PROGRESS IN STACK POWER DENSITY USING THE SOFCONNEX™ CONCEPT

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ABSTRACT

Our SOFC stack development technology is based on the unique SOFConnex™ concept, using flexible gas distribution layers between metal sheet interconnect and thin anode supported electrolyte (ASE) cells. Flexibility is given both in material and design. This ensures proper electrical contact over the whole cell (50 cm² active), without necessitating restrictive cell fabrication tolerances, and allows easy adaptation and evolution of the flow fields. With the presently used configuration, several multiple cell stacks were assembled and tested. Reproducible stack power density (H₂ fuel, λ = 1.5-2, 800°C maximum local temperature) is 0.5 W/cm² at 0.7 V average cell voltage (1.5 kW/L), for 67% fuel utilisation (35% LHV electrical efficiency). Performance with simulated POX-syngas (H₂/CO/N₂) was close to that with H₂. Degradation is the focus of attention now that adequate power density and efficiency using the SOFConnex™ approach have been established and reproduced.

INTRODUCTION

Our approach in SOFC stacking technology is distinct in several aspects: anode supported electrolyte (ASE) cells are thin (<0.25 mm) and tough and made by aqueous cocasting and cofiring; metallic interconnect (MIC) sheets are thin and unprofiled; cathodes are LaSrFeO₃ based; current collection and gas distribution is ensured via the SOFConnex™ interlayer between cells and interconnects. The interlayer can absorb cell imperfections, facilitate gas feeding, reduce Cr poisoning and shows a current collection density superior to other approaches. Moreover, the design is highly flexible: improved configurations are easily developed and tested, manifolding can be internal, external or combined, and sealing is simplified. In combination with the thin components, SOFConnex™ allows compact assembly of stacks with low cost potential and adequate power density, as illustrated in this paper with some recent results.

EXPERIMENTAL

The fabrication of cells and stacks was reported previously (1). HTceramix has now
initiated its production in Yverdon (Switzerland) with a capacity of 200 cells/week. The present configuration uses 8 x 8 cm cells with 50 cm² active area and internal feed holes. Interconnect plates are untreated 0.75 mm thick 1.4760 steel. A stack repeating unit stands 2.3 mm high, while fuel and air pressure drops remain appropriate (typically 4 and 8 mbar). Stacks are spring-compressed between Inconel steel flanges (0.4 kg/cm²), equipped with air preheater and probes for inlet-temperature and pressures of fuel and air. Stack current is taken on the extreme interconnects, adding 5 mΩ per plate (50 mm²) to the circuit. Assembly and installation are fast; an assembled stack is placed as one piece into a top-laden electrical chamber furnace. Heat-up is typically 180 K/h but can be much faster; individual cells resist thermal cycling (RT-800°C-RT) within less than a minute. Test temperatures are typically 700-800°C: of several thermocouples mounted with each stack, the highest recorded value (a thermocouple in contact with a center MIC plate) is taken for i-V registration. Furnace temperature was typically 50 K lower than this value, fuel and air feeds are H₂ flows (humidified, 3%) between 5 and 9 ml/min.cm² (corresponding to 0.72 and 1.29 A/cm² for 100% fuel conversion) and air-λ 1.5 to 2. This paper reports on two identical stacks of 4 and 5 cells, respectively, subjected to similar conditions.

RESULTS

The performance of the 4-cell stack upon start-up (1.5 h after H₂ introduction at 800°C, 5 ml/min.cm²) is given in Fig. 1. (All following graphs consistently show average cell voltage on the Y-axis.) Keeping the stack steadily polarised at 15 A (0.3 A/cm²) for the next days, performance progressed significantly, also shown in the figure (for t = 120 h). Power density reached 0.35 W/cm² at 0.73 V, electrical efficiency was 40% (LHV).

Characteristics were then taken at 750°C and 700°C, and flows increased. Comparative performance at 700, 750 and 800°C is given in Fig. 2, for 8 ml/min.cm² H₂ flow, after 180-190 h of test. At 0.7 V, power density reaches respectively 0.3 W/cm², 0.42 W/cm² and 0.53 W/cm² for the three temperatures. For the latter case (800°C), electrical efficiency reached 37%, and power density is still up substantially from the earlier characteristic (0.4 W/cm², t = 120 h, 5 ml/min.cm² H₂).

The effect of flow is small; the increase in power with higher flow cannot compensate the loss in efficiency. This is exemplified in Fig. 3, where the i-V characteristic at 700°C is given for 4 different flows (H₂ = 5, 6, 7, 8 ml/min.cm², λ_air = 1.6). Performance at 0.75 V for example, is raised from 0.23 to 0.27 W/cm², while efficiency drops from 25% to 19%. It is also worthwhile noticing that performance at 700°C, under constant polarization of 15 A (0.3 A/cm²) at 0.75 V, actually decreased (see curves at t=144 h and t=163 h in Fig. 3), in contrast to the strong activation observed at 800°C for the same current, at 0.77-0.84 V. This could be related to the Ni anode reoxidation potential, 0.7 V at 800°C and 0.75 V at 700°C (2), vs air. From this perspective, it appears more favorable to operate Ni-based ASE cells at the higher of the two temperatures, and 750°C may be a good compromise for both power density and long term stability.
Figure 1. i-V-P output of 4-cell stack (50 cm\(^2\) active area per cell), 800°C, at start-up and after 120 h polarisation at 15 A.

Figure 2. i-V-P output of 4-cell stack (50 cm\(^2\) active area per cell), 700-800°C, at 8 ml/mm.cm\(^2\) H\(_2\) (\(\lambda_{\text{air}} = 1.6\)).
After these first 200 h of operation, the stack was kept at 22 A (0.44 A/cm², 40% fuel conversion, 800°C) for several 100 h, Fig. 4. Voltage decay was uniform on all cells and very important (-3%/100 h), though decelerating after 500 h. Postmortem SEM on cells indicated second phase formation at the cathode-electrolyte interface, which is further investigated. It is noted in the figure that the colder extreme cells (1 and 4) were still activating at 200 h, whereas the hotter center cells already degraded.

A full thermal cycle occurred after 550 h of operation. The stack was reheated for a renewed series of i-V characteristics at different temperature and flow conditions, part of which is reported in Fig. 5. Comparing the performance at 800°C just before the cooldown (546 h) with that after the reheat to the same temperature (but with lower fuel flow of 6 ml instead of 8 ml/min.cm²), it appears that the stack coped well with the thermal cycle: the curves are shifted by around 25 mV which could be accounted for solely by the difference in fuel feed, as observed higher (Fig. 3).

With the aim of simulating behaviour in reformate fuel originating from partial oxidation with air, the anode inlet was now changed, first by dilution with N₂ (50%), then by adding CO (H₂:CO 2:1 and H₂:CO 1:1), while keeping overall fuel flow constant at 6 ml/min.cm². The relevant i-Vs are coplotted in Fig. 5. Anode pressure drop increased from 5 mbar (H₂ only) to 18 mbar (H₂/N₂/CO). It can be seen that performance hardly drops: -12 mV after N₂ dilution, and a further 1-4 mV when adding CO, a larger drop in part being compensated by a higher OCV (0.973 V with only H₂, 0.988 V with second CO addition). However, the drop in efficiency is more significant, as the CO heating value is higher (by 17%) than that of H₂ without a gain in electrical power. Hence, for the

![Figure 3. i-V-P output of 4-cell stack (50 cm² active area per cell), 700°C, at variable fuel flow of 5 to 8 ml/min.cm² H₂ (αair = 1.6).](image-url)
Figure 4. Voltage degradation on each of the cells (4-cell stack) during galvanostatic polarization at 22 A (50 cm$^2$), 800°C, 8 ml/min.cm$^2$ H$_2$ flow, for $t = 200$ to 550 h.

Figure 5. i-V-P output of 4-cell stack before and after a full thermal cycle, then at variable fuel conditions of adding N$_2$ and CO to H$_2$ (total fuel = 6 ml/min.cm$^2$) ($\lambda_{air} = 1.6$).
power density of 0.32 W/cm\(^2\) (0.7 V), efficiency was 29.6% for H\(_2\) fuel, 28.1% for H\(_2\):CO 2:1 and 27.1% for H\(_2\):CO 1:1.

The stack was kept for another week (160 h) at 800°C under the final discussed conditions (N\(_2\):H\(_2\):CO 2:1:1), 0.33 A/cm\(^2\) (40% fuel conversion), during which voltage degradation from ca. 0.77 V to 0.73 V was similar as before (like Fig. 4), again with decelerating tendency. The test was terminated after 820 h for inspection. Cells were demounted intact.

A new 5-cell stack, identical to the one discussed above (same cell batch), was mounted and tested under similar conditions, foremost to probe reproducibility. Its initial performance at 800°C is given in Fig. 6 and compared with that of the 4-cell stack (same flow in both cases, 5 ml/min.cm\(^2\) H\(_2\)). Both characteristics coincide to within 10 mV. Important activation is again observed, the power density at 0.7 V and 800°C progressing from under 0.3 W/cm\(^2\) to above 0.4 W/cm\(^2\) after around 100 h, also shown in the figure (the influence of the larger flow, 7 vs. 5 ml/min.cm\(^2\) H\(_2\), being less pronounced than the activation process itself, as seen higher). However, it is also remarked that the activation at this stage for the 5-cell stack remained well under that of the 4-cell stack (Fig. 2, 0.53 W/cm\(^2\) after 120 h): the difference lies in the fact that the 4-cell stack was kept at 800°C during activation (0-120 h, Fig. 1), whereas the 5-cell stack (Fig. 6) was kept at 700 and 750°C for its time period \(t = 20\) to 92 h. Performance increase is more rapid at higher temperature (3).

At \(t = 100\) h (800°C), the stack was polarized to 0.4 A/cm\(^2\) (40% fuel conversion), from where average cell voltage increased from 0.8 to 0.82 V at \(t = 170\) h, clearly showing continued activation. At this stage, the electrical furnace failed and cooled down to 400°C, with the stack still polarized to 0.4 A/cm\(^2\) (cell voltages dropped to 0.2 V) before current load was finally interrupted and the stack cooled down to room temperature. It was removed, installed to another furnace and reheated. From Fig. 7 can be seen that the stack fully coped with this double treatment (thermal cycle and current load at too low temperature), as the i-V response (\(t = 236\) h) at conditions identical to those before the incident (800°C, 7 ml/min.cm\(^2\) H\(_2\)) remains virtually the same (taking into account activation from 100 h to 170 h).

Raising to the fuel condition of 8ml/min.cm\(^2\) H\(_2\) (\(t = 259\) h, Fig. 7), power density attained 0.5 W/cm\(^2\) (0.76 A/cm\(^2\) @ 0.65 V) and efficiency 34%, close to the values achieved under the same conditions with the 4-cell stack (Fig. 2), despite an appreciably different test history. The spread in cell behaviour is plotted in Fig. 8, at 800°C and 9 ml/min.cm\(^2\) H\(_2\) flow. (Due to the loss of a voltage lead, only the average potential of cells 4 and 5 can be given.) Again performance is lowest for the colder extreme cell (1) and best for the hottest center cell (3), which achieves 0.8 A/cm\(^2\) at 0.7 V. The stack power is nearly 130 W (0.64 V average per cell, 40 A), corresponding to 1.5 kW/L.

As for the previous test (Fig. 4), the 5-cell stack was kept for the next 300 h under constant polarization at 800°C and 0.48 A/cm\(^2\) (40% fuel conversion), Fig. 9. As the two stacks are identical, again voltage decay was severe, approx. \(-3.7%/100\) h (0.77 V to 0.7 V). After 610 h, current was reduced to 0.34 A/cm\(^2\) to restore to original cell voltage of around 0.77 V. Immediately afterwards, an air supply failure occurred (blocking of air.
mass flow controller) for ca. 10 minutes. Current likewise dropped during this event.

Figure 6. i-V-P output of 5-cell and 4-cell stack (50 cm² active area per cell), compared at 800°C and same fuel flow (5 ml/min.cm² H₂).

Figure 7. i-V-P output of 5-cell stack (50 cm² active area per cell), at 800°C, before and after thermal cycle.
Figure 8. i-V-P output of 5-cell stack (50 cm² active area per cell), at 800°C, fuel flow 9 ml/min.cm² H₂ (air λ = 1.6), t = 260 h. Stack power density 1.5 kWₑ/L.

Figure 9. Voltage degradation on each of the cells (5-cell stack) during galvanostatic polarization (800°C, 9 ml/min.cm² H₂ flow) at successive current densities of 0.48 A/cm² (t < 610 h), 0.34 A/cm² (t = 610-775 h), 0.22 A/cm² (t >775 h).
Restoring polarisation to 0.34 A/cm², cells had lost 30-40 mV owing to the incident. Henceforward, decay was ca. -1.4%/100 h, when current was further reduced to 0.22 A/cm² (t =775 h), decay was still ca. -0.5%/100 h. Degradation appears clearly current density related. Improvement in the flow fields is therefore undertaken to homogenize current density over the cell area so as to avoid zones with too high local current density. To further counteract degradation, the cathode has been modified. Figs. 10-11 illustrate the performance and stability on cell tests using the new cathode (1 A/cm² at 0.75 V, 800°C). These data are representative for small test cells (10 cm²) with only 1 cm² of cathode. No clear degradation was seen over the first 1000 h of operation. It is remarked that this test experienced a planned full thermal cycle (t=380h), after which a 15 mV drop in cell voltage was recorded, as well as two unintended cycles owing to generalized grid power cuts (t=650 h), after which a 10 mV cell voltage drop was registered.

![Figure 10. i-V output of small ASE cell (10 cm²) with 1 cm² central cathode, using metal mesh current collector and large gas flows (120 ml/min H₂, 240 ml/ min air).](image)

**CONCLUSIONS**

The pursued SOFC stacking concept (SOFCONNEX™) has by now demonstrated the following performances: 0.5 W/cm² (1.5 kW/L) @ 0.7 V and 800°C (35% LHV efficiency, H₂); reproducibility; little influence from N₂ dilution and CO addition; robust with respect to thermal cycling and gas feed failures. These compare well with published data of other developers using ASE-based stacks (4-7). Degradation (at stack current density 0.4-0.5 A/cm²) is clearly too large. Improvements are scheduled by implementing the newly shown cathode, protecting the metallic interconnects, and modifying cell configuration and flow fields. The concept has clear commercial potential.

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Figure 11. Time dependent behaviour of the cell of Fig. 10, polarized at 1 A/cm².

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