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Research and Development Trends in Solid Oxide Fuel Cell Materials — From the Viewpoint of Electrolyte-Related R&D as Key —

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1 Introduction

Fuel cells (FCs) convert the chemical energy of fuels directly into electrical energy and therefore take on high generating efficiency even in small-scale systems. FCs also produce high quality waste heat which can be utilized effectively. Unlike conventional primary cells, which reach the end of their useful life when the reactant substances are consumed, and secondary cells, which have the drawback of reduced performance due to the buildup of unnecessary products with repeated charging, the generating performance of FCs can be maintained as long as fuel is supplied, and depending on the fuel selected, FCs discharge virtually no air pollutants. Among the various FCs technologies, solid oxide fuel cells (SOFCs) are particularly attractive, as generating efficiency is high, the FCs supply both heat and electricity, and a diverse range of fuels can be used. As a result, this type has been the object of intensive R&D (research and development) worldwide in recent years. In comparison with other types of FCs, SOFCs have high thermal efficiency and excellent long-term performance stability, and assuming the establishment of cell and stack production processes suitable for mass production, a substantial reduction in system cost can be expected. Specifically, because the systems studied to date operate in the high temperature range, it is possible to achieve an electrochemical reaction at the electrodes simply by supplying fuel and air without using an expensive noble

metal catalysts, which mean that low cost component materials can be used in the cell.

This article will review the background and expectations placed on SOFCs R&D and the current status of technologies in Japan and other countries. First, the operating principle and structure of SOFCs are introduced. In particular, the current state of R&D on the electrolyte and electrode materials used in SOFCs and the features required in those electrolytes and related issues in high temperature SOFCs are identified. As a solution to these problems, methods of elucidating the ion conduction mechanism and damage/deterioration mechanism are described. On the other hand, as a completely different approach aimed at solving all of the technical problems of high temperature SOFCs at once, R&D on novel electrolytes with lower operating temperatures than the conventional systems have attracted considerable attention. This article will propose a method of searching for suitable electrolytes in low- and medium-temperature operation.

Background and expectations placed on R&D on SOFCs

2-1 SOFCs as high efficiency generating systems

The relationship between the scale of various types of generating systems and their efficiency is shown at the left in Figure 1.^[1-10] In comparison with gas turbine systems, SOFCs offer higher generating efficiency, as well as molten carbonate fuel cells (MCFCs), polymer electrolyte fuel cells

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Figure 1 : Current efficiency of various power systems and comparison of the output power performance between various power generation systems

PAFC: Phosphate Acid Fuel Cells

Prepared by the STFC based on references [1-10].

(PEFCs), and others. Even higher total efficiency can be realized with an SOFCs combined cycle system (system combining an SOFCs system and a gas turbine), and when hydrogen is used as the fuel, the system discharges only water. Even when using a diverse range of fuels with higher energy densities, such as natural gas or coal gas, emissions of NOx and SOx are lower than systems with other fossil fuel systems. However, with the high temperature SOFCs developed to date, which have operating temperatures exceeding 750°C, heat control is difficult, in that long time is required for starting, etc. For this reason, only limited R&D has been carried out on portable systems which have a high start/stop frequency, and stationary systems have been the main focus of R&D.

As the most representative type of FCs except SOFCs, PEFCs are also an object of R&D. This type has a low operating temperature of 80°C-100°C, enabling easy handling, and because the system is compact, practical application as a power source for moving objects is progressing. However, PEFCs have the disadvantage of high cost because platinum catalysis or the catalysis carrying carbon and fluorine resin mixtures, etc. are used. Among other problems, if a reformed gas other than hydrogen is used as the fuel, cell performance tends to deteriorate due to exposure of the platinum catalyst of the electrode to carbon monoxide, and the performance of the electrolyte film itself may be reduced by long-term operation. A comparison of the relationship between the unit energy density (Wh/kg) and unit output density (W/kg) of various FCs (SOFCs, PEFCs, MCFCs, PAFCs) and other types of cells is shown in Figure 1 at the right. At the current stage, FCs output density is still inferior to that of Li ion cells, but FCs have the advantages of high energy density and quick fuel replenishment. Thus, the challenge for the immediate future is to improve FCs output power density. Currently, application development is progressing in fields where electrical energy is required for extended periods of time, taking advantage of the feature of high energy density.

2-2 Promising fields for application of SOFCs

Promising fields for application of SOFCs are shown in Table 1, classified by the mode of the power source in uses and main applications. A diverse range of applications is expected for SOFCs, from micro systems to medium- and largescale systems. In particular, high expectations are placed on distributed power sources, stationary power sources, and cogeneration systems (simultaneous supply of power and heat or hot water), especially in Europe and North America. Furthermore, steady progress is being made in application development for a home power source, auxiliary power supply for automobiles, and portable power source for mobile electronic devices and the like as well as for an alternative system for conventional power. In particular,

System scale		Purpose Mode of use	Main application	Main issue	
Ultra-small scale	<1kW	Main power source On-board system Stationary	Robots Computer servers Mobile devices	Improvement of output	Compact size Low temperature operation Following load change
Small scale	1~100kW	Moving object (on-site) Stationary Cogeneration Automobile	Home use Automobile auxiliary power unit Electric vehicle Unmanned telecom base Charging device	power density Maintaining long-term performance High reliability Cost reduction	
Medium-to- large scale	>100kW	Installed system Main system Cogeneration Emergency use	Ship Factory Commercial equipment Hospital	(material, manufacturing process)	Medium temperature operation

Table 1 : Fields of application for SOFC systems and main issues for the applications

Prepared by the STFC.

SOFCs is superior to competing technologies. As markets that take advantage of the distinctive features of SOFCs, cogeneration systems which operate steadily at a low capacity of 10kW or less for household and commercial service are considered important. If an economical fuel can be used, it is assumed that these systems will be introduced in commercial markets at an early date.^[10-12]

With regard to the conditions surrounding the introduction of SOFCs in Japan, it is generally thought that introduction as a distributed power source for household service, which is outside the scope of power industry liberalization, is more advantageous than constant load operation as a distributed power source for medium- to large-scale power generation. Due to their high generating efficiency, SOFCs are expected to have market potential even when limited to power generation outside the cogeneration field (i.e., without assuming heat generation). In the initial stage of SOFCs commercialization, it is generally thought that the first priority will be system reliability rather that cost, and introduction will be possible even at a system cost of ¥200,000/kW or less.^[10] SOFCs auxiliary power units (APU) for moving objects have been studied at outputs from the several kW class of automobiles to the several 100kW class for large-scale ships.^[13-17] In particular, because APU are used for steady loads, the number of starts and stops is small and quick starting time is not required. From this viewpoint, application of SOFCs is considered feasible.

3 Current status of R&D on SOFCs systems

3-1 Status of R&D on SOFCs in Japan and other countries

Figure 2 shows the roadmap for the technical development of stationary SOFCs in Japan prepared by the Ministry of Economy, Trade and Industry (METI) and the New Energy and Industrial Technology Development Organization (NEDO), and the roadmap for the development of SOFCs systems in the Solid State Energy Conversion Alliance (SECA) project of the Department of Energy in the United States.^[18-21]

In Japan, R&D on SOFCs began in fiscal year 1981 under the Moonlight Plan. As subsequent NEDO projects, R&D proceeded under Phase I (1989-1991), which focused on cell stack assemblies of several 100W class, Phase II (1992-2000), which prioritized cost reduction and reliability in cylindrical and flat-type SOFCs, and Phase III (2001-2004), which continued R&D on practical SOFCs systems (10+ kW class). The R&D issues for practical application of SOFCs in Phase III included improved cell performance, high output, cost reduction, and high reliability.^[10] Concretely, in order to secure the high reliability and cost competitiveness necessary for full-scale commercial introduction of SOFCs, the mechanism of deterioration of cells and stacks under long-term service conditions was elucidated and countermeasures were proposed, costs were reduced by downsizing,





Prepared by the STFC based on references [18-21].

which was enabled by realizing high output, and element technologies were developed, including technologies for responding to various types of fuels and operating conditions.^[18,19] Figure 2(a) summarizes future technical issues in connection with stationary SOFCs systems and the expected timing of realizing those technologies over a timeframe extending to around 2020.^[18] In particular, from 2004 onward, the priority is attached to technologies for improved reliability and stack technologies for the popularization period, based on a strong presumption that SOFCs will in fact be introduced and popularized. Especially with regard to systems from smallscale distributed power systems to medium-scale thermal power generation substitute systems with high operating temperature of around 950°C, the priority is assigned to the development of systems with an awareness of commercialization

and confirmation of performance by operational demonstration.^[22,23] In parallel with this, the R&D has already been begun on small- and medium-scale systems with medium-range operating temperatures of 700°C-800°C.^[4]

At present, however, the most representative SOFCs R&D project from the global perspective is the SECA Project of the US Department of Energy, which can also be considered the most advanced project. In the United States, development of FCs for use in the electric industry began as the DOE's Vision 21 Program. Subsequently, the SECA Project was launched in 1999, and a strong cooperative system with industry, university, and government organizations was created. At present, the SECA Project is being implemented with all operations carried out by the National Energy Technology Laboratory (NETL). As targets, a low cost technology and technologies related to high performance and high reliability are to be established by 2010. A system with 45-50% HHV efficiency (higher heating value; a standard that considers the latent heat of water vaporization) is to be developed using coal gas, natural gas, or similar fuels, and a cost target of US\$400/kW is to be achieved by around 2015.^[13-17, 20, 21]

In both the United States and Japan, commercialization of SOFCs has not progressed as originally hoped, but nevertheless, the impetus toward practical application is accelerating through confirmation tests and other activities. In both countries, intensive confirmation tests are being conducted with the aims of maintaining power generating performance over the long term, reducing costs, and achieving high reliability, as these are the key issues for practical application, and confirming the effectiveness of the related measures. Furthermore, after confirmation of long-term generating performance and high reliability, both countries plan to introduce this technology in the market beginning with high temperature (operation at 750°C-1,000°C) medium- and large-scale systems.

Private-sector R&D is also being carried out independently of these government projects. Representative examples of recent R&D in the private sector in Japan include the following.^[24]

• A prototype of a SOFCs system operating at 500°C using cells developed by Toho Gas Co., Ltd. and the National Institute of Advanced Industrial Science and Technology (AIST) was

produced on a trial basis, targeting practical application of stationary systems for home and commercial use around 2012.

- Osaka Gas Co., Ltd. is jointly developing a SOFCs cogeneration system for household use with Kyocera Corporation, with commercialization scheduled for fiscal year 2008 or thereafter.
- The Central Research Institute of Electric Power Industry (CRIEPI), in joint work with AIST, is currently developing a SOFCs system for household use with an operating temperature of 500°C-650°C, and has achieved generating efficiency on a practical level with a prototype system.

3-2 Operating principle of SOFCs

SOFCs include the oxygen ion conduction type and the hydrogen ion conduction type. Figure 3 shows schematic diagrams of the operating principles of these SOFCs. As shown on the left, in an oxygen ion conduction type SOFC, oxygen ions are conducted through the electrolyte and combine with hydrogen at the fuel electrode, forming water. Power is produced between the electrodes when electrons are discharged in the water formation process.

On the other hand, as shown at the right in Figure 3, in the hydrogen ion conduction type SOFC, electrons are discharged when hydrogen ions form at the fuel electrode, and power is produced between the electrodes when these



Figure 3 : Operating principles on hydrogen and oxygen ion conduction type SOFCs (schematic diagrams of two-chamber SOFCs)

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hydrogen ions pass through the electrolyte.^[25-27] Because only chemically unstable hydrogen ion conducting electrolytes have been discovered to date, little work has been done on this type of electrolyte. Virtually all R&D has involved SOFCs which utilize an oxygen ion conductor.

Figure 3 showed the operating principle of the two-chamber SOFC. In this type, the fuel and air are supplied to separate chambers. The fuel electrode and air electrode are positioned in the respective chambers and are connected via the electrolyte. In the single-chamber SOFCs, a mixed gas of fuel and air is supplied to a single chamber. When the electrolyte is an oxygen ion conductor, oxygen ions are generated from the air electrode in the mixed gas. These ions are conducted through the electrolyte and react with hydrogen at the fuel electrode, producing an electromotive force between the two electrodes. Because the fuel and air are not separated in the single-chamber SOFCs, research reports have pointed out that this type is superior in various respects, in that a more compact design is possible in comparison with the two-chamber type, mechanical strength and durability are easily secured in the system as the structure does not include a separator, and quick starting is achieved as a result of heat generation at the electrodes.^[28-32] As disadvantages, singlechamber systems are prone to direct oxidation of the air and fuel, and no electrolyte with high ionic conductivity comparable to that used in two-chamber systems has yet been discovered. For these reasons, R&D is overwhelmingly concentrated on the two-chamber type at present. However, assuming the above-mentioned problems can be overcome in the future, the single-chamber type is considered to offer many advantages, particularly in small-scale systems.

3-3 SOFCs stack structure

The cell structure of two-chamber SOFCs systems is categorized as cylindrical or flat. In the cylindrical type, the contact area at the interface between the electrolyte and electrodes is small and as a result, the current path in each cell is long and Joule loss is large. In view of this drawback, R&D on small- and medium-scale systems has focused mainly on the flat type. Figure 4 shows a schematic diagram of a flat stack system and an example of a stack.^[33] In order to obtain high output, the stacks are constructed by connecting single cells in series. The separator where the cells are connected in series performs the functions of transmitting electrons and separating the fuel gas and air. To obtain a satisfactory electrical connection, contact at the interface between the electrolyte and the electrodes is important, while control of the porosity of electrode material is critical for securing the flows of fuel and air at the respective electrodes. Moreover, the electrolyte and the separators must possess sufficient density to prevent mixing of the two gases.

The following will describe the current status and issues in R&D on electrolytes, which have the greatest effect on SOFCs generating performance, as well as possible solutions, for oxygen ion conductors in two-chamber SOFCs, which are currently the main stream in R&D.



Figure 4 : Simplified schematic diagram of flat stack system and an example of a stack The photograph of the stack at the right was reproduced from reference [33].

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Component part	Main material			
Electrolyte	Stabilized zirconia type Cerate type Lanthanum gallate type	$\begin{array}{l} YSZ; Y_2O_3 \mbox{ stabilized } ZrO_2 \\ SSZ; Sc_2O_3 \mbox{ stabilized } ZrO_2 \\ Sm_2O_3 \mbox{ doped } CeO_2 \\ (La, \mbox{ Sr}) \mbox{ (GaMg) } O_3 \end{array}$		
Fuel electrode	Ni/YSZ cermet, Ru/YSZ cermet			
Air electrode	LaMnO₃ type LaCoO₃ type	(La, Sr) MnO_3 , (La, Ca) MnO_3 (La, Sr) CoO_3 , (La, Ca) CoO_3		
Separator	LaCrO₃ type Alloy type	(La, Sr) CrO_3 , (La, Ca) CrO_3 Ni-Cr type, ferrite (Fe) type		

Table 2 : Typical component materials of SOFCs cells developed to date

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Current status and issues in electrolyte R&D

4-1 Current status of electrolyte R&D

SOFCs efficiency improves as the ionic conductivity of the electrolyte used increases. This is explained by the fact that, in many cases, virtually all of the internal resistance in a cell is attributable to electrical resistance loss caused by resistance in the electrolyte. Table 2 shows representative component materials of the SOFCs cells which have been studied to date, classified as electrolytes, electrode materials, and separator materials. ^[1, 25-27] In high temperature (750°C-1000°C) SOFCs, much research has been done on yttria-stabilized zirconia (YSZ: Y₂O₃ stabilized ZrO₂) as the electrolyte, nickelzirconia (Ni-ZrO₂) cermet as the fuel electrode material, lanthanum manganite (LaMnO₃) as the air electrode material, and lanthanum chromite (LaCrO₃) as the separator. However, materials with high oxygen ion conduction, such as scandia-stabilized zirconia (SSZ: Sc₂O₃ stabilized ZrO₂), lanthanum gallate (LaGaO₃), and others, have been the object of intensive research as non-YSZ electrolytes in recent years. In addition to these electrolytes, R&D is also being done on various hydrogen ion conductors, such as barium cerate (barium cerium oxide; $BaCeO_3$), strontium cerate (SrCeO₃), and related materials. Nevertheless, in terms of ion conduction characteristics, chemical stability, cost, and the number of examples of R&D, even today YSZ continues to be the most important electrolyte. SSZ and LaGaO₃ are promising materials for application as electrolytes for

medium temperature SOFCs operating at 750°C and under. YSZ, the cerate based electrolytes, and LaGaO₃ reveal increasingly high oxygen ionic conductivity in that order.

4-2 Features required in electrolytes and the related issues

As conditions for commercialization, SOFCs must possess power generating characteristics and long-term reliability equal or superior to those of competing generating technologies, and their cost must be no higher than that of other technologies. Table 3 shows the main properties required for electrolytes, as well as those of other SOFCs component materials.^[10,26,27,34]

 Table 3 : Main properties required for SOFCs component materials

Component part	Required properties		
Electrolyte	High ionic conductivity, Long-term high temperature performance stability, Density, High long-term reliability (high strength, high durability)		
Fuel electrode	Large reaction field (water formation), Numerous paths for electron and ion conduction, Appropriate porosity (smooth migration of hydrogen and formed water), High temperature stability		
Air electrode	Large reaction field (oxygen absorption, ionization), Numerous paths for electron and ion conduction, Appropriate porosity (oxygen migration), High temperature stability		
Separator	Density, Electron conductivity, High temperature/chemical stability		
Cell stack	Sealability (gas shielding between electrodes), Strong bonding between materials and absorption of differences in thermal expansion, Low reactivity between component parts		

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When designing a cell, first, an electrolyte that demonstrates high ionic conductivity at the operating temperature is decided. This is followed by selection of fuel and air electrode materials capable of extracting the maximum performance from the electrolyte. Therefore, if the electrolyte is not decided, the cell, stack, and SOFCs system structure cannot be materialized. In order to secure reliability, in other words, to secure high performance over an extended period of time, a variety of issues arise in connection with the selection and combination of the electrolyte and electrode materials, the process of manufacturing parts from these materials, the multilayer cell structure, system design, techniques for evaluating performance and durability/reliability, and so on. These issues are discussed below under the headings of generating performance, system structure, and the manufacturing process.

(1) High generating performance and long-term performance stability

When using hydrocarbon fuels, it is necessary to set the reaction temperature of the fuel so as to prevent carbon precipitation and thereby avoid performance deterioration. In order to maintain the original performance of the FCs and minimize performance deterioration over time, it is important to select a combination of materials, including the electrolyte and electrode materials, which is suitable for service environment conditions, including the operating temperature.

There is a possibility that all of the problems of maintaining long-term generating performance, securing high reliability, and the like can be solved at once by reducing the operating temperature. To achieve this, it will be necessary to discover a new electrolyte which has low electrical resistance, in other words, an electrolyte with high ionic conductivity, in the low temperature region.

(2) High strength/reliability system structure

Because the thermal expansion coefficient of electrodes and electrolyte will differ in most cases, the deformation and the damage of cells at operating temperature are problems. The difference in the thermal expansion of the electrolyte and electrodes during the temperature rise from room temperature in a static condition to the FCs operating temperature can easily cause deformation and cracks, either internally in the electrolyte and the electrodes, or at the interface between the two materials. For this reason, structural strength design capable of preventing these problems is indispensable in the cells and the stacks.

Long-term stability of the joints between the separators and cells is also a problem. A fuel cells stack is a multilayered structure comprising layershaped components of heterogeneous ceramics. It is therefore important to establish cells and stack structural designs which guarantee durability and reliability in long-term operation by preventing the crack initiation and propagation in and between these layers, and manufacturing processes for realizing these designs.

(3) Low cost manufacturing process

The cost of high temperature systems which are now in the confirmation test stage cannot still be considered competitive with other power generating systems. In order to reduce the cost of cells and stacks, it will be necessary to minimize the use of expensive materials, simplify the manufacturing process, and establish manufacturing methods suitable for mass production. As the electrolyte and electrode materials are ceramics, various technical issues must be solved in connection with the preparation of the ceramic raw material powders and the powder slurry and the subsequent molding and the sintering process as manufacturing processes peculiar to ceramics.^[35] In forming thin films of the electrolyte, gas phase methods such as pulsed-laser deposition (PLD) can be used in the laboratory, but when considering mass production in the future, a wet process film-forming technology such as the tape casting method is desired.

5 Solutions to electrolyte-related issues

5-1 Necessity of elucidating the ion conduction mechanism in the electrolyte In order to maintain the generating performance of SOFCs systems over the long term, it is necessary to investigate the mechanism which controls performance deterioration in cell component materials. In this, it is important to investigate the ion and electron conduction mechanisms as far back to the nano region of the electrolyte. The following describes an elucidation of the ion conduction mechanism in the electrolyte and the ion conduction mechanism in the interfacial region between the electrolyte and electrodes, and a method of analyzing their behavior in the nano region.

(1) Elucidation of the ion conduction mechanism in the electrolyte

In solid oxides such as YSZ which comprise a crystal structure with numerous oxygen defects, high ion conduction occurs as a result of exchange of the positions of defects and ions. One class of materials which is capable of maintaining a stable crystal structure even though a large number of defects occur is the perovskite oxides*1.[36] With this type of crystal structure, when oxygen ionic conductivity increases, oxygen desorbs easily from the lattice and the material readily manifests electronic conduction, resulting in problems of reduced performance in extended high temperature operation and deterioration due to aging. However, these behaviors are known empirically. The ion conduction mechanism originating in the compound structure of defects at the nanoscale should be investigated while also verifying the phenomena concerned experimentally. Elucidation of these points will make it possible to control the nanostructure of the electrolyte.

Figure 5(a) shows an example of an analysis of the mechanism on ion conduction in YSZ by the laser emission spectrum method;^[37] Figure 5(b) is an example of analysis on the ion conduction path of an LaGaO₃ based compound by the high temperature neutron diffraction method.^[38] Figure 5(a) is an example in which the mechanism on oxygen ion (O₂-) conduction in YSZ was elucidated, showing a schematic diagram of ion conduction in which an O₂- ion occupies a defect as an O₂- ion hole. Figure 5(b) shows conduction in a crystal, in which O2- ions in an LaGaO₃ based compound are distributed continuously and widely in the vertical direction in an arc shape between the stable positions O1 and O2, and the O2- ions maintain bonds with and rotate around cations (positive ions) of a compound of gallium, magnesium, and cobalt (Ga_{0.8}Mg_{0.15}Co_{0.05}). It may also be noted that the results of the analysis on O₂- ion conduction by this high temperature neutron diffraction method showed good agreement with the results of an analysis by the molecular dynamics method.

(2) Elucidation of the ion conduction mechanism in the interfacial region between the electrolyte and electrodes

Together with the ionic conductivity of the electrolyte itself, that in the boundary region between the electrolyte and the electrodes also



Figure 5 : Example of analysis on the oxygen ion conduction mechanisms in YSZ and a LaGaO3 based compound Prepared by the STFC based on references ^[37] and ^[38].



Figure 6 : Schematic diagram on the mechanism of ion conduction in the boundary region between the electrolyte and the air electrode

controls the generating performance of SOFCs. Research to elucidate the mechanism of this type of ion conduction was begun recently. Figure 6 shows a schematic diagram on the mechanism of ion conduction in the interfacial region between the electrolyte and the air electrode.^[39,40] This chart is a model for elucidating the influence of oxygen ion and electron conduction in the interfacial region between the electrolyte and the electrode, the mixed conduction paths of oxygen ions and electrons, and the mechanism involved on the voltage-current characteristics of the FCs. Clarification of the electrochemical behavior on ions and electrons at the interface between the electrolyte and the electrode would provide research guidelines for determining favorable nano- to micro- scale structures in the electrolyte-electrode interfacial region. The DOE's SECA Project is engaged in an analysis of the relationship between electrochemical behavior and material structure in collaborative work by universities and national research institutes.^[39,40] Clarification of the mechanism on the electrochemical reaction in the electrolyteelectrode interfacial region will also contribute to strengthening basic evaluation techniques for SOFCs.

(3) Analytical method for ion conduction behavior in the nano region of the electrolyte

It can be said that to date R&D on cell structural materials, including electrolytes, has been carried out by repeated trial manufacture and evaluation of the materials. However, in the development of materials which manifest

Prepared by the STFC based on references [39] and [40].

revolutionary properties, elucidation of the mechanism responsible for those properties in the nano scale region offers a shorter route than this type of trial-and-error R&D. In other words, an ever-increasing number of examples demonstrate the possibility of realizing dramatic improvement in material properties by the various methodologies for theoretical calculation and experimental verification referred to as nano materials science. Likewise, in R&D on SOFCs electrolytes, it is indispensable to elucidate the atomic and electronic structure by simulations using quantum theory, obtain guiding principles through integrated use of theoretical calculations and experiments, and horizontally systematize various types of materials based on their atomic and electronic structures. Theoretically-based simulation techniques include first-principles calculations, the molecular orbital method, and the molecular dynamics method, while experimental techniques for elucidation of nanostructures include the neutron diffraction method, laser Raman spectroscopy, high resolution transmission electron microscopy (HR-TEM), and electron energy loss spectroscopy (EELS), among others. Figure 7 shows a schematic diagram on methods of creating novel materials by elucidating the mechanism responsible for functions in the nano scale region.

In the future, it will be desirable to carry out research to discover new electrolytes by clarifying the mechanism responsible for material functions through mutual use of empirical and simulation techniques, and to carry out theory-led R&D based on the mechanisms elucidated in this



Figure 7 : New material creation method based on elucidation of the mechanism responsible for functions in the nano scale region

Prepared by the STFC.

manner.

5-2 Elucidation of mechanism on electrolyte damage/deterioration

One current issue in R&D on medium- and high temperature SOFCs in Japan shown in the roadmap in Figure 2(a) is technologies for improving reliability under operating environment conditions. Where this is concerned, it is necessary to investigate the mechanism which controls the damage/deterioration behavior of electrolytes from the macro to the nano scale region. It is considered possible to shorten R&D time as a whole by a procedure of assigning priority to elucidation of the mechanism and actually constructing the system in the final stage, rather than by attempting to achieve R&D targets through a time-consuming process of repeated trial manufacture and evaluation of the system. Analysis of behavior in the nano scale region, which was described in section 5-1 is an effective tool for this purpose.

Analysis of stack destruction behavior has become a top priority topic in the DOE's SECA Core Technology Program, and cell structurerelated destruction criteria have been an object of intensive R&D up to the present.^[21] Placing priority on elucidation of the electrochemical reaction in the electrolyte-electrode interfacial region and analyses of cell and stack destruction behavior, this program is investigating the behavior of materials down to the nano scale region and the related mechanisms, carrying out research on fundamental analytical techniques for these items, and developing simulations and analytical techniques for destruction behavior up to the macro scale.

Long system life is a precondition for SOFCs. However, in the case of medium and high temperature systems, full-scale confirmation tests of long-term performance stability are still in the future. Although development of medium temperature systems is continuing to progress, researchers have relatively little experience with the electrolytes used in these systems in comparison with those in high temperature systems. Therefore, intensive R&D to clarify the mechanical mechanism of defect formation and propagation and expansion of those defects from the nano to the macro scale based on the technologies accumulated with the high temperature type is expected. Elucidation of the mechanism on deterioration and establishment of methods for measuring material life, including materials other than the electrolyte, is indispensable.

5-3 Development of low cost manufacturing processes for cell component materials including electrolytes and low operating temperature systems

The approaches to reducing the cost of high temperature SOFCs systems are (1) adopting low cost electrolytes and other cell and stack component materials and (2) reviewing the manufacturing process for those materials and components. For this, it is necessary to understand the relationship between the electrochemical behavior and the mechanism by which properties are manifested in the

electrolyte in the operating environment and the electrode materials which enable the electrolyte to manifest its properties fully, and the microand macrostructure of these materials at the mass production scale. This manufacturing process technology also includes the selection of low cost ceramic materials, manufacturing processes peculiar to ceramics, and related issues. In the material manufacturing process, wet ceramic processes such as the doctor blade method, spin coat method, and similar techniques are processes for manufacturing low cost ceramics. On the other hand, film manufacturing processes such as physical vapor deposition (PVD), chemical vapor deposition (CVD), pulsed-laser deposition (PLD), and colloidal spray deposition (CSD) are effective methods of film-forming in the material search stage, but are not suitable for mass production of practical cells and stacks. This means that is important to assume, from the first stage of research, an electrolyte film production method which is not limited to simply obtaining small sized samples, but will also enable production of large areas of material and mass production.

Until now, in the initial stage of R&D, R&D on high temperature SOFCs which operate at 750°C-1,000°C was the main stream, but for the above-mentioned goals of securing reliability, reducing cost, and shortening starting time, low temperature operation is desirable. Figure 8 shows the relationship between high reliability, low cost, and high performance, which are reasons why low temperature SOFCs are necessary, and cell component materials, beginning with the electrolyte. Low temperature operation makes it possible to produce low cost cells because heat insulation is easy, freedom of material selection is increased, metal separators can be used, and sealing of the cells and stack is simplified. As a result, a more compact design can be realized in the low temperature system as a whole in comparison with the high temperature type. In addition, high reliability can also be achieved because the temperature differential between the cells is reduced, and performance can be maintained over the long term because deterioration at the electrolyteelectrode interfacial region is prevented, and as a result, a stable reaction surface area can be secured.^[22,35,36,41,42]

On the other hand, if the operating temperature is reduced, generating performance also decreases because electrolyte resistance increases and the reactions at the electrodes become inactive. More specifically, although the maximum theoretical efficiency of conversion from chemical energy to electrical energy increases as the operating temperature decreases, electrolyte resistance and reaction resistance in the electrode interfacial region also become greater, and consequently, generating efficiency is also reduced. This means it is necessary to develop electrolytes which display sufficiently low electrical resistance even at low temperatures.

5-4 Search for new low temperature electrolytes

Figure 9 shows schematically various types of electrolytes, including both inorganic materials (ceramics) and organic materials, from the viewpoint of ion and electron conductivity and



Figure 8 : component materials, focusing on the electrolyte

Prepared by the STFC.



Figure 9 : Classification of electrolytes based on ion conductivity and operating temperature Prepared by the STFC.

operating temperature. Because various problems have become apparent in high temperature operation, intense R&D on electrolytes for medium temperature operation as substitutes for YSZ has been underway since several years prior to this article. This work centers on cerate (CeO₂) based oxides (e.g., SDC: CexSm1-xOy) and LaGaO₃ based oxides. It may be noted that these LaGaO₃ based oxides are a class of high oxygen ion conductors which was developed at a university in Japan.^[36]

R&D on electrolytes for low temperature operation based on these oxides has become active in recent years. Moreover, the search for electrolytes such as LSGM (La_xSr_{1-x}Ga_yMg_{1-y}O₃) which display high oxygen ionic conductivity at temperatures under 500°C has become even more active.^[43,44] The oxides GDC ($Ce_xGd_{1-x}O_2$), ESB ($Bi_{2-x}Er_xO_3$), DWSB ($Bi_{2-(x+y)}Dy_xW_yO_3$), and others have also attracted attention as electrolytes which exhibit high ionic conductivity at low temperatures,^[45] and it is possible that other electrolytes with dramatically higher ionic conductivity in low temperature operation may also be discovered.^[42] Although new low temperature electrolytes with performance equal or superior to that of high temperature materials are still in the search stage, and system technologies which use these electrolytes are in the germinal stage, they have the potential to solve all of the problems confronting high temperature systems at once. Thus, if it proves

difficult to solve the problems limiting the commercialization of high temperature systems, one option is to shift decisively to R&D on low temperature systems. This search for novel electrolytes should be carried out from both the theoretical and experimental directions using nano-level computer simulation and experimental verification techniques, taking advantage of the remarkable advances in nanotechnology in recent years.

Table 4 is a list of the necessary conditions for material design for SDC, GDC, LSGM, etc., which are the main electrolytes for low temperature operation announced in various journals and elsewhere to date. Realizing low polarization or low ohmic resistance by using a thin film electrolyte is an effective approach to improving low temperature oxygen ion conduction characteristics. However, what production process is to be used in producing these thin films is an issue.

At present, electrolytes for medium temperature operation are continuing to be established, and materials for low temperature operation are in the search stage. Determining the optimum electrode material for use with the respective electrolytes is an important technical element for the development of cells with high generating efficiency and high reliability. Figure 10 shows the relationship between electrode materials, types of fuels, operating temperature, and the structure at the cell to stack levels and

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Necessary condition		Hydrogen ion conductor		
in material design	SDC	GDC	LSGM	BCY
Ionic conductivity	0	0	0	0
Insulation property	Our Contract of Contract o	O O O O O O O O O O O O O O O O O O O	0	0
Chemical stability	0	0	\bigtriangleup Ni + SDC addition	_
Chemical stability (oxidizing/reducing atmospheres)			× Under low oxygen partial pressure	 Pr addition
Gas shielding (nonporous)	0	0		O CSD method
Film uniformity (reduction of ohmic resistance)		0		CSD method
Thermal expansion (same as electrodes)	0	0	0	_
Low cost		×	x~△	0

Table 4 : Necessary conditions in material design of low operating temperature electrolytes

Prepared by the STFC.



Figure 10 : Relationship between electrode materials, fuels, operating temperature, and cell and stack structure to electrolytes

 $\begin{array}{l} LSM: La_xSr_{1-x}MnO_3\\ BSCF: Ba_xSr_{1-x}Co_yFe_{1-y}O_3\\ SSC: Sm_xSr_{1-x}CoO_3\\ LSCF: La_{1-x}Sr_xFe_yCo_{1-y}O_3 \end{array}$

the electrolytes developed to date. When an oxygen ion conducting electrolyte is used, the possible reduction in the operating temperature is wholly related to high ionic conductivity in the electrolyte at a low operating temperature and the selection of an electrode material which allows the electrolyte to fully demonstrate this characteristic. In other words, in addition Prepared by the STFC.

to high conductivity in the electrolyte itself, the generating performance of SOFCs is also influenced by easy generation of oxygen ions at the air electrode and their conduction to the electrolyte, high efficiency transfer in the fuel electrode interfacial region, and the electrochemical reaction with hydrogen.

Directions in industrycademia-government collaboration on SOFCs R&D in Japan

Globally, R&D on high temperature SOFCs has reached a stage where wide-ranging confirmation tests of SOFCs systems are underway. Several years ago, it had been predicted that these devices were nearing commercialization, but at present, no SOFCs system is actually being sold commercially in any country. This is due to the failure to solve various problems, including the fact that system reliability cannot be confirmed and cost has not yet reached a level where SOFCs systems can compete with other technologies. Improvement or development of new materials for cells and stacks, beginning with electrolytes, through collaboration among industry, academia, and government in their respective fields of expertise is desired in order to overcome these problems.

One target in the Nanotechnology & Materials area of Japan's 3rd Science and Technology Basic Plan is the development of innovative FCs materials for the purpose of achieving a large increase in energy utilization efficiency through technical innovation.^[47] This is currently in the basic stage and will require considerable time to reach practical application. For R&D which strongly has a social nature and an important impact on environmental protection, it is necessary to develop a new R&D process which frees itself from the methods used in projects in the past and induces innovation by collaboration among industry, academia, and government organizations. In realizing SOFCs which meet the requirements of improved generating characteristics, long-term performance, and reliability in diverse service environments, clarification of the mechanism responsible for functions in the nano scale region of cells component materials is indispensable, centering on electrolytes. The creation of innovative SOFCs should be pursued through collaborative research by industry, academia, and government which prioritizes this type of fundamental and basic technology. In this, universities and R&D Independent Administrative Institutes (IAI) are expected to play a large role in fundamental research. To fulfill these expectations, further improvement and expansion of fundamental research at universities and IAI are needed.

METI's "New Economic Growth Strategy" proposes commercialization and market development through cross-sectoral integration and collaboration in industry, academic, and governmental R&D, which has been carried out independently to date, and suggests a paradigm shift, for example, by returning to basic science as a key point for successful innovation.^[48] In particular, in cases where R&D appears to be blocked, breakthroughs should be realized by temporarily suspending work which is simply a linear extension of the existing R&D and carrying out R&D that return to fundamental scientific principles.

In future national projects related to SOFCs in Japan, a complete strategic study should be made even in the stage before research topics are selected, and these projects should aim at achieving the world's top results when setting target values. In the present context, this means R&D aimed at realizing the world's top SOFCs generating performance. A key part of any effort to develop innovative SOFCs systems must be research that returns to the basic science by elucidating the mechanism responsible for the properties of the cell component materials, and particularly the electrolyte, and R&D on the analytical techniques required to support that research. In this, smoothly linking material technology "seeds," in other words, the results of fundamental research based on materials science by universities and R&D IAIs, and the "needs" of technical development by industry on the system side will be decisive for the creation of innovative technologies.

7 Conclusion

Reasons for the strong interest in SOFCs among the various types of fuel cell systems were discussed from the viewpoint of their features as a high efficiency power generating technology, fields in which application is expected, and the current status of R&D in Japan and other

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countries. In particular, current issues related to SOFCs generating systems were examined from the viewpoint of electrolytes, and methods of solving these problems in the future were proposed.

High temperature SOFCs are now in the confirmation test stage. However, problems related to (1) long-term generating performance, (2) cost reduction, and (3) reliability must be solved in order to achieve practical application. To solve these problems, an elucidation of the mechanism on performance deterioration in cell component materials reaching back to the nano scale region of the materials will be necessary. R&D related to the analytical techniques required to support this type of research are also essential. To secure reliability, in addition to an empirical elucidation of the mechanism on deterioration in component materials, R&D which effectively utilize computer simulation techniques for damage/deterioration are also desirable, particularly from the viewpoint of research efficiency. To reduce the cost of SOFCs systems, it is necessary to adopt economical cells and stack materials and review the manufacturing processes for these materials. It is also important to understand the relationship between the micro- and macroscopic structures of cells component materials in the trial manufacture stage and the structures of same materials at the mass production scale from the viewpoint of the electrochemical behavior in the cell component materials, including the electrolyte, and the mechanism responsible for their properties.

On the other hand, in recent years, electrolytes such as scandia-stabilized zirconia and lanthanum gallate, which display high oxygen ionic conductivity even at low temperature (500°C-750°C and under) have been the object of intensive R&D as alternatives to yttria-stabilized zirconia (YSZ), which is used in high temperature SOFCs. Although R&D on these novel electrolytes and SOFCs systems is still in the germinal stage, these technologies have the potential to solve all of the problems confronting high temperature systems at once.

If the problems limiting commercialization of high temperature system cannot be solved through the current R&D, one option is a decisive shift to R&D on low temperature systems. Considering the remarkable progress which has been achieved in nanotechnology in recent years, the search for new electrolytes for these systems should include both theoretical and empirical approaches, taking advantage of computer simulation and experimental techniques at the nano scale level. It is also important to carry out R&D on materials suitable for cell and stack manufacturing processes, envisioning the practical SOFCs systems of the future, from the material search stage. Moreover, in developing systems which use these new electrolytes, a paradigm shift from the trial-and-error approach used to date to a science-based methodology is desirable. That is, it is important to carry out R&D while clarifying the behavior by which materials manifest properties in the nanostructural region, rather than prioritizing the conventional process for repeated trial manufacture of prototype systems and measurement of their generating characteristics. Finally, in many areas of this R&D work, an approach based on fundamental research by universities and R&D IAI and management methods which effectively enable collaboration among industry, academia, and government and implement an appropriate division of work, can be expected to yield important results.

Glossary

*1 A class of oxide ceramics expressed by a compound of oxygen and metal elements A and B (ABO₃). Virtually all of the metallic elements in the Periodic Table are possible component elements. Depending on the combination of A and B, perovskite oxides reveal a variety of properties, including ferroelectric properties, superconductivity, and ionic conductivity.

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