Challenge for the development of micro SOFC manufacturing technology

— Compact SOFC using innovative ceramics integration process —

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Realization of highly efficient SOFC (solid oxide fuel cell) modules, which are compact and capable of quick startup and shut-down operation, is strongly expected because it would be useful to solve environmental problems. In order to yield new outcomes in new energy production industry market, we have carried out continuous R&D directly linked with the original idea, trial production, and evaluation by using the ceramics integration manufacturing platform. In consequence, original, compact and high-power SOFC modules operable at low temperature have been realized by upgrading of function-structure integration technology. These are drawing attention as products of ingenious technology. This paper presents, in addition to industrial needs, approaches and methods in industry-academia-government collaborative research to overcome tasks toward productization.

Keywords : Ceramics processing, ceramic integration technology, energy conversion, fuel cell, micro SOFC, energy module

1 Introduction

The development of the technology for a low-carbon society by shifting from fossil fuel to clean energy is a global concern for humankind. As shown in the Japanese energy statistics, the demand and use of energy to support the social infrastructure are increasing every year. There is an increasing emphasis on technologies that do not use fossil fuels, such as the unused energy of waste heat as well as recyclable solar cells[1]. Particularly, the fuel cell technologies that enable the use of hydrogen energy is gaining attention as the energy management by electrochemical energy conversion that does not emit CO_2. The principle of the fuel cell technology was proposed by Sir William Robert Grove of the United Kingdom in 1893. With the advancement of electrodes that enable electrochemical reaction and technologies for ion-conducting electrolyte materials, the fuel cell technology was put to practice as the power generation technology, initially for plants in the beginning of the 20th century. The commercialization of home-use cogeneration and automobile generator is starting now. As more facilities will be powered by fuel cells, a drastic reduction in CO_2 emission by 5 million kW level cogeneration is expected by the year 2030[1].

In the development of fuel cell technology, various R&Ds using the electrolyte materials as core technology are being conducted actively as shown in Table 1. Currently, the developments are mainly for polymer electrolyte fuel cells (PEFC) that can be handled easily and solid oxide fuel cells (SOFC) that have high generation efficiency[2].

In the history of the development of materials for SOFC that used ceramic material, the developments were done for electrolyte materials such as zirconium (zirconium oxide) that employed the ion-conducting property of oxides at high temperature range, as well as the cermet electrode materials that combine the catalyst materials and various ceramic electrodes with mixed conductivity. The developments for manufacturing flat or cylindrical ceramic cells and for manufacturing module stacks were led by Japan[3]-[5]. Until now, the nickel electrodes were developed for temperature ranges of 700 °C or higher because the characteristic of SOFC is the utilization of direct reforming reaction of hydrocarbons at high temperature range, and high energy conversion can be achieved with fuels other than hydrogen. Therefore, compared to the low-temperature PEFC, in the conventional SOFC modules, it was necessary to increase the operation temperature and generation surface area by reducing the cell resistance to obtain high power generation. As the module increased in size due to increased generation surface area, a technological issue developed where rapid startups and shutdowns could not be repeated due to thermomechanical stress. On the other hand, since excess generation could be controlled by startup/shutdown depending on the power load, the realization of SOFC module that could be started up or shut down rapidly and was operable at low temperature was highly in demand[2]. If such flexible operation became possible by overcoming the technological issues of downsizing and lower generation temperature, the CO_2 emission could be reduced further. Also, if the operation temperature of the module were lowered, low-cost metal materials could be used.

In this paper, we describe our efforts in solving the various R&D issues that were presented as challenges for the

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Table 1 Types and characteristics of fuel cells

	Electrolyte material	Operating temperature	Characteristics	Generation efficiency
Solid oxide fuel cells (SOFC)	Oxide ion-conducting ceramics	500-1000 ℃ (point is achieving high performance at low temperature range)	Electrode resistance is low since it operates at high temperature, and the cell performance is high. Major improvement in efficiency is possible by altering the fuel quality by using waste heat. It is expected to be future dispersed power source.	40-70 %
Polymer electrolyte fuel cells (PEFC)	Proton-conducting polymer film	Room temperature ~ about 90 ℃	Easy to handle due to low operating temperature. Research is active for use at home, in automobiles, and in portable devices, and commercialization has been achieved in some fields.	~38 %
Molten carbonate fuel cells (MCFC)	Molten carbonate	600-700 ℃	Can be up-scaled easily. Biogas produced from garbage and wood can be used as fuel. Separation of CO_2 is possible.	45-60 %
Phosphoric acid fuel cells (PAFC)	Phosphoric acid	160-220 ℃	Developed for commercial use among fuel cells currently available. It has also been used for dispersed power source at plants.	35-42 %

Reference: J. Larmine and A. Dicks: *Fuel Cell Systems Explained*, Wiley (2003)
[H. Tsuchiya trans.: *Kaisetsu Nenryo Denchi Shisutemu*, Ohmsha (2004) (in Japanese)].

revolutionary micro SOFC manufacturing technology that uses the ceramic integration process technology.

2 Status of the development of energy module technology ~ Expectation of industry for compact fuel cells with high power density and operable at low temperature

As shown in Fig. 1, the industrial demand for highly efficient energy conversion at several W to several kW levels increased due to the diversified use of SOFC in various industries. The expectation is high for the compact SOFC technology that can be easily handled and is space-saving.

In order to promote the use by expanding the operating condition of SOFC as an electrochemical module made of ceramic material, it was essential to develop the micro SOFC technology that would allow rapid startup/shutdown with equivalent performance at lower temperature range of 650 °C or less compared to the conventional operating temperature (700 °C~1000 °C). The micro SOFC is a power generation technology for palm-top cells that is smaller in size compared to the conventional ones, and it enables compact module design that also is space saving. Therefore, we embarked on the micro SOFC technology development to solve the various technological issues. Figure 2 shows the advantages of the micro SOFC. In general, since the oxide ceramic material had smaller heat conductivity compared to metals, a steep temperature gradient occurred throughout the cell during temperature increase (particularly during rapid increase) when the volumes of integrated module and ceramic electrochemical cell increased, and this could cause the destruction of the cell or the module parts. One of the technological solutions was to employ a design that reduced the volume of the cell or the module parts to decrease the relative temperature gradient, as shown in Fig. 2. This reduced the startup energy, and allowed the temperature distribution of the SOFC to be easily controlled at the same time. As a result, high thermal shock resistance was obtained for the cell and the module. In planar cell module, since the generation density per volume of the unit decreased by reducing the cell volume, it was necessary to increase the performances such as the generation efficiency and power density. To do so, it was necessary to develop a new high integration manufacturing technology that could be mass produced and could increase the generation performance per volume, by increasing the electrode surface area and by controlling the unit structure such as the diameter using small tubular cell as shown in Fig. 2. Setting several W to several kW level as our output target, it was important to develop the construction technology of the module that fit within 1 L size even at 2 kW level that surpassed the performance of the PEFC. As the output power increased, the temperature control of the module became difficult. Therefore, the cell integration module technology that enabled low temperature operation and easy control of startup/shutdown was demanded.

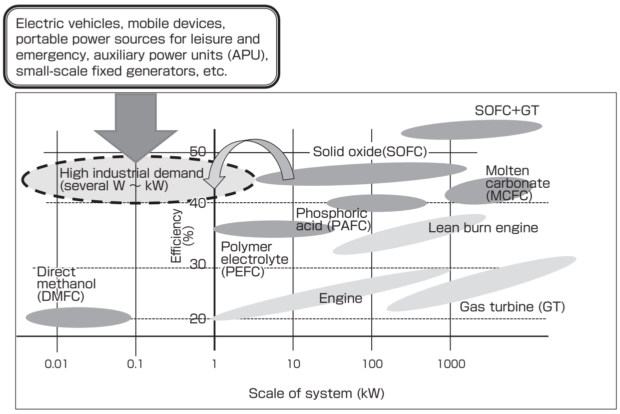

Fig. 1 Industrial development of micro SOFC

The SOFC module is composed of dense oxide ion-conducting ceramics electrolyte, electrodes (anode and cathode) that enhance the electrochemical reaction, and

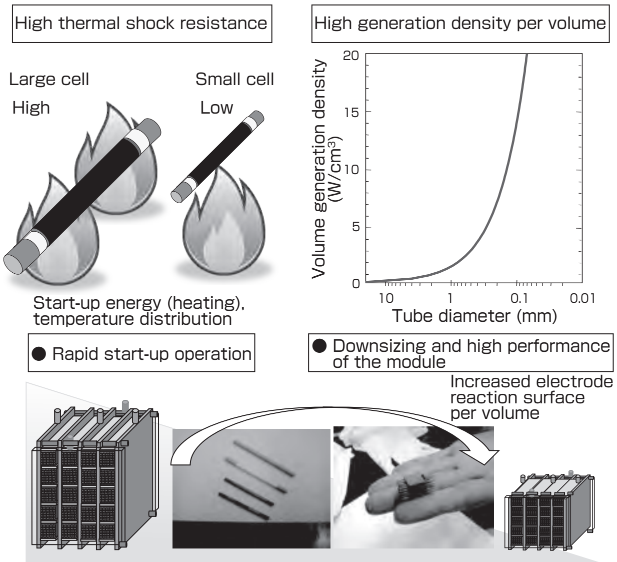

Fig. 2 Advantages of the micro SOFC module and its integration technology

fuel or oxygen (air). To create a unit structure of ordinary SOFC, the ceramic members are manufactured by forming the ceramic powder that exerts various functions into required shapes, by coating and laminating, and then firing. Therefore, according to the shape and size of the cell and integrated module, various ceramics manufacturing process technologies are employed. Moreover, since the cell and integrated modules are manufactured by multiple lamination of various functional materials with different thermal expansion, electric, and strength properties, the design of each material at nano-,micro- and macro-size levels and structural control in the fabrication process strongly affects the final power generation performance.

The challenge for the integrated module using the new micro SOFC was a difficulty of the ceramic manufacturing process technology, and it was necessary to return to the manufacturing of the cell and integrated module as ceramic parts. However, it would take too long a time to develop if we built the individual elemental technologies one at a time. It was necessary to conduct the development of ceramic parts with thermal management properties and ceramic material with electrochemical structure that increased the generation performance at low temperature, as well as the revolutionary manufacturing technology.

With such a background, the "center for the development of functional ceramic manufacturing technology" was established and the R&D was conducted under the "ceramic integration manufacturing process technology" of the "The Advanced Ceramic Reactor Development (subcontracted by NEDO, 2005-2010)" to realize the new product and to solve the issues of the highly integrated micro SOFC manufacturing technology[6][7].

As shown in Fig. 3, the PDCA of design-manufacture-analysis was conducted at the center for the development of functional ceramic manufacturing technology, and the core technology and the product realization technology were developed simultaneously. As a result, the open innovation system where the engineers and researchers of the ceramic manufacturing companies and user companies collaborated was established. As an output, it functioned as the opportunity to train industrial human resource where people could obtain academic degrees as well as produce research results. Specifically, for the development of highly integrated ceramic electrochemical module based on the new concept such as the realization of high-performance SOFC at low temperature, of integration, of high performance, and of mass production, the developments were done from the material selection and cell design (manufacturing design such as structure and size) to the prototype cell and module fabrication (structural control process in clay forming, coating, firing condition, etc.), the establishment of original evaluation and analysis technology for the new cell and module structure (thermal behavior, electrochemical property, generation property), and the general evaluation of the cell and module (improvements in structure and manufacturing process). These were discussed directly among the engineers and researchers of the manufacturer and user companies at the center. Proposals were made for the new process technology, the structural control technology upon discovery of a new phenomenon, and the utilization of new technology in industry. Through collaborations between the researchers and the corporate engineers, a flow was built toward the solution of issues for the new highly integrated module prototype and the development of materials and the elemental process technologies.

In the R&D at the center, the development of fine ceramic manufacturing process was done in collaboration with the ceramics industry in the Chubu region that has traditionally engaged in this industry. They helped build and accumulate the manufacturing technology. An optimal cycle of

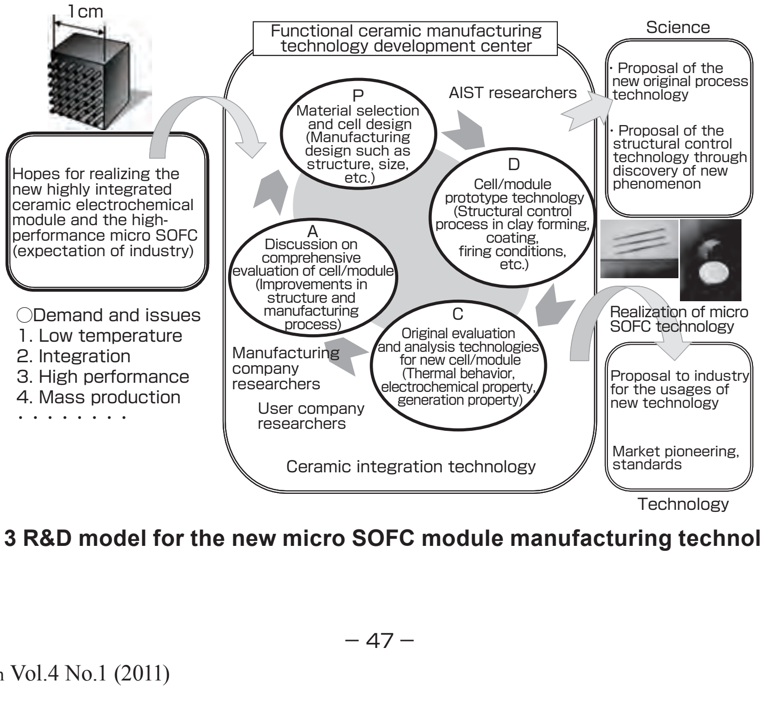

Fig. 3 R&D model for the new micro SOFC module manufacturing technology

prototype manufacture, evaluation, and analysis was set forth toward the mass production of high-performance fuel cell materials that used to be considered unsuitable for the forming technology. Moreover, since the investigation of the structural control that determined the optimal condition was done concurrently with prototype evaluation, the new high-performance micro SOFC and the highly integrated compact module manufacturing were developed in a short time.

3 Issues in manufacturing the micro SOFC for the high-efficiency compact energy module ~ Valley of death in product realization and the solution

For the micro SOFC and integrated module manufacturing technology that was not available before, it was necessary to develop the ceramic manufacturing process that could be mass produced industrially, the electrochemical design of the module of highly integrated micro SOFC, and the technologies for increasing performance. As a new manufacturing process technology under the concept of fusion of function and structure, we shall explain the manufacturing design in the R&D model of Fig. 3 and the development of new structure control process technology.

i) Highly integrated micro SOFC manufacturing and design technology

To increase the performance of the SOFC module, it was necessary to increase the electrode surface area per unit module volume, raise the degree of cell integration, and improve the mechanical strength. For the structure that fulfilled such requirements, it was advantageous to achieve high integration by both the bottom-up manufacturing where the unit cell members were combined and highly integrated, and the top-down manufacturing where the cell structure was built in later using the regularly arrayed micro-channel. To increase the performance of micro SOFC which made use of the conventional manufacturing technology of tubular SOFC, the bottom-up structure of development of the high integration of tubular SOFC was effective. On the other hand, to reduce the cost of module manufacturing and to achieve advanced cell integration structure, it was necessary to develop a new technology where the module with equivalent performance obtained in the bottom-up manufacturing was made by top-down manufacturing. In this R&D, considering the high performance and cost reduction, the R&Ds were conducted for the two types of module manufacturing technology including the tube integration module and the honeycomb micro SOFC.

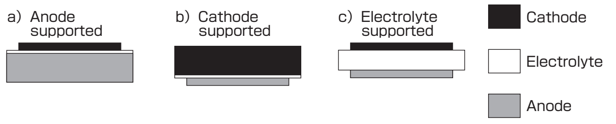

Fig. 4 Structure of various fuel cells

To bring out the advantages of high efficiency and high power density in SOFC power generation, we needed to consider the technologies to improve the reactive surface area of the electrode to enable effective progression of the electrochemical reaction of the supplied fuel, as well as the module structure that allowed the integration of current and gas flow. Ultimately, it was mandatory to select the manufacturing process technology in a form that could be mass-produced, as several cells were needed for high integration. For the reduction of cell resistance that enabled high performance at low temperature range, the support structures of the Anode, Cathode, and the electrolyte were crucial, as shown in Fig. 4. This was because the resistance became minimum in the cermet anode that was partially metalized by reduction. The tubular integrated body with high symmetry of stress distribution was superior to the planar structure as a unit structure that achieved the mechanical strength and also improved the relative surface area of the porous electrode by increasing the degree of integration.

As researches for similar microtube SOFC, there have been studies on rapid startup using the YSZ electrolyte supported SOFC at 2~5 mmφ level with high thermomechanical strength[8][9]. However, there were very few developments for the manufacturing technology for high performance such as achievements at low temperature range of 650 °C or less or the development of small integrated modules. Our challenge was to manufacture high-performance SOFC and integrated modules unseen before, and therefore, we investigated the manufacturing technology of the integrated modules composed of anode supported micro SOFC. Moreover, although there were only a small number of studies since the mechanical strength was low and forming was difficult, we developed the manufacturing process using ceria electrolytes with high oxide ion conductivity at low temperature.

As the technologies for manufacturing and design of the microtube SOFC and integrated module with fuel gas pores

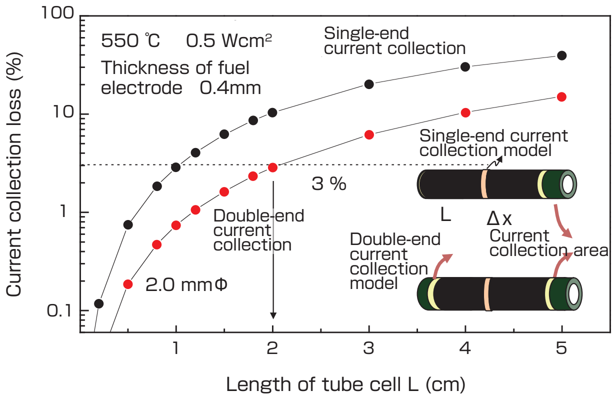

Fig. 5 Design model and the results of current collection loss calculation for the integration of the micro-tubular SOFC design technology

of 2 mmφ or less, the optimization of the cell form (thickness of electrolyte and electrode, optimal cell length, etc.) that affected the final module generation capacity was important. In the anode supported cell, because the electrode has the roles of reactive field in the three-phase boundary for the electrochemical reaction and of the collector of current generated in the reaction, its design greatly affected the generation performance of the cell and integrated module. Figure 5 shows the results of the different power collection methods and the cell form for achieving high performance in the Anode supported microtube SOFC and its integrated module. The results were used to design the length of the cell collector. The collection resistance increased when the long cell design was used to extend the electrode surface area of the single cell structure, and the power generation output decreased (current collection loss).

The design technology needed to manufacture the microtube SOFC with exterior diameter 2 mmφ (interior diameter 1.6 mmφ, electrode film thickness of 0.2 mm) is explained. Assuming the generation performance to be 0.5 W/cm^2 at 550 °C, the current collection loss in the equivalent circuit was calculated using the single-end and double-end current collection models. The relationship of the current collection loss arising from the current collection resistance factor and cell length is shown in Fig. 5. When the length whereby the current collection loss in power generation would be 3 % or less was calculated, it was found that the integrated module must be designed with cell length of 2.0 cm for double-end current collection and 1.0 cm for single-end current collection. This showed that it was necessary to increase the thickness of the double-end current collector and anode in order to increase the surface area of the generation electrode by increasing the cell length[10]. Conversely, since it was necessary to decrease the electrode thickness to increase the cell performance, the cell length optimization was important to improve the integrated module generation performance at low temperature. Under such design guidance, the integrated module manufacturing technology was developed by bottom-up design, and the mass-producible cell manufacturing was developed by improving the film coating technology and the forming precision in the extrusion technology[11]. As a result, the anode supported micro SOFC using the 2.0 mmφ ceria electrolyte achieved a high power density of 1.0 W/cm^2 at 570 °C[11]. Moreover, this high-performance cell (microtube cell) was combined to fabricate the module structure integrated inside porous ceramics, and the optimal cell arrangement within the module was studied by similar equivalent circuit simulation design. As shown in Fig. 6, by calculating the current collection loss in the integrated module model, it was found that the conductivity over 100 S/cm was required at cell interval of 1.0 mm for the current collector members (between cells). The 2 W level generation unit was realized where several microtube SOFC with 2.0 mmφ diameter was integrated in a space the size of a sugar cube. In this investigation, the design and manufacturing technology for the integrated module (cube module) with generation performance over 2 W/cm^3 at 550 °C was developed, and it became possible to fabricate various integrated module structures such as of the serial connection[12].

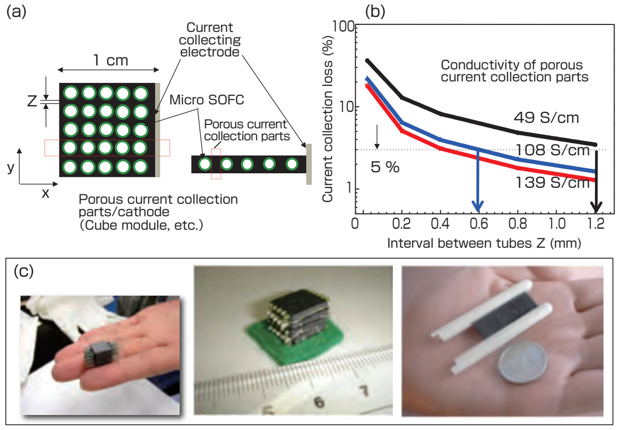

Fig. 6 Cube module design and the developed integrated module
a: Design model of integrated module
b: Result of current collection loss calculation at 650 °C
c: Example of developed module

ii) Cell structure control technology in the advanced coating process

In achieving high performance for the SOFC and integrated module, it was necessary to develop the manufacture process technology that could be applied to macro connections by creating the multilayer structure of different materials such as ceramics electrode and electrolytes based on the electrochemical structure design at nano to micro size. Moreover, it was necessary to develop a simple and mass-producible manufacturing technology such as wet coating that could effectively control the degree of cell integration without being influenced by the composition of base material on which the cells were arranged. For the manufacture of electrodes for the ceramics electrochemical device such as SOFC, new developments of the various cell forms, composition control, and layer structures were necessary, and both the coating technology with high degree of freedom to form functional ceramics and advanced 3D coating technology had to be established. The increased density and formation of the electrolyte film and the structural controllability in cell structure formation had to also be increased. To form the film structure necessary to increase performance, we embarked on the development of the manufacturing process technology that allowed even slurry coating in sub-millimeter 3D space, by advancing the new wet coating manufacturing process technology.

Figure 7 shows the characteristics of the various wet ceramics coating processes. For the wet paste coating on ceramic base material, the control technology for forming the dense electrolyte at single μm thickness was realized for the tubular cell using the dip-coating method[11]. In ordinary dip-coating method, the film formation on the exterior of the base material such as the tube was possible. However, when the electrochemical functional layer had to be formed on the interior wall of the microspace, application of an even coating on the whole surface was difficult, as the slurry would not penetrate deeply due to the balance of viscosity resistance and capillary force. The space could be filled with the slurry by flooding the interior by slurry aspiration method and then spewed out, but the film on the interior wall became thick and uneven, and the coat volume could not be controlled as the number of pinholes increased. To solve these coating process issues, a unique coating process called the slurry injection method was newly developed, where the external force that counteracted the capillary force was added to the coating paste, and the coating volume was controlled by forcefully moving the paste material[13]. With this new top-down manufacturing technology, it became possible to use the microspace in the honeycomb structures with regular 3D pinhole arrays of sub-millimeter diameter, and to form the even multilayer film with controlled film thickness. This method was important for the fabrication of the integrated module structure and for cost reduction by reducing the number of members. In the developed process, the even film coating could be formed on the substrate under the same control condition even in corner areas where the liquid tended to collect during the coating process. The controlled functional layer could be formed in the micropore with sub-millimeter diameter on the ceramics base material, using a simple coating process regardless of the shape of the pore. This developed process technology was adapted to the multilayer coating of the ceramic electrochemical structure of the electrolyte and electrode layers. By utilizing this process for the cell formation in the regular array structure with sub-millimeter diameter, the top-down manufacturing method allowed the fabrication of the electrode unit with regular array of pores in the sub-millimeter space by using the honeycomb extrusion technology, and then later forming the multilayer cell structure such as the dense electrolyte film and porous electrode by combining the coating technology.

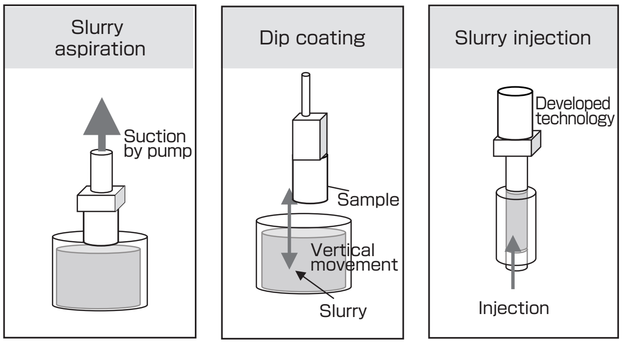

Fig. 7 Wet ceramic coating technology

Table 2 Technological indices for the developed micro SOFC technology

	Cell diameter (mmϕ)	Electrolyte material	Start-up temperature (℃)	Power density (W/cm^2) at 0.7 V	Start-up speed (℃/min)
Developed micro SOFC technology	Exterior diameter: 0.8-2.0 (Interior diameter 0.4-1.6)	ScSZ, GDC	550 – 650	0.5 – 0.8* @ 650 ℃	65 – 217**
Korea Institute of Energy Research (Korea)	Exterior diameter: 10.0	YSZ	750	0.45	20
Adelan Ltd. (U.K.)	Exterior diameter: 2.0	YSZ	850	0.3	200

ScSZ : 10 mol% Scandia-stabilized zirconia, YSZ : 8 mol% Yttria-stabilized zirconia, GDC : 10 mol% Gadolinia doped ceria

Reference) Data updated based on V. Lawlor, S. Griesser, G. Buchinger, A. G. Olabi, S. Cordiner, D. Meissner: Review of the micro-tubular solid oxide fuel cell, Part I. Stack design issues and research activities, *Journal of Power Sources*, 193, 387-399 (2009).

* Data for 2.0 mmϕ ScSZ electrolyte micro-tubular SOFC

** Demonstration data for honeycomb SOFC

Using this technology, the dense electrolyte with thickness 10 μm and the electrode with tens of μm thickness on the bulk body (40 cm^2/cm^3, relative surface area per volume about 20 times the conventional flat SOFC) where there were hundreds of spaces at 0.5-1.0 mmφ diameter were successfully formed, and the new honeycomb micro SOFC was developed[13].

The design-to-manufacturing process technology that was important for the integration of the micro SOFC module was built as the top-down and bottom-up manufacturing technologies, and the new manufacturing technology was presented for the 3D integrated structure in the ceramic electrochemical device manufacture.

4 Realization of the new low-temperature operable micro SOFC manufacturing technology through revolutionary ceramics manufacturing technology ~ Conversion to full-fledged integrated module

We could now fabricate an original unprecedented micro SOFC, based on the new high-performance micro SOFC design and manufacturing technology. As a result, high performance of the micro SOFC technology was achieved in the technical indices such as size, power, temperature reduction, short startup time, and others, as shown in Table 2[14].

To increase generation performance at the low temperature in micro SOFC, the reduction of the structural resistance factors such as the reaction dispersal and ohmic resistance of the cell and integrated module was essential. Much attention was devoted to the film forming technology of the electrolyte layer involved in the reduction of the resistance factors, the analysis of material contraction behaviors in the aforementioned slurry dip coating process, and the co-sintering of laminated material. We succeeded in forming a flawless solid electrolyte film with thickness of single μm in the cycle of the R&D model shown in Fig. 3. Also, the electrochemical reaction resistance and the reaction dispersal were confirmed in detail through the original evaluation and analysis of the micro SOFC prototype using the zirconia electrolyte (ScSZ: Scandia stabilized zirconia) at temperature lower than 650 °C for which there were few precedents. In the low temperature range, it was newly found that the resistance factors of the fuel cell changed according to the operation condition and contributed greatly to the increased generation performance. As shown in Fig. 8, by the optimization of the ceramics manufacturing process, high porosity surpassing 50 % was realized for the anode, and it was found that this greatly reduced the reaction resistance of generation at low temperature range. Figure 8b shows the relationship of the porosity of anode and the cell impedance resistance value at 600 °C. As shown in Fig. 8b, it was confirmed that the reduction resistance values represented as arcs decreased in relation to the increase of porosity of the anode. As a result, the output power performance surpassing 1 W/cm^2 was realized in the low temperature range of 600 °C, as shown in Fig. 8c. From the post-reaction observation of the electrode structure, it was thought that the reduced nickel became nano particles in the electrode structure with high porosity, a high dispersal structure was formed, and this led to the increased number of three-phase boundaries that provided the active sites[15]. For the realization of this technology, the major factor was that the ceramics companies and others were able to achieve high properties at the cell manufacture level through the extrusion and wet coating processes that enabled mass-production and cost reduction. By considering the manufacturing process technology that incorporated the PDCA cycle in the R&D model in Fig. 3, we were able to achieve the micro SOFC manufacturing technology at low temperature range of 600 °C with the same performance as the zirconia electrolyte SOFC with power density 1 W/cm^2 at 700−800 °C[16].

Attentions were drawn in Japan and from abroad to this highly integrated module of fingertip or palm size that was distinctly different from the conventional energy module. Through the manufacture and evaluation in collaboration with user companies, it was demonstrated that these cells could be used to manufacture integrated modules of several hundred W level and were capable of realizing efficiency surpassing 40 % as fuel cells[17]. The future issues will be the development of kW class modules using the integrated modules composed of the developed microtube SOFC, as well as the development of the low-cost manufacturing technology.

In the honeycomb micro SOFC development shown in Fig. 9a, it is necessary to create the electrochemical module of the integrated cell and to ensure the gas seal between the honeycomb SOFCs. As shown in Fig. 9b, a new integrated module technology was developed to handle rapid thermal history utilizing the thermomechanical property that was the stronghold of the honeycomb structure by forming the joint structure using the silver-silica paste as the interconnect. It became possible to manufacture an arbitrary serial structure unit by combining the highly integrated structure of several hundred cell/cm^3 using this SOFC module technology. Also, by utilizing the easily warming property of the micro SOFC structure that has high relative surface area and low relative heat capacity and by confirming the electromotive force and current value, as shown in Fig. 9c and 9d, we proposed the micro SOFC module manufacturing technology that could handle 3-5 minute rapid startup, which was one of the required technical issues[18]. Also, the output power performance per unit volume at 650 °C was 2.8 W/cm^3 or equivalent to the tubular integrated module, and high conversion efficiency could be expected for the SOFC. The

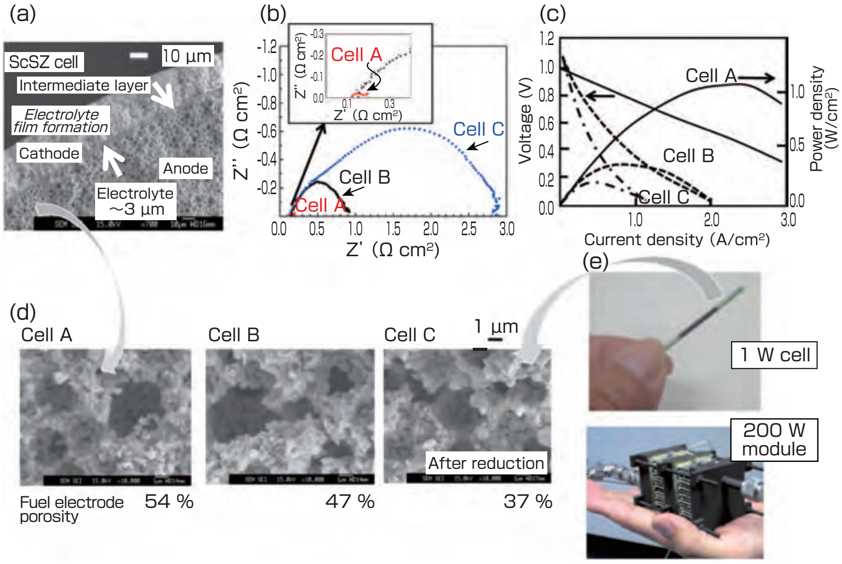

Fig. 8 Realization of zirconia low-temperature micro SOFC module
a: Photograph of cell cross section
b: Relationship of electrode porosity and electrode resistance (600 °C)
c: Generation performance (600 °C, humidified hydrogen)
d: Structure of developed porous fuel electrode
e: Example of developed cell and integrated module

development will continue for easily usable and inexpensive SOFC module utilizing the advantages of high cell integration and rapid startup of the honeycomb micro SOFC, and our aim will be the increased generation performance of the module through seeking solutions for issues such as low temperature.

The developed cell and integrated module technology was a unprecedented totally new ceramic integrated structure composed of small extruded members unseen before, and there were many comparisons with the conventional technology right from the start. Particularly, questions were raised concerning the generator density and generator module structure, that it may not be possible to create a generator module for practical use if the performance is merely the same as the current cells and integrated modules. On the other hand, the micro SOFC achieved the high power density of the same level as 800 °C in the low temperature range of 500−650 °C, and academically significant experimental results were accumulated since the strategic design ~ material and manufacture process technology ~ evaluation technology were rebuilt for the cell design and its realization. This technology realized the module structure for low temperature generation through electrical serial structure and sugar cube size 2 W/cm^3 module. The expectations for a readily usable, compact SOFC module are high.

5 Summary ~ Product realization and creation of a new market

In the future energy-related manufacturing industry, the micro SOFC and integrated module technology is an important technology that utilizes the advanced ceramic materials and manufacturing technology at the level of nano-micro-macro size for which Japan takes the lead. On the other hand, the SOFC technology is mainly geared for the fixed power generation facilities. Our micro SOFC manufacturing technology has cleared the low temperature, rapid startup, sufficient output power, and compact size that are necessary for the module for power generation, future automobiles, portable generator technology, and is geared for new innovations and product development. At this point, the generator module fabrication for several tens W to several hundreds W level have been demonstrated. Currently, the technological issues according to usage are being organized, and developments are done toward high performance with multiple fuel and module fabrication at kW level. It is also possible to propose the usages to industry taking advantage of the characteristics of the micro SOFC. One proposal is the range extender technology using the vehicle-mounted power generator and hybrid technology that uses the internal combustion engine to extend the cruise distance of the electric vehicle that is now being developed aggressively[19]. Our compact generator module can be used in the power source technology as the high-efficiency generator module that can achieve the energy conversion efficiency of 50 % or above (well-to-wheel) that surpasses the limits of the internal combustion engine[19]. The development of fuel cells that do not dependent on hydrogen infrastructure using the advantages of SOFC multiple fuel use is drawing attention. In the future, further improvements must be made for the rapid startup/shutdown properties and the reliability of performance with multiple fuel use, and the technical issues for the requirements of the mobile generator module must be extracted and solved. The development of a safe and low-cost module is an important subject in the future nanotechnology material and manufacture. The priority will be to deliver the easy-to-use, low-cost fuel cell technology to as many industrial fields as possible, for the efficient use of resource and energy and for the realization of a low-carbon society. To do so, it is necessary to continue the development in many industrial fields for the new micro SOFC and its integrated module for which our experience was accumulated by tackling the problems at the center for the development

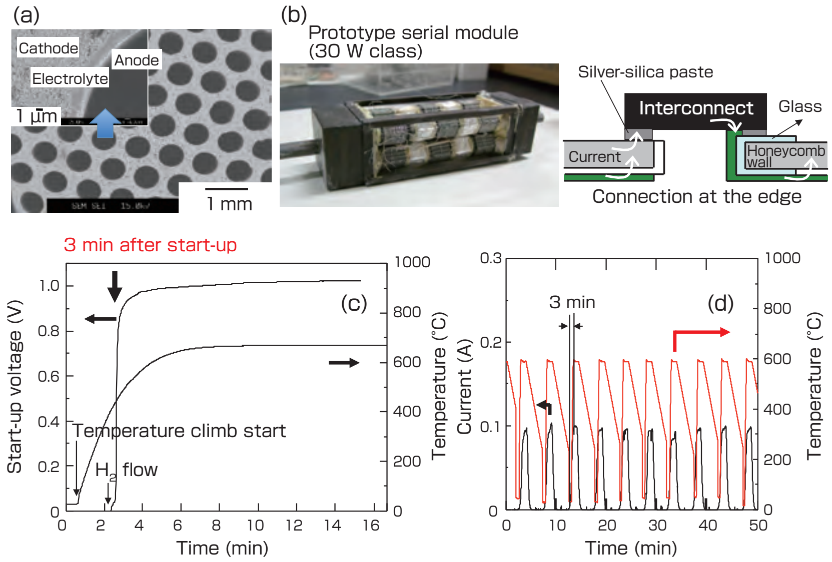

Fig. 9 Honeycomb-type micro SOFC module capable of rapid operation
a: Integrated cell with honeycomb structure
b: Example of prototype module and connecting structure
c, d: Rapid start-up and generation property through thermal history

of functional ceramics manufacturing technology. We aim for the development of an original technology that leads the world in ceramics manufacturing technology, including the standardization of the micro SOFC manufacturing technology.

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Discussions with Reviewers

1 Overall evaluation of the paper

Comment (Toshimi Shimizu, Deputy Director-General for Nanotechnology, Material, Manufacturing, AIST)

This paper is a description of the ideas, the prototype fabrication, and the evaluation results for the power generation module that is compact, has high output, and is highly efficient. It was realized by utilizing the original ceramic integration and manufacturing technology. It contributes greatly to solving the energy issues that are raising social concerns today, and I feel it is appropriate as a paper for *Synthesiology*.

However, in general, the logical construct and the expressions are like project reports or technical manuals. Therefore, while it can be understood by readers who are versed in fuel cells and related technologies, the text including the terminologies and figures is rather difficult for other readers. I think it will be a readable and fulfilling paper by improving the points indicated in the following discussions.

2 Basic positioning of the R&D

Comment (Hiroshi Tateishi, New Energy and Industrial Technology Development Organization)

The points and the flow of technological development are overall appropriately organized but the explanation of the strategy for R&D is weak as a paper of *Synthesiology*. I think the following three points are problems in terms of synthesis, so please consider revising them.

(1) I think there is a lack of explanation on what social significance this development has. If it is a development of the ceramic reactor the content can stand as is, but since you state specifically that it is a development of SOFC, you need to provide a corresponding explanation. The technological goal of realizing characteristics never seen in conventional SOFC is clear, but it is unclear what you wish to accomplish with the results to be obtained. You need to state that in the beginning: “There are A, B, and C that are applications that could not be handled by the conventional SOFC, and we set as our goal such-and-such performance and cost for the specs required.” In reality, the application tends to become visible later in the course of events, but in a paper you should state the objective of the development at the beginning.

(2) You set as the targets for SOFC, the low temperature operation and the speeding up of startup and shutdown. However, these issues are not directly related to the output capacity and aside from the required parameters, they are issues demanded in large-capacity devices as well. This means they are not issues limited to the capacity set as the target in this paper. The creation of a “micro-module” is clearly effective in solving these issues, but in this paper the establishment of the relationship between the above two issues and the output capacity is not sufficiently described. Does the micro-SOFC technology target only the 10~100 W class devices, or will it be extended to large-capacity devices in the long run? You may not be able to achieve this technologically at this point, but what is the strategy you have in mind? I don’t think you will be evaluated highly by electric power users if these points remain unclear.

(3) The relationship between the microtube module and the honeycomb module is not clearly stated in the paper. I think that tubular comes first then honeycomb appears in terms of chronology, but what are their characteristics, will they be used according to different uses in the future, or will you eventually settle on one structure for practical use? What are the future issues for the two structures? You need to explain these points.

Answer (Yoshinobu Fujishiro)

As social significance, when the highly efficient SOFC is used particularly in the homes, significant reduction in CO_2 emission will become possible if it can be operated by DDS (daily start and stop) according to the power load used. To do so, the realization of SOFC that is compact, of high performance, capable of rapid startup/shutdown, and that has high performance at low temperature to allow simple heat management is necessary. Until now, there was no technology to enhance the high performance or high integration by increasing the electrode surface area per volume and to address the issues of resistance of the ceramics material. The greatest significance of the research for the realization of high-performance, compact SOFC is that the ceramics integration technology developed by AIST was utilized to achieve the high integration and high performance at low temperature that were not possible before. We see the research strategy as the development and technological diffusion (through the presentation of readily-usable module) of the functional ceramics member manufacturing technology.

Specifically, we considered the following social significance and modified the text.

(1) We reconsidered the relationship of the issues of output

capacity and the solution by low temperature operation and speeding up the startup/shut-down.

The target is set for generation module, and we assumed the kW class module that is highly in demand as civilian power source. As the module capacity increases, the module volume increases and the exchange of heat rises. The technology to manufacture a module that can withstand low temperature and rapid startup/shutdown using the micro module SOFC will be effective from the aspect of solving the heat control issues.

We added the description of the target for developed output capacity and the thoughts on solutions by low temperature operation and startup/shut-down control.

(2) We reconsidered and modified the paper in response to your indication that the flow from microtube to honeycomb SOFC is unclear, and that there is a need for future issues.

3 Current situation, issues, and strategy for solution of the micro SOFC technology

Comment (Toshimi Shimizu)

I understand that there is importance in the development of compact, high-output, low-temperature fuel cells to meet the demands of industry and society from the descriptions in the first half of the paper. However, the main part or the development trend, comparison of performances, problems, and other things about the micro SOFC technology are not described. However, in the latter half of the paper, you abruptly bring out the comparison chart of performances, and conclude that you realized an original micro SOFC never seen before. The readers want to know the details of the table, and the strategy of this research and the synthesiology. I think the table should be utilized more effectively in the first part.

Answer (Yoshinobu Fujishiro)

As you indicate, we understood that the descriptions of development trends, comparison of performances, and problems of the micro SOFC technology are insufficient in the text, and also that the discussion of the research strategy for the solution is weak. On the other hand, the micro SOFC technology and its module development have not progressed very far in Japan and abroad among the various SOFC fields. It is not a generally major technological field, and the research is carried out with the advantage of AIST, and we think it is a field for which the technological development should be pushed by defining the issues. We wish to emphasize that, along the technological strategy road map, one of the solutions for the compact, high-output, low-temperature fuel cells is the realization of the micro SOFC module technology, and I hope the readers understand this. There is no technological index for micro SOFC, but we presented the comparison table to clarify the positioning of our technology and to show the world benchmark. For the above reason, we shall reconsider if it is necessary to review the synthesis. We added some description of the technological issues and explanation to emphasize that the technological issues for SOFC is the realization of low-cost, readily usable, high-output, and low-temperature fuel cells, and this is a strategy for the CO_2 reduction technology.

Comment (Hiroshi Tateishi)

This point is related to Discussion 2. In Table 2, there is no explanation of the technological indices shown here and I am unable to see how the technology shows “high performance”. Also, it is unclear whether the results shown in the table are the results of the microtube or those of the honeycomb integration.

Answer (Yoshinobu Fujishiro)

The technological indices are values achieved in microtube SOFC and honeycomb SOFC for the cell form, material, and operating temperature. For the generation density, since the anode thickness is thinner in honeycomb than in microtube SOFC, the maximum value is the value for microtube SOFC. We added the output density for honeycomb SOFC and included an explanation. The rapid startup (217 °C/min) in 3 min was demonstration data for honeycomb SOFC, and this will be stated in the note. Even with the microtube SOFC, startup can occur in a few minutes for a single cell, it is about 10 min (65 °C/min) at 200 W level by burner startup in the module.