Membrane Electrode Gasket Assembly (MEGA):
A Novel Technology for PEFCs

A. Pozio a*, L. Giorgia, S. Galli, M. De Francesco a, R.F. Silva a,
R. Lo Prestia a and A. Danzi b

aENEA, C.R. Casaccia, Via Anguillarese 301, 00060, S. Maria di Galeria (Rome), Italy
bRoen Est Fuel Cell, Via S. Isaia 24, 40100 Bologna, Italy

(Received March 3, 2003; received revised form August 20, 2003)

Abstract: A new technology for the production of a membrane electrode gasket assembly (MEGA) for polymer electrolyte fuel cells (PEFCs) is defined. The MEGA system was prepared by sealing a previously prepared membrane electrode assembly (MEA) in a moulded gasket. For this aim, a proprietary silicone based liquid mixture was directly injected into the MEA borders. Gaskets obtained in different shapes and hardness grades are stable in a wide temperature range. The MEGA technology shows several advantages with respect to traditional PEFCs stack assembling systems: effective membrane saving, reduced fabrication time, possibility of quality control and failed elements substitution. This technology was successfully tested at the ENEA laboratories and the results were acquired in laboratory scale, but industrial production appears to be simple and cheap.

Key words: polymer electrolyte fuel cell, membrane electrode assembly, gaskets, stack

1. INTRODUCTION

Because of their high power density performance at low temperature (70-90°C), polymer electrolyte fuel cells (PEFCs) are very promising as power source for vehicular transportation and other applications requiring clean, quiet and portable power. Nevertheless, the cost of a PEFC stack is still prohibitive for mass production and the introduction of low-cost materials and/or processing is necessary. Moreover, industrial production needs a further quality control to guarantee the every PEFC stack will have well defined performance. This goal can be achieved only when single cell substitution in a stack is allowed. Stack disassembly and replacement of failed cells are possible only upon introduction of a new production process that permits: 1) quality control, 2) long-term use without any change or decrease of performance, 3) long-term storage and 4) low cost.

In a PEFC stack the individual cells are placed side by side so as to form a series connection in order to obtain the desired value of overall voltage. The MEGA (Membrane Electrode Gasket Assembly) technology, developed by ENEA and Roen Est Fuel Cell, takes into account the problems concerning the connection between the membrane/electrode assembly (MEA) and the current collectors. The production of electric current takes place thanks to the high proton conductivity of a perfluorinated sulphon membrane that prevents the reactant gases from coming into physical contact, permitting only H+ ions to pass through it. The gases are evenly distributed over the surface of the MEA by flowing inside channels grooved on the current collectors. Of extreme importance is the absence of gas leaks between the MEA and the plates. In this sense, a seal (i.e. a gasket) is placed between the MEA and the plates. However, the gasket must not come into contact with the porous electrode, since it would cause the gases to escape. For this reason, the membrane overhangs the four sides of the assembly. Due to its non-porous nature, the membrane provides an adequate connection surface for the gasket.
Some drawbacks may arise when a PEFC stock is to be assembled: a) difficulty to guarantee the gas sealing as the number of individual MEAs is increased; in fact, two gaskets are required for each individual MEA assembly; b) due to the need to make the membrane exceed the edges of the electrode, each MEA is made separately; c) three operations for each individual cell are foreseen for assembling the stack (first, positioning the gasket; second, positioning the MEA, and third, positioning the second gasket); d) there is a membrane waste because the membrane area overhanging the electrode is not used for the ionic exchange since it is left for connection with the gaskets.

The MEGA technology introduced in this work has the purpose of eliminating, in principle, these drawbacks by using a “drowning” process of the MEA in the gasket, according to a known process of material injection moulding. This aim is attained by a device characterised as being made basically like a MEA assembly without the overhanging membrane. A bi-component silicone based liquid mixture is injected directly into a special homemade mould on the perimeter of each MEA, so as to create the MEGA assembly.

The perfect seal of the gas is guaranteed by the fact that the gasket, which is injected and not only placed, is inserted in the porosities of the electrode in order to form a perfectly impermeable cell. As already pointed out, the MEGA assembly is basically one single component after the moulding operation has been completed and, therefore, we no longer have three components of the traditional system, that is, the two gaskets and the MEA. Due to this fact, the number of operations needed for assembling the stack is considerably reduced, i.e. a unique operation is needed instead of three. In summary, as far as the manufacturing system is concerned, we get a substantial time saving both in the making of the MEGA assemblies and in the complete stack assembly. Furthermore, the whole membrane is used for electric current production and a substantial saving of material (i.e. membrane) is achieved with this system. Finally, the MEGA assembly proves to be of superior quality as it takes advantage of the electrode porosity for the gas sealing process. The final product has all the characteristics listed above, which are considered to be necessary for mass production.

2. EXPERIMENTAL

2.1. Materials

Commercially available 20 wt% Pt/C catalyst powders on carbon black (Vulcan XC72) were obtained from E-TEK Inc. Three-layer (substrate/diffusive layer/catalyst layer) gas diffusion anodes and cathodes (106 cm²) were prepared using a spray technique described in detail in previous works [1-3]. The substrate was carbon paper (Toray TGP1090). The weight composition of the diffusion layer was 85 wt% of carbon (Super P – MMM Carbon Belgium) and 15 wt% of PTFE, with carbon loading of 1.93 mg cm⁻². The catalyst layer was prepared by mixing appropriate amounts of carbon supported catalyst (24.5 wt%), 5 wt% Nafion ionomer solution from Aldrich (14.9 wt%) and glycerol (60.5 wt%). The platinum loading in all anodes and cathodes was kept constant at 0.68 mg cm⁻² on MEGA1 and MEGA2 and at 0.34 mg cm⁻² on MEGA3. Nafion 115 membrane (Du Pont) was used after purification treatment in 5 w/v% H₂O₂ solution at 80°C for 1 h, followed by treatment in 1 M H₂SO₄.

The membrane electrode assemblies were made by hot pressing the electrodes (106 cm²) onto the membrane at 130°C for 1-5 minutes and 50-100 kg cm⁻².

The MEAs were inserted in a heated (50°C) aluminium mould provided with holes for injection of a Roen Est’s own proprietary silicone based mixture [4]. The mixture, injected directly on the MEA borders by means of a pressure gun, solidifies in a few minutes and no substances are emitted during the solidification process. The aluminium mould was formed to originate a well-defined shape in accordance with the cell hardware (Fig.1). At the lateral borders of the assembly, manifolds holes for gas feeding are visible. All complex shapes can be easily formed and a wide range of hardness grades is achievable (30-90 shore) by using different mixture compositions. However, silicone with shore level of 50 was found to have the best features able to provide adequate sealing and mechanical resistance to the compaction force. The gasket materials have physical-chemical and mechanical characteristics suitable for the PEFC operative conditions, as follows: 580 psi of tensile strength and 400% of elongation. The thermal stability was tested in the range from −50° to +180°C and the gas sealing successfully tested up to 10 bars. Other important features of our silicone-based gaskets include their non-adhesive, water repulsive, electric insulating and fire extinguishing properties. Three membrane electrode gasket assemblies (namely, MEGA1, MEGA2 and MEGA3) were prepared and stored fully immersed in distilled water. Graphite plates were assembled with a typical parallel channel configuration using commercially available BMA5 graphite produced by SGL Carbon Group (Germany).

2.2. Electrochemical characterisation

Full-cell electrochemical tests were separately carried out on the MEGA1, MEGA2 and MEGA3 systems using a 106 cm² single cell incorporated in a Globe Tech Inc mod. 890 test station. Two aluminium end plates and two graphite current collectors composed the single cell. The MEGA system and the other components of single fuel cell are schematised in Fig.1. After the single cell measurements, MEGA1, MEGA2 and MEGA3 were stored in distilled water for some weeks. Subsequently, simply by adding a middle graphite bipolar plate to the single cell hardware, the two characterised MEGA1 and MEGA2, were assembled and tested in a two-cell stack. Successively, by adding a second middle bipolar plate to the bi-component stack, MEGA3 was inserted and a three-cell stack was assembled and tested.
Cell voltage vs. current density measurements were galvanostatically performed with a programmable power supply interfaced with a computer for data acquisition. All the measurements were carried out in the same operative conditions reported in Table 1. To obtain a steady state galvanostatic polarisation curve, the time for each measurement was found to be at least 0.5 min/point. The overall ohmic resistance was measured by electrochemical impedance spectroscopy (EIS). The fuel cell was connected to an Ecochemie potentiostat/galvanostat mod. PGSTAT30 having frequency response analyser expansion modules interfaced with a personal computer. EIS measurements were conducted in the frequency range from 10 kHz to 0.1 Hz at open circuit potential (OCP). The amplitude of the ac signal was always 10 mVpp.

**Figure 1.** Scheme of a single fuel cell showing the location of the components: A) aluminium end plates, B) gaskets, C) graphite current collectors, D) MEGA (draw and photographic detail).

**Table 1.** Operative conditions used in cell tests.

<table>
<thead>
<tr>
<th>Operative parameters</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell Temperature/°C</td>
<td>75</td>
</tr>
<tr>
<td>H₂ pressure/bar abs</td>
<td>1.1</td>
</tr>
<tr>
<td>Air pressure/bar abs</td>
<td>1.1</td>
</tr>
<tr>
<td>H₂ flow/scc/min/A/cell</td>
<td>14 (2 S.R.)</td>
</tr>
<tr>
<td>Air flow/scc/min/A/cell</td>
<td>52.5 (3 S.R.)</td>
</tr>
<tr>
<td>Anode humidifier temperature/°C</td>
<td>80±85</td>
</tr>
<tr>
<td>Cathode humidifier temperature/°C</td>
<td>50±70</td>
</tr>
</tbody>
</table>

Losses at different airflows were measured using a pressure drop indicator Spriano connected in parallel with the flow air inlet and outlet (DP_max=160 mbar).

**3. RESULTS AND DISCUSSION**

Steady-state galvanostatic polarisation data of the MEGA1, MEGA2 and MEGA3 systems are shown in Figure 2. The cells show the same trend with an increase of power density with current. At low current densities, all different cells show the same behaviour while at high current densities MEGA1 and MEGA 2 show slightly better performance. The difference between MEGA1 and MEGA2 can be attributed to ohmic or diffusive factors probably due to irreproducibility, which it is inherent to the electrodes and/or the MEA production process. A worse performance of MEGA3 with respect to MEGA1 and MEGA2 assemblies can be ascribed to the lower Pt loading.

**Figure 2.** Cell voltage vs. current density for MEGA1, MEGA2 and MEGA3, at \( T_{\text{cell}}=75^\circ \text{C} \) under \( \text{H}_2/\text{air} \) flux at 1.1/1.1 abs bar, S.R._\text{cathode}=2 and S.R._\text{anode}=3.

At 566 mA cm⁻², voltage values of 0.563 V for MEGA1, 0.508 V for MEGA2 and 0.43 V for MEGA3 were obtained in the operative conditions presented in Table 2. Two and three-element stack polarisation data are shown in Figure 3.

**Table 2.** Maximum absolute power and power density for single cells and stacks.

<table>
<thead>
<tr>
<th>System</th>
<th>( P_{\text{max}}/\text{W} )</th>
<th>( P_{\text{max}}/\text{kw} \text{m}^{-2} )</th>
<th>( P ) @ 472 mA cm⁻²/kW m⁻²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single Cell MEGA1</td>
<td>32.3</td>
<td>3.04</td>
<td>2.75</td>
</tr>
<tr>
<td>Single Cell MEGA2</td>
<td>30.3</td>
<td>2.86</td>
<td>2.64</td>
</tr>
<tr>
<td>Single Cell MEGA3</td>
<td>25.5</td>
<td>2.41</td>
<td>2.37</td>
</tr>
<tr>
<td>Stack (2 elements*#)</td>
<td>62.2</td>
<td>2.93</td>
<td>2.67</td>
</tr>
<tr>
<td>Stack (3 elements#)</td>
<td>67.2</td>
<td>2.93</td>
<td>2.11</td>
</tr>
</tbody>
</table>

The stack voltage at maximum current density of 566 mA cm⁻² was 1.043 V for the two-cell stack and 1.12 V for the three-cell stack. Table 3 compares the data obtained for all systems and evidences that the absolute maximum power of the stack is the sum of those of the single cells only for the two element system. The power density is about 3 kW m⁻² and the performance appears to be interesting considering the low operative pressure (\( \text{H}_2/\text{air}=1.1/1.1 \) bar abs), stoichiometric...
ratio ($H_2=2$, air=3) and platinum loading. As the number of cells is increased to three, we observe a decrease of performance. We address this behaviour to a non-optimised fluid dynamics, which is explained by uncontrolled diffusive phenomena taking place as the current density increases. In fact, up to 200 mA cm$^{-2}$, the two and three cell stacks show the same specific power. For this reason, the following tests were carried out only on the two-cell stack.

Short-term stack performance stability was tested maintaining the stack at high current density (562 mA cm$^{-2}$) for about 210 min in the operative conditions of Table 1 and measuring the stack voltage (Fig. 4). It was observed that the cathode humidifier temperature plays a very important role in the working current densities and in the amount of produced water. In fact, the water produced in the catalytic layer at the cathode side can either spread in the membrane, keeping it wet and avoiding an increase of ohmic resistance, or diffuse into the electrode flooding it. Therefore, a control of the air humidifier temperature is extremely necessary in order to equilibrate these two different phenomena.

At low current density, the rate of water production is low and saturation of the inlet air flow is needed to prevent the membrane from drying. The cathode humidifier temperature must be set at a temperature close to that of the cell (75-80°C). The rate of water production increases with current and, in this condition, membrane humidification is guaranteed, but electrode flooding can occur. For this reason, it is necessary to reduce the water saturation in the inlet airflow. This can be achieved by decreasing the cathode humidifier temperature. For example, at current density of 562 mA cm$^{-2}$, the best performance was obtained with an humidifier temperature of 62°C. Long-term durability and stability tests were carried out on the MEGA3 assembly at 70°C and 150 mA cm$^{-2}$ in H$_2$/O$_2$ flux. Fig.5 shows the cell potential vs. time curve for ca. 3400 hours. No material degradation or leaks were observed and the cell potential was 0.74±0.02 V with 3.3% variation due to uncontrolled but reversible phenomena such as lowering of the humidification levels and/or electrode flooding.

Figure 3. Cell voltage vs. current density for two and three-cell stack (MEGA1 + MEGA2 + MEGA3), at $T_{cel}=75$°C under H$_2$/air flux at 1.1/1.1 abs bar, $S.R_{cathode}=2$ and $S.R_{anode}=3$.

Table 3. Ohmic resistance obtained by EIS for MEGA1 and MEGA2 single cells and stack (MEGA1+MEGA2).

<table>
<thead>
<tr>
<th>System</th>
<th>$R_O$/ohm cm$^2$</th>
<th>$R'_O$/ohm cm$^2$/cell</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single Cell MEGA1</td>
<td>0.258</td>
<td>0.258</td>
</tr>
<tr>
<td>Single Cell MEGA2</td>
<td>0.254</td>
<td>0.254</td>
</tr>
<tr>
<td>Stack</td>
<td>0.570</td>
<td>0.285</td>
</tr>
</tbody>
</table>

Short-term stack performance stability was tested maintaining the stack at high current density (562 mA cm$^{-2}$) for about 210 min in the operative conditions of Table 1 and measuring the stack voltage (Fig. 4). It was observed that the cathode humidifier temperature plays a very important role in the working current densities and in the amount of produced water. In fact, the water produced in the catalytic layer at the cathode side can either spread in the membrane, keeping it wet and avoiding an increase of ohmic resistance, or diffuse into the electrode flooding it. Therefore, a control of the air humidifier temperature is extremely necessary in order to equilibrate these two different phenomena.

At low current density, the rate of water production is low and saturation of the inlet air flow is needed to prevent the membrane from drying. The cathode humidifier temperature must be set at a temperature close to that of the cell (75-80°C). The rate of water production increases with current and, in this condition, membrane humidification is guaranteed, but electrode flooding can occur. For this reason, it is necessary to reduce the water saturation in the inlet airflow. This can be achieved by decreasing the cathode humidifier temperature. For example, at current density of 562 mA cm$^{-2}$, the best performance was obtained with an humidifier temperature of 62°C. Long-term durability and stability tests were carried out on the MEGA3 assembly at 70°C and 150 mA cm$^{-2}$ in H$_2$/O$_2$ flux. Fig.5 shows the cell potential vs. time curve for ca. 3400 hours. No material degradation or leaks were observed and the cell potential was 0.74±0.02 V with 3.3% variation due to uncontrolled but reversible phenomena such as lowering of the humidification levels and/or electrode flooding.

Figure 4. Two-cell (MEGA1 + MEGA2) stack voltage and power density vs. time at constant current density of 562 mA cm$^{-2}$; $T_{cel}=75$°C, H$_2$/air flux at 1.1/1.1 abs bar, $S.R_{cathode}=2$ and $S.R_{anode}=3$.

Figure 5. Cell voltage vs. time for MEGA3 at 142 mA cm$^{-2}$, $T_{cel}=70$°C, H$_2$/O$_2$ flux 1.1/1.1 abs bar, $S.R_{cathode}=2$ and $S.R_{anode}=3$.

Figure 6 shows EIS data obtained for the three systems. In particular, the real part of the impedance at high frequency represents the ohmic resistance ($R_O$) which is the sum of the proton exchange membrane and electronic resistances of the single-cell/stack test fixtures. Table 3 reports the active area normalised data of $R_O$ for single cell and stack, showing that MEGA1 and MEGA2 assemblies have similar resistance values. The stack resistance is about twice that of the single
cell and the difference between the specific cell resistances ($R'_W$ in Table 3) is ca. 31-35 mohm cm$^2$. This increase of resistance in the stack can be due to the presence of the middle graphite bipolar plates. An influence of clamping torque force on the resistance values in the cell/stack assembly was found. The best performance was obtained by applying a torque force of about 45-50 kgf cm corresponding to a compaction force of 220-240 N cm$^{-2}$, which is close to the values reported by Davies et al. [5].

Figure 6. Real impedance vs. frequency for MEGA1, MEGA2 and two-cell stack at OCP, 75°C, under H$_2$/air flux.

Figure 7 shows the cathodic pressure loss ($\Delta p$) vs. airflow plot for the three systems in the operative conditions (75°C and open circuit voltage). For all the systems $\Delta p$ was less than 100 mbar at high airflow, evidencing for a good design of collector plate flow field in the single cell and also in the stack.

A twenty-cell stack (106 cm$^2$ each cell) was assembled at the ENEA laboratories with this new technology (Fig. 8). The stack was fully immersed in a tank containing a non-conductive, circulating liquid continuously pumped into a cooling system in order to keep the stack temperature constant.

One MEGA assembly was electrochemically tested prior to its insertion in the stack in order to verify its performance stability (Fig. 9). It is worth pointing that the reason for lower voltages at 5A was due to the constant value imposed to the cathode hydration temperature (62°C), which was inferior to that of the cell temperature (75°C) and optimised for high current density as already explained above.

Figure 8. ENEA/Roen Est twenty-cell (106 cm$^2$) stack.

In addition, the stack was opened several times to control the integrity of the assemblies and to test the possibility of substitution and storage. Figure 10 shows the average cell power in the same operative conditions after four different disassembly operations have been made. Once again, we cannot compare these data with those registered for MEGA1 and MEGA2 assemblies, because the operative conditions were different and the stack itself was not yet optimised to reach the same high current density, as we showed for the three-cell system. However, the single MEGA assembly in Fig. 9, kept in the operative conditions of Table 1, provided a similar voltage value to those of MEGA1 and MEGA2 (0.59 V at 566 mA cm$^{-2}$).
A second optimised thirty-cell stack (106 cm² each cell) was assembled and tested in the same operative conditions of Table 1. In this stack the graphite plates were fabricated with a lower number of parallel channels with respect to the twenty-cell stack and the MEGAs were produced using Nafion 112 membranes. Also in this case one MEGA assembly was electrochemically tested to compare its performance with those of the cells in the stack. Figure 11 shows the power density for each cell in the stack at 293 mA cm⁻² and for the single cell tested in the same operative conditions. The results show that about 57% of the cells in the optimised stack gave performance above the mean value and several cells worked better than the single one. The single MEGA assembly in Fig. 11, kept in the operative conditions of Table 1, provided a higher voltage value (0.62 V at 566 mA cm⁻²) than MEGA1 and MEGA2. The better results are due to the low ohmic resistance of thinner Nafion 112 (0.21 ohm cm⁻²) with respect to 115 (see Table 3) and to the optimised graphite plates fluid-dynamics.

4. CONCLUSIONS

The paper shows that the MEGA technology looks promising for large-scale fuel cell diffusion offering several advantages with respect to the traditional assembling systems:

1) Cost reduction due to a better use of expensive electrolyte membrane (reduction cost ca. 5-10%): the membrane is not used as a gasket support; instead, it is present only in the active area.

2) Manufacturing time reduction: the MEGA assembly preparation as well as its insertion/extraction in/from the fuel cell stack can be considered as an easy and quick process.

3) Quality control and simple storage before assembling: the MEGA system can be characterised in a single fuel cell configuration and stored before utilisation in a stack with maintenance of performance.

4) Operation and maintenance: it is possible to disassemble stack for failed element substitution. MEGA can be used several times without any change or decrease of performance.

REFERENCES


Despite the difficulty to compare the data, it was verified the possibility to disassemble the stack, moving and/or replacing cells, without any change or decrease of performance. This technology evidences for very promising, simple and cheap industrial production scale.