000 h operation are shown in Fig. 5. The fuel was town gas ‘13A’, which is the condition used for practical application. One short interruption occurred after 1000 h of operation but the system was restarted immediately. The electrical efficiency was higher than 45% [ac/LHV] after 1000 h of operation. In order to evaluate the performance of the module, the fuel flow rate and dc current were periodically set to the prescribed values and the dc voltage was measured (Fig. 6). There is no degradation in stack voltage during the 1000 h of operation with an ac output of 1 kW. The degradation ratio was about 0.5%/1000 h after 2000 h of operation. The overall operation

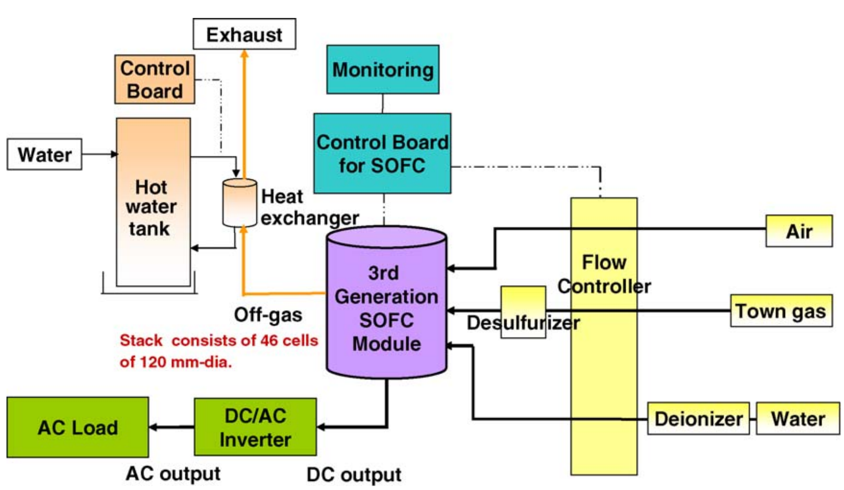

Fig. 4. System diagram of 1 kW-class system.

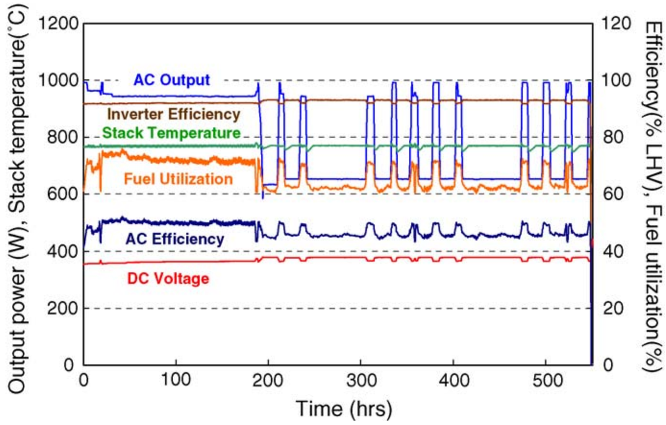

Fig. 7. Characteristics during load change operation.

period was 2084 h. Next, the system was restarted after some cells were replaced by the new ones. The change in the characteristics during the next operation is given in Fig. 7. The operation was performed for 550 h and an electrical efficiency of 51% [ac/LHV] was obtained with an ac power of 992 W a fuel utilization of 75%, and an average stack temperature of 766 °C. A load change between 650 and 1000 W (ac) was repeated 11 times during the period. There was no noticeable degradation in stack voltage. The problems involved in achieving an improvement in performance were defined through these stability tests.

3.2. *Long-term stability of a single cell stack unit*

The terminal voltage during a long-term stability test of a single-cell stack unit is shown in Fig. 8. The stability test was continued for over 10,000 h at a H_2 flow rate of $3\,ml\,min^{-1}\,cm^{-2}$. No decrease in the terminal voltage is observed until 2000 h. Afterwards, degradation ratio is 1 ∼ 2%/1000 h. The sudden change in voltage at around 9000 h is due to a black-out of the power supply. Good stability of the stack unit during 10,000 h of operation is obtained. Nevertheless, further improvement in stack stability is needed since the performance target is 40,000 h.

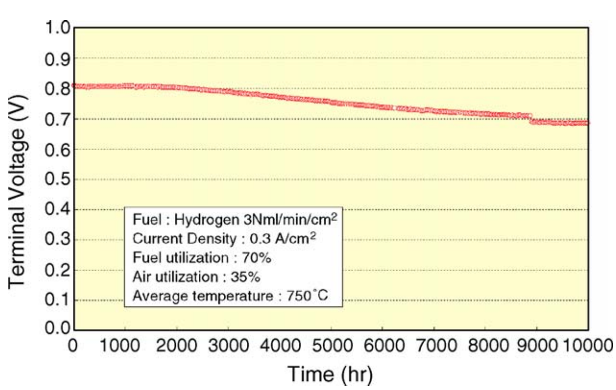

Fig. 8. Terminal voltage during long-term stability testing of a single-cell unit.

Table 1
Results of the heat-recovery test

Temperature of hot water (°C)	Overall efficiency (%, LHV)[a]		
	FY 2003	FY 2004	
	Test model[b]	Improved model[b]	Advanced-plate type
90	55 [44]	73 [45]	77 [44]
60	80 [44]	88 [46]	85 [44]
Volume (L)	1.67	1.53	0.3

Cold water temperature at heat exchanger inlet: 13–19 °C; [ ]: electrical efficiency at heat recovery test (%, LHV).
[a] Overall efficiency = electrical efficiency + heat recovery efficiency. The temperature of off-gas: FY 2003—155°C, FY 2004—250 °C.
[b] Plate type.

4. Heat recovery of 1 kW-class system

The system diagram of the heat recovery unit, which contains a heat exchanger, a 370 L hot water tank and controller of water flow is shown in Fig. 9. The unit can make hot water at 60–90 °C by utilizing the high-temperature exhaust gas of the SOFC module. The temperature of the exhaust gas is about 250 °C. The heat-exchanger transfers heat energy from the exhaust gas of the SOFC module to water. The heat energy utilized includes not only sensible heat but also latent heat. The overall efficiency of the heat recovery test and the volume of the heat-exchanger is shown in Table 1. The heat recovery was carried out at the rated output operation in FY 2003 and FY 2004. The test and the improved models in Table 1 were plate-type heat-exchangers. Data for a new type of heat exchanger, the advanced-plate type, which has a smaller size is also given in Table 1. On obtaining 90 °C hot water, the overall efficiency was higher than 70% [LHV], and the overall efficiency was higher than 85% [LHV] for 60 °C hot water. Those results clearly show that the intermediate-temperature SOFC system can produce hot water with high efficiency. The information gathered heart recovery test will be reflected in the development of several 10 kW-class co-generation system.

5. Fourth-generation 1 kW-class module

In parallel with the 1 kW-class system using the third-generation module, a fourth-generation module was developed. This had an improved temperature profile and a better module heat-flux distribution within the module; it is built for cost reduction and downsizing. Both types of module are shown in Fig. 10. The third-generation module is equipped with an external manifold, a larger sized pre-reformer, etc. On the other hand, the fourth-generation module has an internal manifold, a smaller sized pre-reformer, etc. The number of parts and assembly time for the internal manifold are both less than those for the external manifold. The pre-reformer of the fourth-generation module has a simpler structure and the reforming efficiency is higher. The performance of the fourth-generation module is shown in Fig. 11. A dc power output of 1146 W was recorded under thermally self-sustained conditions against the target output of 1120 W. In addition, an electrical efficiency of 60% [dc/LHV]

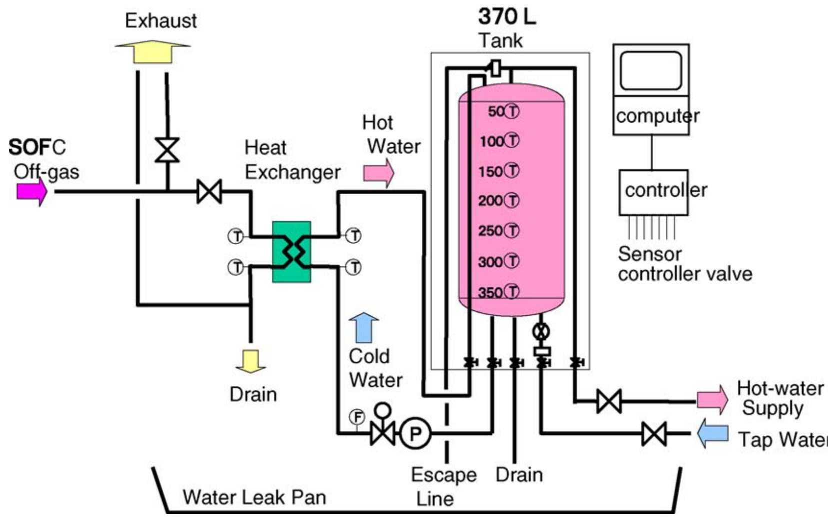

Fig. 9. System diagram of heat-recovery unit.

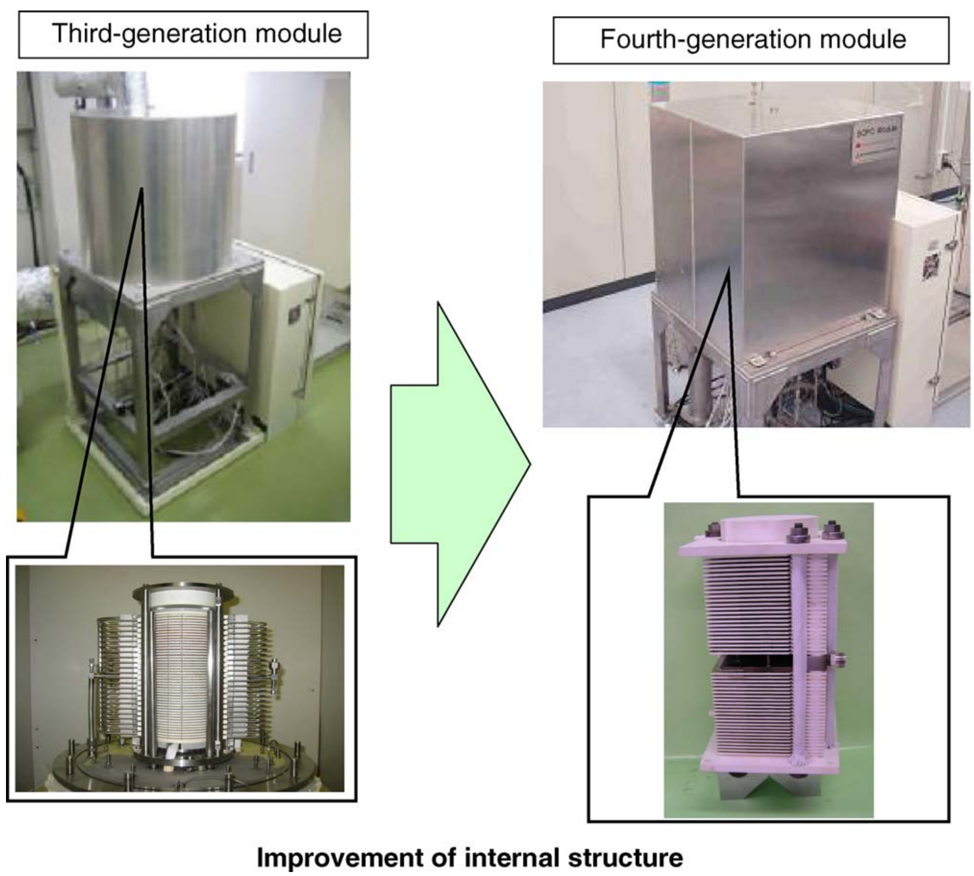

Fig. 10. Comparison of third-generation and fourth-generation modules.

was obtained with town gas as a fuel. In this case, fuel utilization was 80% and the average temperature of the stack was about 750 °C and the steam:carbon ratio was 3.4.

6. Development of several 10 kW-class co-generation system in NEDO project

For initial commercialization in 2008, NEDO setup the SOFC system development (several 10 kW- to 100 kW-class) project from FY 2004 to FY 2007. The objective is to evaluate the durability and reliability of the co-generation system. MMC and KEPCO have been jointly developing several 10 kW-class co-generation system based on disc-type intermediate-temperature SOFCs as one part of the NEDO project. The target of development is demonstration of highly efficient, inexpensive and compact 10 kW-class SOFC co-generation system. In addition, the conceptual design of several 10 kW-class SOFC system is planned with based on the 10 kW-class system results. A

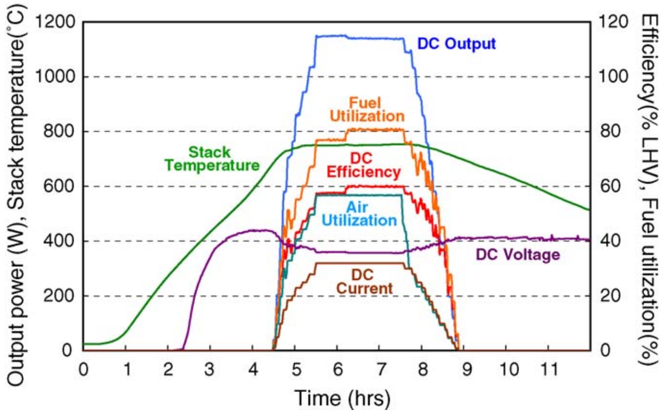

Fig. 11. Performance of fourth-generation 1 kW-class module.

schematic illustration of the 10 kW-class module is shown in Fig. 12. The module is composed of multi-stacks (16 stacks: 2 rows × 2 lines × 4 stages) each with a starting burner, pre-reformer, heat-exchanger and thermal insulator. The stack is equipped with an internal manifold, which was developed in the design of the fourth-generation 1 kW-class module. The 10-kW-class module will be integrated into a co-generation system. MMC designs and fabricates the module, while KEPCO constructs the system in cooperation with the Daihen Corporation. The performance target by the end of FY 2007 is given in Table 2. The target electrical efficiency is more than 40% [ac/HHV], and that for the overall efficiency is more than 80% [LHV]. A voltage degradation ratio lower than 0.25%/1000 h during an operation of 3000 h is also targeted. The detailed design is planned by use of numerical simulation. The schedule for development is as follows: (i) FY 2004: basic design of a module; (ii) FY 2005: detailed design, manufacturing and performance testing of the demonstration module and basic design of the systems; (iii) FY 2006: detailed design and manufacturing of the co-generation system with the improved module; (iv) FY 2007: field testing and conceptual design of several 10 kW-class systems. A scaled-down test module was fabricated for the preliminary evaluation of the 10 kW-class module, as shown in Fig. 13. The module is composed of 8 stacks (2 rows × 2 lines × 2 stages), a starting heater, etc. The module has been operated twice in FY 2005. The targeted dc power output of 6.25 kW was obtained, and basic data was collected to assess the support structure of the stack, the thermal performance of the components in the module, and the distribution of gas. The tasks for further improvement will be based on, the test and investigation results and will be reflected in the detailed design of the module for the system.

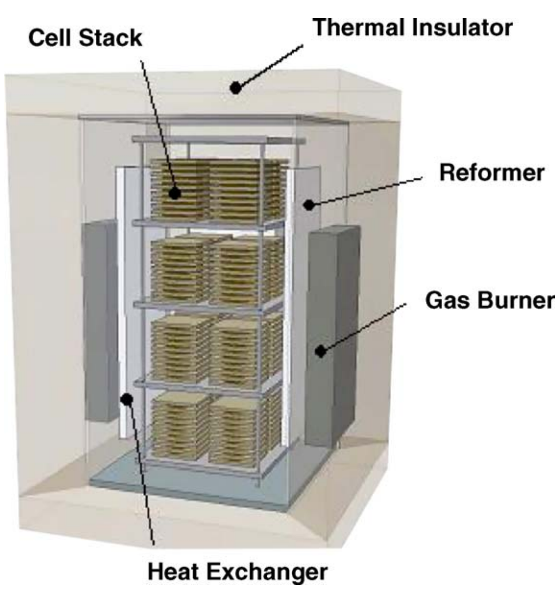

Fig. 12. Schematic of 10 kW-class module.

Table 2
Performance targets in NEDO project by the end of FY 2007

	Target
Power output	10 kW-dass (net ac)
Electrical efficiency	More than 40% (net ac/HHV) (at the rated output)
Overall efficiency	More than 80% (HHV) (with 60 °C hot water)
Long-term stability	Voltage degradation ratio 0.25%/1000 h (at the rated conditions >3000 h)

Fig. 13. Exterior view of scaled-down module.

7. Conclusions

(1) A 1 kW-class demonstration co-generation system was operated with a stable output for 2000 h. A long-term sta-

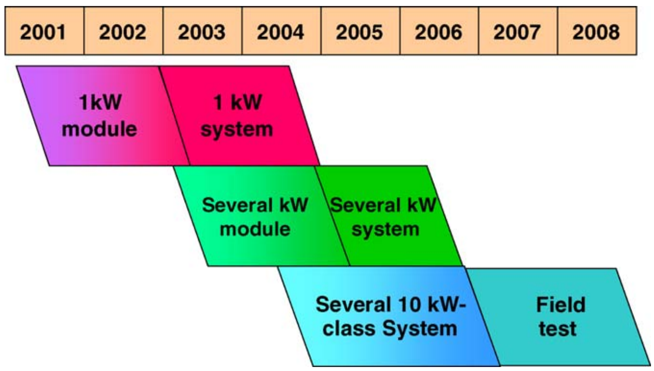

Fig. 14. Schedule of development.

bility test for over 1000 h gave no decrease in stack voltage. The tasks for improvement were defined through these stability tests.

(2) The long-term stability test of a single-cell stack unit was continued for over 10,000 h. No decrease in terminal voltage was observed over 2000 h and the degradation ratio was (1–2%/l000 h for operation up to 10,000 h. The various degradation modes are becoming clear and will be resolved in the near future.

(3) Hot water of 90 °C was obtained by the heat-recovery unit using the high-temperature off-gas from the SOFC module. The overall efficiency was above 70%. It was clearly demonstrated that the intermediate-temperature SOFC system could produce hot water with high efficiency.

(4) The electrical efficiency of the fourth-generation module with an internal manifold was 60% (dc/LHV), using town gas as the fuel at an average stack temperature of about 750 °C.

(5) MMC and KEPCO have been jointly developing several 10 kW-class co-generation system based on a disc-type intermediate-temperature SOFC as one scheme in a NEDO project since 2004.

(6) The first milestone will be completion of the development of several 10 kW-class SOFC system by the end of FY 2006, as shown in Fig. 14.

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