Progress towards realizing distributed power generation with highly efficient SOFC systems

— Development and standardization of performance evaluation methods targeting early market entry of SOFC systems —

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Solid oxide fuel cells (SOFC) are expected to serve as the power source for highly efficient distributed power generation systems. To achieve early market entry and fair competition, it is essential that performance evaluation methods for SOFC be established and standardized. For this purpose, we have developed high-precision performance evaluation methods and test apparatuses to measure the power generation performance of SOFC cells, stacks, and systems. These methods were achieved by a combination of elemental technologies and tools, such as commercial measuring instruments, precision flowmeters and reference materials that are traceable to national standards, and catalysis technology. We evaluated the performance and usefulness of the test apparatuses developed in cooperation with SOFC manufacturers, and presented our standards activities, focusing mainly on the preparation of a Japanese Industrial Standard (JIS) for electrical efficiency test methods.

Keywords : Solid Oxide Fuel Cell, SOFC, highly efficient distributed power generation, performance evaluation methods, standardization

1 Introduction

The 3.11 Great East Japan Earthquake has prompted a debate on alternative ways to secure a stable supply of electricity and other forms of energy in Japan. One of the most urgent issues facing the nation is the need to reduce the country's overdependence on large-scale central power stations constructed in coastal areas. Among the proposals made, the Smart Community project is attracting increasing attention as a means to achieve efficient use of energy by installing small distributed generation systems near high-energy demand areas.[1][2] Renewable energy sources such as solar energy are expected to play the key role in the Smart Community; however, photovoltaic systems are not always able to provide the necessary electricity at night and in some weather conditions. While electricity storage systems are deployed to solve this issue,[3] high-efficiency distributed power generation systems using fuels are also considered an effective alternative.

Recently, a 500,000 kW-class combined-cycle system using natural gas achieved an electrical efficiency of 60 %, and its installation has begun in a coastal area near major cities where there is heavy demand.[4] For small-to-medium cities in the interior regions, transmission loss is a serious issue given their long distance from thermal power stations in the coastal areas. A high-efficiency power system of no more than a few 10,000 kW is therefore more appropriate for these regions.

Fuel cells have long been a promising candidate to provide power to high-efficiency distributed power generation systems because of their ability to convert fuel's chemical energy directly into electricity. Since the turn of the century in particular, research and development efforts have increasingly focused on the polymer electrolyte fuel cell (PEFC) operating around 80 ºC and the solid oxide fuel cell (SOFC) operating in the range of 700 to 1000 ºC. As Fig. 1 shows, no systems other than stand-alone SOFC systems and combined systems integrating an SOFC and a gas turbine can achieve an electrical efficiency of 50 % or higher when generating power in the range of 1 to 10,000 kW. Ultimately, SOFC systems are expected to achieve an electrical efficiency of 70 %.[5]

As will be discussed below, an SOFC system comprises a stack, which serves as the power generation unit and is formed by stacking multiple single cells (the smallest units) together, and the balance of plant (BOP) such as flow controllers. Hence, in conducting research and development for SOFC, it is important to accurately evaluate and improve the performance of each of the single cell, stack, and system.

In Japan, as part of the Demonstrative Research Project on Solid Oxide Fuel Cell Systems conducted by the New Energy Foundation (NEF) and the New Energy and Industrial Technology Development Organization (NEDO), a 700-W residential system using an SOFC demonstrated an electrical

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efficiency of 45 %.[6] In October 2011, JX Nippon Oil & Energy Corporation became the first company in the world to market a cogeneration system using city gas or liquefied petroleum gas (LPG) with an electric output of 700 W. Recently, the development of commercial SOFC systems of 5 kW to 250 kW has begun, and there is also a plan to develop a triple combined-cycle system integrating a gas turbine and a steam turbine (targets: capacity of up to 800 MW and electrical efficiency of 60 % or higher).[5] Overseas, a 2-kW residential system has achieved an electrical efficiency of 60 %.

Incidentally, it is recently common to express the electrical efficiencies of SOFC systems and thermal power stations using the lower heating values (LHV) (i.e. the latent heat of vaporization is not included), and the efficiencies described so far in this paper are expressed using the LHV standard. The same will be used below unless it is noted that a high heating value (HHV) is used. In the cases of city gas and natural gas, efficiencies based on the LHV standard are 11 % higher than those based on the HHV standard.

With assistance from the Ministry of Economy, Trade and Industry (METI) and NEDO, our group has been engaged in the research and development of high-precision performance evaluation methods since around 2001 for all SOFC units from the smallest component to the complete product, namely the single cell, stack, and system. The objectives are to facilitate the early market entry of SOFC systems by assisting the research efforts in the private sector and to establish the standards for performance testing methods that are essential for promoting fair competition among manufacturers of SOFC systems once they become available for sale.

Recent IEC and ISO standards recommend that the concept of uncertainty replace error, accuracy, and other concepts as the indication of reliability of measurements.[7] Given this trend, we have collaborated with the Fluid Flow Division of the National Institute of Advanced Industrial Science and Technology (AIST), private corporations, and others to ensure that the performance measurement methods are traceable to national standards. Uncertainty measurements are estimated as the range of population means with a standard confidence level of 95 % based on the mean of observed values (measurement values) and the variability of all parameters, assuming that the true value is unknown.[8] For further details on uncertainty, see reference material [7].

This paper reports on our research and development activities for SOFC single cell/stack performance evaluation methods, which are essential for the development of SOFC systems, as well as our research/development and standardization work for the measurement methods of SOFC system electrical efficiency.

2 Research objectives and approaches aimed at early market entry of SOFC

2.1 Overview of SOFC systems and research objectives

The main components of an SOFC system, the complete SOFC product, include flow controllers, an evaporator (water vapor generator), a reformer, an SOFC stack, and an inverter, as shown in the schematic in Fig. 2. The following ingredients are supplied to the system: a raw fuel for the anode such as city gas, an oxidizing agent (water, oxygen) for the reforming of the raw fuel at the reformer, and air for the cathode. Reforming converts the raw fuel into hydrogen, carbon monoxide, methane, water vapor, and carbon dioxide, which are then distributed as anode gas to the anode of each cell forming the stack.

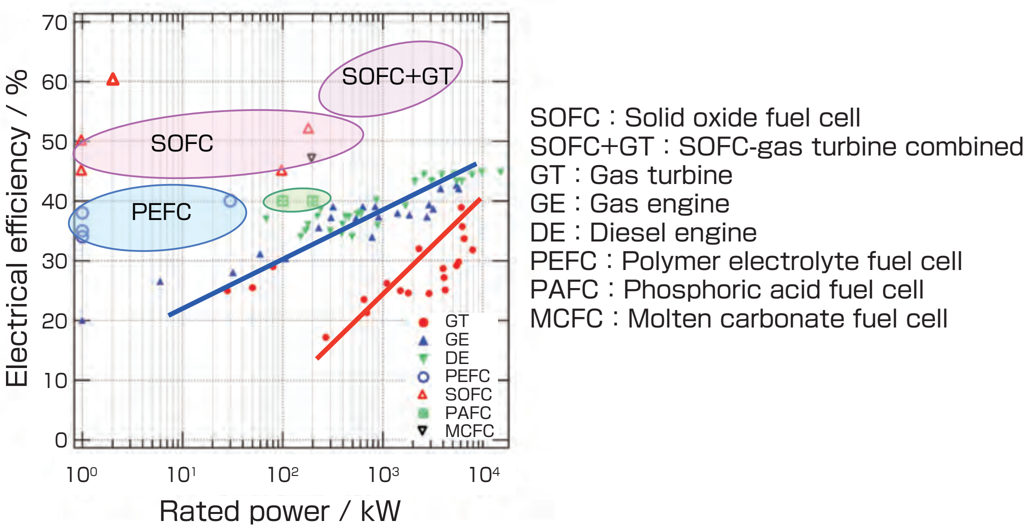

Fig. 1 Rated output and electrical efficiency of small power generation systems

At the anode, hydrogen and carbon monoxide react with oxygen ions (O^{2-}), which migrate from the cathode through the electrolyte. The reaction generates water vapor and carbon dioxide while electrons migrate to the external circuit to produce direct-current (DC) power (output). Generally speaking, the DC output is converted into alternating-current (AC) output using an inverter or another type of power conditioner. Some of that AC output (about 5 to 10 %) is consumed internally by the SOFC system while the remainder is available as the net AC output.

Based on this mechanism, the research and development of an SOFC system is generally conducted in the following order: the development of the materials and structure of the single cell; the development of the stack; and the design and development of the reformer and other system components. To achieve an optimal design and development of the stack, as shown in Fig. 3, it is important to first establish a good understanding of the performance of the single cell and, based on such understanding, evaluate the variability in performance among cells arising from such factors as temperature and uneven fuel distribution within the stack. For the purpose of system development, the important performance determining factors include stack performance, along with thermal design and reformer performance, while electrical efficiency is the most important performance indicator.

We believe that by developing the performance evaluation technologies for SOFC single cells, stacks, and systems, respectively, AIST is fulfilling its roles as a public institution. Such roles include not only assisting the private sector's research efforts and facilitating the early market entry of SOFC systems, but also establishing the international and national standards for the performance testing methods that are essential for promoting fair competition once SOFCs become commercially available.

The power generation performance of SOFC cells and stacks is affected substantially not only by the characteristics of the SOFC per se, but also by the testing conditions, including gas flow rate and composition, temperature, and fuel utilization (i.e. the ratio of fuel used for power generation to the supplied fuel). However, no performance evaluation methods in the past have specified the degree of accuracy required for measurements of gas flow rate/composition or other parameters. In addition, approximately 10 % of the anode gas consists of highly toxic carbon monoxide. Typically, therefore, gases such as pure carbon monoxide and pure hydrogen are sourced separately and mixed for testing. This requires stringent safety measures for the handling of pure carbon monoxide, thereby complicating the testing equipment and raising the testing cost.

As for the measurements of electrical efficiency, uncertainty of ±1 % or below (relative uncertainty of ±2.5 %) is

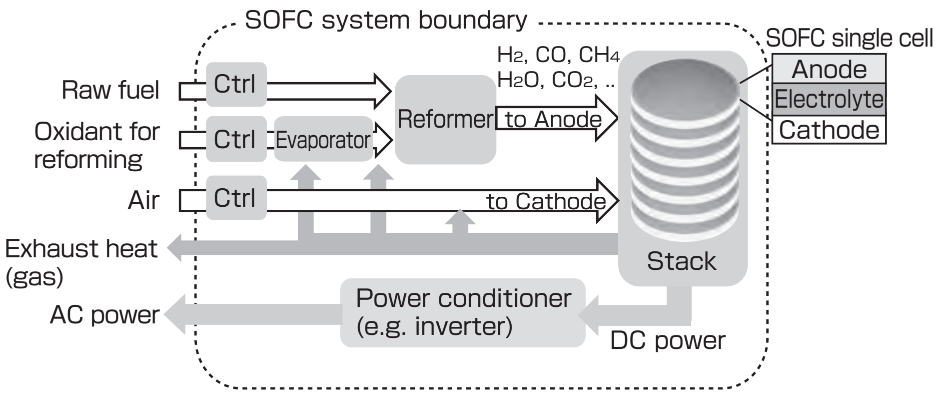

Fig. 2 Configuration of a standard SOFC system (Ctrl represents flow controller)

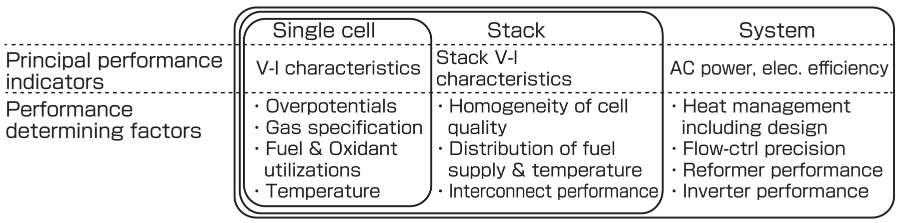

Fig. 3 Relationships between performance indicators and performance determining factors Vand I stands for voltage and current, respectively.

considered appropriate for an electrical efficiency of 40 % (for rated, transmission-end output in HHV), the target value set for cogeneration systems in the NEDO project 2004-2007.[9] At the time, however, there were no standards that prescribed the measurement precision of the performance evaluation methods for phosphoric acid fuel cells (PAFC) or PEFC, both of which were developed earlier than SOFC. JIS B8122, the then existing standard for cogeneration units, only prescribed ±1.5 % for the wattmeter and ±3 % for the fuel gas flowmeter as the instrumental precision for the type tests.[10] As a result, the uncertainty of electrical efficiency measurements was estimated to be approximately ±5 %, which was unsatisfactory.

In addition, because it is difficult to transport an SOFC system of 10 kW or more to a well-equipped laboratory for measurement purposes, the best solution, we believed, was to perform on-site measurements at private SOFC installation locations. Hence we believed it was vital to establish a transportable method for measuring SOFC electrical efficiency.

As for the standardization of the performance testing methods, we believed that the standards for SOFC systems should be given highest priority, since cell manufacturers and system developers were working on the development of 1 to 200 kW-class SOFC systems at the time, and their market entry was imminent. Recently, however, a number of corporations in and out of Japan have begun offering SOFC cells and stacks for sale. An increase in commercial transactions involving these products is foreseeable, and the development of the relevant performance testing standards is therefore vital. We are currently proposing IEC and JIS standards for SOFC cell/stack testing methods, and the details of our activities were reported in a previous issue.[11]

Against this background, the following development objectives were established for the purpose of evaluating SOFC single cells/stacks: a measurement method for the power generation performance of SOFC cells and stacks, in which a test operator can easily simulate the operating conditions of an SOFC system and measure the voltage and other parameters as safely as possible at the relative uncertainty of ±1 %; and a stack performance evaluation method that allows for the evaluation of the variability of stack performance and the factors causing the variability.

As part of our goal to develop a high-precision measurement method for electrical efficiency, we established ±1.0 % as the target value of the relative uncertainty of on-site electrical efficiency measurements that is attributable to the measurement system. To achieve this goal, we developed various technologies, mainly focusing on the technologies for the highly accurate measurement of the flow rate and the composition of raw fuels such as city gas and liquefied natural gas (LNG). At the same time, we pursued the standardization of the testing methods for electrical efficiency.

2.2 Specific approaches to achieving research objectives

One of the goals of this study was to minimize uncertainty in measurement values. For this purpose, we worked on high-precision calibration techniques for commercially available measurement instruments with excellent repeatability and linearity, such as thermal mass flowmeters, gas chromatographs, and voltmeters, while ensuring traceability to the national standards.

Figure 4 shows a schematic of the component technologies developed and combined in this project. In the area of flow rate measurement, for example, our group worked with the

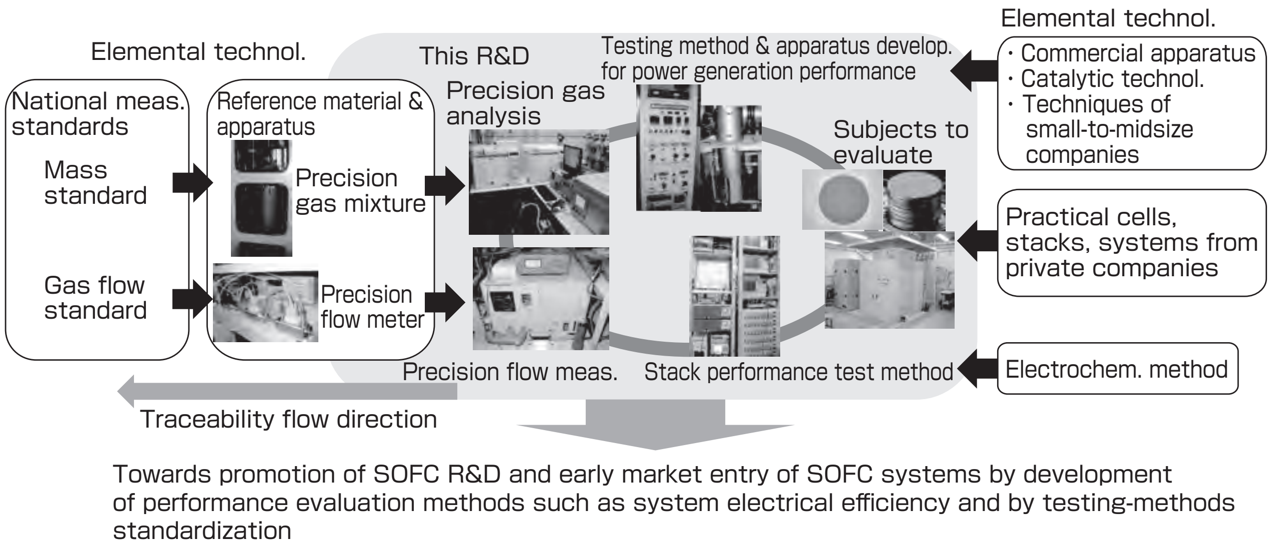

Fig. 4 Elemental technologies incorporated into the research and development project and combined results

Fluid Flow Division of the Metrology Institute of AIST, which developed the standard and the working standard used for the measurement of the flow rate of city gas and hydrogen. Our group was responsible for the development of the flow calibration method and the high-precision flow measurement method/apparatus for the measurement of electrical efficiency.

For gas composition analysis, we cooperated with a standard gas manufacturer to develop an analysis method using a standard gas mixture obtained by the gravimetric method in order to reduce the level of uncertainty. We prepared a high-precision gas composition analysis system as well in collaboration with a manufacturer of precision analysis instruments.

To establish an evaluation method for power generation performance, we developed a method that combined an existing catalytic technology and the aforementioned new high-precision flow rate measurement method, as shown in Fig. 4. In a joint effort with a instrument manufacturer, we developed a testing system capable of testing privately developed, full-size SOFC cells/stacks.

For a stack performance evaluation method, we worked with a manufacturer of electrochemical instruments to prepare a prototype of a measurement apparatus capable of making simultaneous measurements of different factors causing variability in stack performance by applying the AC impedance method, a basic electrochemical evaluation method.

Thus, in this study we developed new, high-precision SOFC performance measurement methods and systems by combining commercially available measurement instruments, high-precision flowmeters and standard gas mixtures that are traceable to measurement standards, and an existing catalytic technology. At the same time, we evaluated the performance and applicability of the systems we developed in collaboration with SOFC manufacturers and others.

3 Performance evaluation and measuring methods

3.1 Development of SOFC cell/stack performance evaluation methods

3.1.1 Development of power generation performance evaluation methods

The voltage of a single SOFC cell is obtained by subtracting the following from the electromotive force, which is determined by the partial pressure of oxygen and the temperature within the anode and cathode gases: resistance overpotentials resulting from ohmic resistance, activation overpotentials arising from electrode reactions, and concentration overpotentials caused by the low gas diffusion rate near the electrode.

The partial pressure of oxygen within the anode gas mixture (e.g. H_2-H_2O-CO-CO_2-CH_4) is dependent on factors such as the composition and pressure of the anode gas mixture and fuel utilization. It is important, therefore, to specify these parameters for the cell/stack performance test. In addition, the degree of overpotentials mentioned above varies according to the type of material and the structure of SOFC; minimizing these factors would help improve the performance of SOFC.

A variety of fuels, such as city gas and LPG, can be used for an SOFC system, and are reformed before they are fed to the anode side, as shown in Fig. 2. In other words, anode gas composition varies significantly depending on the types of feedstock fuels used and the reforming conditions.

There are a number of problems associated with the testing method of power generation performance, examples of which are: (1) The testing conditions of a simplified test using humidified hydrogen differ significantly from the actual operating conditions of an SOFC system; and (2) to supply simulated reformate gas, it is necessary to use highly poisonous pure carbon monoxide, and the difficulty of generating a stable supply of water vapor at a low flow rate increases the fluctuation of cell voltage.

Incidentally, there is very low risk arising from carbon monoxide in an SOFC system (final product), since there is no possibility that anode gas would leak in its original form; unused fuel components such as carbon monoxide within the anode exhaust gas are combusted by the oxygen within the cathode exhaust gas.

Against this background, our research efforts on SOFC power generation performance evaluation focused on the development of a method of generating and feeding simulated reformate gases that would allow SOFC developers to conduct the power generation test safely and easily while achieving reliable results.

Figure 5 shows the method of supplying reformate gases that we developed for this study. Water vapor is generated by using a catalytic combustor filled with a commercially available platinum catalyst and reacting excess hydrogen with oxygen at 200 ºC or above. This method makes it possible to generate a stable supply of water vapor even at a low flow rate and prepare an H_2-H_2O gas mixture.

In addition, to avoid the use of pure carbon monoxide, we developed an equilibrium reactor using a commercially available nickel or ruthenium catalyst for methanation. This equilibrium reactor is capable of producing carbon monoxide from carbon dioxide just before the gas mixture reaches the anode by using the reverse-shift reaction ($H_2 + CO_2 \rightarrow H_2O + CO$) while a methanation reaction ($4H_2 + CO_2 \rightarrow CH_4 +$

$2H_2O$) simultaneously proceeds to produce methane.

In sum, by using hydrogen, oxygen, carbon dioxide as gaseous ingredients and the newly developed catalytic combustor and equilibrium reactor, our method is capable of simulating reformate gases and feeding them to the anode, while maintaining their composition and flow rate identical to those generated by the reforming reaction at equilibrium.

In addition, even if carbon monoxide leaks during the test, it is possible to cut the supply of carbon dioxide and stop the production of carbon monoxide, thereby making it possible to devise safety measures for the test.

Next, we developed a test apparatus for SOFC power generation performance in collaboration with Eiwa Corporation, as shown in Fig. 5 (patent application 2008-045311), by applying the know-how we acquired from the new anode gas supply method. We successfully produced a compact catalyst combustor (10 mL) and equilibrium reactor (20 mL) for testing 100 W-class SOFC systems.

We controlled the flow rate of the simulated reformate gases using a thermal mass flow controller (MFC), and achieved an uncertainty of ±0.5 to 1.0 % by calibrating the flow rate using a high-precision flowmeter (uncertainty: ±0.2 %).[12]

An examination of the performance of this test system revealed that the precision of the composition of the simulated reform gases was ±0.5 to 1.0 mol% (Fig. 5) of the equilibrium value, while the stability of the gas composition was ±0.04 mol% of the same, demonstrating that with this apparatus it is now possible to conduct a highly accurate power generation performance test.[13]

In addition, when we tested an actual single SOFC cell developed by a private corporation, it became clear that the fuels and reform conditions can be modified easily and quickly by changing the flow ratios of the raw gases. The cell voltage stabilized within 30 seconds, significantly reducing the testing time and also keeping the fluctuation of the cell voltage at ±0.1 % or below.

In summary, the SOFC power generation evaluation method and the testing apparatus we developed in this study are capable of feeding a stable and precise supply of simulated reformate gases at a low flow rate to the anode while avoiding the use of highly poisonous pure carbon monoxide, thereby making the testing of the power generation performance of SOFC easier, safer, and more precise.

Given that several SOFC manufacturers have recently acquired this testing system, we intend to continue providing technical assistance to private testing-instrument manufacturers to contribute to the overall research and development efforts for the commercialization of SOFC. Currently, we are using the findings from this study to prepare a draft for single cell/stack performance testing methods and proceeding with the JIS and IEC standardization efforts at the same time.[11]

3.1.2 Development of stack performance evaluation methods

The performance of an SOFC system is dictated largely by the performance of its stack, which is formed by connecting multiple cells (Fig. 6). Minimizing the performance

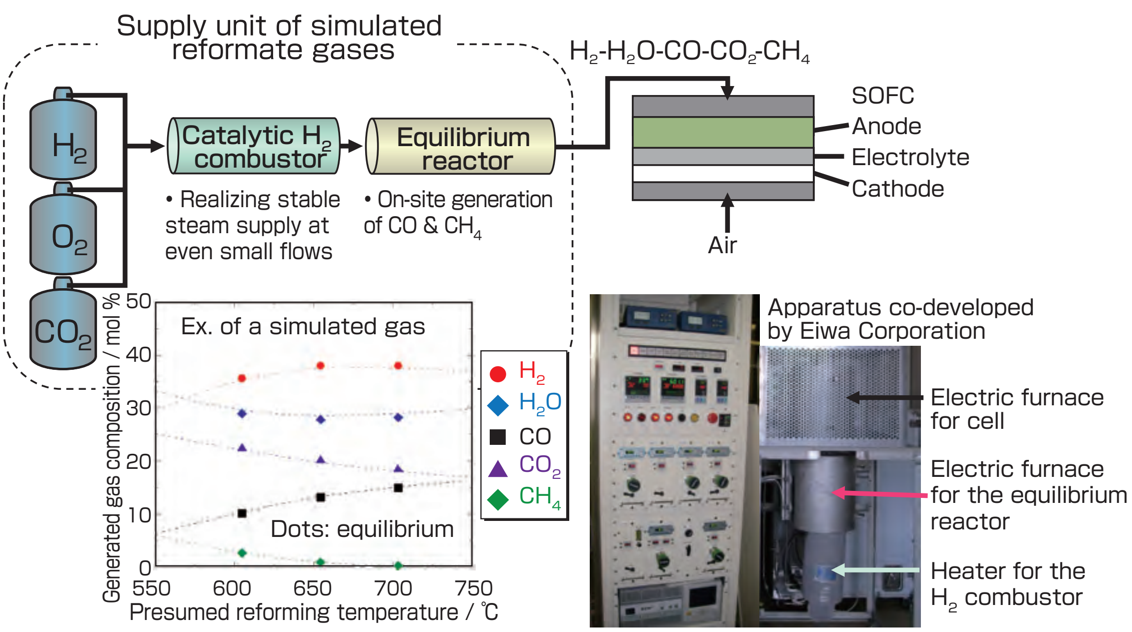

Fig. 5 SOFC power generation measurement method/apparatus developed in the project

variability among the cells in the stack makes it easier to control the performance of the system as a whole. In addition, because the cells are normally connected in series, the stack would stop functioning if any one of the cells fails or degrades significantly.

For these reasons, it is desirable to evaluate the performance of each cell within the stack as part of the research and development of the SOFC system. As shown in Fig. 6, examples of factors causing performance variability among the cells include individual variations among the cells and surrounding components, inconsistency in fuel distribution, temperature distribution in the stack, and partial degradation in performance. However, no method has been established in the past for evaluating the performance of SOFC based on different parameters such as those described above.

Applying the AC impedance method, which is commonly used for the electrochemical evaluation of single cells, we developed an evaluation method capable of making separate and simultaneous measurements of each of the factors contributing to the variability of cell performance in a stack. As shown in Fig. 6(b), we prepared a prototype of a multi-impedance analyzer capable of measuring the impedance of 47 cells simultaneously.[14]

In this test system, the AC current is superimposed on the DC current (with a frequency range of several tens of kHz to 0.01 Hz). The voltage responses of each cell are Fourier transformed to simultaneously obtain multi-impedance spectra as a function of the time constant of an electrode reaction or other parameters.

Using the prototype, we tested a 1-kW stack (46 cells in series) developed by our joint research partners, Kansai Electric Power Co., Inc. (KEPCO) and Mitsubishi Materials Corporation. Figure 6(c) exhibits an example of the impedance measurement results (cells #41 to 46) in a Cole-Cole plot.

The x-axis is the real part of impedance. As shown by the spectrum for #46, the resistance in an SOFC stack is categorized into the following: A: Ohmic resistance; B: activation overpotentials; and C: resistance corresponding to the change in gas concentration due to fuel consumption. For example, the results for cells #41 and #42 show that C is larger than for other cells, and we can conclude that the actual fuel utilization has increased due to insufficient fuel supply. Feeding back such measurement results into SOFC research efforts can help accelerate development progress.

In summary, the study demonstrated that by developing a stack performance evaluation method capable of separately measuring performance variability and each resistance

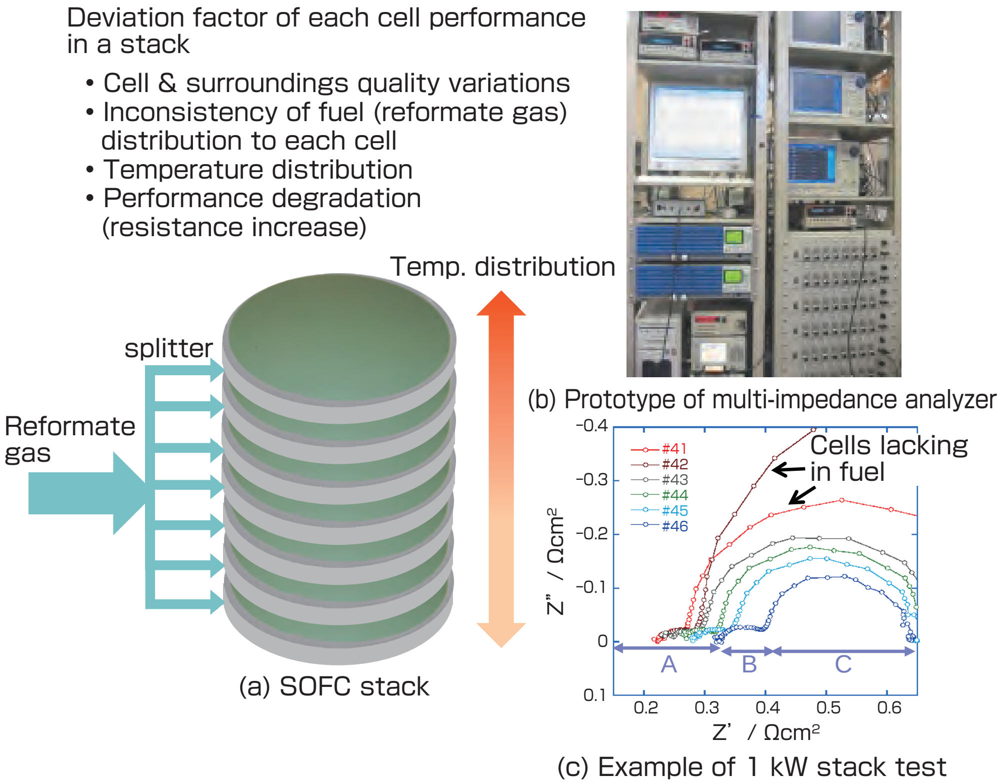

Fig. 6 Development of a stack performance evaluation method and a measurement example of a privately developed 1-kW stack

factor that causes the variability in cell performance and by applying this method to measure the performance of a privately developed 1-kW stack, it is possible to make a multifaceted evaluation of parameters such as inconsistent fuel distribution and thereby contribute to overall SOFC research efforts. In fact, the results from this study proved useful in the development of a 10-kW cogeneration system by our research partner corporations. We believe that the method also has applicability for conducting durability tests.

3.2 Development of measurement methods for electrical efficiency of SOFC systems and standardization efforts

3.2.1 Development of a high-precision measurement method for electrical efficiency

As part of the NEDO project in 2004-2007 “Research on the Technology for the Evaluation of System Efficiency Measurement,” we conducted a research and development project in a joint effort with the Fluid Flow Division of the Metrology Institute of AIST, mainly focusing on the development of a high-precision measurement method for the electrical efficiency of the SOFC system. The Fluid Flow Division worked on the development of the standard and the working standard for measuring the flow rate of city gas and hydrogen.

In the meanwhile, our group developed a transportable electrical efficiency measurement method. We used this method to take an on-site measurement of the electrical efficiency of a 10-kW cogeneration system developed by Kansai Electric Power Company (KEPCO) and Mitsubishi Materials Corporation as part of the aforementioned NEDO project, and conducted uncertainty analysis. As mentioned in subchapter 2.1, the HHV standard was used to express electrical efficiency.

As shown by equation (1), the net electrical efficiency of an SOFC system, η_e, is defined by the following: heating value of city gas or other types of raw fuels supplied to the SOFC system, H; flow rate of the fuel, f; and net AC output (power to export), P.

$$\eta_e = \frac{P}{H \times f} \qquad (1)$$

In calculating an electrical efficiency, it is important to measure each of these parameters in such a manner that the target uncertainty value can be achieved. The sources of uncertainty include not only the uncertainties arising from the measurement instruments used, but also those uncertainties attributable to deviation of the parameters themselves (e.g. the composition of city gas, power output).

Hence, if the target relative uncertainty of electrical efficiency attributable to the measurement system is set at ±1.0 % and is assigned equally to H, f, and P, each parameter must be measured at the relative uncertainty of ±0.6 %.

Given the above, in this research project we established the target value of ±0.6 % for the relative uncertainty of the measurement instruments used to measure each of the heating value, flow rate, and output. In order to minimize uncertainty, we developed high-precision measurement methods, including calibration methods of measurement instruments that are traceable to standards. The measurement methods are described below.

3.2.1.1 Development of a heating value measurement method

The most commonly used method for obtaining the heating values of gaseous fuels such as city gas is to measure the gas composition using a gas chromatograph and to calculate the heating value based on the measurements, as specified by JIS K 2301 and ISO 6974 and 6976 (the ISO standards are applicable only to natural gas).

In this study, we developed measurement methods based on the method prescribed by the ISO standards that also include uncertainty calculation methods. Gas samples were taken from the SOFC system on site using a portable gas sampler (patent application 2008-045311). As illustrated in Fig. 7, the gas samples were then transported to our laboratory and measurements were taken using gas chromatographs with specifications in compliance with the aforementioned JIS or ISO standards. We also deployed a measurement method using a highly portable micro gas chromatograph with the capability of making on-site quick analysis within a few minutes (Fig. 7).

While ISO 6976 proposes an uncertainty of approximately ±0.1 % for the heating value of each city gas component, the standard does not incorporate this value into uncertainty calculations. In this study, however, this uncertainty value was taken into consideration.

For high-precision analysis of fuel gas composition, it is important to reduce the uncertainty of the standard gas mixture (i.e. a mixture that has predetermined concentration levels of gas components) to be used for the calibration of the gas chromatographs. We examined standard gas mixture products for city gas having a defined uncertainty value and commercially available in Japan. It was found that the concentration of methane, the main component, had a relative uncertainty of ±1.0 %, such as 88.47±0.89 % (k=2). This value was insufficient for this study.

With cooperation of Sumitomo Seika Chemicals Co., Ltd., we filled a 47-L gas cylinder with each component of city gas using the gravimetric method. We took three repeated measurements of the mass of each gas in the cylinder using a large balance scale that is traceable to the national standard on mass measurements and prepared a standard gas mixture. Based on the uncertainties of the mass, the scale, and other factors, we calculated the mole fraction of each component of the standard gas mixture, and the uncertainty of each in accordance with the method described by ISO 6142. The result, in the case of methane, was 88.002±0.016 %, or a relative uncertainty of ±0.02 % (k=2), demonstrating that we were able to reduce the uncertainty of the standard gas mixture to 1/50 of the existing uncertainty value. Then, we calibrated the gas chromatographs and prepared a calibration curve using several of such high-precision standard gas mixtures.

Additionally, in collaboration with Kimoto Electric Co., Ltd., we developed a high-precision, automated heating value measurement apparatus (Fig. 7) capable of the following: repeated analyses of three types of standard gas mixtures before and after the sample measurement; automated analysis of the city gas sample in a separately developed portable gas sampler; and output of analysis data of parameters including the flow rate, gas pressure, and temperature at the time when samples are taken.

Thus, the above demonstrates that we were able to develop a system capable of measuring the heating value of city gas at an uncertainty of ±0.12 %.

3.2.1.2 Development of a measurement method of city gas flow rate

To measure the flow rate of city gas or other raw fuels supplied to the SOFC system, it is necessary to install a flowmeter designed for efficiency measurement upstream of the SOFC system. While a wet-type volumetric flowmeter designated by the Measurement Act is used as the reference instrument for the inspection of residential gas meters,[15] no industrial standard has been established for the flow rate of city gas in Japan and hence traceability has not been properly addressed in this regard. In this project, therefore, the Fluid Flow Division developed the standard and the working standard for measuring the flow rate of city gas.

Flowmeters that emit oil and water vapor such as the one mentioned above are not suitable for efficiency measurement, as they may affect the SOFC system. In contrast, thermal mass flow controllers (MFC), with easy maintenance and excellent repeatability, are often used in SOFC system prototypes. For these reasons, in this study we developed a city gas flow measurement and calibration system using a commercially available thermal mass flowmeter (MFM).

When the flow measurements of methane by commercial MFMs and MFCs were calibrated using a high-precision mass flowmeter, results showed an instrument error of up to 8 %. To achieve the target relative uncertainty of ±0.6 %, it is necessary to calibrate the flowmeter using an identical gas under efficiency measurement. Because of this, it is important to minimize the uncertainty of standard flowmeters for city gas or other fuels used for calibrating the MFM.

In addition, while gas composition has little impact on the volumetric flowmeter, the sensitivity of the MFM is influenced by the type of gas under measurement.

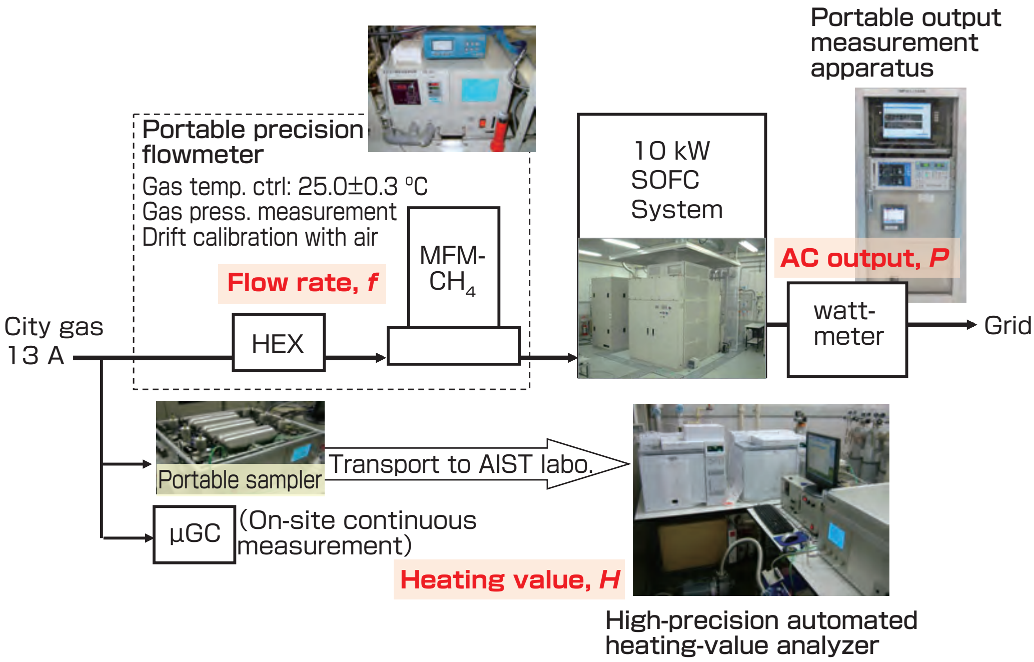

Fig. 7 Overview of transportable high-precision generation efficiency measurement system
HEX: Heat Exchanger; MFM-CH4: thermal MFM for methane; μGC: micro gas chromatograph

Therefore, corrections must be made using each gas type (mixture) under measurement. Moreover, to achieve a highly precise flow rate measurement, the impact of pressure and temperature must be taken into consideration even for measurements using the MFM.

In order to minimize the impact of temperature on the MFM, we developed a transportable flow rate measurement apparatus capable of recording data and controlling gas temperature with a fluctuation of ±0.3 ºC using a heat exchanger and others, as shown in Fig. 7. In addition, in order to prepare for the durability testing of the SOFC system, we installed on this measuring equipment a city gas bypass line, a compact air compressor, and a high-precision laminar flowmeter (molbloc from DHI), so that it is possible to calibrate the MFM and evaluate its sensitivity drift with ambient air on a regular basis (e.g. once a month) without causing any impact on the test subject.

The mass flow rate (g/min) measured by the MFM was converted into volume flow rate (l min^{-1}, m^3 s^{-1}) using the volume of the ideal gas in the standard state (0 ºC, 101.325 kPa), and is expressed in Nl min^{-1} and Nm^3 s^{-1}.

To calibrate the flow rate measurement of the MFM, an MFC was prepared for each of the main components of city gas: methane (concentration: approximately 89 %), ethane (6 %), propane (3 %), and butane (2 %). Using the MFCs, we developed a prototype of a city gas flow calibration apparatus that calibrates the flow of each component based on the mass flow rate, mixes the gas components using a static mixer, and generates simulated city gas whose temperature has been adjusted by a heat exchanger. We used this calibration apparatus to calibrate the MFM and conduct MFM characteristic tests to study the impact of city gas composition, temperature, pressure, and other factors on the MFM.

This calibration apparatus can calibrate the flow of city gas up to 70 Nl min^{-1}, which corresponds to the flow rate of a 20-kW SOFC. Details are available from a separate volume of this journal.[16]

In addition, it was confirmed that the gas component correction factor (CF_{mix}) of the MFM used for methane in this study can be calculated based on multiple linear regression analysis using the polynomial equation shown in equation (2).[16] The uncertainty of the calculated value was found to be ±0.15 %.

$$1/CF_{mix} = \sum x_i \,/\, CF_i \qquad (2)$$

where x_i is the mole fraction of component i and CF_i is the relative sensitivity of gas component i to methane in a thermal mass flowmeter. It was also confirmed that the repeatability, linearity, temperature dependence, pressure dependence, and sensitivity drift over time are approximately identical to those found in the catalogue specifications. It was determined, therefore, that catalogue specifications can be used for the analysis of uncertainties attributable to these factors.

In sum, we developed a high-precision, transportable city gas flow measurement/calibration system traceable to flow standards and supporting SOFC systems of up to 20 kW. By doing so, we achieved a relative uncertainty of ±0.44 % for the gas flow rate. We also identified the MFM properties necessary for the uncertainty analysis of the flow measurement of city gas. For example, we established a prediction equation to obtain the effect of gas compositions on the sensitivity of a thermal mass flowmeter.

Thus, it is now possible to make gas flow measurements whose traceability is ensured by calibrating the measurements using a high-precision flowmeter traceable to the standard or the working standard developed by the Fluid Division or a flowmeter certified by the Japan Calibration Service System (JCSS) that recently became available on the market.

3.2.1.3 Development of an output measurement method

For the output measurement of 10 kW-class SOFC systems, a wattmeter having an accuracy of ±0.1 % is available commercially, so that you can convert the accuracy into uncertainty with type B evaluation of uncertainty.[7] Similarly, a precision calibration method certified by the JCSS has been the established method for the measurement of electric power. In addition, the Japan Electric Meters Inspection Corporation (JEMIC) also offers general calibration services with an uncertainty of ±0.04 % (JCSS's best measurement capability is ±0.02 %). It is possible, therefore, to achieve the target relative uncertainty of ±0.6 % for output measurement.

It must be noted, however, that the AC output of the SOFC system is obtained by converting the DC output produced by the stack into the utility frequency AC power using an inverter. The AC output includes not only the fundamental wave of the utility frequency, but also different harmonics as well as the carrier of the inverter. Hence, for the purposes of this study, we defined net output, P, to include only the active power of the fundamental wave.

The evaluation of higher harmonic waves and other frequencies other than the fundamental is essential, as they may have a major impact on the uncertainty of the electric power measurement. Currently, however, there are no national standards that cover AC power containing higher harmonics, and general commercial wattmeters only cover

Table. 1 Measurement values and their uncertainties for the initial characteristic test for net electrical efficiency of a 10-kW cogeneration system

Parameters	Average	Relative uncertainty	(JIS regulation example[1)])
Higher heating value of city gas, H (MJ Nm^{-3})	44.69	±0.12 %	No regulation
Flow rate of city gas, f (10^{-4} Nm^3 s^{-1})	5.507	±0.58 %	±3.0 %
AC output, P (kW)	10.14	±0.46 %	±1.5 %
Net electrical efficiency, η_e (%)	41.2	±0.74 %	No regulation

1) Type test in JIS-B8122 (2001) Test methods for measuring performance of cogeneration unit

AC power with higher harmonics that fall within the error range of a given accuracy class.

For these reasons, we developed a compact, transportable output measurement apparatus as shown in Fig. 7. This apparatus is based on a commercially available precision power analyzer (Yokogawa WT3000) capable of making the precision measurement of AC power, the analysis of higher harmonics, and the measurement of DC power when needed.

Other than active power, this apparatus measures various parameters, such as voltage, current, power factor, higher harmonics, and inverter carrier waves under output conditions. The apparatus is also designed to measure the environment temperature and the instrument temperature at the time of the measurement for the purpose of uncertainty analysis.

3.2.1.4 Electrical efficiency measurement of a privately developed 10 kW-class system

Using the transportable high-precision efficiency measurement system (Fig. 7) we developed as described in sections 3.2.1.1 to 3.2.1.3, we tested the initial characteristics of the net electrical efficiency of a 10 kW-class cogeneration system and took on-site measurements of its electrical efficiency during a 3000-hour durability test. As mentioned above, this power system was developed by KEPCO and Mitsubishi Materials as part of the NEDO project and had the following specifications: AC output of 60 Hz; three-phase three-wire system of 200 V; maximum current of 38 A; and power factor of 0.99.

To connect the 10-kW system to the transportable output measurement apparatus, we employed a three-phase four-wire output measurement method, whereby a virtual neutral point generated by a starpoint adapter is used for the connection, in order to minimize uncertainty.[16]

Figure 8 shows the flow rate and output measured during the initial characterization test. Because the fuel utilization (the ratio of fuel used for cell reactions) of this 10-kW system was kept constant, the city gas flow rate stayed relatively stable with a fluctuation of ±0.14 %. In contrast, the AC output fluctuated ±0.45 % due to the fluctuations of auxiliary power consumed internally by the SOFC system.

The fluctuation of the heating value of city gas was analyzed using a micro gas chromatograph at the installation location of the 10-kW system. City gas samples were also taken using the portable sampler before and after the test, and the heating value fluctuations were analyzed at the AIST laboratory. The results showed that the fluctuation range of the heating value during the initial characteristic test is sufficiently small, at ±0.02 %.

For measurement values we used the mean values of the HHV and flow rate of the city gas, and output (active power of 60 Hz), which were, respectively, 44.69 MJ Nm^{-3}, 5.507×10^{-4} Nm^3 s^{-1}, and 10.14 kW, as shown in Table 1. Using equation (1), an HHV-based net electrical efficiency of 41.2 % was obtained.

When we combined the fluctuation ranges of the parameters mentioned above, the uncertainties attributable to the measurement conditions at the time of efficiency

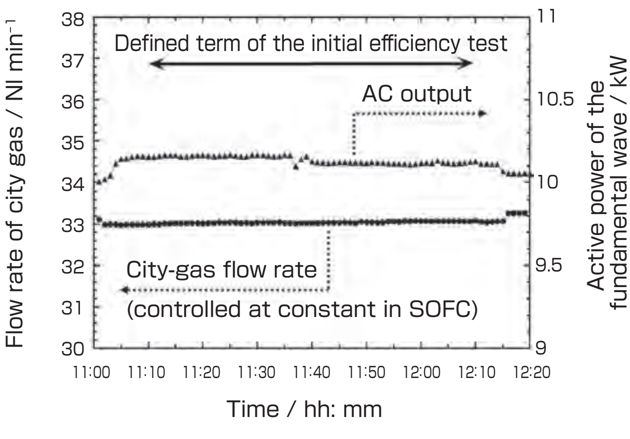

Fig. 8 City gas flow rate and AC output at the time of the initial characterization test for electrical efficiency of a 10-kW cogeneration system

measurement such as the temperature and pressure of city gas, and the uncertainties deriving from measurement instruments as described in subsections 3.2.1.1 to 3.2.1.3, we found that the relative uncertainties of the heating value, flow rate, output, and electrical efficiency were, respectively, ±0.12 %, ±0.58 %, ±0.46 %, and ±0.74 %, as shown in Table 1. The electrical efficiency is estimated to be 41.2 ±0.3 % (HHV), which meets the target uncertainty level of the NEDO project of ±1.0 % while demonstrating that this 10-kW system exceeded the electrical efficiency of 40 % (HHV), NEDO's development target value.

In sum, in this study we developed a transportable, high-precision efficiency measurement method and apparatus traceable to national measurement standards. It was shown that the apparatus was capable of measuring electrical efficiency on-site at a relative uncertainty of ±1.0 % or below.

In other words, even if net electrical efficiency improves to 50 to 70 % in the future, it will be possible to measure the efficiencies at an uncertainty of ±0.5 to ±0.7 %. In addition, this method is likely applicable not only to SOFC systems but also to efficiency measurements of systems operated by other fuel cells or other energy technologies.

3.2.2 Standardization of electrical efficiency measurement testing method

While IEC 62282-3-2 has been published as the international standard for the electrical efficiency testing methods for fuel cell systems, no JIS has been established in Japan for the electrical efficiency testing methods for SOFC systems. Furthermore, when SOFC is ready for practical and commercial applications, a testing method whose measurement uncertainty should be comparable to that of AC utility power (i.e. uncertainty of output measurement at the ±0.1 % level) is required.

Therefore, we applied the findings of the high-precision electrical efficiency measurement method described in section 3.2.1 to investigate the impact that the transportation of the measurement instrument to the SOFC installation location has on the instrument uncertainties. Based on the results, we prepared a JIS draft for electrical efficiency testing methods that achieve effective comparisons of electrical efficiency levels, the most important performance indicator of an SOFC system. With the cooperation of the Japan Electrical Manufacturers' Association (JEMA), the draft was then examined by a deliberating committee consisting of cell/stack manufacturers, system manufacturers, and neutral research institutions.

Based on the results of the committee deliberations and our surveys of existing JIS and calibration systems, we prepared a draft of a JIS Technical Specification (TS) prescribing electrical efficiency measurement methods for SOFC systems at an uncertainty level of less than ±1 % while ensuring that the methods are traceable to national measurement standards. Following the deliberation of the draft by the Japan Industrial Standards Committee (JISC), JIS TS C0054 was published in 2010 with the title of "Testing method of power generation efficiency for solid oxide fuel cell power systems fueled with gas in which methane is main component," well in advance of the release of residential SOFC systems in October 2011.

4 Summary and future prospects

We developed new SOFC performance evaluation methods for all SOFC units, cell, stack, and system (complete product), by combining commercial measurement instruments and catalyst technologies while ensuring traceability to national measurement standards. Our objectives have been to promote the early commercialization of SOFC systems and to develop domestic and international standards for performance testing methods that are essential for ensuring fair competition among makers of SOFC systems.

The establishment of performance evaluation methods for SOFC cells/stack is crucial for the research and development of SOFC systems. We devised easy, safe, and high-precision power generation performance testing methods for SOFC cells/stacks and developed a testing apparatus in collaboration with private corporations. We developed a measurement method and apparatus as well that can simultaneously measure different factors causing cell-to-cell performance variability in the stack. Through these projects we have contributed to the research and development efforts for SOFC systems in the private sector.

As for the measurement of electrical efficiency of SOFC systems, we established a transportable method capable of making high-precision measurements of electrical efficiency even at SOFC installation locations. When actual measurement was made on a 10-kW system developed by private manufacturers, the result was 41.2 ±0.3 % (HHV), which demonstrated that the NEDO project target value (net electrical efficiency of 40 % or above and uncertainty of ±1.0 %) was achieved. This method will be more than adequate even when the electrical generation efficiency of SOFC systems improves in the future. Furthermore, we prepared a JIS (TS) draft and were able to publish the final version prior to the commercial release of SOFC systems.

We will continue with our efforts to accelerate the development of JIS and IEC standards for SOFC cell/stack performance test methods. At the same time, by taking advantage of the methods developed in this study, we will make further improvements on the electrical efficiency of SOFC systems, contribute to the research and development of SOFC systems operating on novel fuels (e.g. dimethyl ether), biomass, and coal, and commit our efforts to the development of portable SOFC systems, which have the potential to serve as a supplementary power source for automobiles and other applications. Our goals also include improving the acceptance level and applicability of high-efficiency distributed SOFC systems and the standardization of durability testing methods, which are essential for the commercialization of SOFC.

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The project team would like to thank METI and NEDO for providing funding and assistance for this research and development project and the standardization of performance testing methods. We would also like to thank KEPCO, Mitsubishi Materials Corporation, Sumitomo Seika Chemicals Co., Ltd., Kimoto Electric Co., Ltd., Eiwa Corporation, and the Fluid Flow Division of the Metrology Institute of AIST and all others who have offered their generous time and cooperation for our research and development project. We would also like to thank JEMA and all SOFC cell manufacturers, SOFC system developers, and neutral research institutions that offered their collaboration for our standardization work.

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Discussions with Reviewers

1 Clarity

Comment (Hiroshi Tateishi: AIST)

Because the paper is quite long, some parts need to be simplified. In particular, much of the chapter entitled, "1 Introduction" is about the general background/objectives of the paper, so I think it should be kept to a minimum for the benefit of the general readers. Please provide a simple description of the need to fight against global warming and adopt distributed power sources, ways in which SOFC can fulfill this need, the SOFC development progress, and the objectives and significance of the authors' R&D activities (especially in regards to the current state and importance of the evaluation methods). In addition, it is questionable how many readers can understand the description regarding '*uncertainty*' towards the end of the paper and it would be helpful to include a reference material.

Comment (Yasuo Hasegawa: AIST)

We ask the authors to write their reports in an easy-to-understand manner so that readers from other fields can read *Synthesiology* without difficulty. For this reason, while a reference material on uncertainty is included, I think it would be even clearer if a simple explanation is included in the paper and the reader is directed to the reference material for details. Because uncertainty is a fairly common term, we need to be careful that readers are not misled into thinking that they understand the concept without actually doing so.

Answer (Yohei Tanaka)

I tried to make the introductory part simpler. I removed the detailed description of the history of SOFC development and included a brief description of the current state of system development efforts as well as the future plans. I also touched upon the characteristics of the SOFC system as a highly efficient generation system and the importance of high-precision performance evaluation technologies, our group's project.

As for the significance of SOFC, while it is an effective solution against global warming, we feel that it has an even more important role to play as a highly efficient distributed power generation system providing a stable supply of electricity and heat in the interior regions (i.e. through efficient use of energy) in the future, in contrast to centralized power stations found in coastal areas. Therefore I did not make very many corrections in this regard. Regarding uncertainty, I did add a brief explanation as suggested and added a note to refer to reference material [7].

2 Performance evaluation framework

Comment (Hiroshi Tateishi)

The paper does not provide any systematic description of the specific factors concerning each of the cell, stack, and system that need to be evaluated for the development of performance testing methods. As a result, it would be difficult for a non-expert to understand the objectives of individual R&D projects. It would be helpful to add a well-organized discussion of the specific factors that need to be measured for a comparative evaluation of fuel cell performance, the specific testing methods and the pros and cons of each, and the differences among the evaluation methods for the cell, stack, and system, etc.

Answer (Yohei Tanaka)

I discussed the importance of the performance evaluation of the single cell, stack, and system in chapter 1 and subchapter 2.1. I added Fig. 3 to explain how the evaluations of each unit level relate to one another and also included a list of the performance indicators and determining factors. I also added an explanation of evaluation parameters (performance indicators) for each unit level and their relevance.

3 Hierarchy of technology development projects

Comment (Hiroshi Tateishi)

Because there is very little analysis of the relationships between the individual technology projects described in chapter 3, they appear to be completely independent from one another. This relates to Issue 2 as well.

3.1.1 The development of performance evaluation methods for single cells

3.1.2 The development of performance evaluation methods for stacks

3.2 Development and standardization of system performance evaluation methods.

I believe it is necessary to explain the relationships between these parts and the overall flow of the projects. For example, how are the evaluation methods for the single cell useful for the evaluation methods for the stack or system?

Answer (Yohei Tanaka)

Thank you very much for the valuable comment. As I answered partly in my reply concerning Issue 2, I provided a brief description of the general flow of the performance evaluation/R&D work, from single cells to stacks to systems, along with the performance indicators and evaluation technologies for each level in chapter 1, and elaborated in subchapter 2.1.

In regards to the question of how cell performance evaluation is utilized for stacks or systems, the stack—which is formed by multiple single cells stacked together—serves as the power generation component of the SOFC system. Therefore it is important to understand the performance of single cells, evaluate the variability of cell performance due to temperature fluctuation and inconsistency of gas distribution, and establish an optimal design. For the system design, in addition to stack performance, thermal design and the design of the flow controllers and other peripherals play important roles, but stack performance especially has a direct impact on the overall performance and of the system, including its electrical efficiency.

4 Order of standard development

Question (Hiroshi Tateishi)

What are the reasons for developing the standards for system evaluation methods first before developing the standards for cell/stack performance evaluation methods? It seems to make more sense to start with the standardization of the components, and leave the systems to the end.

Answer (Yohei Tanaka)

As you suggested, the development and standardization should proceed with the cells and stacks first, before the systems. However, because cell manufacturers and system developers were working on the development of 1 to 200-kW SOFC systems at the time, and their market introduction was imminent, we determined that the standards for system testing methods took priority.

Recently, some companies in and out of Japan are starting to sell single cells and stacks, and there will likely be an increasing number of commercial translations involving these products as well as international collaborations for SOFC development. For this reason the development of standards for single cell/stack performance tests is becoming increasingly important. Currently, we are proposing IEC and JIS standards for testing methods of single cells/stacks and we reported our efforts in Vol. 5, No. 4 of *Synthesiology* (reference material [11]). I added this point in subchapter 2.1.