**Title: An analysis of the combined bipolar plate concept: Gas diffusion layer soldered to a bipolar plate using metallic tin, for use in Proton Exchange Membrane fuel cells**

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**Abstract**

A novel investigation was performed into the electrodeposition of metallic tin onto a stainless steel bipolar plate for use as a combined BPP/GDL in PEM Fuel Cells. The process of plating the tin onto a SS316L bipolar plate was optimised to the following specifications: A tin layer of thickness 30 µm was deposited onto the pre-cleaned BPP, before hot pressing with a pre-cut GDL at 230 oC for 20 minutes at a pressure of 0.5 bar, and cooling slowly to room temperature. This procedure obtained the lowest contact resistance of 5.45 mΩ cm2 at 140 N cm-2, and was tested both in-situ, and ex-situ, to simulate conditions over the lifetime of the PEMFC. The in-situ testing produced excellent results, with a minor increase in ex-situ contact resistance from 8.8 to 9.2 mΩ cm2 and a decrease in cell voltage from 0.714 to 0.667 V over the course of 200 hours. Therefore, the combined Sn/GDL bipolar plate concept is a promising low-cost alternative for use in PEMFCs.

**1. Introduction**

In recent years, a greater demand for substitutes to fossil fuels has prompted the development of Proton Exchange Membrane (PEM) Fuel Cells. PEMFCs provide a portable energy source, and are a viable alternative to petrol and diesel for use in the automobile industry1. A number of large companies, including Toyota2, Honda3 and Hyundai4 have set up long term plans to develop fuel cell technology for this industry, however a number of problems have to be overcome before a reliable alternative to oil and gas is developed. These problems include intrinsic limitations concerning durability, longevity and costs, particularly with the bipolar plate (BPP), which contributes 11-45% of the cost5 and 45-80% of the stack weight6 of the PEMFC.

The bipolar plates (BPPs) act as the anode and cathode for the cell, allow even distribution and separation of the fuel gasses, and facilitate removal of the waste products7. They also provide support for the stack and manage heat produced during the fuel cell reactions6, 8. As they have many functions within the cell, the BPP must meet a strict series of criteria. The plates must be lightweight, have high electrical conductivity and low interfacial contact resistance to minimise ohmic losses9, as well as being mechanically supportive10. They must also be thermally conductive to remove excess heat, and impermeable to the reactant gasses to minimise fuel crossover11.

In order to obtain these properties, a number of different materials and coatings for the production of BPPs have been investigated. Due to the inherent limitations of graphite12 and other carbon based BPPs6, metal plates are now considered the most promising candidates, with stainless steel found to be more suitable than titanium13, nickel 14 or aluminium 15 alloy plates. Stainless Steel has a significantly lower manufacturing cost and a higher strength than graphite16, as well as a high electrical conductivity and low gas permeability. The main drawback of a metallic BPP is its struggle to maintain both low contact resistance and high corrosion resistance inside the fuel cell environment. When undergoing corrosion processes, the stainless steel releases metal ions that could lead to poisoning of the membrane and catalyst 6. Another issue is the formation of a non-conductive chromium oxide (Cr2O3) layer on the surface of the plate upon contact with air 17. In order to limit the formation of such oxides and ions, a series of protective coatings have been developed for the stainless steel BPPs. A large amount of research has been done into different coating materials, including carbon based coatings18–20, metal nitrides21–23, carbides24–26 and noble metals27,28, many of which have improved the corrosion resistance and the ICR of the BPP to the standards set by the US Department of Energy29. However, the coating methods for many of these BPPs, such as physical vapour deposition (PVD) or arc ion plating, are costly, so if stainless steel bipolar plates are to become commercially viable, a cheaper alternative must be found.

Tin is widely used as a solder in the electronics industry due to its high electrical conductivity30, good wettability and reliability31. It is also easy to produce a layer of tin by electrodeposition, a technique that is popular due to its low cost, simplicity and good controllability of coating morphology and thickness.32,33

As similar conditions need to be met for fuel cell applications, it can be considered a good candidate for further research. Tin has also been proven to improve the corrosion properties of stainless steels through the formation of a dense SnO2 passive layer, which inhibits further corrosion 34–36. Despite this, Sn has been thought unsuitable for PEM applications due to the potential poisoning of the membrane by Sn2+ ions. This has proven to not be the case by X et al, who reported no negative effect on membrane performance when using tin as a catalyst carrier in PEM FCs.

A novel concept is that metallic tin will be electroplated onto a stainless steel bipolar plate in a uniform layer of a known thickness, in order to protect it from the corrosive environment of a fuel cell as well as significantly improving the electronic conductivity. In this case, tin is deposited onto the bipolar plate before hot pressing with a pre-cut gas diffusion layer (GDL) at a temperature around the melting point of tin. This process softens the tin, and the addition of pressure forces it into the pores of the GDL. It is soldered to the carbon fibres once solidified, and produces a conduction pathway from the BPP through to the GDL.

When exposed to a PEM environment, including slightly acidic conditions and voltages up to 1.4 VSHE during start up and shut down37, the outer layer of the deposited tin will oxidise to form a SnO2, layer38, as indicated by the Pourbaix diagram39. This passive layer remains conductive so will protect the underlying tin and stainless steel substrate from further oxidation whilst maintaining a good conductivity and low contact resistance for the BPP. This concept is demonstrated in Figure 1.

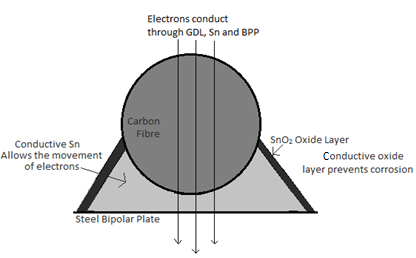


Figure . The combined Sn/GDL concept

In this work, we present a new and novel method for combing a stainless steel bipolar plate and a carbon based gas diffusion layer using tin electrodeposition followed by hot pressing at temperatures close to the melting point of tin. This method produces improved through-plane cell conductivity, yielding extremely low contact resistances.

The process and quality of the joined BPP/GDL material are studied with respect to conductivity and durability through a series of measurements, including interfacial contact resistance (ICR), ex-situ chronoamperometry and electron microscope imaging with elemental analysis. Finally, long-term in-situ testing in a simulated fuel cell environment is performed. The produced plates show good adhesion and low contact resistance, even after being exposed to a simulated fuel cell environment. The method described in this work brings a move towards simple, yet reliable, coating methods for BPPs in PEM systems.

**2. Experimental**

AISI 316L bipolar plates were used as received from Elring Klinger for all tests. The total surface area of the plates was 39.2 cm2, with a land area of 6.2 cm2, for ICR measurements. Potentiostatic and ICR measurements were recorded ex-situ, as well as Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS) analysis using a Hitatchi S-3400N. In-situ analysis of the plates was also performed using a Greenlight G-40 Fuel Cell Test Station, with a Gamry Reference 3000 Potentiostat.

## 2.1 Construction of combined Bipolar Plate with GDL

Prior to all experimental work, the stamped SS316L BPP was etched in HCl (12.8 wt. %, 3.47 M, room temperature, 15 minutes) to remove any oxide layer, followed by rinsing in deionised water and drying under nitrogen at room temperature. A coating of Micro super XP 2000 stop-off lacquer was then applied to the backside of the BPP with a paintbrush. The electroplating solution was prepared as follows:

Tin(II)Sulfate (36.17 g, purity ≥ 95%, Sigma Aldrich) was dissolved in type 2 distilled water (700 ml) produced by a Milli-Q integral system. H2SO4 (98 %, 100 ml) was added dropwise, and left to cool. SLOTOTIN 71™ (Schlötter, 20 ml) and SLOTOTIN 72™ (Schlötter, 3 ml) were added and the mixture topped up to 1 L with distilled water. The stainless steel bipolar plate was immersed in the electroplating bath and connected to the negative pole of the current source. A pair of tin electrodes were used as the counter electrode and a current density of 1.5 A dm-3 was applied to obtain a deposition rate of 0.7 µm min-1.

After removal of the plate from the solution, it was cleaned with distilled water and the lacquer removed from the back side. It was placed on top of a pre-cut GDL in a Teflon envelope, and hot-pressed at a set pressure, temperature and time before cooling slowly to room temperature under pressure. This process was repeated for three plates at each set of conditions.

ICR and electrochemical tests were then performed.

## 2.2. Interfacial Contact Resistance Measurements

ICR measurements were taken before and after each electrochemical test using a setup adapted from H. Wang et al 40. The sample was placed between two gold-coated copper conducting plates, and a current of 2 A was passed between the bottom plate and the top plate. The voltage between a spring-loaded gold pin and the top plate, through the sample, was recorded as the compaction force was increased from 70 to 650 N cm-2. All quoted values for ICR are recorded at 140 N cm-2, which is the standard in-situ compaction pressure 40. It was assumed that the bulk resistance of the gold and copper plates, as well as the bulk resistance of the SS316L BPP and GDL were negligible.

## 2.3 Ex-situ Electrochemical Testing

The specimens were evaluated for their corrosion behaviour using potentiostatic measurements taken by a Gamry Ref 600 potentiostat. The combined BPP/GDL was used as the working electrode, and was submerged in Na2SO4 (0.1M) /H2SO4 pH 5.5 at 80 oC along with a Pt mesh counter electrode. Before testing, electrolyte was injected into the channels of the bipolar plate with a syringe. The reference electrode (Hg/Hg2SO4/K2SO4Sat, 0.64 VSHE) was connected to the electrolyte via a salt bridge. The electrolyte was heated to 80 oC and purged with nitrogen prior to testing.

For potentiostatic measurements, the samples were first stabilised at open circuit potential before applying a voltage of 1.4 VSHE for 1 hour.

## 2.4 In-situ Fuel Cell Testing

A Greenlight G-40 Fuel Cell Test Station equipped with a Gamry Reference 3000 potentiostat was used for in-situ fuel cell measurements. The cell had an active area of 15 cm2. A GORE® PRIMEA® Membrane Electrode Assembly (MEA) of thickness 15 µm, Pt content of 0.1 mg cm-2 at the anode and 0.4 mg cm-2 at the cathode, was used. H23C641 GDL (Freudenberg FFCT) was used throughout this study.

The fuel cell was constructed with two of the combined BPP and GDL plates, as an anode and a cathode, being placed with a MEA in between two gold current collectors, and then clamped together. The clamping pressure was piston-regulated and separated from the sealing pressure at 200 kPa over the area of the bipolar plate, equivalent to 140 N cm-2. The cell was operated at 70 oC, with synthetic air supplied in excess at the cathode (AGA 99.999 %, stoichiometry 5) and hydrogen in excess at the anode (AGA 99.999 %, stoichiometry 3). The excess of gas ensured a stable performance and minimum variation in reactant concentration throughout the entire length of the bipolar plate. Back pressures of 20 kPa and 30 kPa were applied to the anode and cathode, respectively.

After the assembly of the cell, the test procedure was initiated. The catalytic layers and membrane interactions were activated before a cycling procedure of high and low current densities (1.2 and 0.5 Acm-2), interspaced with shut-downs and start-ups, as seen in Figure 2, was performed for 150 hours.



Figure The cycling procedure during in-situ testing

After completion of the initial 150 hours test procedure, the cell was shut-down and left at open circuit overnight before the test was resumed for another 50 hours.

Over the course of the in-situ testing, impedance measurements were taken at the high current density, over a frequency range of 10 kHz to 100 mHz, and a single data point at 1 kHz was used for the High Frequency Resistance (HFR). Cyclic voltammetry of the fuel cell was conducted with H2 on the anode and N2 on the cathode to provide an estimate of the change in catalytic surface area.

The same procedure was implemented for two control plates, stainless steel coated Galvanic gold and stainless steel coated TiN+C, provided by Elring Klinger.

# 3. Results & Discussion

## 3.1 Optimisation of procedure: Combining BPP and GDL with Sn

The procedure for combining the metallic plate with the GDL through Sn impregnation via hot pressing was optimized firstly with respect to lowest possible ICR and then to highest corrosion resistance. The effects on interfacial contact resistance and corrosion rate of various preparation parameters such as hot press pressure, temperature and time, as well as deposition charge (coating thickness) and cooling method, were investigated. Before the Sn/GDL plates could be tested in-situ, each of these parameters was optimised to increase performance.

### 3.1.1 Hot Pressing

When hot pressing, the pressure was set at 140 Ncm-2, equivalent to the in-situ pressure. Increasing the pressure could cause flattening of the BPP channels, compromising the structure. Blocking of the channels by the GDL could also occur, reducing the cell performance by hindering the transport of reactants into and waste products out of the cell. Lower pressures reduce adhesion between the GDL and BPP as permeation of the tin into the GDL is lessened, reducing the length of the conduction pathway and the conductivity.

Temperatures around the melting point of tin (231.9 oC 42) were selected to investigate how the degree of melting of tin could produce a best possible electronic contact between the metallic plate and the carbon based GDL. Table 1 shows the ICR values recorded after pressing at temperatures between 226 and 232 oC. At temperatures above 230 oC, the tin becomes ductile enough to penetrate the GDL more sufficiently, ensuring an increased contact area and a better pathway for current, lowering the average ICR from 8.0 mΩ cm2 at 228 oC down to 5.4 mΩ cm2 at 230 oC. In addition to an improved through-plane electronic conductivity, the plates prepared with a hot press temperature of 230 oC offered an improved adhesion due to the infiltration of GDL fibres into the tin, something that did not occur at temperatures lower than 230 oC. For this reason, a temperature of 230 oC was used for all further plates.

Table Contact resistance (mΩ cm2) at various hot press times and temperatures

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Press Temp (oC)  Press time  (min) | 226 | 228 | 230 | 232 |
| 1 | 7.9 | 6.5 | 6.6 | 5.4 |
| 10 | 8.3 | 8.4 | 5.4 | 7.6 |
| 60 | 6.0 | 9.2 | 4.4 | 5.6 |

The time required for hot pressing was also investigated. Varying the hot-press time from 1 to 60 minutes produced variations in contact resistance with no clear pattern, as seen in Table 1. The best press time varied with temperature, so 20 minutes was chosen as a suitable time to allow the temperature to equalise after the initial temperature drop from the addition of the cool bipolar plate.

### 3.1.4 Thickness of Tin Layer

The thickness of the tin coating was also optimised. This layer must be thick enough to allow the tin to permeate into the GDL and form a good contact. However, if it is too thick, the channels of the BPP can become blocked, hindering the transport of reactants and waste products. Tin was deposited in a series of thicknesses from 10 to 60 μm, and the corrosion resistance was tested in the ex-situ setup, the results in Figure 3.

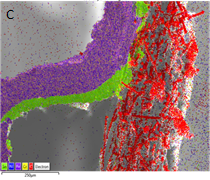
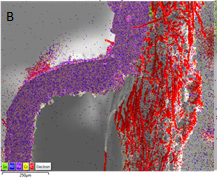
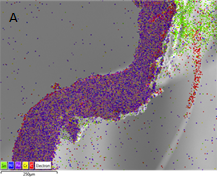
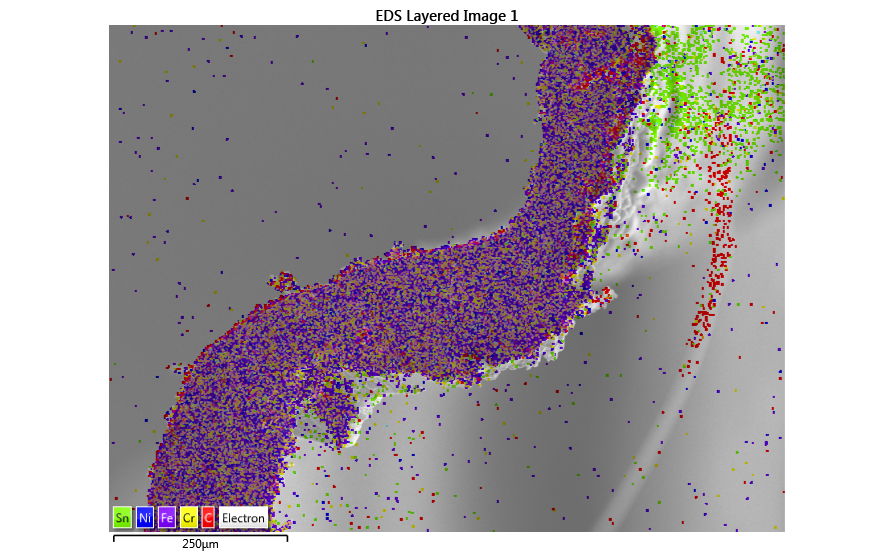
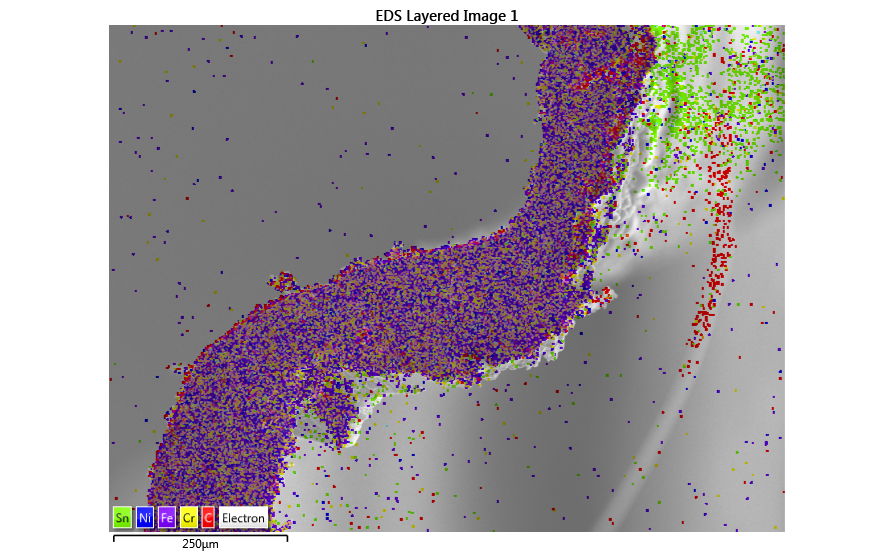
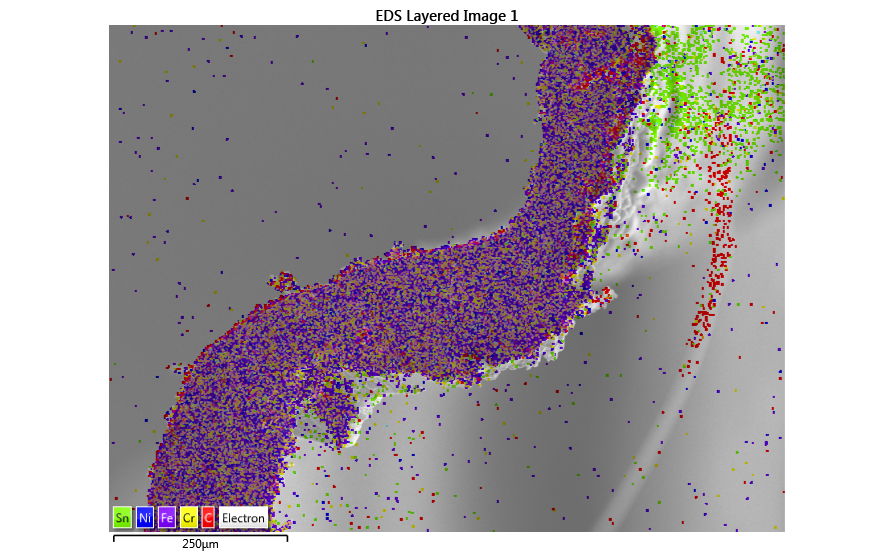
Before corrosion, all ICR values were lower than DoE requirements, and decreased with an increasing plating thickness, due to greater permeation of the tin into the GDL and increased electrical contact between the layers. The bulk resistance of tin is low (1.09 x10-7 Ω m 43), so a thicker layer does not impact negatively on the conductivity.

Initially, the contact resistance measured was lower than values obtained from gold and stainless steel plates using the same equipment. This is the case as hot-pressing the BPP with the GDL produces a better conduction pathway than simply placing the GDL on top of the BPP.

Figure Comparison of coating thickness against ICR before and after ex-situ testing

After corrosion testing, the ICR of the thinner coatings increased vastly, by up to 560 %. This is because approximately 20 µm of tin was corroded away from the BPP, as is observed in the SEM/EDS images in Figure 4. The Sn is oxidised to SnO2, which delaminates from the Sn and falls off the plate into solution instead of protecting the underlying Sn, as hypothesised.

After ex-situ testing, the deposited tin layer in Figure 4 A has reduced from a thickness of 10 µm to being non-existent. In the EDS images, green is Sn, the purple colour represents the SS316L BPP, and the red is the GDL, which is also not present in Figure 4 A. The removal of Sn during corrosion testing reduced adhesion between the GDL and BPP enough that the GDL entirely delaminated from the plate. From Figure 4 B, there is also very little of the tin layer remaining, but just enough to continue to adhere the GDL to the BPP.



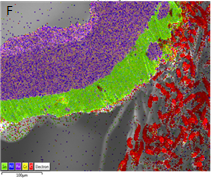
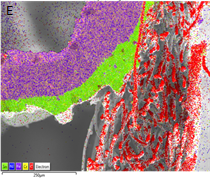
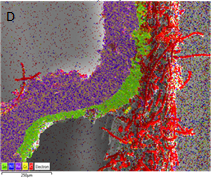
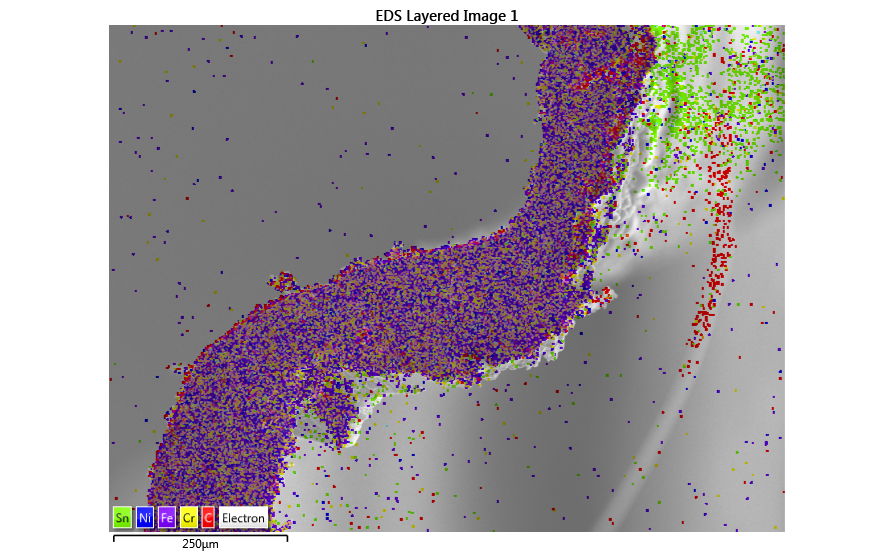
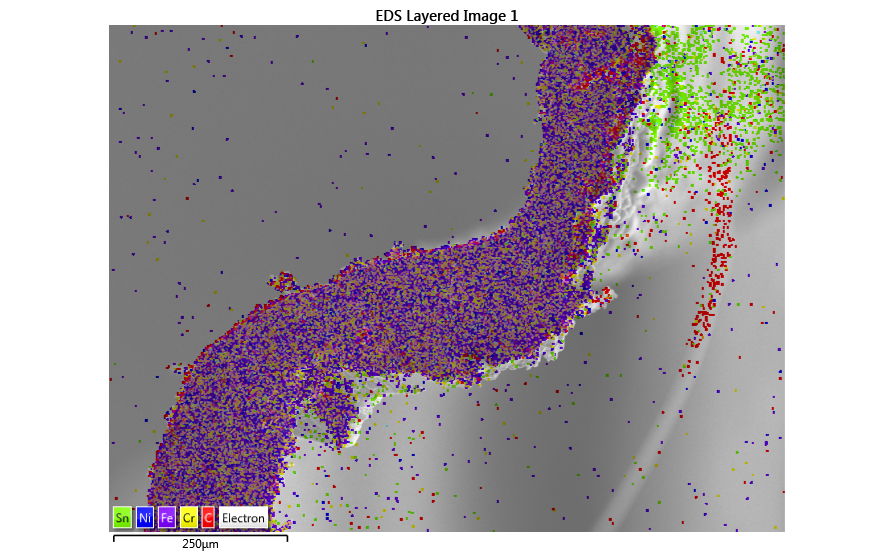
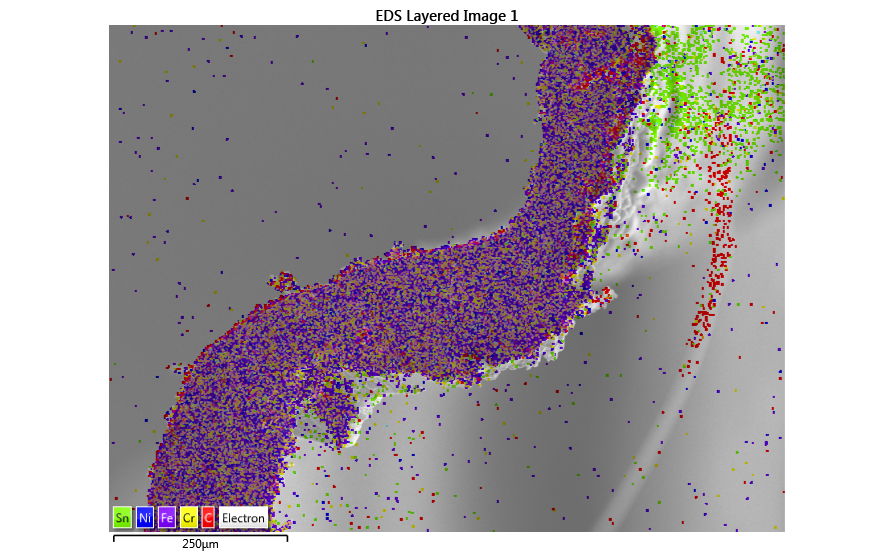


Figure A-F. EDS images of BPPs & GDL with varying thickness of tin after ex-situ testing, purple representing the bipolar plate, green the tin and red the GDL. The thickness of deposits were initially 10, 20, 30, 40, 50 & 60 µm on figure A to F, respectively

As the thickness of the tin layer increases, in Figure 4 C, D, E and F, there is more tin left after corrosion and so more adhesion. The ICR of the thickest coatings, 50 and 60 µm, also increased by a large amount, up to 570 %, after polarization to 1.4 V for 1 h. This is possibly due to the larger volume of SnO2 formed during the corrosion process. SnO2 has a lower conductivity than metallic Sn42, contributing to the observed increase in ICR. Additionally, the SnO2 produced after testing has a larger volume than Sn, so falls off the surface of the Sn and causes delamination between the remaining Sn layer and the GDL. This excess SnO2 is not visible in the EDS images, as it is removed by the washing process after ex-situ electrochemical testing.

From the series of electrochemical tests, the best performing bipolar plate was found to have a tin layer thickness of around 30 µm, which was then used for all subsequent plates. From the images in Figure 4 (C and D), it is obvious that the tin has permeated into the GDL during the hot-pressing stage, so despite the loss of tin that took place during the high voltage procedure, there is still a good conduction pathway and the ICR remains low. The ICR increased by around 260 % after corrosion testing. Despite this being less than for the other thicknesses, all of which were more resistant than bare SS316, there is still a necessity to improve the deposition process or stabilise the SnO2 layer that is formed after exposure to the PEM environment.

### 3.1.5 Cooling Method

Initially, after hot-pressing, the plates were immediately removed from compression and left to cool rapidly to room temperature. After corrosion testing, this gave a large and undesirable increase in ICR, so in an effort to improve this as well as the adhesion between the GDL and tin, the plates were cooled whilst still under pressure. This slows down the cooling process, increasing grain size and producing fewer grain boundaries and therefore fewer active sites for corrosion 44. It also forces the Sn to stay in contact with the GDL whilst solidifying.

The plates were left in the hot press to cool to room temperature, before being removed. The ICRs for both cooling methods were comparable before corrosion, at 6.8 and 7.1 mΩ cm2 at 140 N cm-2 (Figure 5), but after polarisation to 1.4 V for 1 hour, a significantly improved ICR was observed for the slowly cooled plate, 13.2 compared to 17.9 mΩ cm2 at 140 N cm-2 when the plate was rapidly cooled.

Figure Comparison of the quickly vs slowly cooled plates before and after corrosion testing

The plate that was cooled slowly provided a significantly more corrosion resistant coating than the rapidly cooled one. Although the increase in ICR after testing is still significant, it shows the importance of precision control processing parameters. The slower cooling procedure was used in the production of all further plates.

## 3.2 In-situ Analysis

An in-situ study of the optimised Sn/GDL BPP was performed and compared to identical tests on Au and TiN+C coated SS316 BPPs. Both before and after the 200 h cycling procedure, a full series of standard measurements including catalytic surface area (ECSA), H-crossover and high frequency resistance (HFR) were undertaken to ensure normal operation of the cell. Minimal differences were seen between the Sn/GDL and control plates.

During cycling, the cell voltage and HFR were monitored. A visible drop in cell voltage and an increase in high frequency resistance, as seen in Figure 6, occurs due to the degradation of the membrane and a reduction in catalytic surface area. In addition, on the bipolar plate, some of the tin coating is converted into tin oxide, and removed with the waste water in the cell. This in turn reduces the conductivity of the coating and the number of electrons that can be transferred across the membrane to be converted to current.



Figure In-situ performance of BPPs over 200 hours

Looking at the decrease in cell voltage over time provides an insight into the longevity of the cell for real operating conditions. As seen in Figure 6, all plates perform well over the 200 hour test. The voltage drops are minimal, with the average drop in cell voltage over 200 hours being around 0.06 V. The overnight shutdown at 150 h simulates real-world operation and provides a regeneration in the voltage. This is because the cell is allowed to cool and all waste is expelled from the cell, increasing the effectiveness of the membrane and the transport of electrons.

Comparing the performance of the three bipolar plates, the BPP that ended up with the smallest drop in cell voltage was the Sn/GDL BPP, with a drop from 0.714 to 0.667V, indicating that it would have the longest lifetime in the cell. More long term testing is needed to confirm this.

After testing for 200 hours, the plates were removed from the cell and their ICR was tested ex-situ, as seen in Table 2.

Table Contact resistance measurements before and after in-situ testing

|  |  |  |  |
| --- | --- | --- | --- |
| Plate | Ex-Situ ICR before testing (mOhm cm2) | Ex-Situ ICR after testing - Anode (mOhm cm2) | Ex-Situ ICR after testing - Cathode (mOhm cm2) |
| Au | 7.7 | 7.2 | 7.4 |
| TiN+C | 8.4 | 9.3 | 10.3 |
| Sn/GDL | 8.8 | 9.2 | 19.7 |

The Sn/GDL combined plate performed well when compared with the control plates, with a very small increase in ICR being observed at the anode side. However, a significant increase was observed on the cathode side of the cell. This indicates some instability in the coating, as seen from the SEM images. Instead of producing a stable oxide, the Sn flakes off the plate and is washed from the cell, which could produce contamination issues downstream. Therefore, work must to be done to stabilise the tin and produce a coating with a longer lifetime in-situ.

# 4. Conclusions

An optimised electrodeposition process was successfully used to produce a combined Sn-GDL protective layer on an SS316L bipolar plate. Very low ICR values were obtained before ex-situ electrochemical testing, due to the uninhibited conduction pathways connecting the GDL and BPP through the soldered tin. If maintained in-situ, these pathways will provide easy conduction through the lifetime of the cell. However, the ICR increased after corrosion testing due to the instability of SnO2 on the surface of the coating leading to the breakdown of the conduction pathways.

The optimised conditions for the deposition of tin onto the SS316L BPP are as follows: A plating thickness of 30 µm was deposited onto the pre-cleaned BPP, before hot pressing with a pre-cut GDL at 230 oC for 20 minutes at a pressure of 0.5 bar, and cooling slowly to room temperature. This procedure obtained the lowest contact resistance of 6.5 mΩ cm2 at 140 Ncm-2, well below U.S DoE targets, and was tested both ex-situ and in-situ. The contact resistance increased to 13.2 mΩ cm2 at 140 Ncm-2 after ex-situ testing in Na2SO4 at pH 5.5 and 80 oC, with 1.4 VSHE applied for 1 h. In-situ testing produced results equivalent to high-cost PVD coatings, with interfacial contact resistance values increasing from 8.8 to 9.2 mΩ cm2 on the anode side and 19.7 mΩ cm2 on the cathode side, and the cell voltage decreasing from 0.714 to 0.667 V over the course of 200 hours.

Future work will focus on enhancing the stability of the SnO2 oxide layer, and introducing alloying elements to optimise performance. More long term in-situ testing will also be done.

**Acknowledgements**

The research leading to these results has received funding from the European Union's Seventh Framework Programme (FP7/2007-2013) for the Fuel Cells and Hydrogen Joint Technology Initiative under grant agreement n° 303449 10.

KM thanks the Department of Materials Science and Engineering at NTNU for the award of a scholarship.

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