Figure 3.35 — ARPA-SCE SOFC Generator Terminal Voltage, Current and Power.

3.7.2 PMF Units

3.7.2.1 100 kWe EDB/ELSAM SOFC Experimental Field Unit

The world's first 100 kWe class Solid Oxide Fuel Cell power generation system has been fabricated by Westinghouse and is sponsored by EDB/ELSAM, a consortium of Dutch and Danish power companies. This natural gas fueled experimental field unit is being installed near Arnhem, The Netherlands, at an auxiliary district heating plant (Hulp Warmte Centrale) at the Rivierweg in Westervoort, a site provided by NUON, one of the Dutch participants and will supply ac power to the utility grid and hot water to the district heating system serving the Duiven/Westervoort area. Performance objectives for the 100 kWe SOFC system are an electrical generation efficiency approaching 50% [net ac/LHV] and two years of operation.

The 100 kWe SOFC generator module utilizes tubular Air Electrode Supported (AES) SOFCs of nominally 22 mm diameter by 1500 mm active length. These cells are fabricated using a plasma spray process for the interconnection and the Electro-chemical Vapor Deposition (EVD) process for application of the electrolyte and attachment of the fuel electrode.

The 100 kWe SOFC generator module or stack is of seal-less design and employs 1152 tubular SOFCs oriented vertically and arranged in twelve bundle rows. See Figure 2.2. Each bundle row consists of four bundles, with each bundle a rectangular cell array having three cells in parallel and eight cells in series. The bundle rows are connected in electrical series yielding a serpentine current path. The stack design for the 100 kWe unit differs from previous Westinghouse practice in that the natural gas reformers are integral with the insulation barriers between bundle rows, with heat supplied by thermal radiation directly from the SOFCs. In the 25

kWe-class SOFC units, the reformers are hydraulically integrated with the cell stack, but heated by exhaust gas. As in prior Westinghouse practice, spent anode gas is recirculated and mixed with fresh fuel (desulfurized natural gas) using an ejector with pressurized natural gas as the primary fluid. The stack operates at nominally ambient pressure. The outer canister of the 100 kWe generator module is cooled with process air to limit dissipation to the ambient and to limit canister temperature.

The 100 kWe SOFC power generation system is composed of five discrete skid mounted assemblies or "skids." The generator skid supports the SOFC stack and the electrically powered process air heater used for startup. The Thermal Management Skid (TMS) supports the recuperators, the air movers (blowers), air and exhaust piping and air control valves, and the Electrical Distribution System (EDS), a shallow set of enclosures which houses all electrical distribution and electronic hardware including the control computer. The Fuel Supply System (FSS) skid supports the fuel and purge gas control valves, the desulfurizers, and the small steam generator used during startup along with a small water supply tank. These three skids are arranged in a rectilinear package measuring 8.42 m long by 2.75 m wide with a maximum height of 3.58 m. The power conditioner and the hot water heater are also skid mounted, but supplied by EDB/ELSAM. The system was designed to be in conformity with applicable European Economic Community Directives (codes and standards) and to qualify for the "CE" mark.

The Factory Acceptance Test (FAT) of the 100 kWe SOFC took place at the Westinghouse Science & Technology Center in Churchill, PA during the period of 2 October 1997 to 30 October 1997. The FAT demonstrated that the EDB/ELSAM 100 kWe SOFC Power Generation System could be started, operated, stopped and cooled in a controlled manner. It also included tests that satisfied the requirements for approved system certification per Fuel Cell Power Plant Standard Z21.83.CGA12.10.

The FAT was performed after a significant battery of Process and Control (PAC) tests and other de-bugging operations had been completed and all Balance of Plant (BOP) sub-systems had been demonstrated to be functioning per the design intent. The PAC tests were performed without the SOFC generator module by routing, using special extra piping, the hot process air flow exiting the air heater back to the high temperature recuperator exhaust gas inlet. Following the integration of the SOFC module for the FAT, critical subsystems were checked to provide assurance that the integration was performed correctly.

During the FAT, most of the safety functions were tested to confirm the inherent integrity of the control system. Exhaust gas was checked for emissions of NO_x, CO, and C_xH_y. Noise levels were measured during operation and after shutdown. Electromagnetic compatibility tests were performed to provide data for addressing the European EMC directive. The electrical efficiency was determined at different loadings.

The EDB/ELSAM 100kWe SOFC field unit factory acceptance test (FAT) was completed at the end of October in 1997 after 335 hours of power generation. Figure 3.36 is a photograph of the unit at the FAT. The unit was disassembled, packaged for shipment, and sent to the NUON test site in Westervoort, the Netherlands by mid November where the re-assembly of the unit is taking place. The start-up for the unit in the Netherlands is expected to be in December, 1997.

Figure 3.36 — EDB/ELSAM 100 kW SOFC Power System at Westinghouse PMF.

3.8 TASK 8.0 — MWE CLASS POWER SYSTEM DEMONSTRATION

3.8.1 3 MWe PSOFC/GT Power System: Fort Meade MW-Class Demonstration

One of the sites under consideration for the first MW-Class SOFC/GT demonstration is the EPA's Fort Meade laboratory near Washington DC. Being a site controlled by EPA, a site owned and managed by DOD, it promises to be a highly visible site for a major DOE/inter-agency fuel cell demonstration project. Being also a project aimed at improving fuel efficiency and reducing emissions, it would address two urgent issues that are very relevant for all three federal agencies. A map of the proposed site is shown in Figure 3.37. The EPA would make available the necessary space and required infrastructure to house the SOFC power system as well as laboratory and administrative space for equipment operation monitoring.

There is significant interest in the project at EPA, as they have published a brochure concerning the project. Extensive discussions have also been held regarding EPA providing funds for the project. These however are proceeding slowly. EPA is willing to commit minor funds for the site related work but has so far been reluctant to commit major funds for the project, despite encouragement from Congress to put funds into their budget request. Responsibility for finding the funds rested initially with EPA's Office of Administration, but their budget request was turned down by EPA management. Since the fourth quarter of 1997 and the Kyoto conference there seems to be broader support within EPA and responsibility for finding the funds was transferred to the Office of Atmospheric Programs.

Meetings have been held with DOD/Fort Meade executives to win their support. Several meetings have also been held with Baltimore Gas and Electric (BGE) the local gas/electric utility who is willing to cooperate. BGE is also willing to provide nominal support for the project but is not yet willing to provide major support.

New proposals to EPA and BGE are being discussed to get the project moving.

Westinghouse is taking the position that the first MW-Class demonstration will be initiated when sufficient funds for a particular site are identified and committed by the interested parties. To this end we are pursuing other opportunities in Canada, Europe and the U.S.

Figure 3.37 — Environmental Science Center, Ft. Meade, Maryland.

4. Conclusions

Many significant advances have been made during the seven year Westinghouse/U.S. DOE Cooperative agreement program entitled "High-Temperature Tubular Solid Oxide Fuel Cell Generator Development:"

- The Westinghouse tubular SOFC has successfully evolved to a commercial class geometry, increasing from an experimental 16 mm diameter, 50 cm length cell with a peak power of 1.27 watts/cm to a commercial scale 22 mm diameter, 150 cm length cell with a peak power of 1.40 watts/cm.
- The Westinghouse state-of-the-art air electrode supported solid oxide fuel cell design (AES-SOFC) is thermally rugged, having shown the ability to thermally cycle from 1000°C to room temperature and back more than 100 times without any performance loss.
- The Westinghouse AES-SOFC is stable at elevated pressures having shown improved performance up to 15 atmospheres, thereby facilitating integration with gas turbine systems.
- The Westinghouse AES-SOFC has excellent voltage stability with a demonstrated degradation rate of approximately 0.1% per thousand hours of operation at 13,000 hours.
- The Westinghouse has made great strides in reducing cell cost by replacing two expensive EVD processing steps with simple less expensive processes without compromising life or reliability.
- Westinghouse SOFC Module cost has been reduced via a new insulation system design and material, and improved stack assembly procedures.
- The Westinghouse SOFC has the potential for efficiencies in the 60 to 70% range in MW class pressurized SOFC/gas turbine hybrid cycle systems, as predicted by performance analysis coupled with pressurized single cell test data.
- The Westinghouse SOFC Experimental Field Units are highly successful with a number of 25 kWe-class units having been built and operated for thousands of hours during the seven year Cooperative Program.
- Westinghouse is the recognized world leader in SOFC technology with the first 100 kWe class SOFC power generation system completed, factory acceptance tested, and shipped to the customer's site in the Netherlands.

5. Acknowledgments

This report is a compilation of information developed and documented by many investigators, and summarizes the work of approximately 100 people in the Westinghouse SOFC organization.

Westinghouse acknowledges the guidance and assistance of Mr. William C. Smith, SOFC Project Manager, Gas Power Systems Division, U.S.-DOE-FETC, in the course of this Cooperative Agreement, spanning the period from December 1, 1990 through November 30, 1997.

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3.4.2 Pressurized Cell Testing

Westinghouse in conjunction with Ontario Hydro Technologies has tested AES cells at pressures up to 15 atmospheres on both hydrogen and natural gas fuels. Figure 3.18 shows the comparative voltage-current density curves and Figure 3.19 the power output curves at 1, 3, 5, 10 and 15 atmospheres for a 22 mm diameter, 150 cm active length AES cell at 1000°C. Operation at elevated pressures yields a higher cell voltage at any current density due to increased Nernst potential and reduced cathode polarization, and thereby permits higher stack efficiency and greater power output. With pressurized operation, SOFCs can be used to supplant combustors in combustion turbines; such integrated SOFC-combustion turbine power systems are expected to reach efficiencies approaching 70%, and thus result in reduced fuel consumption and reduced capital per unit power output. As of November 30, 1997, pressurized cell tests had accumulated 4800 hours of operation without deleterious effect.

3.4.3 Pressurized Bundle Testing

A bundle test designated PBT#1.0 has been designed and built in preparation for installation and testing at Ontario Hydro in Toronto, Canada. The bundle test contains 48 cells, each with an active length of 150 cm and 22 mm diameter. The test article has been designed to perform component verification tests at elevated pressures:

- **Reforming:** Investigate effects of temperature and catalyst variations. Different catalyst depths will be examined to determine mixing effects such as increased channeling in shorter bed lengths and perturbations in flow distributions due to density changes in the fuel mixture.
- **Insulation stability:** Investigate the effects of pressure on fuel bypassing and channeling within and between insulation material commonly used in the submodules. Also, the geometry of the material itself will be examined to determine if there is a change due to pressurization.

Figure 3.18 — Effect of Pressure on AES Cell Performance at 1000°C.

Figure 3.19 — Power Output of an AES Cell at 1000°C as a Function of Pressure.

3.5 TASK 5.0 — PRESSURIZED SYSTEM TECHNOLOGY DEVELOPMENT

3.5.1 Pressurized SOFC/Gas Turbine Power System Conceptual Design @ 5 MW

The basic building block for an SOFC generator module is the 100 kW (atmospheric pressure) SOFC stack (1152 cells, each 22 mm diameter by 1500 mm active length). Please see Section 3.7.2.1 and Figure 2.2. Using two such building blocks, each with an additional cell row (96 cells), an SOFC submodule was configured with 2496 tubular AES SOFCs in a common canister. See Section 3.4.1. SOFC generator modules housing four such submodules comprise the SOFC systems in the 5 MW pressurized SOFC/GT power plants discussed in the following.

An operating pressure of ten atmospheres will yield about a ten percent increase in cell voltage, hence efficiency relative to atmospheric pressure operations. At three atmospheres, the voltage increase is about half that at ten atmospheres. Pressurizing the SOFC submodule results in a nominal capacity of 600 kW dc at nine atmospheres.

A pressurized SOFC/GT power plant utilizing a highly efficient two shaft intercooled and recuperated gas turbine [Heron] was evaluated (see Figure 3.20). In the gas turbine, the first shaft functions as a zero-net-power hot-gas generator, and ac power (1.4 MW) is produced by the power turbine on the second shaft. The gas generator is equipped with two stages of intercooled compression, and the compressor turbine has an inlet temperature requirement (861°C) that is very close to the temperature that is available at the exhaust of an SOFC module. The power turbine design inlet temperature is 863°C, which is normally achieved by burning natural gas fuel at a reheat combustor. To configure the PSFOC/GT cycle, two pressurized SOFC

modules, each containing four 2496 cell submodules, are placed upstream of the normal combustors, one at high pressure and the second at low pressure.

Figure 3.20 — SOFC/Heron Power Cycle.

The SOFC operating point can be selected such that no fuel is required by either combustor. This leads to a PSOFC/GT plant rating of approximately 5 MW net ac. Performance estimates for the 5 MW power plant are summarized in Table 3.4; based on a 93% effective recuperator.

Table 3.4
PSOFC/GT (Two-Shaft) Power Plant Performance Estimates

Plant	5 MW-class
Current Density, mA/cm ²	410/344 *
SOFC Fuel Utilization, %	90
SOFC Power, MW ac	3.98
Gas Turbine Power, MW ac	1.40
Plant Net ac Power, MW	5.36
Plant Efficiency, Net ac/LHV	71.5

* HP module/LP module

3.5.2 Pressurized SOFC/Gas Turbine Power System Conceptual Design: MW-Class

Utilizing a single-shaft gas turbine basis, and a pressurized SOFC module housing only one submodule, a MW-class PSOFC/GT power plant can be configured that will also achieve very attractive performance (see Figure 3.21). Figure 3.22 shows design-point performance for the plant operating with an SOFC fuel utilization of 85%. At each point on the curve the SOFC system configuration is fixed as indicated above, but the gas turbine sizing is allowed to change from point to point in response to changes in the SOFC operating point and the air mass flow requirement. Given a cell current density at each point on the curve, the SOFC stoichs requirement (3.5 minimum) was adjusted to maintain the recuperator effectiveness at 93%. The

selection of a particular design-point on the curve for a specific application will of course depend on economic considerations.

Figure 3.21 — SOFC/Combustion Turbine Power Plant Cycle.

Figure 3.22 — MW-Class PSOFC/GT Power Plant Performance Estimates.

For the Figure 3.22 analysis, the distribution between SOFC power and gas turbine power ranges from 81%/19% at the low-power point, to 66%/34% at the highest power.

This same small power plant could also be equipped with a heat recovery steam generator (HRSG) and a hot-water heater for cogeneration application. Plant performance estimates are provided in Figure 3.23 for the case in which the HRSG/water-heater combination is positioned at the recuperator exhaust exit. Again, these estimates are the result of a design-point analysis in which the SOFC system consists of a single SOFC submodule, but the gas turbine and heat recovery system sizing is variable. The features of a fixed plant design would depend on economics. HRSG steam conditions at each point on the curves were fixed at 10 atm (abs), 10°C superheat, and the exhaust temperature at the water heater exit at 70°C. Hot water temperatures in the 120 to 140°C range will be achieved depending upon the water mass flow rate. It is noted that the thermal performance of the heat recovery system is affected by the arrangement of equipment. For example, higher steam temperatures could be achieved by placing the superheater ahead of the recuperator. An air bypass around the recuperator would also affect thermal performance.

Figure 3.23 — MW-Class PSOFC/GT Cogeneration System Performance Estimates.

Since the PSOFC/GT combined cycle can be configured to achieve very high levels of efficiency at capacity levels two orders of magnitude smaller than non fuel cell systems, the Westinghouse PSOFC/GT is the ideal distributed power generator, and the concept has attractive cogeneration potential. Widely dispersed and located near load centers, Westinghouse PSOFC/GT systems could contribute to a world wide reduction in carbon dioxide production because of their inherent efficiency. Using desulfurized fuel, and with minimal fuel combustion, the PSOFC/GT system would also produce significantly less NO_x and SO_x than non fuel cell alternatives. The addition of the gas turbine as the bottoming cycle to the SOFC adds an increment of output without the consumption of additional fuel thereby yielding the higher efficiency. More importantly perhaps, the relatively low technology level of the turbine required results in the addition of this capacity and efficiency increment at far less cost than would be possible with fuel cells alone.

3.6 TASK 6.0 — MANUFACTURING FACILITY

3.6.1 Pre-Pilot Manufacturing Facility

The first Westinghouse cell manufacturing facility was the Pre-Pilot Manufacturing Facility (PPMF) located in Monroeville, PA. This 30,000 square foot plant was designed to produce cells up to a length of 1 meter in batch mode and longer length cells on an individual basis. The PPMF was shut down and all applicable equipment moved to a new PMF facility (below) over the course of two years (1995-1996).

3.6.2 Pilot Manufacturing Facility

A major milestone of the Westinghouse commercialization program was the completion in 1996 of the 40,000 square foot Pilot Manufacturing Facility (PMF) dedicated to SOFC technology (Figure 3.24 and Figure 3.25). The centerpiece of the PMF is an EVD reactor capable of processing 108 AES cells, 22 mm in diameter and 1500 mm long in a single batch. The capacity of the PMF is 4 MW of SOFC per year. The PMF provided the opportunity to complete the manufacturing and process development and enables Westinghouse to put in place the processes and quality control programs that will be required to commercialize the SOFC technology.

Figure 3.24 — PMF.

Figure 3.25 — PMF.

3.6.2.1 PMF Cell Processing

Cell Processing Summary. Incoming air electrode tubes are inspected, cleaned and have plasma sprayed interconnections applied using vendor supplied doped lanthanum chromite powder (Figure 3.26). Interconnection assemblies are prepared for EVD electrolyte processing (Figure 3.27) after which fuel electrodes are applied to electrolyte assemblies in the HVLP spraying area (Figure 3.28) prior to EVD fuel fix processing. Fuel fix assemblies are nickel plated to complete fabrication. Finally, finished cell assemblies are heat cycled (Figure 3.29) and inspected to determine compliance to drawing requirements before being released for generator use.

Figure 3.26 — Plasma Spray Processing.

Figure 3.27 — EVD Processing Area.

Figure 3.28 — HVLP Area.

Figure 3.29 — Heat Cycle Furnace.

Processing Problems: Cell production for the EDB/ELSAM 100 kWe generator was scheduled to start in October of 1995 and to be completed by June of 1996. Actual cell production did not begin until March, 1996 and took over a year to complete. Most delays were related to slippage of the aggressive design/fabrication/installation/shakedown schedule or due to delays in receipt of vendor supplied materials or components.

Plasma Spray Booth: No major problems occurred during the relocation of the booth from the PPMF to the PMF at STC, however, delays caused by improper installation of support hardware (exhaust ducts and compressors) occurred.

EVD: The most significant delays in the relocation process occurred in the EVD area. Many delays were the result of poor equipment design and quality control on the part of hardware vendor. Also, large tolerances resulted in non-uniform grinding/feeding characteristics which created unacceptably high pressures during metal chloride feeding. Also, an ashless paper chosen for use in pressing chloride bars was later found to be clogging the EVD. A large percentage of the rejected cells during this campaign were associated with EVD anomalies.

Fuel Electrode Application (HVLP): During fuel electrode processing, delays were encountered due to spray gun nozzle clogging, seal and leak/spillage problems in the pumping system, water curtain pump train upsets and motion control system program memory losses caused by electrical power surges.

Nickel Plating of Interconnection: Poorly attached nickel deposits were produced during the 100 kWe generator production campaign. The problem was most noticeable after heat cycling. After an extensive investigation was completed, the following were identified as the main causes contributing to poorly attached or peeled nickel electroplating: variability of interconnection electrical resistance along the length, plating deposit thickness, variability of interconnection surface roughness and plating apparatus positional effects affecting deposit quality. Each cell nickel plated after October of 1996 was subjected to a plating integrity test to identify

poorly attached electroplating deposits. Cells that failed were replated with modified equipment using improved surface preparation techniques, pre-plating processing techniques, plating parameters and drying parameters. Nickel plating requirements were altered to accentuate functional requirements rather than visual appearance characteristics. Since the described changes were implemented, peeling has not been a problem and plating yield has been above 90%.

Waste Treatment: Waste treatment system problems were confined to the sand filter used to remove fine particulates from liquid wastes and in the ability to maintain consistent additions of chemicals needed to achieve proper flocculation. Additional waste processing difficulties were caused by the addition of HVLP waste (which was not expected to require treatment in order to meet regulatory discharge requirements) and to EVD processing upsets which placed unforeseen burdens on the waste treatment system.

3.6.2.2 Commercial Manufacturing Facility

As part of the commercialization plan for SOFC, a Commercial Manufacturing Facility (CMF) was sized and a conceptual design was performed. The CMF design consists of three duplicate production lines, each with an annual capacity of 100 MWe of SOFCs (~150 MWe of systems) assuming 3 shifts per day and 330 production days per year. The process flow diagram for the "1997 AES Type" cell is as specified in Figure 3.30, and the above production rates are the basis for the CMF manufacturing equipment selection and sizing. The manufacturing equipment cost estimates were based upon quotations from equipment suppliers and scale-up of known PMF equipment costs accounting for size and cost savings associated with the purchase of multiple machines. In addition, no credit was taken for ongoing work to reduce the capital cost of the EVD process while maintaining its high levels of product quality and equipment reliability. The equipment costs for the first 100 MWe per year cell production line was estimated at \$45 million while the second and third production lines were estimated at \$40 million each.

In order to determine the building size, a layout of the three cell production lines was performed. In addition, space was allocated for SOFC module assembly and system assembly assuming the output of the CMF to be skid-mounted, fully packaged 3 MWe PSOFC/GT Power Systems requiring a minimum of site installation. The cost of the manufacturing building and property was estimated assuming both new construction and the purchase of an existing building. It was concluded that utilizing an existing building was advantageous to minimize the initial capital investment for the CMF as well as to take advantage of an abundance of vacant sites. Preliminary estimates require approximately 250,000 ft² of 35 ft high bay space per production line of 150 MWe per year of 3 MWe power systems for a total floor space of 750,000 ft². The building purchase price for an existing building of this size was estimated at \$45 per ft² or \$35 million.

In the cost study the manufacturing equipment was depreciated over 10 years and the building was depreciated over 20 years. These annual costs were included as part of the cell manufacturing cost.

Figure 3.30 — SOFC Process Flow Diagram (CMF).

3.7 TASK 7.0 — POWER SYSTEM DEMONSTRATIONS

3.7.1 PPMF Summary of Generator Units Delivered

3.7.1.1 The UTILITIES 25 kWe-Class SOFC Experimental Field Unit

The "Utilities," a consortium of the Kansai Electric Power Company (KEPCO), The Tokyo Gas Company, and the Osaka Gas Company sponsored a 25 kWe-class experimental field unit tested at facilities operated by KEPCO. The generator modules used in the 25 kWe-class Utilities Unit utilized cells of 500 mm active length, and consisted of two independent dc generation systems (modules A and B), each incorporating a 576-cell, 20 kW class stack.

Assembly of the unit was completed in November 1991 and after factory start-up and operational tests, the unit was shipped via overland truck and air freight to the Rokko Island Test Center for Advanced Energy Systems (near Kobe, Japan) operated by The Kansai Electric Power Company. Testing was initiated in February 1992, following installation and three weeks of pre-operation on-site software and hardware verification directed toward licensing. This unit was the first high temperature fuel cell certified by Japan's Ministry for International Trade and Industry (MITI) as being in compliance with all applicable regulations for fossil fuel power plants.

Module A operated for 2529 hours at an average output of 16.8 kW dc, achieving a maximum output of 17.7 kW dc. The system was started from ambient temperature and cooled back to ambient temperature four times and as a result of various system difficulties endured nine partial cool-downs. The final shutdown was induced by current stability problems with the dissipator. A restart was not possible because of damaged cells and Module A was returned to the factory.

Module B operated for 1576 hours at an average output of 17.4 kW dc and achieved a maximum power of 19.2 kW dc. The system was started from ambient temperature twice and experience two shutdowns to ambient temperature and three partial cool-downs. The final system shutdown occurred during a period of operational probing for maximum output. Subsequent post operation investigation showed that the unit was operated at excessively high fuel utilization as evidenced by a detachment of the fuel electrode from a few cells in quadrant 4. Module B was returned to the factory for repair. At the factory, quadrant-4 was replaced, but the remaining quadrants were not examined in great detail. A subsequent restart in October 1992 was aborted because of excessive internal temperatures. Subsequent post operation examination revealed that approximately six percent of the cells had cracked.

Module B was rebuilt with new cells (Module B-2) and returned to Rokko Island without factory start-up. Installation and start-up took place during the last days of March 1993. Start-up was successful, but hampered by a lower than expected cell voltage and an instability in the output of the natural gas mass flow meter. Field modification of the control software during operation permitted completion of an extended start-up process. Module B-2 operated for 7064 hours on PNG spanning the period from 3/31/93 to 3/10/94. Nominal output during this period was 17 kW dc. Shutdown was caused by a failure of the air supply system. Drive belts for the blower failed, and the motor bearings failed the day after belt replacement. The operational period was vexed by inadequate ambient temperature compensation in the air and PNG flow meters. Analysis of cool-down data indicated the existence of anomalous temperatures and a restart was not attempted.

A photograph of The Utilities 25 kWe-class SOFC is shown in Figure 3.31 and a plot of performance versus time is shown in Figure 3.32.

Figure 3.31 — The UTILITIES 25 kWe-Class SOFC Experimental Field Unit.

Figure 3.32 — The UTILITIES 25 kWe-Class SOFC Module B-2 Performance.

3.7.1.2 Joint Gas Utilities (JGU) 25 kWe-Class SOFC Experimental Field Unit

The Joint Gas Utilities (JGU), a consortium of the Tokyo Gas Company and the Osaka Gas Company, sponsored in 1992 a 25 kWe-class SOFC cogeneration system that utilized two SOFC generator modules in a single packaged enclosure. Each generator module used 576 cells, 16 mm diameter by 500 mm active length, of the now obsolete porous zirconia support tube (PST) design. As this unit did not perform satisfactorily, it was modified and repaired.

The JGU cogeneration system was modified to accept a single generator module utilizing 576 air electrode supported (AES) cells of 16 mm diameter by 500 mm active length in place of the previously used pair of PST type modules. This repaired unit was originally scheduled to be shipped to Osaka, Japan for application testing, but the great Hanshin earthquake which struck the Kobe area in January 1995 disrupted the operations of Osaka Gas to such an extent that this proved infeasible. An extended factory test of the JGU air electrode supported (AES) SOFC Cogeneration system was initiated on 22 March 1995. One anomaly apparent early in operation was that there was a much more marked spread in the control temperatures for the four quadrants, approximately 50°C, than had ever been observed in the past. Except for this quadrant to quadrant temperature anomaly, the stack appeared to be in excellent condition with good internal voltage balance and good internal temperature distribution.

Since the SOFC units are controlled for temperature based upon the maximum of the quadrant control temperatures, the coldest quadrant limited the maximum achievable current and constrained operation. For approximately 500 hours the cells continued to improve and current was increased. A maximum power of 21.4 kW was achieved at 180 amperes, and 80% fuel utilization. Since the bottom of the cells in the coldest quadrant were operating much colder than desired, the current was decreased and the unit operated at maximum air flow so that with the aid of the electric air heater, the cells would achieve a more uniform and higher average temperature. Cell voltage improvement was observed until about 850 hours into operation, after which it was observed the cells in three quadrants were declining in voltage. Various diagnostic activities were undertaken, and at approximately 1200 hours of operation the unit was shut down since it became clear that the reason for the voltage decline was a combination of deficient fuel flow in the hottest quadrant due to a fuel leak and reformer deactivation as evidenced by a change in the temperature distribution and an increased burden on the air heater to maintain temperature—i.e. cooling of the cells.

After shut down of the JGU unit, cold flow tests were conducted to confirm balance in air and fuel flows and corrective measures taken to ensure fuel flow balance and eliminate fuel leaks from the reformers. In addition, the reformer catalyst was replaced and the control algorithm designed to ensure adequate oxygen to carbon atom ratio in the reformer revised.

The JGU 25 kWe-class AES-SOFC Cogeneration system was moved from its site of manufacture, the Westinghouse Pre-Pilot Manufacturing Facility (PPMF) in Monroeville, PA to the new Pilot Manufacturing Facility (PMF) located at the Westinghouse Science & Technology Center in mid July 1995. The unit was tested at the PMF from August 7, 1995 through February 10, 1997 and accumulated 13,194 hours of operation along with ten complete thermal cycles on local desulfurized natural gas. The unit achieved a power output of 24.9 kWe without steam co-generation, and less than 2% power degradation was measured for the entire 13,193 hour test period for the unit operating at 85% of its maximum power. A photograph of the JGU Cogeneration System is shown in Figure 3.33 and a plot of performance versus time for the AES-SOFC modified unit is shown in Figure 3.34.

Figure 3.33 — JGU 25 kWe-Class SOFC Cogeneration System.

Figure 3.34 — JGU kWe AES-SOFC Power System.

3.7.1.3 Southern California Edison (SCE) 25 kWe-Class SOFC Experimental Field Unit

A 25 kWe-class SOFC system unit was delivered to the Highgrove Generating Station of SCE in Grand Terrace (near San Bernardino), CA in March 1994, and in late May 1994 a PST-SOFC stack was installed. The unit was successfully started on June 7, 1994.

For a program period of 8000 hours, the 25 kWe-class PST-SOFC system operated for 6079 hours. Shortly after startup, the unit suffered two unscheduled shutdowns. The first was due to inadequate natural gas pressure into the gas compressor, and the second was due to an inadvertent interruption of UPS sourced power to the unit. For the first 1700 hours of operation, cell performance improved with time and SCE operators exercised the system over a range of operating parameters. The maximum power achieved was 18.9 kW at 85% fuel utilization and a generator control temperature of 1020°C. After about 1700 hours, system performance began to decline. The last 4000 hours of operation was not continuous, but was interrupted by three site power outages at the SCE test site, at 1900 hours into the test, and at 4550, and 5100 hours. An attempt was made to restart at 5700 hours into the program. Unfortunately, the electric air heater used for startup failed at this point and the restart was aborted. The spare heater was installed, but programmatic difficulties within SCE prohibited restart until approximately 6400 hours into the program, on March 2, 1995. From this date until final shutdown at 8000 program hours, (6015 hours of power generation) operation went smoothly although the power capability of the stack was declining. On May 2, 1995, the unit was shut down in order to permit installation of a new stack incorporating the air electrode supported (AES) tubular cell technology.

3.7.1.4 SCE/ARPA Generator

In late May 1995, a Westinghouse field service team removed the PST type stack from the 25 kWe-class SOFC system located at the Southern California Edison (SCE) Highgrove Generating Station in Grand Terrace, CA and returned it to Westinghouse manufacturing facilities in Pittsburgh for post test evaluation. A replacement stack utilizing air electrode supported (AES) cells was installed. This new stack was fabricated as part of a project funded by the United States Department of Defense (DOD) Advanced Research Projects Agency (ARPA) administered by the National Aeronautics and Space Agency (NASA) under contract NAS3-27022. Under the NASA contract, the SOFC system was modified so as to be able to accept as fuel either natural gas or reformat from a logistic fuels (DF-2 diesel or JP-8 jet turbine fuel) processor located externally to the SOFC system. The NASA contract also supported the development of a "brassboard" logistic fuels processor. This LFP was developed by Haldor Topsoe, Inc. under a subcontract.

The ARPA/SCE AES SOFC system was successfully started on May 23, 1995 on natural gas fuel and shifted to LFP fuel on July 25, 1995. Through February 26, 1996, the AES-SOFC operated for a total of 5582 hours, 766 hours on JP-8 fuel, 815 hours on low sulfur DF-2 diesel fuel, 740 hours on high sulfur DF-2 and 3261 hours on natural gas. During this time, the unit endured six thermal cycles to ambient temperature, with no apparent degradation. Peak power of 27 kWe-dc was achieved using both natural gas and DF-2 fuel. See Figure 3.35 for a plot terminal voltage and current vs. time.

YSZ over porous air electrode tube followed by sintering is also being investigated. If successful, this will result in further reduction in the cost of manufacturing SOFCs.

3.1.2 Cell Manufacturing Development

Two Westinghouse cell manufacturing facilities have been dedicated solely to SOFC technology. The first plant was the 30,000 square foot Pre-Pilot Manufacturing Facility (PPMF). The equipment in the PPMF was designed to produce cells up to 100 cm in length in batch mode, with the capability of making individual cells of lengths up to 150 cm. Aside from the 100 kWe EBD/ELSAM Generator, all field test demonstration units were built in the PPMF.

The facility which was most recently completed is the Pilot Manufacturing Facility (PMF) and has completely replaced the PPMF as the cell and generator manufacturing facility. The PMF was completed in 1996 and is comprised of 40,000 square feet of high bay space. The capacity of the PMF is 4 MW per year of commercial size cells of 22 mm outside diameter, 150 cm of active length. The largest and most recent SOFC power system, the 100 kWe EDB/ELSAM Generator, was built in the PMF.

3.1.2.1 Pre-Pilot Manufacturing Facility (PPMF)

Air Electrode Supported (AES) Cell Production. Electrical tests successfully demonstrated that self-supported air electrode tubes could replace the calcia-stabilized zirconia porous support tube/air electrode assemblies used previously. Performance enhancements due to improved diffusivity characteristics and lower resistivity of the one-piece air electrode tube were realized in addition to decreased cell production costs. NGK¹ tubes, delivered in 8/91, were the first commercially fabricated calcium-doped lanthanum manganite self-supported air electrode tubes to be manufactured, processed into finished cells and successfully electrically tested. Bundle Tests demonstrated that 15.8 mm diameter one meter active length air electrode supported (AES) cells could be bundled and utilized in applications.

Doping of Calcium Stabilized Lanthanum Manganite Air Electrode Tubes. The first self-supported air electrode tubes were composed of calcium doped lanthanum manganite. This material exhibited an unacceptably high level of thermal cyclic shrinkage during electrical operation. In order to fabricate several hundred dimensionally unstable 15.8 mm one meter length AES tubes into finished production-quality cells, a series of experiments were conducted to determine the feasibility of dimensionally stabilizing the air electrode material by taking advantage of small cation (B-site) substitution techniques. "X" was shown to be an effective dopant that improved dimensional stability of the air electrodes while maintaining other important refractory properties of the perovskite material. The technique of incorporating "X" into the lattice structure was optimized and qualified by single cell and bundle-type electrical tests before being used in generator units.

Cell Optimization Study. In 1992, a study was initiated to determine the optimum cell size for commercial generator applications. During the course of the investigation, cell features such as interconnection width, fuel electrode width, cell diameter, cell length and component thickness were explored. Over 90 different combinations of cell characteristics were analyzed to select the optimum cell configuration that would improve performance and meet other requirements.

¹ NGK Insulators, Ltd., 2-56 Suda-cho, Mizuho-ku, Nagoya, 467 Japan was developed as a qualified vendor for lanthanum manganite tubes for fuel cells to Westinghouse exclusively.

A small group of cell designs were selected based on their predicted electrical performance improvements. The mechanical strength of each preferred candidate cell design was estimated to ensure a high probability that it could survive fabrication and generator assembly without failure. Additional analyses were also performed on candidate cell configurations to determine suitability for the generator operating environment based on air feed pressure drop calculations. From this investigation, the current 22 mm diameter 150 cm active length cell design was selected.

HVLP Fuel Electrodes. At the PPMF, green fuel electrodes of 50 and 100 cm active length cells were applied using a vertical dipping process in a confined work area. This work area was not large enough to process 150 cm and longer cell designs and, based on the results of one meter cell development work, it was believed that maintaining uniform end-to-end fuel electrode properties to meet engineering requirements on a production basis was not achievable. A high volume low pressure (HVLP) spray application technique was selected as a replacement for fuel electrode dipping. Electrical tests demonstrated that acceptable performance could be obtained using the new technology. When the PMF was constructed, the HVLP spraying process was incorporated into the cell production line. All cells manufactured for the EDB-ELSAM 100 kWe generator were fabricated using the HVLP spraying technique.

Production of 1.81 (overall length) Meter Cells at PPMF. Activities associated with converting from small to large cell diameter production began in 1995 at the PPMF. Major processing changes in the areas of plasma spray interconnection application, EVD processing, nickel plating, heat cycling and electrical qualification testing had to be implemented to accommodate the new cell design. The most difficult challenges were associated with modifying EVD electrolyte and fuel fix process parameters and equipment. In April of 1995 the first process development EVD electrolyte run was completed using eighteen 22 mm diameter 1.8 meter over all length cells.² During the next four months, approximately 250 large diameter cells were processed through the PPMF to gain further understanding of processing capabilities and requirements associated with the new cell design. An electrical test was conducted to confirm the electrical and thermal cyclic performance of the 22 mm diameter 1.8 meter overall length cell design. Much of this information was used to assist in the design of the next generation pilot manufacturing facility (PMF) located at the Science & Technology Center (STC).

Demonstration of Fuel Electrode Sintering Capability at PPMF. Eighteen large diameter 1.8 meter overall length cells were processed with sintered fuel electrodes in August of 1995 at the PPMF. The EVD reactor was modified to perform the fuel electrode sinter cycle at one atmosphere to verify proof of concept. Based on the success of that activity, a decision was made to refurbish the PPMF EVD reactor as a fuel electrode sintering furnace for use at the SOFC PMF.

3.1.2.2 Pilot Manufacturing Facility (PMF)

Seamless AES Tube Qualification. A one-piece or seamless closed end design conceptually offered the potential to decrease tube integrity concerns associated with the plugged tube design because the closed end and tube would be extruded as a single unit from the same material. In addition, costs of producing the seamless tube were expected to be reduced due to the simplified closed end forming technique and decreased finished machining requirements associated with the as-extruded near net shape of the closed end. A 30 cell pilot production run

² Note: A cell with an active length of 150 cm, as measured by the length of the interconnection, will have an overall tube length of 1.8 m.

was completed to qualify large diameter 1.8 meter length NGK seamless air electrode tubes with the WPC3 composition for use in SOFC generators. Two sub-groups, consisting of 15 tubes each, were processed. One sub-group was fabricated with a higher elastic modulus. The second sub-group had the standard maximum elastic modulus. The new design AES tubes were fabricated, inspected, processed, electrically tested and microcharacterized. The results of the qualification activity confirmed that both the low modulus and higher modulus seamless air electrode tubes met Westinghouse engineering requirements and were acceptable for generator applications. Seamless air electrode tubes were first used in the EDB-ELSAM 100 kWe generator.

Plasma Spray Powder Development for Interconnection Application. During the course of developing doped lanthanum chromite powders for plasma spray application of the interconnection, different slurry compositions were formulated. Each slurry formulation was given a different designation (A1 through A9). The A4 - A9 type powders were considered superior to the others based on ease of preparation and finished component quality. The final results of microcharacterizations, vacuum leak rate tests, standard electrical and accelerated interconnection degradation tests indicated that the A6 powder chemistry was more robust than the others. As a result, the A6 powder composition was specified for use during the EDB-ELSAM 100 kWe production campaign. Over 30 different A6-type powder batches were consumed throughout the fabrication effort. A review of powder receiving inspection and cell processing/testing data associated with each powder batch produced recommendations to further improve powder and interconnection quality. These recommendations included: modifications to powder feedrate, agglomerate size distribution, chemistry, flowability, benefits of sieving, binder content and vacuum leak rate requirements. Experiments are continuing to confirm the validity of these recommendations with the objective of defining optimum plasma spray processing parameters, powder properties and interconnection requirements for SOFC applications.

Dipped Fuel Electrodes. Green fuel electrodes were applied using the HVLP spray process during the 100 kWe production campaign. Significant difficulties were encountered trying to maintain throughput due to spray gun nozzle clogging and extensive cleaning and work area maintenance issues. In addition, approximately $\frac{3}{4}$ of the slurry was wasted as over-spray. Yield, throughput capability, waste treatment and cost reduction issues prompted an investigation to replace the HVLP process. Experiments using modified nickel slurry dipping techniques confirmed that fuel electrode application by slurry dipping was more efficiently and easily controlled as compared to HVLP spraying. Subsequently, trial fuel electrode dipping runs using a newly installed dipping apparatus established that 1.81 meter long cells with green fuel electrodes applied using the dipping process consistently met applicable engineering drawing and specification requirements. The fuel electrode dipping process using a water based nickel slurry has been implemented to apply green fuel electrodes for the SCE 250 kW PSOFC/MTG generator production campaign.

A Comparison of EDB-ELSAM and SCE Generator Production Yields. Due to feedstock quality and process or equipment shakedown difficulties associated with production startup of the new SOFC pilot manufacturing facility (PMF) at STC, the overall production yield for the EDB-ELSAM production campaign was approximately 50% which included a high number of rejected air electrode tubes. Problem areas were primarily confined to air electrode tube quality, variability of plasma spray powder, EVD processing, nickel plating attachment and heat cycling. The overall yield through the first half of production (excluding the air electrode

tubes) for the SCE generator stands at approximately 85%, and the yields in the individual areas where the most problems occurred are all now greater than 90%.

Chloride Recovery. Only about 20% of the metal chlorides currently fed into the EVD reactor are utilized to form the electrolyte deposit. Recent tests verified that approximately 30-40 weight percent of the chlorides fed can be recovered and reused. This offers the potential to reduce chloride costs significantly. Follow-on tests are underway to verify predictions and determine if this approach is cost effective relative to other efforts to reduce chloride costs.

Reductions in Chloride Consumption. Two other approaches have been investigated to reduce metal chloride consumption during EVD processing and thereby reduce costs. The first is to reduce the length of the reaction phase itself, and the second is to increase the utilization rate of metal chlorides so that less material is wasted. Investigations into reaction phase reductions have shown that chloride deposition times can be decreased by 35% or more with little change in electrolyte quality. The most noticeable effect of reducing deposition time is decreased electrolyte thickness. The data accumulated so far indicate that noticeable reductions in electrolyte thickness can be tolerated with no loss of performance or endurance characteristics during electrical operation.

Lowering the metal chloride feedrate to increase utilization is another way to reduce chloride consumption. If the theoretical weight of metal chlorides required to produce the desired deposit thickness are divided by the total weight of chlorides delivered during the reaction phase, a bulk utilization percentage is established. Recent data suggest that bulk utilizations as high as 60% can also be tolerated without sacrificing electrolyte quality.

Qualify Alternate Chloride Vendors. Investigations are underway to identify and qualify additional suppliers of metal chlorides so that projected raw material requirements necessary to meet future production objectives can be satisfied. To date, several prospective vendors have been contacted and have supplied samples of materials that meet compositional requirements. Costs for these materials are competitive with current suppliers.

3.2 TASK 2.0 — CELL VOLTAGE STABILITY, DURABILITY, AND LIFETIME

3.2.1 Atmospheric Testing

A tubular SOFC, manufactured using the porous support tube technology of the 1980's, has surpassed seven years of power operation with a cell voltage degradation rate of less than 0.5% per 1000 hours of operation (see Figure 3.3). The test was terminated on December 23, 1996 after completing 69,000 hours due to a sudden decrease in voltage. Figure 3.4 shows the number of AES cells and the total operating hours through and including the most recent field unit of 50 cm cells.

Figure 3.3— Total Time on an SOFC Performance Test.

Figure 3.4— AES-SOFC Operating Time.
(50 cm active length)

A large number of AES cells with plasma sprayed interconnections have been electrically tested for up to about 7,000 h. These cells have shown the ability to perform for extended periods of time under a variety of operating conditions with little performance degradation. The performance degradation has decreased from about 0.5% per 1,000 h for PST cells to less than 0.2% per 1,000 h for AES cells. Figure 3.5 shows the voltage output of two AES cells (15.6 mm diameter, 50 cm active length) as a function of time at 1000°C and 450 mA/cm² current density with 89% H₂ + 11% H₂O fuel (85% utilization) and air as oxidant (6 stoichs). The voltage shows no degradation over time. The voltage-current characteristics of the AES cells (15.6 mm diameter, 50 cm active length) at 900, 950, and 1000°C, with 89% H₂ + 11% H₂O fuel (85% fuel utilization) and air as oxidant (4 stoichs) are shown in Figure 3.6. These voltage-current characteristics at 1000°C are compared with those of the thick-wall PST and the thin-wall PST cells, under the same conditions in Figure 3.7. The change from PST cells to the AES cells has resulted in a power density increase of about 33%.

In addition to improved performance, the AES SOFCs have shown the ability to thermally cycle to room temperature over 100 times without any mechanical damage or performance loss as shown in Figure 3.8. This ability to sustain thermal cycles is essential for any SOFC generator to be commercially viable. The thermal cycles in Figure 3.8 are for temperature ramps from ambient temperature to 1000°C within 5 h or less. This ramp rate of 200°C/h would allow a generator to move from a hot standby condition at 600°C to full power at 1000°C within 2 hours.

A long cell testing station supplied to the Kansai Electric Power Company (KEPC) recently completed the exercise of a test article holding 4 cells of 150 cm active length. The test article generated power for over 10,000 hours and was thermally cycled 100 times (see Figure 3.9).

Figure 3.5 — Voltage Stability of Two AES Cells at 1000°C and 450 mA/cm².

Figure 3.6 — Voltage-Current Characteristics of an AES Cell at Different Temperatures.

Figure 3.7 — Comparison of the Voltage-Current Characteristics of the Thick-Wall PST, the Thin-Wall PST, and the AES Cells at 1000°C.

Figure 3.8 — Thermal Cycling Capability of AES Cells.

KEPCO Test Article #2 Performance
[10,700 hours of power generation, 100 thermal cycles]

Figure 3.9 — Thermal Cycles on Commercial Length Cells.

3.2.2 Materials Evaluation and Microstructural Analysis

A representative micrograph of the plasma sprayed interconnection is shown in Figure 3.10, which shows the interconnection to be uniform in thickness and dense with no open porosity.

Figure 3.10 — Representative Micrograph of the Plasma Sprayed Interconnection Over Air Electrode.

Sintered fuel electrode polarization is lower than the EVD fuel electrode polarization. This is believed to be due to a larger contact area and a greater number of electrochemically active sites at the electrolyte/sintered fuel electrode interface. Representative micrographs of the two types of fuel electrode are shown in Figure 3.11.

Figure 3.11 — Representative Micrographs of an EVD Fuel Electrode and a Sintered Fuel Electrode.

3.2.3 Reliability Enhancement

Figure 3.12 shows the number of AES-SOFCs which have been thermally cycled versus the number of cycles experienced. The figure shows that out of the AES-SOFCs completing between one and 106 thermal cycles to room temperature, eight have resulted in failure, all of which can be traced to either known pre-existing flaws or erroneous operation. The cells with pre-existing flaws, determined from extensive quality control inspection, would not have been acceptable for generator applications.

Figure 3.13 shows a typical quality control thermal cycle endured by selected production cells to verify integrity of cell batches. The plot shows two cycles in temperature to 600°C followed by a drop to under 300°C with the cells going to open circuit on the decrease in temperature maintaining air on the oxidant side and a reducing environment on the anode side, reloading during the temperature increase above 600°C and reaching a current density of 335 mA/cm² at 1000°C. It should be noted that the cells return to their previous operating point after each current reload.

AES - SOFC Thermal Cycle Endurance

Through 1/15/97

Figure 3.12 — Number of Cells vs. Thermal Cycles for 50 cm AES Cells.

Figure 3.13 — Response of an AES-SOFC to Rapid Thermal Cycling.

3.3 TASK 3.0 — MODULE COST REDUCTION

3.3.1 Module Component Development

Module Description

The SOFC Module is made up of closed end cylindrical cells assembled into arrays. Nickel felts are used between the cells to form electrical interconnections. Arrays of cells are combined to produce generator modules of various sizes. Westinghouse devised the cylindrical cell with the closed end and axial interconnect strip in the early 1980's and developed the seal-less module design to accompany it (Figure 3.14).

Each cell is gas tight with air introduced via a feed tube to the bottom or closed end of the interior of the cell. The air exits the feed tube, turns, and flows back up in the annular area formed by the feed tube outer diameter and the cell inner diameter.

Prior to feeding fuel to the cells, it is partially reformed to eliminate any potentially damaging higher hydrocarbons. Internal stack reformers were developed to completely reform the fuel before it is fed to the cells and also to help maintain an even cell temperature profile over the active length of the cells. The water for the reformation process is obtained internally by using a spent fuel recirculation loop. Once the fuel has been reformed it is introduced at the bottom (closed) end of the cell and flows over the exterior surface.

The fuel and airflow both flow upward toward the open end of the cell. Typically, 85% of the fuel is consumed electrochemically within the SOFC module. Due to the closed end, gas tight design of the cells, this array of vertically oriented gas tight cells requires no high integrity seals to separate fuel from oxidant.

Figure 3.14 — Cross Section Showing SOFC Generator Module and Flow Paths.

3.3.2 Module Cost Reduction

The following is a partial list of cost reduction efforts during this period:

- **Low cost canister material:** The canister material was changed from Inconel 600 to Stainless 304 at a substantial cost savings. High temperature oxidation corrosion tests were conducted in dual fuel-oxidant exposure conditions to qualify the use of the new material.
- **Inner insulation vendor evaluation:** A low cost supplier for the high purity all alumina inner insulation board has been identified and the facilities evaluated. Alumina insulation board costs have been cut in half as a result of the new supplier.
- **Cell bundling operation:** A sintering and bonding agent has been developed to facilitate bundle sintering at lower temperatures. The use of the sintering agent could eventually eliminate the need for very high temperature furnaces. The previous bundle sintering process took place at high temperatures. Also, several intermediate steps involved in the current bundling process may be eliminated by using the new agent.
- **Net shape insulation:** Experiments have been conducted to evaluate the fabrication of inner insulation into net shape in a process somewhat like casting, called net shaping. Net shape fabrication reduces machining costs and mate-

rial waste otherwise seen from buying boards and cutting away material to obtain a final desired shape.

- **Metallic liner:** A concept for using a thin metallic liner was developed to replace the canister wall as a fuel boundary. Oxidation resistant materials such as Haynes 214W Nickel Aluminide have been identified for this application. The use of a liner results in a lower cost fuel boundary around the cell stack.
- **Low cost insulation:** A type of silica abatement material is being considered for use in the generator along with lower purity insulation. The intention is that the abatement system will prevent Silicon contamination of the fuel cells and the peripheral system components.
- Even with the excellent reliability shown by AES-SOFCs and the extensive quality control inspection used to avoid placing defective cells into generators, the possibility of a cell failing over the life of a commercial generator exists. In response to this potential problem, Westinghouse has demonstrated a bundle repair procedure allowing for any number of cells to be replaced with minimal shutdown time. Also, tests of cell bundles have demonstrated that cells adjacent to failed cells show no damage and can be restarted once the failed cells are replaced. To demonstrate this feature, Westinghouse ran a test containing two 3×6 cell bundles of which some cells were known to have pre-existing flaws. This test operated on both natural gas and hydrogen for 2000 hours before three defective cells were found to have failed. At this point, the test was shut down and the failed cells were removed from the bundle and replaced with new cells. Though the cells adjacent to the failed cells showed some oxidization of the fuel electrode, they were subsequently reduced upon returning to fuel gas and performed without any sign of permanent degradation. The test was then restarted and ran for an additional 4300 hours.

3.3.3 Bundle Test Articles

3.3.3.1 Atmospheric Tests

BTA# 4 was a DOE sponsored test to evaluate, in a generator environment, the 50 cm active length porous support tube (PST) cell produced at PPMF. Operating time of the test, which concluded in December of 1990, was 6840 hours and included eight thermal cycles. The test performance showed no significant voltage degradation although high cell leak rates were measured after disassembly of the test article.

BTA # 4.1 - 4.3 was a series of tests performed as a part of the development effort for the new air electrode supported (AES) cells. The tests were used to verify the performance of new cell designs and new manufacturing processes for 50 cm active length cells which were used in the JGU and SCE/ARPA 25 kWe-class generators. The tests all contained 36 cells of 16 mm diameter arranged in two three by six bundles.

BTA# 5.0-5.1B was the first series of test articles to incorporate two larger, longer bundles each composed of 24 AES cells (three by eight bundles) of 100 cm active length, 16 mm diameter. The test objective was to evaluate new generator components and assembly processes intended for implementation in the 100 kWe generator. More specifically, testing the internal reformer design, the cell string and the generator assembly procedures were the main objectives of the

tests. The internal reformer design was developed and verified for generator use during this series of test articles.

BTA # 6.0 and 6.0A were used to evaluate the performance of EDB/ELSAM 100 kWe generator cells and generator components. The tests were composed of two bundles of cells (48 cells total) each 22 mm diameter with 150 cm active length and contained prototypic generator components. BTA6.0A was used to verify the rapid repair process for removing and replacing individual cells within the stack, using two cells that were manufactured in the PMF.

3.3.3.2 Pressurized Tests

PBTA # 1.0 is the first test in a series of pressurized bundle test articles. The test contains 48 cells of prototypic design, 150 cm active length and 22 mm diameter. The objective of the test will be to verify component operation at elevated pressures, including cell performance, internal reformation and fuel flow paths through insulation.

3.4 TASK 4.0 — PRESSURIZED MODULE TECHNOLOGY DEVELOPMENT

3.4.1 Pressurized Module Conceptual Design

A conceptual design was performed for a multi MW PSOFC generator, consisting of multiple SOFC submodules contained within a common pressure vessel as illustrated in Figure 3.15. Each submodule consists of 2496 SOFCs arranged in 26 bundle rows, each consisting of 96 cells, as shown in Figure 3.16 and Figure 3.17, 16 air plenum assemblies which include 2496 alumina air feed tubes, 24 stack reformer sections which are sandwiched between bundle rows, a spent fuel recirculation loop (two ejectors and one pre-reformer), insulation, instrumentation, and 1 stack container. Finished cells are assembled into bundle rows with the use of nickel felts. The stack is then assembled by arranging the bundle rows and stack reformer sections in an alternating pattern on a support assembly which also distributes the reformed fuel to each cell. The spent fuel recirculation loop and the air plenum assemblies are then installed. The stack is then surrounded by insulation and inserted into a stack container completing the module assembly. Multiple modules are inserted into the pressure vessel as shown in Figure 3.15 forming a multi MW PSOFC generator. Power output will be a function of operating pressure, temperature, fuel utilization, and current density. At nine atmospheres pressure, 1000°C average temperature and 85% fuel utilization, the maximum power for a 2496 cell module is approximately 600 kWe.

Figure 3.15 — Multi MWe PSOFC Generator.

Figure 3.16— Cell Bundle Row.

Figure 3.17— 600 kWe Cell Stack.

High-Temperature Tubular Solid Oxide Fuel Cell Generator Development Final Report

for

**United States Department of Energy
Federal Energy Technology Center
DOE Cooperative Agreement DE-FC21-91MC28055
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Abstract

During the Westinghouse/U.S. DOE Cooperative Agreement period of November 1, 1990 through November 30, 1997, the Westinghouse solid oxide fuel cell has evolved from a 16 mm diameter, 50 cm length cell with a peak power of 1.27 watts/cm to the 22 mm diameter, 150 cm length dimensions of today's commercial prototype cell with a peak power of 1.40 watts/cm. Accompanying the increase in size and power density was the elimination of an expensive EVD step in the manufacturing process. Demonstrated performance of Westinghouse's tubular SOFC includes a lifetime cell test which ran for a period in excess of 69,000 hours, and a fully integrated 25 kWe-class system field test which operated for over 13,000 hours at 90% availability with less than 2% performance degradation over the entire period. Concluding the agreement period, a 100 kW SOFC system successfully passed its factory acceptance test in October 1997 and was delivered in November to its demonstration site in Westervoort, The Netherlands.

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1. Executive Summary

The state-of-the-art solid oxide fuel cell called the air electrode supported solid oxide fuel cell (AES SOFC), developed by Westinghouse, has shown substantially improved electrical performance over the previous zirconia porous support tube (PST) design with the ability to thermally cycle from 1000°C to room temperature and back more than 100 times without any performance loss. This new cell design has also shown enhanced and stable performance at elevated pressures up to 15 atmospheres which will facilitate integration with gas turbine systems. The AES-SOFC has been manufactured and tested in lengths of 50, 100, and 150 cm active length. Westinghouse's AES cells have also shown the ability to operate with fuel utilization as high as 88% and with air flow as low as 3.5 times stoichiometric without decreases in performance.

In the early 1990's, the shift was made from a SOFC built around a porous zirconia support tube to one supported by the air electrode itself. This shift resulted in a power density increase of close to 50% over the thin PST and a simpler manufacturing process. During the development of this AES-SOFC, development of the doped lanthanum chromite interconnection and nickel cermet fuel electrode have allowed these elements to be manufactured through non-EVD processes. The interconnection was the first cell element removed from the EVD process resulting in a two EVD step cell. The second cell element removed from EVD processing was the fuel electrode. This removal of two EVD steps from the AES-SOFC manufacturing process resulted in needed cost reductions without the compromise of life or reliability.

Along with the increase in power density, the cell length was scaled up from 30 to 150 cm in active length resulting in increased net power per cell. AES-SOFC's of 150 cm active length have demonstrated power levels of 210 watts per cell at atmospheric pressure compared to 20 watts for the original 30 cm thick-PST or 63 watts for a 50 cm AES cell. This 330% increase in power of the 150 cm active length AES-cell over the 50 cm AES-cell results in fewer cells to manufacture for a given capacity and major cost reductions per kilowatt of electricity.

Recent performance of Westinghouse's tubular SOFCs and other technical achievements have been extraordinary:

- Demonstrated SOFC lifetime — in excess of 69,000 hours
- Excellent voltage stability — degradation approximately 0.1% per thousand hours at 13,000 hours for AES-SOFCs
- Thermal cycle toughness — over 100 hot/cold cycles without deleterious effect for AES-SOFCs
- System integration — eleven fully integrated systems field tested by customers with the last 25 kWe-class unit having operated for over 13,000 hours at 90% availability.
- Successful completion of the 100 kWe SOFC Factory Acceptance Test — demonstrating that the EDB/ELSAM Power Generation System could be started, operated, stopped, and cooled in a controlled manner.
- Successful pressurized AES-SOFC operations for over 4800 hours without deleterious effect.

In addition, conceptual designs for dry (no steam or rankin cycle) hybrid cycles that employ a pressurized SOFC (PSOFC) coupled with a gas turbine (GT) indicate that electrical generation efficiencies in excess of 70% should be achievable for systems 3 to 5 MW in capacity. The cycle showing this extraordinary efficiency employs a two shaft intercooled, recuperated, and reheated GT in which both high pressure and low pressure GT combustors are supplanted by PSOFC generator modules. This high level of efficiency (70%) is the highest postulated to date for natural gas fueled power systems. Hybrid cycles employing recuperated single shaft GT's should be able to achieve an efficiency greater than 60% at capacity levels as low as one MW. PSOFC/GT hybrid cycles will exhibit the highest electrical generation efficiency across the capacity range from a few hundred kW to several hundred MW.

These accomplishments have spawned increasing development activity worldwide and affirmed Westinghouse as the recognized world leader in SOFC technology.

2. Introduction

This report is a summary of the accomplishments made by Westinghouse under Cooperative Agreement No. DE-FC21-91MC28055 between Westinghouse and the U.S. Department of Energy — Federal Energy Technology Center (DOE-FETC) for the development of tubular SOFC technology. This seven-year program, titled "High-Temperature Tubular Solid Oxide Fuel Cell Generator Development," commenced on November 1, 1990, and ended on November 30, 1997.

The cooperative program yielded the successful factory acceptance test (FAT) of a commercially prototypic power system of nominally 100 kWe electrical generating capacity, and the completion of a pilot manufacturing facility for the non-commercial production of SOFC power systems.

2.1 BENEFITS

Fuel cells are electrochemical devices that directly convert the chemical energy contained in fuels such as hydrogen, natural gas, and coal gas into electricity at high efficiency with no intermediate heat engine or dynamo and without conventional combustion and concomitant emission of pollutants such as NO_x . The solid oxide fuel cell (SOFC) is distinguished from other fuel cell types by its all solid-state ceramic structure and its high operating temperature, nominally 1000°C . By virtue of its high operating temperature, the natural-gas-fueled SOFC, when operated at elevated pressure and integrated with a conventional gas turbine/generator, can achieve a system electrical generation efficiency in excess of 70%. In this configuration, the SOFC is an electrochemical topping cycle that supplants the gas turbine's combustor, with the gas turbine/generator functioning as a bottoming cycle. In addition, when configured for the cogeneration of process steam and/or hot water, overall system fuel effectiveness can exceed 85%. The SOFC thus has the potential to generate electricity from hydrocarbon fuels (as well as from hydrogen) at efficiencies higher than any known alternative power generation system, thereby significantly reducing the consumption of fuel and the production of CO_2 — the greenhouse gas, with negligible (less than 2 ppmv) production of air pollutants such as NO_x that cause acid rain.

2.2 TECHNICAL BACKGROUND

Because of its high operating temperature, the SOFC relies upon ceramics for its structure. The development of suitable materials and processes for the manufacture of an SOFC of practical geometry at commercially acceptable costs is a formidable challenge that has occupied researchers world-wide for the past several decades. Pioneering work to address this challenge was initiated at Westinghouse over three decades ago with corporate strategic funds. Over the years, SOFC development at Westinghouse has continued with support from Westinghouse and the U.S. Department of Energy and its predecessor agencies. Westinghouse pioneered the tubular geometry for SOFCs and has developed this technology to the point where it is universally acknowledged as the most advanced SOFC in the world.

Figure 2.3 — Cross Section of the 100 kWe Class SOFC Stack with 1152 Cells Showing Gas Flows.

Natural gas fuel enters the stack at the top through a fuel nozzle/ejector assembly. The ejector extracts a portion of the moisture rich depleted fuel gas exiting the electrochemically active region of the stack and mixes it with the incoming fresh fuel in order to humidify the fuel prior to the reformation process. The fuel mixture (650 to 700°C) enters a small pre-reformer which converts the higher hydrocarbons in the natural gas to CH_4 and H_2 and CO . The partially reformed fuel mixture then flows through fuel manifolds positioned underneath the stack between bundle rows and directly beneath each stack reformer. This fuel mixture is directed into the stack reformers where the CH_4 is essentially 100% reformed. The stack reformer is maintained at a sufficient temperature for the reformation reaction (700 to 800°C) by direct radiation from the surrounding cells.

The stack reformers are designed such that the reformation rate is uniformly distributed from top to bottom resulting in uniform heat extraction along the cell length. This design fosters a near uniform cell temperature distribution and provides a very compact footprint for the stack. The reformed fuel exits the stack reformers near the bottom of the stack and is distributed to the closed ends of the cells. The reformed fuel flows upward in the fuel channels between cells within the stack. Typically, the equivalent of 80 to 90% of the incoming natural gas is electrochemically utilized prior to exiting the active portion of the stack. A portion of the depleted fuel gas is recirculated and the remainder enters the combustion zone above the stack where it reacts with the depleted air exiting the cells. This combusted gas mixture forms the exhaust gas that preheats the incoming air inside the air injector tubes (which hang vertically, passing through the combustion zone) and is then discharged from the stack at a temperature between 800 and 850°C. The Westinghouse tubular SOFC stack design is distinguished from others by the complete absence of high integrity seals between cell elements or between cells and manifolds. Further, the reformer and the cell stack are thermally and hydraulically integrated.

3. Results and Discussion: Task Reports

3.1 TASK 1.0 — CELL COST REDUCTION

Cell Design/Materials

The SOFC exploits the phenomenon that stabilized cubic-phase zirconia is an excellent conductor of oxygen ions while remaining impermeable to the molecular flow of gasses at elevated temperatures near 1000°C (1832°F). In concept, the SOFC is simple. To generate a voltage, a thin film zirconia electrolyte only needs to be sandwiched between gas permeable, electron conducting electrodes and exposed to a very low concentration of oxygen at the anode or fuel electrode, and a relatively high concentration of oxygen at the cathode. Electricity will be driven through the external circuit connecting the electrodes in direct proportion to the flow of oxygen ions through the electrolyte, as long as a continuous flow of fuel is supplied to consume oxygen at the fuel electrode, and a continuous flow of air is supplied at the cathode to provide the oxygen.

This simplicity in concept is deceiving, however, because the compatibility requirements for the materials of construction and the technology required for the fabrication of a viable cell represent formidable challenges.

Electrolyte: Zirconia doped with about 10 mole percent yttria is used as the electrolyte. The high ionic conductivity of yttria-stabilized zirconia is attributed to oxygen ion vacancies along with low activation energy of oxygen ion migration. The thermal expansion of 10 mole percent yttria-stabilized zirconia is about $10 \times 10^{-6}/^{\circ}\text{K}$; materials for all other cell components are chosen to have thermal expansion near this value.

Air Electrode: For the air electrode (cathode), calcium or strontium-doped lanthanum manganite is generally used. The material can have oxygen excess or deficiency depending upon the cathode gas oxygen partial pressure and temperature.

Fuel Electrode: For the fuel electrode (anode), due to the reducing atmosphere of the fuel gas, a metal such as nickel can be used. However, the thermal expansion coefficient of nickel is considerably larger than that of the yttria-stabilized zirconia electrolyte; this large thermal expansion mismatch can cause delamination of the anode from the electrolyte surface. Also, nickel can sinter at the cell operating temperature resulting in a decrease in anode porosity. To circumvent these problems, a skeleton of yttria-stabilized zirconia is formed around the nickel particles. This skeleton of yttria-stabilized zirconia supports the nickel particles, inhibits sintering of the nickel particles during cell operation, provides adherence to the electrolyte, and also provides an anode thermal expansion coefficient closer to that of the electrolyte. Such nickel/yttria-stabilized zirconia anodes show very low diffusion and activation polarization losses during cell operation. Such anodes have also been shown to possess sufficient catalytic activity at 1000°C to reform natural gas and other hydrocarbons in situ.

Interconnection: For the interconnection, doped lanthanum chromite is used. The defect chemistry, oxidation-reduction behavior, and the thermal expansion behavior of these chromites have been extensively studied.

3.1.1 Accomplishments

Cell Design and Materials. The calcia-stabilized zirconia support tube (PST) has now been completely eliminated and replaced by a doped lanthanum manganite tube in state-of-the-art SOFCs. This doped lanthanum manganite tube is extruded and sintered to about 30 to 35 percent porosity, and serves as the air electrode onto which the other cell components are fabricated in thin layer form. The latest technology tubular cells are designated as WPC3 air electrode supported (AES) cells; the schematic design of such a cell is shown in Figure 3.1. A series of Westinghouse proprietary compounds, designated WPC-n, were developed. Early compounds had excessive shrinkage which was eliminated with the WPC3 compound finally adopted for use.

In addition to eliminating the calcia-stabilized zirconia support tube, the active length of the cells has also been continually increased to increase the power output per cell. A greater cell power output decreases the number of cells required in a given power size generator and thus improves SOFC power plant economics. The active length has been increased from 30 cm for pre-1986 thick-wall PST cells to 150 cm for today's commercial prototype AES cells. Additionally, the diameter of the tube in longer length AES cells has been increased from 15.6 mm to 22 mm to accommodate larger pressure drops encountered in longer length cells. Figure 3.2 shows the power output from different cells illustrating the many-fold increase in power output from the latest technology AES cells.

Figure 3.1 — Air Electrode Supported (AES) Type Tubular Solid Oxide Fuel Cell Design.

Figure 3.2 — Power Output of Different Type and Length Cells.
 $T = 1000^{\circ}\text{C}$, Fuel = 89% H_2 & 11% H_2O , Oxidant = air, Oxidant utilization = 16% typically

Table 3.1 shows the cell design evolution.

Table 3.1
Cell Design Evolution

Year	1983-87	1987-89	1990	1991	1993	1994	1995-97	1997+
Cell Type	Thick PST	Thin PST	AES	AES	AES	AES	AES	AES
Cell I.D. (mm)	8.9	10.4	11.8	11.8	11.8	11.8	17.6	17.6
Cell O.D. (mm)	15.2	15.9	15.9	15.9	15.9	15.9	22.3	22.3
PST Thk (mm)	2.0	1.2	0.0	0.0	0.0	0.0	0.0	0.0
AE Thk (mm)	1.0	1.4	1.9	1.9	1.9	1.9	2.2	2.2
Cell Length (cm)	30, 36	50	50	50	50	100	150	150
Peak Power (W/cm)	0.67	0.95	1.27	1.27	1.27	1.27	1.40	1.40
EVD Steps	3	3	3	3	2	2	2	1

Interconnection Fabrication. The fabrication of PST cells involved three electrochemical vapor deposition (EVD) steps, one each for the doped LaCrO_3 interconnection, the yttria-stabilized zirconia (YSZ) electrolyte, and the Ni-YSZ fuel electrode. Though EVD provides very high quality thin films, it requires capital intensive equipment making the process rather expensive. Investigations on alternate processing techniques have been underway at Westinghouse for several years to replace one or more EVD steps with a more cost-effective approach. The interconnection is now deposited by plasma spraying calcium aluminate containing lanthanum chromite powder over the porous, doped lanthanum manganite air electrode tube. Calcium aluminate facilitates densification during plasma spraying and subsequent heat treatment. The plasma sprayed interconnections are a single phase, perovskite structure. These interconnec-

tions have a thermal expansion coefficient much better matched to that of the electrolyte than the previously-used EVD Mg-doped lanthanum chromite interconnection. Plasma spraying of interconnections has now been implemented in the manufacturing of all AES SOFCs. This has resulted in reduced process cycle time, increased yield, and a major reduction in cell fabrication cost. Plasma spraying will allow continuous, fully automated deposition of interconnection films in commercial SOFC production. The materials and fabrication processes for the state-of-the-art AES cells are summarized in Table 3.2.

Table 3.2
Materials and Fabrication Processes for AES Cells (1996).

Component	Material	Thickness	Fabrication Process
Air Electrode Tube	Doped LaMnO_3	2.2 mm	Extrusion-sintering
Electrolyte	$\text{ZrO}_2(\text{Y}_2\text{O}_3)$	40 μm	Electrochemical vapor deposition
Interconnection	Doped LaCrO_3	85 μm	Plasma spraying

Air Electrode Fabrication. Over 90% of the weight of an AES cell is that of the doped lanthanum manganite air electrode tube. Presently, the air electrode material is synthesized using high purity component oxides such as La_2O_3 and MnO_2 . Over 70% reduction in the cost of air electrode raw materials is possible if mixed lanthanides are used instead of pure lanthanum compounds to synthesize the air electrode powder. These mixed lanthanides contain Nd, Pr, Ce and Sm in addition to La. AES cells have now been fabricated using air electrode powder synthesized using mixed lanthanides. Figure 3.3 compares the performance of a cell fabricated using mixed lanthanides for the air electrode to that of a cell fabricated using pure lanthanum oxide instead. The performance of the cell with air electrode fabricated using mixed lanthanides at 400 mA/cm^2 is only about 8% lower, primarily due to slightly higher resistivity of the mixed lanthanides air electrode. Further adjustments in the composition of the air electrode material synthesized using mixed lanthanides are expected to result in lower air electrode resis-

Figure 3.3 — Effect of Air Electrode Material Purity on the Voltage-Current Characteristics of AES Cells at 1000°C.

tivity and a cell performance equivalent to that of cells using high purity air electrode material synthesized using pure lanthanum oxide. The substitution of mixed lanthanides for lanthanum in air electrodes results in a major reduction in the cost of materials used in the manufacture of tubular SOFCs.

Elimination of one of the cell components (calcia-stabilized zirconia support tube) and replacement of one EVD step by plasma spraying (for depositing interconnection) has resulted in a major reduction in the cost of the AES cells and the cost of electricity (\$/kW) produced by using such cells.

Fuel Electrode Fabrication. Investigations are underway to further reduce the cost of cells by replacing another EVD step (for depositing fuel electrode) by a more cost-effective sintering approach. Investigations to deposit Ni-YSZ fuel electrode by a non-EVD process have also shown great promise. Deposition of a Ni-YSZ slurry over the YSZ electrolyte followed by sintering has yielded fuel electrodes that are equivalent in electrical conductivity to those fabricated by the EVD process.

Through the reduction of EVD steps, major manufacturing cost reductions can be realized. Fortunately, the electrical performance of cells processed with one EVD step have shown little performance loss over cells processed with two EVD steps. Figure 3.4 shows the voltage vs. time and Figure 3.5 shows the performance (voltage-current characteristic) of a cell processed with one EVD step versus a cell processed with two EVD steps.

Figure 3.4— A Comparison of the Voltage-Current Characteristics of the One-EVD Step and the Two-EVD Step Cells at 1000°C.

Figure 3.5 — A Comparison of the Voltage-Current Characteristics of the One-EVD Step and the Two-EVD Step Cells at 1000°C.

Both cells were operated on humidified hydrogen fuel at 85% fuel utilization. Even at this high fuel utilization, the performance of the non-EVD fuel electrode is comparable to that of the EVD fuel electrode. In fact, the fuel side polarization with a non-EVD fuel electrode is actually better than the EVD fuel electrode. This has been proven through electrical test as well as through microcharacterization (see Section 3.2.2). Upon review of microstructure, the non-EVD fuel electrode shows a greater contact percentage of nickel to zirconia than the EVD fuel electrode. This increased number of active sites allows for lower anode polarization.

To electrically test the fuel side polarization, diagnostic tests are performed where the fuel is diluted with nitrogen in a 1:1 and 2:1 ratio. Table 3.3 shows the impact on performance of these dilution tests.

Table 3.3
Comparison of Fuel Side Polarization Between EVD and Non-EVD Fuel Electrodes

Cell Type	Average EVD Fuel Electrode Cell	Non-EVD Fuel Electrode- Cell 1	Non-EVD Fuel Electrode- Cell 2
1:1 Dilution	7mV	1mV	4mV
2:1 Dilution	15mV	4mV	10mV

From Table 3.3 it is clear that the increased number of active reaction sites result in a 50% reduction in fuel side polarization of the non-EVD fuel electrode over the EVD fuel electrode.

In the very near future, the AES cell production process will use EVD for only the electrolyte. The EVD process deposits very thin (20 to 40 μm thick), gas-tight electrolyte film over the porous air electrode, reliably, uniformly, and in acceptable cycle time. Nonetheless, deposition of the YSZ electrolyte film by a non-EVD technique such as colloidal/electrophoretic deposition of