CHEMICAL ENGINEERING TRANSACTIONS
VOL. 41, 2014
A publication of AIDIC
The Italian Association
of Chemical Engineering
www.aidic.it/cet
Guest Editors: Simonetta Palmas, Michele Mascia, Annalisa Vacca

ISBN 978-88-95608-32-7; **ISSN** 2283-9216
DOI: 10.3303/CET1441030

The Use of Additive Manufacture for Metallic Bipolar Plates in Polymer Electrolyte Fuel Cell Stacks

Richard J. Dawson*, Anant J. Patel, Allan E.W. Rennie, Simon White

Engineering Department Lancaster University, Lancaster LA1 4YR, UK

*r.dawson@lancaster.ac.uk

The bipolar plate is of critical importance to the efficient and long lasting operation of a polymer electrolyte fuel cell (PEMFC) stack. With advances in membrane electrode assembly (MEA) design greater attention has been focused on the bipolar plate and the important role it plays in performance and durability. Although carbon composite plates are a likely candidate for the mass introduction of fuel cells, it is metallic plates made from thin strip materials (typically 0.2 mm thick stainless strip) which could deliver significant advantages in terms of part cost, electrical performance and size. However, there are some disadvantages. Firstly, interfacial stability of the metal interconnect is difficult to achieve leading to migration of ions into the MEA and also an increase in contact resistance. Secondly, and the issue addressed here, is the difficultly and cost in developing new plate designs when there are very significant tooling costs associated with manufacture. The use of selective laser melting (SLM: an additive manufacturing technique) was explored to produce metallic bipolar plates for PEMFC as a route to inexpensively test several plate designs without committing to tooling. Crucial to this was proving that, electrically, bipolar plates fabricated by SLM behave similarly to those produced by conventional manufacturing techniques. This research presents the development of a small stack to compare the short term performance of metallic (316L stainless steel) plates made by machining against those made by SLM. Polarisation curves and impedance experiments were conducted. These demonstrate that the cell performance was unaffected by the manufacturing method used and that the pure resistive content of the impedance spectra, a proportion of which could be attributed to contact resistance between the MEA and plate, was very similar. It is concluded that additive manufacturing could be a very useful tool to aid the rapid development of metallic bipolar plate designs. However, when making direct comparisons with very space efficient designs, some challenges exist in the generation of very thin planar forms which would be most representative of sheet metal parts.

1. Introduction

1.1 Bipolar Plates

The three main functions of bipolar plates are:

1. to provide stable, low resistance electrical interconnection between cells;
2. to transport reactants and products to and from the MEAs allowing efficient and safe operation;
3. to aid with the thermal management of product sized stacks.

The bipolar plates most frequently used in PEMFCs can be classified into three broad types: non-porous graphite; conductive filled – polymer composites; and metallic plates. Non-porous graphite has been a traditional choice for bipolar plates as they have good electrical properties and can be machined easily on a small scale. The US Department of Energy (DoE) targets 30 USD / kW at a volume cost of 500,000 units per annum, each containing perhaps up to 100 bipolar plates, (DoE (2013)). At these volumes, non-porous graphite is prohibitively expensive. This is where polymer composites seek to provide a solution which maintains the same level of functional fulfilment as the non-porous graphite but allow scalable manufacture through known processes such as injection moulding. To this end, many researchers have investigated exact filler and polymer combinations seeking optimum performance. Hermann et al. (2005)

Please cite this article as: Dawson R., Patel A., Rennie A., White S., 2014, The use of additive manufacture for metallic bipolar plates in polymer electrolyte fuel cell stacks, Chemical Engineering Transactions, 41, 175-180 DOI: 10.3303/CET1441030

listed several combinations of polymer matrix composites with metallic or carbon based fillers, but it is carbon based plates which have received the most attention due to cost and strength issues. Kakati et al. (2010) demonstrate that a good performance plate can be made using a combination of natural graphite, carbon black and carbon fibre fillers. Although these plates could fulfil the targets, the presence of high filler content renders the moulding process difficult and the end product is susceptible to low strength and anisotropic properties. The potential advantages of pressed metal plates are outlined in Table 1.

Table 1: Basic comparison of bipolar plates which can be used for volume production of PEMFC stacks. Specific mass is based on the mass per plate area for a generic design 1.5 mm thickness plate moulded or pressed from 0.2 mm Stainless Steel.

Property / Function	Polymer – carbon composite	Metallic (316L SS)
Plate specific mass / g cm^{-2}	0.2	0.2
Bulk conductivity / S cm^{-1}	25-300	13,500
Strength	Low	High
Dimensional accuracy	Moderate	High
Post machining requirement	Likely	Unlikely
Cycle time (s)	Several	<1
Part complexity vs cost	Increasing complexity such as fineness of features increases costs through more runners, extra heated plates and increased cycle time to ensure mould is filled.	Unlike injection moulding, steel strip is not formed in a single operation but at several stations in a progression tool. Increasing complexity increases the tool cost (length) but does not increase cycle time.
Recyclability	Difficult	Easy

The major disadvantage of metallic bipolar plates which has largely stopped them being a properly competitive selection is corrosion resistance, particularly in the acidic and oxidising environment of the cathode compartment. Antunes et al. (2010) presents an extensive review of candidate steel corrosion data in acidic conditions showing that bare metal corrosion rates are significant. At the interface between the gas diffusion layer (GDL) and bipolar plate, this causes two unwanted effects. These are an increase in contact resistance due to passivating scale build up and a release of metal ions which migrate into the membrane reducing its conductivity. Progress is being made in the development of surface treatments such as electrochemical treatments to enrich the Cr and Ni content of the surface as demonstrated by Gabreab et al. (2014) and physical treatments such as plasma nitriding as demonstrated by Omrani et al. (2012) who show significant improvement in both corrosion resistance and interfacial contact resistance. A possible promising commercial solution to the problem is precoating strip steel to form a corrosion resistant coating which also has a low interfacial resistance. Commercial coating lines for continuous strip processing are available at major steel producers such as Sandvik AG and are much more cost effective than post-coating individual complex parts when volumes are significant. Care has to be taken to avoid coating damage particularly at the contact points during the pressing process, however; with careful press tool design this can be avoided. When developing a fuel cell stack, simulation can be used to guide the designs but confidence, particularly for the purposes of investment in further development, is gained through successful demonstration. To commit to a design and produce tooling for bipolar plates usually costs in excess of 200,000 €. Processes such as continuous sheet metal hydroforming as being developed by Borit BV can reduce development costs but are still expensive. This tooling cost also precludes researchers in academia from being involved directly with pressed metal bipolar plates without significant industrial input, which could limit the scope of research. In the research reported here, the use of selective laser melting (SLM, an additive manufacturing technique) is investigated to produce metallic bipolar plates for PEMFC as a route to provide inexpensive prototypes of several plate designs without committing to tooling. It is important to prove that electrically, bipolar plates made by SLM behave similarly to those made by conventional techniques.

1.2 Selective Laser Melting (SLM)

SLM is an additive manufacturing technique whereby the energy from a laser is directed to the surface of a bed of powder and thereby used to build up a three-dimensional (3D) structure in a layerwise manner. It is particularly suited for bipolar plates as 316L and other stainless steels are commonly used to produce net shape products which are 98-99% dense. Kruth et al. (2007) provide a good overview for a number of metallic AM processes. Apart from the time to build, the major disadvantage of the process is the roughness of the surface finish produced which is of a size order dependent on the powder particle size used. This can be improved by a surface remelting process as shown by Yasa et al. (2011) or also by mechanical finishing methods Morton et al. (2012). No tooling or jigging is needed to fabricate the part, but dependent on exact build configuration and geometry, support structures may be required for overhanging features. The SLM machine employed in this study was a Realizer SLM 100, using a 316L SS powder with a nominal particle size 45 μm giving an approximate layer thickness of 50 μm during the build process.

2. Stack Design

It is possible to build sub-mm plate-like structures with SLM most easily by orienting the plate end-on (standing on an edge) although this is at the expense of build time as it increases the z-height of the build envelope. However, since metal press tools and pressings are not financially viable in this study, a comparison between the performance of 316L SS milled plates of a suitable design are compared with the SLM version. The surface finish and porosity achieved in the resulting investigation is independent of the geometry and so the results of the comparison will still be valid against the overall objective. The stack design illustrated in the exploded side view of the 3D CAD model (designed using SolidWorks^{TM}) in Figure 1 is based on 5 cm^{-2} active area cell, on 25 cm^{-2} section of 50 μm thick Nafion membrane as supplied by Paxitech (Echirolles, France, product code MEA S5-3L). The catalyst loading on these MEAs is 0.5 mg Pt cm^{-2} for both anode and cathode.

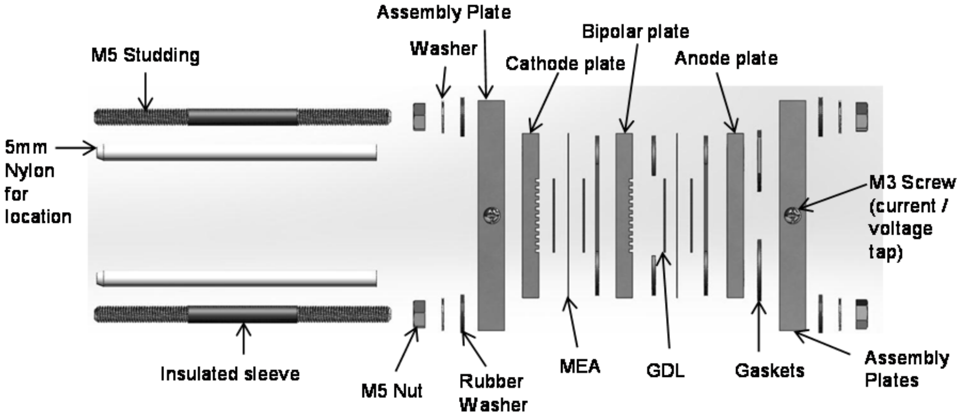
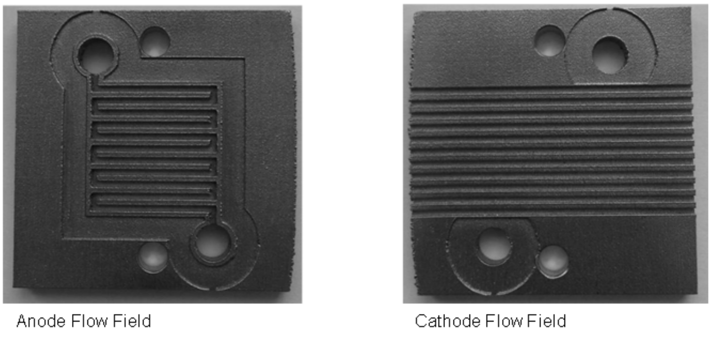


Figure 1: Side view of a 2 cell PEMFC prototype stack design in exploded view.

Carbon fibre based GDLs are pressed into the active area to aid current collection and gas distribution. Gas seals are made from peroxide cured EPDM rubber sheet. Stack compression is maintained by external mild steel compression plates which also serve as current collectors from the cathode and anode endpoles. Shorting is avoided through the use of rubber sleeved studding and rubber washers. Nylon assembly pins are included within the perimeter of the bipolar plate design to provide repeatable location for the main stack components, though gasket placement and compression is assured through the gasket rebate in the design. The features of the anode and cathode plates can be seen in Figure 2.


Anode Flow Field

Cathode Flow Field

Figure 2: Images of anode and cathode side of the bipolar plate made by SLM.

After considering various designs compatible with the overall cell size, a single serpentine with channel section 1 mm square was selected. Even gas were predicted for both cells in the stack using Ansys Fluent^{TM} computational fluid dynamics (CFD) which also demonstrated that there should not be any hydrogen maldistribution between the two cells even when both hydrogen flow and return were through the same compression plate. Ignoring consumption, the CFD predicted that a pressure drop of 11.2 mbar would be experienced across the plates at a flow rate of 21 ml min^{-1} with an imperceptible drop in the larger headers. The milling operation to produce the machined versions of the plates largely determined the overall plate thickness (5 mm) and the achievable channel size (1 mm).

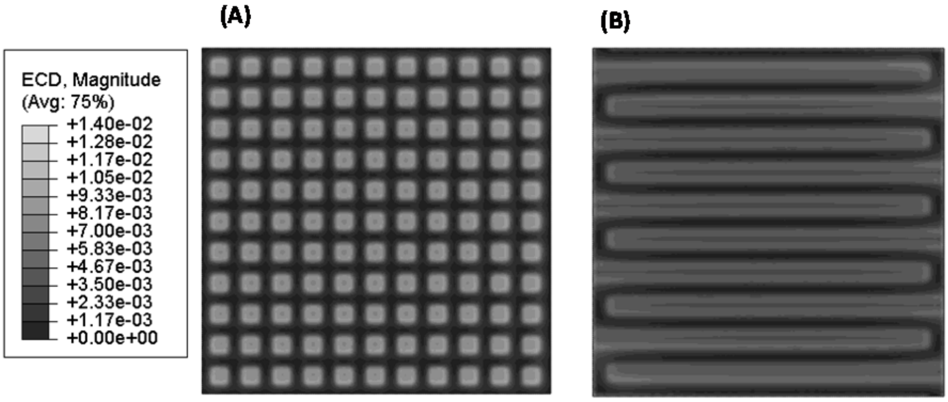


Figure3: Current density distributions in A mm^{-2} through the GDLs as predicted by Abaqus^{TM} Finite Element Analysis (FEA) for a uniform cell current density of 250 mA cm^{-2} and a 316L SS current collector material for two designs. (A) dimpled current collection and (B) serpentine ribbed current collection.

The expected current collection losses were simulated using Abaqus^{TM} FEA software assuming a uniform cell current density and an idealised GDL. The GDL used was an Optiveil (Technical Fibre Products Ltd, Kendal, UK) carbon fibre laminate at 20 g m^{-2} sheet weight. This was treated with 3 %wt PTFE suspension and into which a particulate carbon-based layer (50 %wt carbon black, 12.5 %wt activated carbon and 37.5 %wt PTFE) was calendared giving an overall layer thickness of approximately 0.4 mm. This was modeled as a homogenous layer of 0.4 mm with conductivity of 813 S m^{-1} based on the sheet resistivity value quoted for the Optiveil product, although in practice the presence of the carbon particles are likely to enhance this. An interfacial contact resistance of 250 mΩ cm^2 was taken between the 316L SS current collection features and the GDL. This was based on the work of Gabreab et al. (2014) for untreated 316L SS. The local current densities which dictate local resistive losses are shown in Figure 3 for the serpentine design built and another dimple based design. The difference in the losses between the two designs is particularly clear when the resistive heating losses integrated for the whole current collector are considered. This gives an area specific resistance (ASR) contribution of 0.47 Ω cm^2 for the serpentine design versus 0.96 Ω cm^2 for the dimple design. For the air side rib current collection, the contribution is predicted to be around 0.46 Ω cm^2. These cases are dominated by the ratio of the contact area difference for the designs since this affects both the losses at the interface but more significantly the mean current path length through the comparatively resistive GDL. If the DoE targets of 0.02 Ω cm^2 for the interfacial losses (DoE (2013)) are to be achieved by 2017, then coatings or surface preparations such as those described by Gabreab et al. (2014) which can achieve more than this target, will be required if low cost base steels such as 316 are to be used.

3. Experimental Methods

The experimental bipolar plates were manufactured and surfaces carefully cleaned (SLM plates were wire brushed to remove loose powder particles and any high spots removed by emery paper). Before each experiment, the plates were gently sonicated in a small volume of acetone for 5 minutes to remove particles and any organics, thoroughly rinsed in 18 MΩ ultra-pure water (Millipore) and sonicated for a further 5 minutes. Plates were then dried for at least 20 minutes in a recirculating lab oven at 50 ºC before assembly. The MEA and GDL combination was prepared by lamination at room temperature at a pressure of 2.4 MPa after the holes for the location pins had been carefully cut. The cells were then clamped together with a consistent torque of 3 Nm applied to each nut leading to a consistent and uniform contact pressure. The rig was then connected to a hydrogen supply flowing an excess of humid hydrogen from a

PEM water electrolysis unit (35 sccm). For all experiments, a small sized fan from a computer CPU was used with a specially manufactured plenum and manifold to direct a consistent volume flow rate of air across the cathode side. Electrochemical measurements were made using a PGstat 30 potentiostat equipped with a frequency response analyser module (Metrohm, Autolab BV). A stable open circuit potential was obtained each time and the cells then stabilised running at a constant current of 200 mA for 5 minutes. After this, a polarisation curve was run at a current scan rate of 2 mA s^{-1} down to a current of 1 A. Two polarisation curves were conducted since no easily discernible change was observed between the second and subsequent scans. The cell was then returned to a constant load of 500 mA and allowed to stabilise before electrochemical impedance spectroscopy (EIS) measurements were taken at current point of 500 mA with a perturbation of 10 mA. This ensured that the cell was running on the linear part of the polarisation curve, although linearity of response with perturbation depth was not specifically assessed.

4. Results and Discussion

Two single cell experiments were conducted for machined plates and SLM plates using fresh MEAs each time. At the end of the single cell tests, a two cell stack was constructed from the SLM plates using the previously run MEAs. Two holes were cut in cell number 1 to allow hydrogen to flow through to cell number 2. In all experiments, to reduce the likelihood of electrical shorting across the stack, current collection was via Ni strips direct onto the flow plates rather than from the compression plates. Voltage monitoring was conducted directly on the flow plates with separate wire connections to remove contributions from unwanted series resistances. Figure 4 shows the polarisation curves and Nyquist plots for the cell and stack that were obtained.

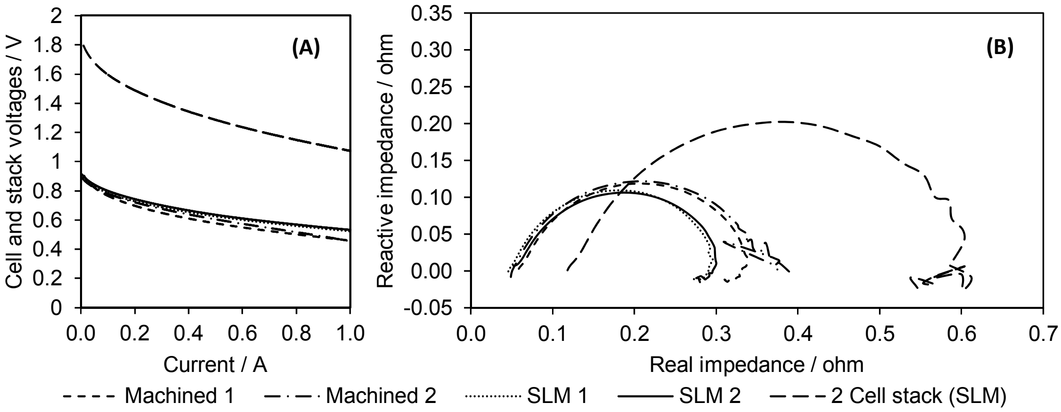


Figure 4: (A) Polarisation curves for single cells using both machined plates and SLM plates and for a 2 cell stack made of SLM plates. Current scan rate at 2 mA s^{-1}. Curves shown are for second measurement taken. (B) Nyquist plot of cell and stack impedance from 10 kHz to 0.1 Hz taken at current point of 500 mA with a perturbation amplitude of 10 mA. Fuel flow rates were 35 sccm in all cases.

Good consistency between the measurements was achieved. The plates fabricated using SLM appear to perform as well as those made by machining with a very similar pure resistive content to the impedance indicating that the contact resistance achieved was very similar in each case. The single result for the two cell stack also shows twice the cell voltage and approximately twice the impedance giving confidence that the small stack design is working well without excessive problems with sealing or contact. The comparatively low fuel flow rate (still around 5 times excess at 1 A) which was the same for both the single cell and 2 cell stack experiments did not appear to cause any significant fuel starvation problems which would have been expected to be aggravated when running two cells. When considering the contact and GDL series resistances predicted ASR contibutions for a single cell, 0.93 Ω cm^2, the experimental results are considerably better than expected giving a purely resistive ASR content of ca.0.26 Ω cm^2 each time. This will be largely due to the improved GDL conductivity from the addition of carbon black and the improved conductivity of the GDL under compression and it is also likely that the freshly cleaned contact surface has considerably less resistance contribution than assumed. Looking particularly at Figure 4B, it is apparent that the performance of the cells when using the SLM plates was slightly enhanced, evident in the smaller size of the arc. Given the standard procedure used each time and assuming that the cells supplied and assembled with GDL are consistent, as the results would indicate, then it is likely that the subtle differences in the plates have had an effect on reactant transport. The rough surface of the SLM plates, which tend to give a slight corrugation across the rib since the plate is built in layers, will produce an easier reactant diffusion path under the contact ribs when the plates are assembled into the cell. This is clearly shown in Figure 5. There could also be an affect due to the rough channels causing turbulence and so greater transport rates, however, Reynolds numbers are low (<10) so this is less likely.

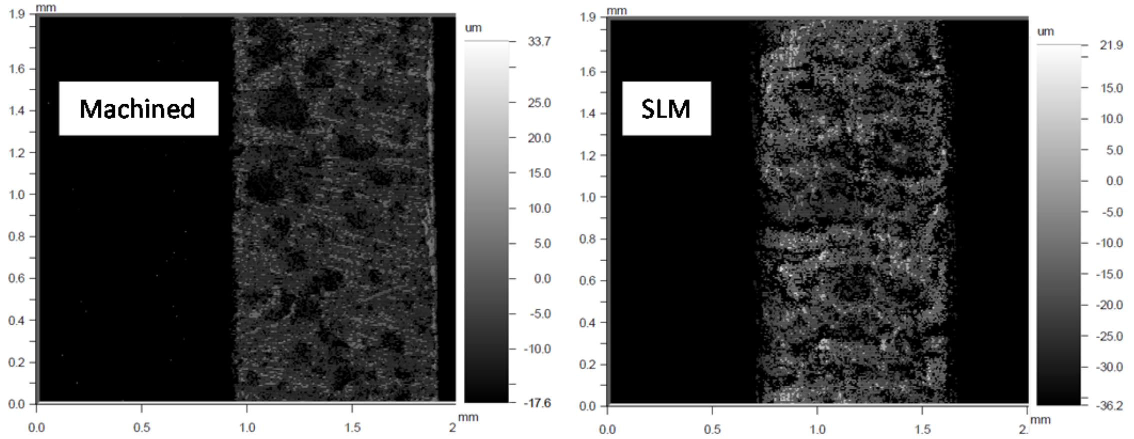


Figure 5: White light interferometer (Veeco Wyko NT1100) images for surface roughness for machined and SLM plates showing a section across an air side contact. Ra values are 2.3 μm and 4.9 μm respectively.

5. Conclusions

The experimental results reported here demonstrate the feasibility of using SLM for the production of metallic bipolar plates for PEM fuel cells. The contact resistance and performance are shown to be the same as or slightly better than experienced when using plates of the same design but manufactured by a conventional machining process. The use of SLM and other additive manufacturing techniques will allow different designs to be fabricated quickly with no tooling cost to give representative cell and stack performance. Further work will be required to investigate the use of thinner light-weight designs representative of processes such as metal pressing.

Acknowledgements

The authors would like to thank the technical staff at Lancaster University, The Lloyds Register lab for loan of equipment and particularly Hassan Abou Ghazala for manufacturing the GDLs.

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