STATUS OF SOFC DEVELOPMENT IN JAPAN

Hiroaki Tagawa
Institute of Environmental Science and Technology
Yokohama National University
Tokiwadai, Hodogaya-ku, Yokohama, Japan, 240

ABSTRACT

Many companies in several industries are interested in developing solid oxide fuel cells. Over ten companies have performed the development under the financial support of New Energy Development Organization, Petroleum Energy Center, or some utility companies of electric power and gas, and other companies by themselves. In the Moonlight Project conducted by NEDO, the first phase was carried out in 1989-1991 and the second phase of a 6 year program started in 1992fy. PEC has conducted a 5 year program since 1990. Basic research on component materials, reactivity of the materials and kinetics and mechanism on the electrodes has been performed at several universities and national laboratories.

INTRODUCTION

Fuel cells have been developed as an electricity generating technology with high energy conversion efficiency for conserving fossil fuels and moreover for aiming at the restraint of the CO2 emission into the atmosphere. In particular high-temperature solid oxide fuel cells (SOFC) are thought to be the most promising, electric power generating system because the principle is simple, and the requirement for component materials is not so severe as compared with that of other technologies as MHD and high-temperature gas turbine electric power generation systems.

In Japan, research and development on SOFC have been carried out by universities, national and private laboratories, and companies in many industries such as ceramic, electrical and plant engineering and oil refinery. Utility companies of electric power and gas especially have had high R&D activities. This review describes the present status of the development on SOFC performed by public and private organizations with emphasis on the national projects.

MOON LIGHT PROJECT

R&D on fuel cells as one of the so-called Moonlight Project was begun from phosphoric acid fuel cells (PAFC) for aiming at energy conservation in the beginning of 1970s. Under the Moonlight Project,
basic research on SOFC was started out by Electrotechnical Laboratory, one of the national laboratories, in 1981 for utilizing hydrogen as an energy carrier. They intended to establish the manufacturing technology for tubular-type cells using plasma-spraying. National Chemical Laboratory for Industry also started research on the high-performance cell materials and development of fundamental technology for composing planar-type SOFC in 1986.

In parallel with R&D by the national laboratories, New Energy Development Organization (NEDO) started a 3 year R&D program on SOFC in 1989. NEDO, as an executive organization under the Agency of Industrial Science and Technology, has actually conducted the Moonlight Project by financially supporting private companies and laboratories for PAFC since 1981 and for SOFC since 1989.

The objective at the first phase of the Moonlight for SOFC was to develop suitable materials or basic technologies given in the following six subjects, and further to fabricate one or two hundreds watt class stacks by use of these technologies. The specific technologies were as follows:
- Developing ceramic separator
- Developing alloy separator
- Developing co-firing technology for making tri-layer (anode/electrolyte/cathode) single cells
- Developing sealing technology
- Developing plasma-spraying technology for making single cells
- System analysis for SOFC

The target of SOFC fabricated by using each subject was as follows:
- Output of a stack : one or two hundreds Watt
- Specific power density : 0.5kW/dm³ (expected value)

Supplemental specification for single cell
- Cell voltage : 0.7V at 300mA/cm²
- Specific power density : 0.2W/cm²
- Unit cell area : 100cm² (in the case of planar-type)

The budgets for developing SOFC during 1989-1991 period were

| Fiscal year | million yen |
|------------|-------------|
| 1989       | 72          |
| 1990       | 124         |
| 1991       | 261         |
| Total      | 457         |

Following organizations accepted to participate in the project, where the order of organization corresponds to that of the specific technologies above shown, respectively: Fuji Electric Corporate Research and Development Ltd., NKK Corp., Murata Manufacturing Co., Sanyo Electric Co., Fujikura Corp, and Institute of Applied Energy (IAE). IAE has performed reviewing the SOFC technology, analyzing a SOFC system,
and finally making a conceptual design for the SOFC system.

The next stage of the NEDO program on SOFC started in the 1992 fiscal year. The items for the 6 year program are as follows:

I. Development of a planar-type modules
(1) To develop multi-kilowatts (1-3kW) modules using steam-reformed natural gas by 1995.

Target specification:
- Current density 0.18W/cm² (at the first run of the module)
- Fuel utilization 70% (expected value)

(2) To develop multi-ten kilowatts (10-30kW) modules using steam-reformed natural gas by 1997 after check and review of the 1-3kW modules.

Target specification:
- Current density 0.2 W/cm² (at the first run of the module)
- Fuel utilization 75% (expected value)
- Degradation less than 1%/1000h

II. Development of fundamental technology for materials and stack fabrication

(1) To develop high-performance and long durable cathode materials and to establish cathode manufacturing technology
(2) To develop high-performance anodes for internally steam-reforming natural gas in the cells
(3) To develop gas-sealing materials and to establish co-firing technology using the sealing materials

III. Establishment of SOFC engineering and system

(1) To conduct engineering studies covering SOFC electric power generating system from several to hundreds of megawatts using natural and coal gas as fuels
(2) To establish SOFC processes and systems that will be necessary to commercialize the SOFC technology
(3) To develop SOFC-related equipments such as valve, heat exchanger, blower which are usable at high temperatures as high as 1000°C

The budget in the 1992 fiscal year was 274 million yens.

The following ten organizations accepted to participate in the three groups of the second stage of the NEDO project.

Group I. Development of a planar-type modules
- Fuji Electric Corporate Research and Development Ltd.
- Sanyo Electric Co.
Group II. Development of fundamental technology for materials and components and module fabricating
- Mitsui Shipping Co.
- Japan Fine Ceramics Center
- Fujikura Corp.
- Mitsubishi Heavy Industries Ltd.
- Murata Manufacturing Co.

Group III. Establishment of SOFC engineering and system
- Central Research Institute of Electric Power Industry
- Electric Power Development Co. and Mitsubishi Heavy Industries Co.
- Metal Materials Research Center.

PETROLEUM ENERGY CENTER

In order to effectively utilize light petroleum fuels with C3 or more hydrocarbons, such as naphtha and kerosene, the Petroleum Energy Center (PEC) started a 5 year program for developing high-temperature fuel cells in 1990. They are conducting R&D on SOFC for developing 1kW module using light petroleum as fuels. Their goals are as follows:

1. To fabricate high power density SOFC
2. To establish steam reforming technology for naphtha and kerosene
3. To establish a system in which the hydrocarbons are internally reformed.

The following companies join the project: Idemitsu Kosan Co., Nippon Oil Co. and Tonen Corp.

R&D CONSORTIUMS CONTAINING UTILITY COMPANIES

There are many public and private organizations which are interested in R&D on SOFC, but do not participate in the national projects. These companies have performed R&D on SOFC by themselves funds. Utility companies of electricity and gas have carried out R&D on fuel cells by themselves funds.

Of the public utility companies, each Electric Power Co. of Tokyo, Chubu (at Nagoya) and Kansai (at Osaka), and Tokyo Gas Co. and Osaka Gas Co. have large activities for the development on SOFC. They have individually, or as a group of two or three companies, organized consortiums with some SOFC developers such as Westinghouse Electric Co., Mitsubishi Heavy Industries Ltd., and have made verification tests of electric generating or on-site cogeneration systems in collaboration with the developers. They have also carried out basic research on component materials and the SOFC system by themselves.

The activities in some consortiums are as follows:
Three companies have cooperatively developed tubular-type SOFC. They demonstrated to operate a 1.2kW module with the maximum output of 1.3kW for over 1000 h in 1991. The degradation of the stack was less than 3% for the entire operation period. They also intend to operate a 10kW module in 1993.

Planar-type SOFC has been cooperatively developed. A single cell is fabricated by co-firing each component, where the materials of YSZ, 90wt.%La(Sr)MnO3±10wt.%YSZ, and 60wt.%Ni+40wt.%YSZ, as electrolyte, cathode and anode, respectively, are prepared by doctor-blade method. A stack which consists of 10 disk-shape 100cm² single cells has generated 100W power output for over 1000 h.

Both companies have developed a 1kW plate-type monolithic integral layer built stack, so-called MOLB, and achieved to run the MOLB with at full power in the fall season of 1992. The MOLB used consisted of three stacks connected in series, and one stack was made of 40 single cells (single cell area of 225cm²).

Three companies have cooperatively performed a verification test of a 25kW SOFC power generation system, manufactured by Westinghouse Electric Corp., for over 1,500hr in the spring of 1992. The system consists of two 20kW modules using internally steam-reformed natural gas. The system is operated at 1000°C and atmospheric pressure.

They have individually tested a 3kW tubular-type module fabricated by Westinghouse Electric Corp. and achieved a long operating time in excess of 3000 and 5000 h, respectively, in 1987-1988.

Natural gas fueled SOFC cogeneration system has jointly developed with Westinghouse Electric Corp. The design concept is essentially the same as that in a joint project with Kansai Electric Power Co. A verification test is scheduled at an Osaka Gas test facility in 1993.

Many companies in different industries such as ceramic, electrical engineering, heavy industry, petroleum industry, and utility of electric power and gas have performed R&D on SOFC. The following organizations presented R&D activities in the reference[1,2,3,4,5]:

Fabrication of thin 8YSZ sheets (less than 0.1mm in thick) by extrusion has been developed.
A substrate-type, planar SOFC with a disk-shaped center-manifold structure has been developed. A single cell consists of two substrates. One substrate is a thick porous Ni+YSZ anode, on which 8YSZ electrolyte plasma-sprayed, and A (La,Sr)MnO₃ cathode is screen-printed on the electrolyte. Another substrate is made of the cathode material on which an interconnection (La,Sr)CrO₃ is plasma-sprayed on the cathode. They attained 410W at 0.5V and 80A using a 10 cell stack, where the diameter of a single cell was 200mm, and the effective area 200cm².

Fujikura Corp.
They have developed tubular-type SOFC with emphasis on plasma-spraying conditions such as making porous cathode and dense electrolyte. The structure of a single cell is similar to that of Westinghouse Electric Corp. A stack has an array of 4 x 4 single cells. The effective area of a single cell is 105cm². The maximum output was 103W.

Idemitsu Kosan Co.
The development of tubular-type SOFC has been performed by using plasma-spraying. They have tried to find out plasma-spraying conditions for fabricating high-performance single cells with thin impermeable electrolyte and interconnection layers, and porous electrodes.

Ishikawajima Harima Heavy Industries, Ltd.
They have extensively developed molten carbonate fuel cells under Moonlight project. A planar-type SOFC has been developed on a small scale. R&D of the following subjects is performed:
- Co-firing of single cells
- An effect of morphology of the electrodes on polarization

Mitsubishi Heavy Industries Ltd. at Kobe Works
Planar-type SOFC has been developed. The development of the so-called MOLB was described in collaboration with Chubu Electric Power Co. They have developed several stacks with different sizes of single cells. They have also carried out the research for raising the performance of the electrolyte and electrodes, and for finding out suitable sealing materials with/in their Takasago Laboratory.

Mitsubishi Heavy Industries Ltd. at Nagasaki Works
Both tubular- and planar-type SOFCs have been developed these 10 years. The development of the former has worked in collaboration with TEPCO and EPDC, and the latter with TEPCO. MHI have also carried out development of plasma spraying technology and its application to SOFC, especially for densification of the electrolyte, with their Nagasaki R&D Center.

Mitsui Engineering & Shipbuilding Co.
A planar-type SOFC has been developed, where the electrolyte, YSZ, of 0.2mm in thickness is fabricated by tape casting, and the anode, 40 vol.%+YSZ, and the cathode, (La,Sr)1-x(Mn,Cr)O₃, are screen-printed on the green electrolyte sheets. Chemical stability of the interconnection
(La,Ca)CrO₃ with other components has been also studied.

Murata Manufacturing Co.

Planar-type SOFC has been developed using co-fired trilayer single cells. The single cell consists of the layer of (La,Sr)MnO₃/YSZ/Ni+YSZ. A stack is composed by repeating a unit with an array of interconnector/composite sealing material/current collector/co-fired trilayer single cell/current collector/composite sealing material/interconnector. A composite material of glass and ceramics and Ni-Cr based alloy are used for sealing and interconnection, respectively. The maximum output was 117W for a 6 cell stack with a 110cm² single cell.

NGK Insulator, Ltd.

Component materials for Planar-type SOFC have studied with emphasis on reducing electric contact resistance between co-fired single cells when a stack is composed of. Use of Ni-metallized LaCrO₃ is proposed as interconnection.

Nippon Oil Co.

They have developed catalysts which steam-reform naphtha at low steam/carbon ratio without carbon deposition. A verification of a high-efficient electric power generation system has been tested using naphtha. Direct internal steam-reforming has been developed.

NKK Corp.

Planar-type SOFC has been developed with emphasis on metallic separator. Ni-alloys coated with an electric conductive oxide (Cr₂O₃) was selected as the results of several tests. The coated alloy showed excellent efficiency in maintaining conductivity and in raising heat resistance. A 8 cell stack with an 144cm² single cell area generated 220W (5.1V, 300mA/cm²).

Osaka Gas Co.

Tubular-type SOFC with the same design concept as Westinghouse Electric Corp. has been developed. The development for the following R&D subjects are performed:
- Fabricating high-performance Ru/YSZ anode with low overvoltage and no sinterability
- Fabricating single cells Ru+YSZ/YSZ/(La,Sr)MnO₃(self-support tube)
- The maximum power density obtained is 1.55W/cm² at 0.5V and 3A/cm².
- Fabricating interconnection by use of laser-ablation method instead of EVD
- Developing non-isothermal simulation code for getting homogeneous temperature distribution in a module and for executing fuel-recycling system by use of oxygen as oxidant.

Sanyo Electric Co.

Development of component materials and fabrication of stacks have been performed. The following subjects are studied:
- Improvement of the anode by choosing a mixing ratio and particle size for Ni and YSZ
Using glass as a so-called melt-seal material
Fabrication of a stack of 20 layers of 125 cm² single cells (415 W at 22 V and 0.3 A/cm²).

Tokyo Gas Co.
Planar-type SOFC has been developed. The SOFC-related materials and stack manufacturing technology, and basic research have been carried out. They have fabricated single cells of several sizes (the maximum size of 400 cm²). A 25 cell stack constructed from 100 cm² single cells showed an output of 420 W at 26 V and 16 A. The following subjects are studied:
- EVD of YSZ on a sintered, porous Ni+YSZ anode plate
- Kinetics and mechanism on air electrode reaction
- Kinetics and mechanism of steam reforming reaction on Ni+YSZ anode
- Chemical diffusion in LaCrO₃.

Tonen Corp.
R&D of a planar-type SOFC has been performed. Single cells were fabricated by painting Ni+YSZ and (La,Sr)MnO₃ as anode and cathode materials, respectively, on both side of a thin of Al₂O₃-added 3YSZ plate, which was prepared by doctor blade method. They attained 1.3 kW at 17.5 V and 75 A, using a 30 cell stack constructed by using 225 cm² single cells. They have also developed as
- Developing LaCrO₃-based oxides as an interconnection material
- Comparison of alloys and LaCrO₃-based oxides as interconnection
- Co-firing of single cells

TOTO Ltd.
Tubular-type SOFC using wet process has been developed. Porous air electrode of (La,Sr)MnO₃, fabricated by extrusion, is used as support tube. (La, Ca)CrO₃ interconnection, YSZ electrolyte, and Ni+ YSZ anode are fixed on the support tube in the order by repeating the procedures of slurry coating and firing. The thickness of the electrolyte is 0.2-0.3 mm. An output of the 10 cm² single cell was 0.44 W/cm² (0.7 V and 0.63 A/cm²).

BASIC RESEARCH

For a long time in Japan, basic studies in the field of solid state ionics and ceramics have been conducted in universities for the purposes of establishing phase relations in systems containing zirconia, finding new electrolytes, and analyzing crystal structures of electrolytes and perovskite-type oxides. The present activities of the basic research on SOFC are based on this history.

Basic research on fuel cells, including SOFC, for achieving high conversion efficiency has been carried out in several universities, three national laboratories, and public and private research organization.
Basic research on SOFC-related subjects is carried out at the following universities:

- Muroran Institute of Technology:
  Reaction mechanism on Pt/YSZ electrode.
- Akita University:
  Kinetics and mechanism in the reactions between component materials
- University of Tokyo:
  Establishment of plasma-spraying technology for fabricating a single cell
- Tokyo Institute of Technology:
  Fabrication method of thin-film-type SOFC
- Yokohama National University:
  Reaction kinetics and mechanism on electrodes, thermodynamics and physicochemical properties of component materials
- Yamanashi University:
  Development of component materials for low temperature SOFC
- Nagoya University:
  Protonic conducting electrolytes and its application to SOFC
- Mie University:
  New zirconia-based electrolyte and phase relations of component materials
- Kyoto University:
  Vapor phase electrolytic deposition of component materials
- Kyushu University:
  Ceria-based electrolyte and its application to SOFC, and reaction kinetics and mechanism on electrodes
- Oita University:
  Fabrication of perovskite-type oxides as cathode materials for low temperature SOFC

**Electrotechnical Laboratory**
ETL started R&D on tubular-type SOFC, fabricated by plasma-spraying, under the Moonlight Project in 1981. The laboratory has developed the plasma-spraying method for tubular cells, and succeeded in fabricating and running a 1kW module in 1986. After established that technology, R&D at ETL has turned toward to be basic research. The present works are:
- Determining the reaction mechanism on the cathode
- Chemical vapor deposition

**National Chemical Laboratory**
NCLI started R&D on component materials and a planar-type stack in 1986. Following subjects are studied:
- Co-firing green sheets of single cell, anode/electrolyte/cathode
- Chemical stability and reaction containing (La,Ca)CrO3
- Phase relations and thermodynamics of component materials
- A route to fabricate planar-type cells

**Government Industrial Research Institute, Osaka**
GIRIO has developed SOFC operated at low temperatures. The
following subjects are studied:
- Fabrication of a thin YSZ-based electrolyte sheet by wet process and a YSZ thin film by ion-beam spattering
- Development of a ceria-based electrolyte, and an application of ceria-based material to anode

Central Research Institute of Electric Power Industry
CRIEPI has performed R&D on component materials and kinetics and mechanism on the electrodes, and also cost estimation for fabricating tubular- and planar-type SOFC.

Japan Fine Ceramics Center
JFCC has performed basic research on component materials and the related reactions:
- Modification of cathode materials such as (La,Sr)MnO3 and (La,Sr)MnO3 +YSZ
- Modification of porous structure for anode materials
- Relationship between performance and microstructure of the electrodes

SOFC-RELATED SOCIETIES

The following societies organize academic meetings on solid state ionics and SOFC:
- The Electrochemical Society of Japan
- The Solid Oxide Fuel Cell Society of Japan
- The Solid State Ionics Society of Japan
- The Committee of Battery Technology.

Over one hundred papers on SOFC are annually presented in meetings held by these societies.

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