The SOFCRoll (1, 2) is a new solid oxide fuel cell design being developed at the University of St Andrews. Tape casting, cheap and easily scalable, is utilised as the production technique, and co-firing as a single unit further reduces the cost and time of the cell production. The SOFCRoll geometry eliminates the need for thicker support components, resulting in higher power densities and small space requirements. This paper describes an overview of current SOFCRoll development, together with some predicted power output expectations.

INTRODUCTION

For solid oxide fuel cells (SOFCs) to achieve their full commercial potential, various issues need to be addressed. It is important to keep the cost as low as possible without sacrificing the performance of the fuel cell. This can be achieved by development of new materials and production techniques, which must be viable for mass production. High power densities are required, for which development of new materials is relevant, also changing fuel cell geometry can affect the available power density. In addition, good reliability is important – reduction of thermal stresses associated with thermal expansion differences between components and minimal degradation of fuel cell components. The new SOFC design proposed at the University of St Andrews - the SOFCRoll - aims to address many of the above issues.

Figure 1. Schematic of the SOFCRoll design.
The SOFCRoll design shown in Figure 1 removes the requirement of thicker support components associated with the two leading designs – tubular and planar. This decreases the mass of the fuel cell without decreasing the active surface area of the electrodes and hence increases the power density. The gases are introduced at the two centre ‘tubes’, they flow both down the length of the fuel cell similar to the tubular design, and in addition, travel along the spirals of the fuel cell. The ‘spirals’ – layers of fuel cell materials wrapped around the centre in this way therefore not only give the fuel cell structural support, but are an active part of the fuel cell.

The production technique for all components of the SOFCRoll is tape casting (1, 2), which is both cheap and easily scalable for mass production. The fuel cell is made in the green state, and all components are co-fired as a single unit, reducing production time and hence further lowering the production cost. The problems of high temperature seals associated with the planar design are not experienced with the SOFCRoll.

COMPONENT DEVELOPMENTS

Microstructure of Components

Currently the materials used in each component are as follows: 8YSZ electrolyte; NiO/YSZ anode (reduced in situ) and LSM/YSZ cathode. Figure 2 shows the microstructure of a typical fuel cell produced by co-firing all 3 tape cast components. The density of each component was estimated from geometry measurements of singularly fired components. The electrolyte has a density in excess of 94%, the minimum density required to prevent fuel cross-over in the fuel cell (3).

![SEM image showing the microstructure of the co-fired SOFCRoll fuel cell produced by tape casting. From left to right: cathode LSM/YSZ, electrolyte YSZ, anode Ni/YSZ.](image)

Figure 2.

Variation of the ceramic particle sizes and organic additives in the green tape act to add porosity to the electrodes. The addition of graphite pore formers has been shown to be the most effective method of introducing porosity (2), and for both electrodes the porosity
was further increased in this manner. The porosity of both the anode (after reduction) and cathode meet the required 50% porosity of the SOFCRoll with a porosity of approximately 60%.

**Shrinkage of Components**

The ‘state of the art’ materials used in the SOFCRoll have similar thermal expansion coefficients to each other and so don’t cause cracking or warping on heat treatment of the sintered fuel cell. However, various organic additives required in the slurries for tape casting mean the quantity of powder in a cast tape can be as low as 40 vol% after solvent evaporation and in the absence of any pore former. When the tapes are heated a large shrinkage occurs due to the removal of this organic material. As with the matching of the thermal expansion coefficients of fuel cell materials, it is important to match the shrinkage profiles of all components in order to avoid delamination, warping and cracking of the fuel cells on firing.

Previous work showed that increasing the organic concentration of the tapes and addition of a pore former had the effect of increasing the shrinkage when fired, whereas increasing the ceramic particle sizes decreased the shrinkage (2). Using this information the composition of the tapes could be tuned to allow the shrinkage of each component to be as close as possible. Figure 3 shows how the shrinkage of the anode was tailored in order to match that of the electrolyte.

![Shrinkage profiles before Tailoring of the Slurries](image1)

![Shrinkage profiles after Tailoring of the Slurries](image2)

**Figure 3. Shrinkage profiles of anode and electrolyte before and after modification of anode composition.**

**POWER DENSITY PREDICTIONS**

While the SOFCRoll design addresses many issues, like many other designs, it involves long current paths. In SOFCRoll the electrical connection can only be made at the centre of the roll or outer edges of the spiral, therefore the current will have a long path along the current collectors, which ideally could be just the electrode materials. As these would preferably be reasonably thin, their resistance would be quite high inducing a significant IR drop over the electrode. At the same time the partial pressures of the gases will change as the fuel is utilised while passing from the inlet in the centre along the spiral. Both of
these effects will act to lower the overpotential and so lower the current density. A simple model was produced to establish appropriate design parameters.

The model is based on a strip of fuel cell 1 cm wide (see Figure 4(d)) with a contacts along one edge, where the gas is also inserted. This strip is obtained by cutting a 1 cm section from the spiral (Figure 4(a)), and unrolling (Figure 4(b)), and then cutting it in half (Figure 4(c)). The power density near the current collector was taken to be 0.5 W cm\(^{-2}\) (i.e. similar to a fairly standard planar cell) at an output voltage of 0.7 V. The current density was taken to drop linearly with the overpotential \(\Delta V = V_{oc} - V\) for the cell, where \(V_{oc}\) varies with the partial pressures along the strip according to the Nernst equation. The current must be zero at the opposite end from the current collector, and increases as the sum of the current density at that point plus the sum of all the current before that.

![Figure 4. Schematic of model fuel cell and relation to SOFCRoll. (a) 1 cm section from roll, (b) unrolled to form sandwich, (c) halved and flattened to give single cell, (d) with current collectors at end. Gas flow along strip from current collectors.](image)

The voltage increases as IR along strip:

\[
V(x) = V_o + \int_0^x R I(y) dy
\]  

[1]

and the current decreases as \(J(V_{oc}-V)\) along strip:

\[
I(x) = I_o - \int_0^x J(V_{oc} - V(y)) dy \quad \text{where} \quad I_o = \int_0^x J(V_{oc} - V(y)) dy
\]  

[2]

where \(V_{oc}\) is given by the Nernst equation

\[
V_{oc} = E^\circ + \frac{RT}{nF} \ln \left( \frac{pH_2(x)\sqrt{pO_2(x)}}{pH_2O(x)} \right)
\]  

[3]

The simultaneous integral equations were solved numerically by transforming [1] and [2] into a discrete form:

\[
V(x+dx) = V(x) + R_{\text{cathode}} I(x) \ dx + R_{\text{anode}} I(x) \ dx
\]  

[4]

\[
I(x+dx) = I(x) - \Delta J \left( V_{oc}(x) - V(x) \right) \ dx
\]  

[5]
This forms a model as shown in Figure 5, where $V_{oc}$ depends on the partial pressures of the reactant gases which are also calculated discretely based on the flow rate of gas:

$$pH_2(x + dx) = pH_2(x) - \frac{\Delta I}{2F} \cdot \frac{1}{\text{flow}}$$  \hspace{1cm} [6]

$$pO_2(x + dx) = pO_2(x) - \frac{\Delta I}{4F} \cdot \frac{1}{\text{flow}}$$  \hspace{1cm} [7]

$$pH_2O(x + dx) = pH_2O(x) + \frac{\Delta I}{2F} \cdot \frac{1}{\text{flow}}$$  \hspace{1cm} [8]

where $\Delta I = \Delta J (V_{oc}(x)-V(x)) \, dx$,

$\Delta J$ = reference current density / reference overpotential,

$\text{flow}$ = gas flow rate in mol of gas/s

$E^o = \Delta G/2F$  \hspace{1cm} [12]

with $\Delta G$ from JANAF tables (4).

Figure 5. Equivalent circuit of (discrete) model. $V_{oc}$ varies with gas partial pressures.

The above equations [3-9] were placed into a spreadsheet and the initial conditions, e.g. temperature, flow rate, electrode resistance, gas partial pressures etc, were set. The only unknown is the initial current $I_0$, which can be obtained by an iterative process, to give the correct limiting current of $I = 0$ at end of strip. The value of $I_0$ was obtained either by hand or the faster ‘Solver’ function of Excel if possible.

An example plot is shown in Figure 6 from the model for a 5 cm long strip with a 0.1 $\Omega$ cm$^{-1}$ resistance cathode current collector and 0.01 $\Omega$ cm$^{-1}$ anode current collector. The current drops quickly as the distance increases from the current connection, while the voltage rises to the open circuit voltage. With a cathode material resistance of 0.1 $\Omega$ cm$^{-1}$ a typical power output of about 30% of the same area ideal (no Nernst losses) planar electrode. The results from this model are sensible, with smooth curves and a singular result, and numbers produced are physically possible.
Figure 6. Voltage and current profiles across a 5 cm strip of fuel cell, this corresponds to an overall power output per unit area of 0.15 W cm\(^{-2}\).

The only unrealistic assumption used in the model is the linearity of the current density with overpotential, over a range of partial pressures. However, as this is only a simple model which ignores many other possible losses, e.g. thermal gradients, pressure drops etc, this approximation is close enough to give a guide to the upper limit of current output and so to optimise geometry and material selection.

The most important parameter is current collector (electrode) resistance, and the variation of output current for a 5 cm strip with cathode resistance is shown in Figure 7. The two traces show the difference between just the IR drop (without Nernst calculation) and that including both IR drop and partial pressure changes along the strip. The current drops very quickly with increasing resistance, dropping below 20% of an ideal planar fuel cell. This shows that 0.1 \(\Omega\) cm\(^{-1}\) is a sensible maximum for the electrode resistance for this cell design, and any improvements in conductivity will give a significant increase in power output.

If the electrode material has a bulk conductivity of 200 S cm\(^{-1}\), this gives a resistivity of about 0.1 \(\Omega\) cm\(^{-1}\) for a 50 \(\mu\)m thick dense layer. The ability to vary the composition and structure of the electrode, i.e. thicker, or adding extra current collection layers can be used to lower the resistance of the current path, without significantly modifying the production process.

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Figure 7. Percentage capacity relative to an ideal planar electrode (0.5 W cm$^{-2}$) as a function of electrode material resistance. The upper trace shows the effect of not including partial pressure changes in calculations.

Figure 8 shows the change in output current with strip length for a 0.1 $\Omega$ cm$^{-1}$ electrode which can be used to get the optimum length for efficient usage of material. This shows that over 90% of the possible current is obtained for a 4 cm long strip, with any portion of strip (or length from centre of spiral) over 10 cm from the current collector not electrochemically active. Spiral lengths in excess of this would increase the mechanical strength, however this would decrease the power density of the system.

Figure 8. Output current variation with fuel cell length for a 0.1 $\Omega$ cm$^{-1}$ electrode.
It is interesting to note that for resistances of 0.1 $\Omega$ cm$^{-1}$ or below, and using air as the oxidant gas (initial partial pressure 0.23), the model cell is primarily limited by the oxygen availability, as well as IR drop, with about the same decay length. This is an important limitation to follow as well.

**CONCLUSIONS**

The development of the SOFCRoll is very promising. It is possible for each component of the fuel cell to be produced by tape casting and co-fired as a single unit without cracking or warping of the cell. This was achieved by tailoring the organic additives concentrations and the relative ceramic particle sizes in the green tape to ensure the shrinkage profile of all components was a close match. The high porosity required for the electrodes was obtained by addition of a graphite pore former, yielding electrode porosities in excess of 50%.

Modelling of the voltage and current distribution along the spiral of the SOFCRoll showed that the minimum practical resistance of the current collectors is 0.1 $\Omega$ cm$^{-1}$. Furthermore, for a 0.1 $\Omega$ cm$^{-1}$ total resistance current collector, a spiral length between 5 and 10 cm would produce maximise power output with out wasting material. These dimensions are in the range of practically useful sizes.

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