

Membrane electrode gasket assembly (MEGA) technology for polymer electrolyte fuel cells

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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 injected directly 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.

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1. Introduction

Thanks to their high power density performance at low temperature (70–90 °C), polymer electrolyte fuel cells (PEFCs) are very promising as energy source for electric vehicles. Nevertheless, the cost of a PEFC stack is still prohibitive for mass production and the introduction of low-cost materials and/or process is necessary. Moreover, industrial production needs a further quality control to guarantee that 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-range use without any change or decrease of performance, (3) long-range 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 membrane electrode gasket assembly (MEGA) 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 sulphonic membrane that prevents the reactant gases from coming into physical contact, permitting only H⁺ ions to pass through. 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 assembly and the plates. For this reason, 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 once 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.

The former discussion brings about a variety of drawbacks which are listed as follows: (a) difficulty to guarantee the gas sealing as the number of individual MEA 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

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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 home made 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's porosity for the gas sealing process. The final product has all the characteristics listed above, which are considered to be necessary for massive 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 cm^2) were prepared using a spray technique described in detail in previous works [1–3]. The substrate was carbon paper (Toray TGPH090). The weight composition of the diffusion layer was 85 wt.% of carbon 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 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% H_2O_2 solution at $80\text{ }^\circ\text{C}$ for 1 h, followed by a second treatment in 1 M H_2SO_4 .

The membrane electrode assemblies were made by hot pressing the electrodes (106 cm^2) onto the membrane at $130\text{ }^\circ\text{C}$ for 1–5 min and $50\text{--}100\text{ kg cm}^{-2}$.

The MEAs were inserted in a heated ($50\text{ }^\circ\text{C}$) 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 seconds 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. Thermal stability was tested in the range from -50 to $+180\text{ }^\circ\text{C}$ and 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 graphite materials BMA5 produced by SGL Carbon Group (Germany).

2.2. Electrochemical characterisation

Full-cell electrochemical tests were separately carried out in the MEGA1, MEGA2 and MEGA3 systems using a 106 cm^2 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 and MEGA2 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 MEGAs, were assembled and tested in a two-cell stack.

Cell voltage versus current density measurements were galvanostatically performed with a programmable power supply interfaced with a computer for data acquisition. All measurements were carried out in the same operative

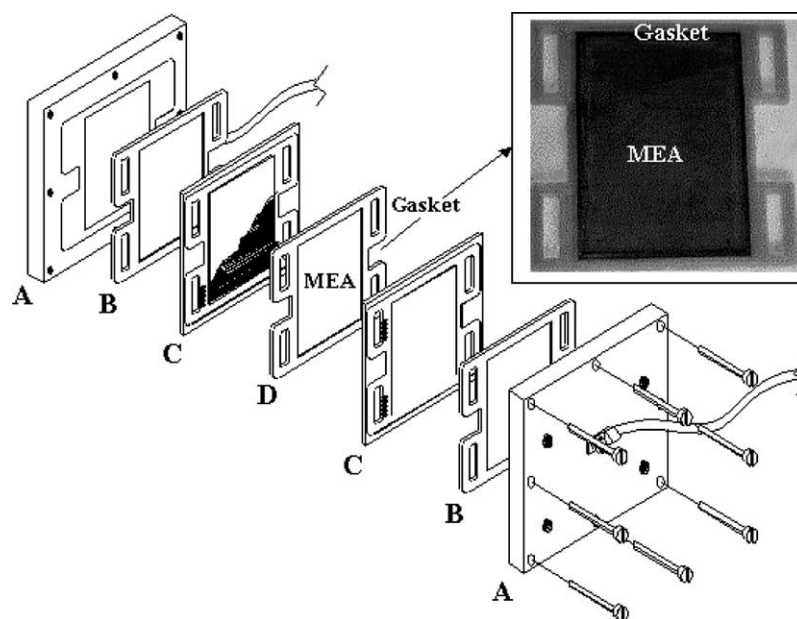


Fig. 1. Schematic of single PEM fuel cell showing the location of the components: (A) aluminium end plates, (B) gaskets, (C) graphite current collectors, (D) MEA (draw and photographic detail).

conditions reported in Table 1. 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.

Losses at different airflows were measured using a pressure drop indicator Spriano connected in parallel with the flow air inlet and outlet ($\Delta P_{\max} = 160$ mbar).

3. Results and discussion

Steady-state galvanostatic polarisation data of the MEGA1 and MEGA2 systems are shown in Fig. 2. Both cells show the same trend with an increase of power density with current. At low current densities, the two different cells show the same behaviour while at high current densities MEGA1 shows lightly better performance. The difference

can be attributed to ohmic or diffusive factors probably due to un-reproducibility which it is inherent to the electrodes and/or the MEA production process. At 566 mA cm^{-2} , voltage values of 0.563 V for MEGA1 and of 0.508 V for MEGA2 were obtained in the operative conditions presented in Table 1. Stack polarisation and power density data are shown in Fig. 3. The stack voltage at maximum current density of 566 mA cm^{-2} was 1.043 V. Table 2 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. 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 ($\text{H}_2 = 2$, air = 3) and platinum loading.

Table 1
Operative conditions used in cell tests

Operative parameters	
Cell temperature ($^{\circ}\text{C}$)	75
H_2 pressure (bar abs)	1.1
Air pressure (bar abs)	1.1
H_2 flow (scc/min/A/cell)	14 (2 S.R.)
Air flow (scc/min/A/cell)	52.5 (3 S.R.)
Anode humidifier temperature ($^{\circ}\text{C}$)	80–85
Cathode humidifier temperature ($^{\circ}\text{C}$)	50–70

Table 2
Maximum absolute power and power density on MEGA1 single cell, MEGA2 single cell and stack (MEGA1 + MEGA2)

System	P_{\max} (W)	P_{\max} (kW m^{-2})	P (at 472 mA cm^{-2})
Single cell MEGA1	32.3	3.04	2.75
Single cell MEGA2	30.3	2.86	2.64
Stack (MEGA1 + MEGA2)	62.2	2.93	2.67

Table 3
Ohmic resistance obtained by electrochemical impedance spectroscopy on MEGA1 and MEGA2 single cells and stack (MEGA1 + MEGA2)

System	R_{Ω} (ohm cm^2)	R'_{Ω} (ohm cm^2/cell)
Single cell MEGA1	0.258	0.258
Single cell MEGA2	0.254	0.254
Stack (MEGA1 + MEGA2)	0.570	0.285

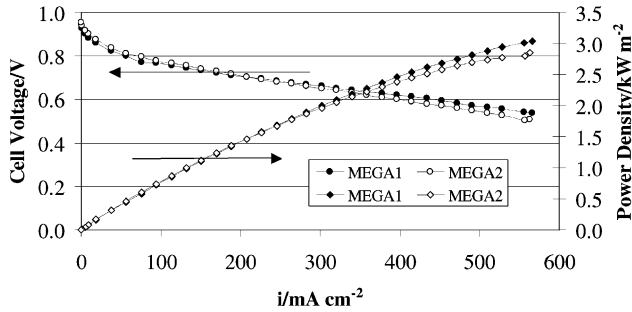


Fig. 2. Cell voltage vs. current density for MEGA1 and MEGA2, at $T_{\text{cell}} = 75^\circ\text{C}$ under H_2/air flux at 1.1/1.1 bar abs.

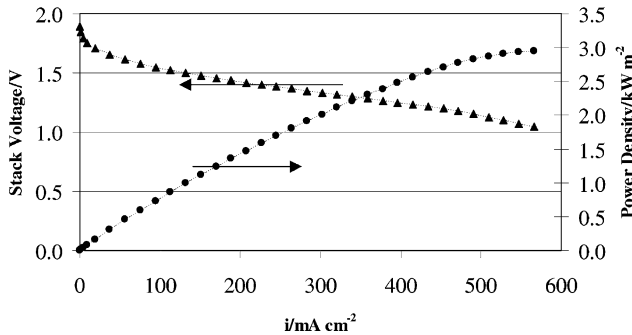


Fig. 3. Cell voltage vs. current density for two-cell stack (MEGA1+MEGA2), at $T_{\text{cell}} = 75^\circ\text{C}$ under H_2/air flux at 1.1/1.1 bar abs.

Short-range 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 to 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 to prevent the membrane from drying is needed. The cathode humidifier temperature must be set at a temperature close to that of the cell ($75\text{--}80^\circ\text{C}$). As the current increases, the rate of water production raises also; 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 a current density of 562 mA cm^{-2} , best performance was obtained with an humidifier temperature of 62°C . Long-range 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 versus time curve over the specified duration. No degradation was observed and the cell potential was $0.74 \pm 0.03\text{ V}$ with 4% variation due to uncontrolled but reversible phenomena such as lowering of the humidification levels and/or electrode flooding. This result cannot be compared with those of the MEGA1 and MEGA2 assemblies, because the operative conditions are different. Nevertheless, the MEGA3 assembly maintained in the operative conditions of Table 1 furnished a voltage value of 0.43 V at 566 mA cm^{-2} . The worse performance with respect to MEGA1 and MEGA2 assemblies can be ascribed to the lower Pt loading for MEGA3. Fig. 6 shows EIS data obtained for the three systems. Particularly, the real part of impedance at high frequency represents the ohmic resistance (R_Ω) 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_Ω for single cell and stack, showing that the 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'_Ω in Table 3) is ca. $31\text{--}35\text{ m}\Omega\text{ cm}^2$. This increase of resistance

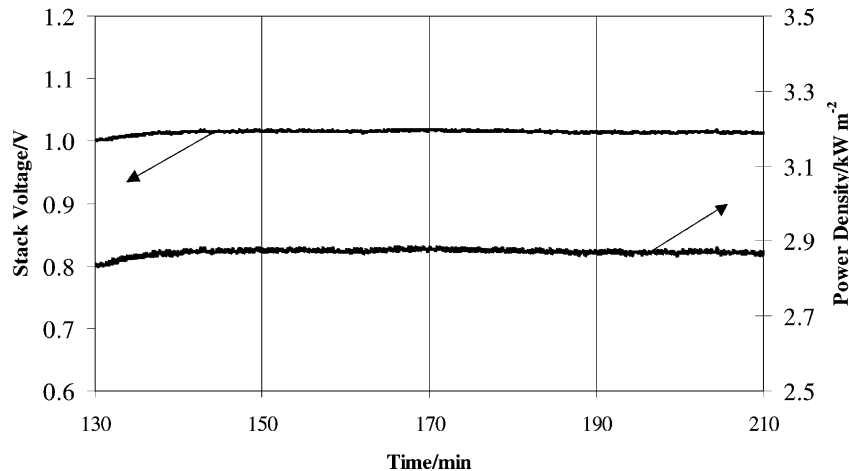


Fig. 4. Two-cell stack voltage and power density vs. time at constant current density of 562 mA cm^{-2} ; $T_{\text{cell}} = 75^\circ\text{C}$, H_2/air flux at 1.1/1.1 bar abs.

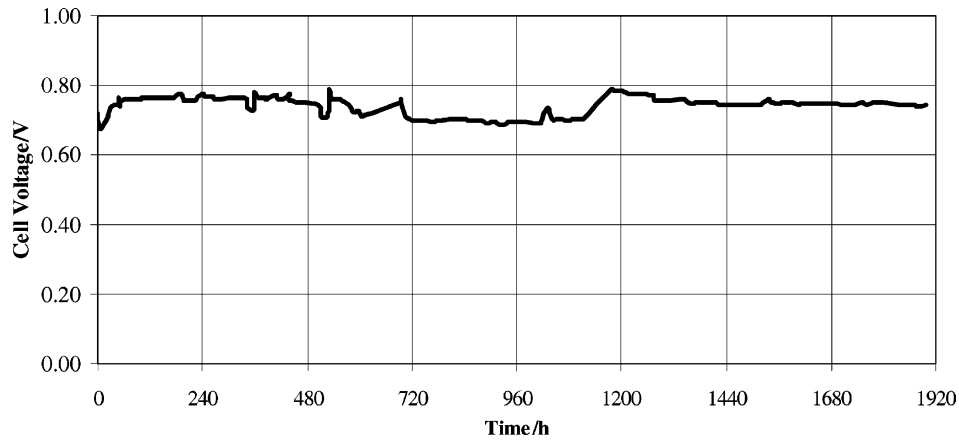


Fig. 5. Cell voltage vs. time for MEGA3 at 142 mA cm^{-2} , $T_{\text{cell}} = 70 \text{ }^\circ\text{C}$, under H_2/O_2 flux 1.1/1.1 bar abs.

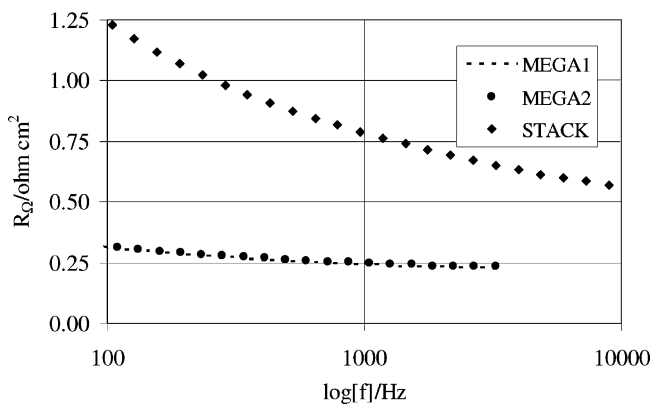


Fig. 6. Real impedance vs. frequency for MEGA1, MEGA2 and two-cell stack at OCP, $75 \text{ }^\circ\text{C}$, under H_2/air flux.

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. Best performance was obtained by applying a torque force of about $45\text{--}50 \text{ kg f cm}$ corresponding to a compaction force of $220\text{--}240 \text{ N cm}^{-2}$, which is close to the values reported by Davies et al. [5].

Fig. 7 shows the cathodic pressure loss (Δp) versus airflow plot for the three systems at operative conditions ($75 \text{ }^\circ\text{C}$ and open circuit voltage). For all systems, Δp was less than 100 mbar at high airflow, evidencing for a good design of

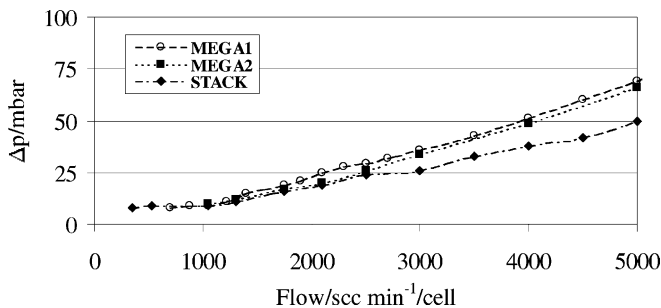


Fig. 7. Cathode pressure loss for MEGA 1, MEGA2 and two-cell stack in operative conditions.

collector plate flow field in the single cell and also in the stack.

A 20-cell stack (106 cm^2 each cell) was assembled at the ENEA laboratories with this new technology (Fig. 8). One MEGA assembly was electrochemically tested prior to its insertion in the stack in order to verify its performance stability (Fig. 9). Additionally, the stack was opened several times to control the integrity of the assembly and to test the possibility of substitution and storage. Fig. 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 the 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. However, the single MEGA assembly in Fig. 9, kept in the operative conditions of Table 1, provided a similar voltage value to MEGA1 and MEGA2 (0.59 V at 566 mA cm^{-2}).

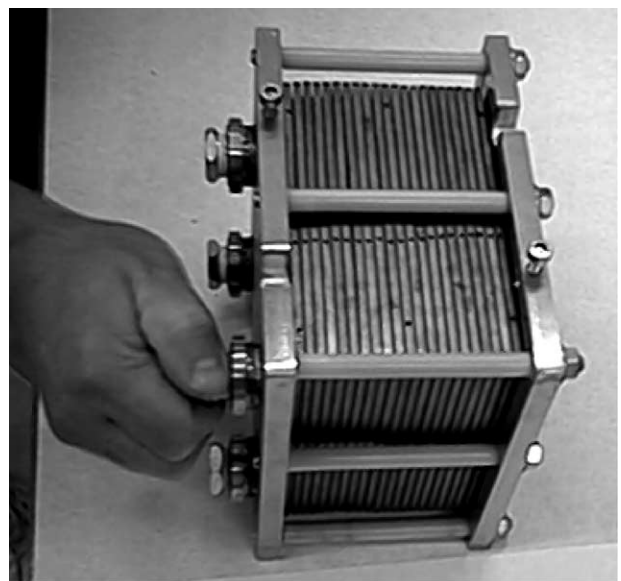


Fig. 8. ENEA/Roen Est© twenty cell (106 cm^2) stack.

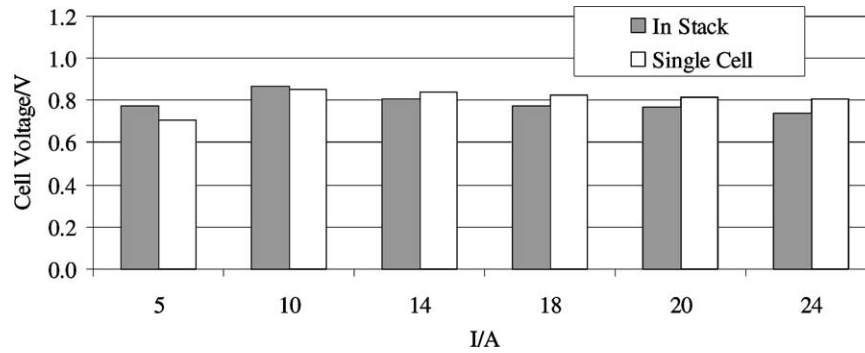


Fig. 9. Cell voltage vs. current for a MEGA in single cell configuration and in 20 cell stack: $T_{\text{cell/stack}} = 70^\circ\text{C}$, under H_2/O_2 flux 1.5/1.5 bar abs.

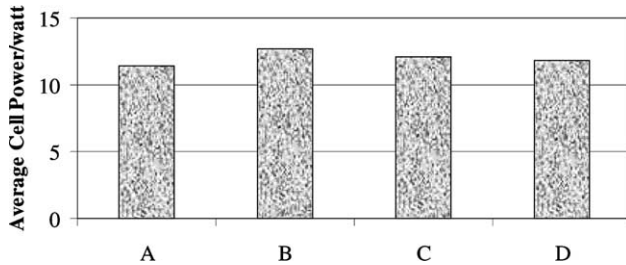


Fig. 10. Average cell power in stack after four different disassemblies: 18 A, $T_{\text{stack}} = 70^\circ\text{C}$, under H_2/O_2 flux 1.5/1.5 bar abs.

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.

4. Conclusions

The paper shows that the MEGA technology offers several advantages with respect to the traditional assembling systems:

1. Effective membrane saving of ca. 5–10% (cost reduction): the membrane is not used as a gasket support; instead, it is present only in the active area.

2. Fabrication time reduction: the MEGA assembly preparation as well as its insertion/extraction in the fuel cell stack can be considered an easy and quick process.
3. Quality control: the MEGA system can be characterised in a single fuel cell configuration and stored before utilization in a stack with maintenance of performance.
4. Failed element substitution: it is possible to disassemble the stack and replace only the failed cell. MEGA can be re-used several times without any change or decrease of performance.

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