GLOBAL THERMOELECTRIC'S INTEGRATED CELL MANUFACTURING OF PLANAR SOFCs

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ABSTRACT

Global Thermoelectric Inc. has been actively engaged in integrated solid oxide fuel cell (SOFC) manufacturing technology research and development for the past five years. Global’s current cell manufacturing process has three major operation units for fabrication of its anode supported planar cells. Anode substrates approximately 1 mm in thickness are produced by a single step tape casting process. The anode, electrolyte and cathode functional layers are screen-printed directly on the green anode substrate tape. The multi-layer green cells are sintered into complete cells through a continuous co-firing process. The tape casting, screen-printing and co-firing (TSC) processes evolved from the laboratory research stage to the pilot plant production stage over three years. Global is able to produce cells at a volume equivalent to 5 MW/year with greater than 90% yield. Global attributes this progress to the combination of advanced materials technologies, engineering principles and adherence to manufacturing discipline. Cells made by this integrated manufacturing process are capable of thermal and load cycling. Global’s cell can produce over 1.4 W/cm² at 750°C. A long-term degradation rate approximately 1% per 1000 hours at a current density of 0.55 A/cm² was demonstrated over 6000 hours of testing. Global’s cell performance can be attributed to the optimized microstructures developed by the integrated cell manufacturing process. In this paper, the process technology, microstructure and characteristics of Global’s planar SOFCs will be presented in detail.

INTRODUCTION

Global Thermoelectric, Inc. is a leading developer of planar SOFC systems. Since 1997, Global has developed leading technology in thin film, high power density anode supported SOFC technology (1, 2). Global presently has approximately 160 people working on SOFC commercialization. Global’s (80,000 ft²) SOFC development facilities include offices, research laboratories, and a state-of-the-art pilot plant.

Global’s unique anode supported, thin film cell design provides very high power density. At an operating voltage of 0.7 V and temperature of 750°C, a power density of 1.4W/cm² was achieved. The lower operating temperature allows for improved integration with
fuel processing functions and the use of non-exotic standard materials in stack assembly, thereby facilitating a lower system cost.

Global has also developed a proprietary SOFC stack (as shown in Figure 1a) design utilizing metallic interconnect plates and a patent pending compressive sealing system to provide reliable stack performance. This stack design, based on 10 cm x 10 cm cells, has demonstrated power outputs of up to 1600 Watts for a 20-cell stack (over 1 W/cm²) at 750°C on hydrogen fuel. Global has integrated its stacks into a number of natural gas fuelled prototype systems. Over 2000 hours of continuous operating has been achieved in the current RP2 system (Figure 1b). Global has positioned itself to reach SOFC product commercialization by 2005.

Figure 1a and 1b. Global's stack and RP2 system.

Along Global's commercialization path, significant cost and performance improvements are required to transform today's SOFC technology into one suitable for low cost, mass production of small systems for multi-market applications. Global has identified the key technical issues that must be resolved to achieve the low cost commercial SOFC system of the future and is executing a carefully structured technical and engineering approach that will lead to the resolution of these issues. One of the successful examples is the development of an integrated cell manufacturing technology capable of producing high performance planar SOFCs.

GLOBAL'S INTEGRATED CELL MANUFACTURING PROCESS

Global Thermoelectric Inc. has been actively engaged in developing an integrated cell manufacturing technology for the past three years (3). A strong cell process development team had been developed with a mandate of reducing cost and improving volume and yield. Each dimension has stretch targets with aggressive schedules.

Three generations of cell manufacturing processes have been successfully developed; S31E, TSCI, and TSCII. In September 1999, a set of well-established ceramic processing technologies was selected as Global's new cell manufacturing technology; tape casting, screen-printing, and co-firing (TSC). In November 2000, a 3,000 m² (32,000 ft²) pilot plant facility was developed and all major production equipment was installed and commissioned. The major equipment included a continuous tape caster, screen printers with continuous dryers and a continuous pusher plate tunnel kiln. In June 2001, the
The TSCI process replaced the S31E process as Global's platform cell manufacturing process. Two major advancements in the TSCI process are screen-printing directly onto unfired (green) anode tape and co-firing the thin electrolyte layer/anode assembly. Since production equipment is involved, there is a significant reduction in labor, which shortens the production pipeline from 30 days to 8 days. As a result, 50% cost reduction was achieved. Furthermore, a production yield of 80% was obtained using tighter specifications. In late 2001, a significant number of materials research breakthroughs and process improvements further streamlined the process by elimination of the separate cathode-firing step. The second generation of the TSC process, TSCII, replaced TSCI as Global’s current cell manufacturing process in April 2002.

The TSCII process is a fully integrated cell manufacturing process. All major processing steps have been demonstrated as cost effective manufacturing processes for the semiconductor packaging and multilayer capacitor industries. The TSC process can be further engineered into repeatable mass manufacturing modules to meet the technical and cost targets. Furthermore, the flexibility of this process has been demonstrated by its ability to scale to larger cell sizes (up to 25 cm x 25 cm) and alternate geometries (round, hexagonal, internal manifold, etc.)

As shown in Figure 2, cell cost per kW (direct labour and materials) has been significantly reduced by 65%. In the meantime, cell production volume capability increased from 100 cells/week to 2500 cells/week. Furthermore, the production pipeline reduced from 30 days to 4 days and production yield improved from 50% to 90%.

![Figure 2. Cost, volume, and yield achieved by the S31E process (1999), TSCI process (2001), and TSCII process (2002).](image)

Cell process development at Global follows a structured stage-gate process. Advanced materials technology has been transferred into cell products through four stages: research, development, engineering, and pilot production.

At the research stage, advanced materials and process technologies are developed based on lab-scale experiments. At the development stage, advanced cell process technologies are evaluated at pilot plant scale. A process integration campaign is conducted for validation. At the engineering stage, all production procedures are completed. A full-
scale process standardization campaign is performed to reveal the capability of the process, equipment and operating staff. A formal engineering report is generated to benchmark the process. Finally, at the production stage, manufacturing disciplines are applied to ensure the stability of the process as well as the reliability of the cells produced.

As shown in the process diagram (Figure 3), Global’s platform cell manufacturing process has three major unit operations: tape-casting, screen printing and co-firing.

![Tape Casting](image)

**Figure 3.** TSCII cell manufacturing process diagram.

The tape casting operation unit includes the quality assurance of all anode raw materials, slurry preparation, tape casting, controlled drying, and tape cutting. Global’s continuous tape caster is 20 m in length. A single step casting yields anode substrates of approximately 1 mm in thickness and, if required, can be adjusted to yield thinner substrates. Tape casting yields consistent and high quality tapes with thickness variations less than 2% (< 20 μm).

The screen-printing operation unit includes paste preparation, subsequent layer screen-printing, continuous drying and unfired (green) cell punching. Anode, electrolyte and cathode functional layers are printed directly on the green anode substrate tape and dried in belt driers 6 m in length. All operations are conducted in a clean-room environment. One-step co-firing of all cell components is one of the main features of the TSCII
process. The combined binder burnout, high temperature co-sintering and cooling cycle in a 20 m continuous pusher plate tunnel kiln are less than 24 hours.

Sintered cells undergo an intensive quality assurance program that measures physical, mechanical, microstructural, and electrochemical properties of the finished products. At the current cell volume, quality assurance not only provides reliable cells for stack manufacturing, but also yields a great amount of data for studying the process capability and for assisting future process development. Data is the critical link among the cell’s manufacturing process, microstructure, mechanical and electrochemical properties. In order to form a closed loop at the high volume process engineering and production stages, Global has developed an effective bar code tracking system to record process related data in parallel with cell process advancement. This bar code tracking system tracks the raw material used, equipment applied, and operator involved at each unit operation. Custom screens were designed, which allows for data entry directly into a SQL database. These custom screens improve data integrity by confirmation, validation and policing of data entry.

A web browser was designed to navigate the database. The web application is an excellent tool for searching information regarding a particular cell and provides its processing history with its microstructure and properties. A query page can be used to access the database and output data for further statistical analysis. Formal reports, such as a WIP (working in progress) report, have been built to report real time operating information.

**THE CHARACTERISTICS OF GLOBAL’S PLANAR CELL**

Development of a fully integrated cell manufacturing technology not only reduces manufacturing cost, but also offers better cell microstructure and consequently the performance of the Global cell has been greatly improved over the past three years.

Due to continuous cell technology and process improvements, a well-engineered cell microstructure has been developed. Figure 4 is an SEM image of a Global cell after reduction.

![Figure 4. Cross section SEM photomicrograph of a TSCII cell (after reduction).](image-url)
The microstructure improvement enabled the Global cell to operate at a high power density at reduced operating temperatures. Figure 5 illustrates the power density improvement from 1999 to 2002. A power density of over 1.4 W/cm² was achieved at 0.7 V and 750°C. In order to simulate the stack operating condition better, these tests were performed in a single unit short stack configuration as following:

- Common stainless steel test jigs with cross flow fuel and air delivery.
- Standard production cells were used (10 x 10 cm for 2002 test, and 5x5 cm for the previous tests with an active area of 81 and 16 cm² respectively).
- The same class of materials and components such as compression seals were used in the assembly.
- H₂ with 3% H₂O was used as fuel. Air was used as the oxidant.

![Global Thermoelectric Inc. - Single Cell Performance Curves](image)

**Figure 5. Cell electrochemical performance improvement.**

Although Global’s cell consists of the same class of materials, the significant cell microstructure improvement has been achieved through incorporating both materials technology advancement and manufacturing process optimization. In order to achieve co-firing in Global’s current platform cell manufacturing technology, materials research and process development efforts have been focused at following areas:

- Slurry and paste formulation were optimized for both substrates and thin functional layers.
- Interactions among all component layers were tailored at green stages.
- Component sintering kinetics was studied in detail to provide the optimized co-firing profile.
- Electronic and ionic conducting paths, as well as porosity of each component are well engineered.

The optimization of the cell microstructure has enabled higher power densities and reduction in area specific resistance. Interfacial integrity and stability were greatly
enhanced through component interactions at green stage and during co-firing. This is reflected in the mechanical and electrochemical properties of a cell. Improvement in interfacial strength has been verified by standard thin film adhesion tests.

The excellent interfacial integrity also provides a Global cell with thermal cycling and load cycling capabilities. Figure 6 demonstrates a cell that went through 20 thermal cycles between 750°C and room temperature. The total degradation of approximately 4% was achieved after 20 thermal cycles.

Figure 6. Thermal cycling of a planar Global SOFC.

Figure 7 shows a cell that went through rapid electrical load cycling from open circuit to 0.49 A/cm² and back to open circuit, with a frequency of 1 minute. After 100 hrs and approximately 6000 cycles, both the average cell open circuit voltage and operating voltage at 0.49 A/cm² improved slightly.

Global’s cell microstructure provides long term stability at high current density. Figure 8 shows a long-term testing of a single unit short stack. The overall degradation rate of approximately 1% per 1000 hours is achieved at a constant current density of 0.555 A/cm² and 50% fuel utilization.

CONCLUSIONS

Global has developed an integrated planar SOFC manufacturing technology. Well-engineered cell microstructure has been achieved by applying Global’s current platform process, TSCIL. As a result, a number of improved cell characteristics, such as higher power density, reduced steady-state degradation, capability of thermal cycling, and capability of electric load cycling, have been demonstrated.
Load Cycling Test; Glob 10673; 0-40-0 Amp Ramp Load (1 minute intervals)
Voltage Response 50% Fuel Utilization, 25% Air Utilization

Figure 7. Electrical loading cycling of a planar Global SOFC.

Voltage Trend Data
Glob 10645; January 9 - October 17, 2002

Figure 8. Long-term degradation test of a planar Global SOFC.
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