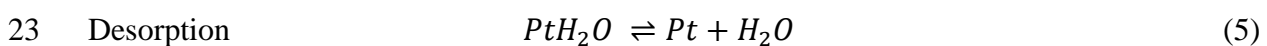
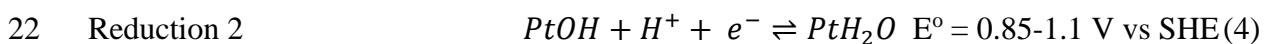
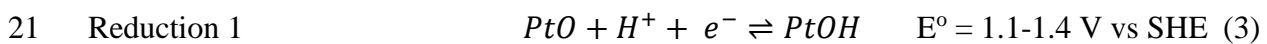
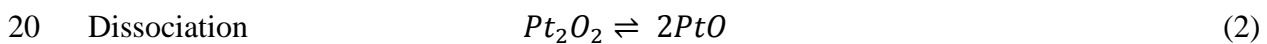
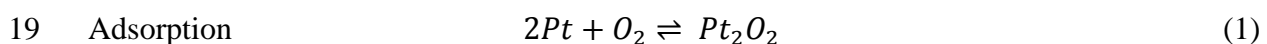


1 Introduction

2
3 Proton exchange membrane fuel cells have been present for many decades however large scale
4 commercialisation has still yet to be achieved. The major factors that are hindering this are the
5 low cathodic catalytic activity, lower lifetime of the fuel cells and use of expensive Pt as
6 catalyst which makes it more expensive to compete in the current market [1-3]. A lot of
7 research has been focussed on discovering new catalyst materials ranging from Pt based alloys
8 to non Pt based alloys to improve the catalytic activity, lower the use of Pt to reduce the cost
9 of the system while at the same time trying to improve the lifetime of the fuel cell [4-13].
10 However, one of the major factors which is often overlooked is the performance loss of the
11 fuel cell during operation. This can increase the cost of the system as the system has to be
12 oversized to accommodate the performance loss. One cause of this performance loss is due to
13 the formation of oxide layers on the platinum (Pt) catalyst which reduces the current density at
14 a given potential. The oxidation of Pt occurs as a natural intermediate in the oxygen reduction
15 reaction (ORR) and hence cannot be avoided entirely, as it is why platinum is such a good fuel
16 cell catalyst. The reactions can be summarised below, although it should be noted there is a
17 wealth of detail and some controversy in the academic literature [2, 3, 13-16] as to the exact
18 reaction pathway in practice:



24 Each of the electrochemical reactions, (3) and (4) have an equilibrium potential, and so
25 there will be an equilibrium coverage of species on the surface that is a function of the operating
26 cathode potential. Hence at different potentials there are different equilibrium concentrations

1 of species, meaning that there are more or less free sites available for oxygen to adsorb on the
2 platinum (i.e. reaction (1)). Thus, the reaction intermediates can also be considered poisons to
3 the reaction, as if their concentration builds up too high they can limit the current density at a
4 given potential. Fortunately, because all the species adsorbed on the surface are in dynamic
5 equilibrium with each other, the equilibrium concentration for a given potential takes time to
6 be established, and therefore, it is possible to achieve a time averaged concentration of
7 intermediate species that is different from the equilibrium concentration for a given potential,
8 by periodic perturbation of the system (i.e. short circuiting).

9 There are various reports which highlight the formation of H_2O_2 as an intermediate
10 during the oxygen reduction reaction [2, 3, 13, 17, 18]. This H_2O_2 formation not only reduces
11 the efficiency of the fuel cell system but also accelerates degradation of the Nafion membrane
12 by the formation of free radicals [14, 17-22]. It may be possible to avoid the formation of the
13 peroxides if the average coverage of OH can be reduced by short circuiting, thereby reducing
14 the degradation due to low voltage idling conditions.

15 There are a very limited number of publications on the use of short circuiting as a
16 method to improve the performance of fuel cell systems. In early 2000, a patent by Fuglevand
17 et. al [23] was published on the use of short circuiting with fuel cells for the performance
18 improvement of the system and they attributed this performance increase to the increase in
19 proton conductivity of the membrane because of two reasons : 1) more water production and
20 2) increase in temperature to evaporate water and increase air flow to the cathode. Koschany
21 et.al [24] also recommended the use of short circuiting to improve the performance of the fuel
22 cell by maintaining the water content. In contrast Pearson et.al [25, 26] secured a patent on the
23 method and apparatus for improving the performance of fuel cells by periodically short
24 circuiting. They believed the performance improvement was due to the stripping of oxides and
25 other adsorbed species from Pt. This patent led to the commercialisation of a short circuiting

1 controller by Ballard systems with their fuel cell stacks. Other fuel cell system suppliers, such
2 as Horizon fuel cells, also use a short circuiting controller which does a short circuit for 100
3 ms every 10 s of operation but does not vary with the condition of the stack. Some studies on
4 the use of short circuiting has been reported by researchers in academia as well where Kim
5 et.al [27] used this technique to humidify the stack for unmanned aerial vehicle and Zhan et.al
6 [28] reported improvement in performance and lifetime for use of fuel cell in hybrid
7 applications. Both studies reported the production of excess water during the short circuiting
8 as the reason for performance improvement and also the moisture evaporation from the GDL.
9 Despite the fact that it works, there is still a lack of clear evidence for the mechanism for
10 improving the performance and there are two competing theories. There is also no published
11 strategy on how frequently and how long the short circuiting should be done.

12 Examining the two theories for why short circuiting improves performance, it is easy
13 to quickly discard the water/thermal management theory. Consider that a typical short circuit
14 period might be 100 ms every 10 s, and that during the short circuit the current and therefore
15 amount of water produced during this time might double. However, the time during short
16 circuit represents only 1% of the total operating time. Thus only 1% more water in total will
17 be produced by the short circuit. We consider this to be highly unlikely to have a significant
18 effect on the water management of the fuel cell system, except perhaps the local concentration
19 of water in the catalyst layer. Secondly the argument that short-circuiting will increase the
20 temperature and hence evaporate water and increase air flow to the cathode, can also be
21 discarded. Consider that during the short circuit, the system runs at 0 V, and so it is true that
22 all energy generated will be dissipated as heat internally. This could indeed lead to a sudden
23 increase in temperature at localised points which could evaporate water at those points.
24 However, again the short circuit represents only 1% of the total operating time. In addition, the
25 purpose of the short circuit is that the average voltage increases, and hence the efficiency of

1 the fuel cell increases and the heat generated during the remaining 99% of the time is less. This
2 means less total heat overall will be produced, and thus the stack should be cooler, which could
3 increase the risk of flooding. Therefore by Occam's razor [29, 30] we assume that the oxide
4 stripping mechanism, as the simplest theory, is correct.

5 It is very important to understand the relationship between the kinetics and
6 thermodynamics of oxide formation in order to predict the mechanism of oxide removal. Pt
7 oxidation is severe at the higher potentials and thus should need more frequent short circuiting
8 and less at lower potentials. The short circuiting time will depend on the discharging of the
9 double layer capacitance and thus the minimum short circuiting time should exceed the time
10 needed to discharge the double layer, but not too long to cause fuel or air starvation, and thus
11 there should be an optimum time depending on the performance enhancement as well as
12 application. There has been a recent study by Rinaldo et. al [31] wherein the relationship
13 between kinetics and thermodynamics of oxide formation and reduction was studied using
14 cyclic voltammetry. They used a potentiodynamic model to study the different stages of oxide
15 formation and reduction by including reaction rates that are dependent on the species and
16 adsorption energies. They were able to model the behaviour of Pt (111) and predict the
17 irreversibility of the CV and relate it to the two-site non-electrochemical surface reaction taking
18 place at high potential. However, the model was only validated for the Pt (111) surface and
19 failed to predict the behaviour in case of polycrystalline Pt which is normally the catalyst used
20 in fuel cell.

21 In summary the short circuiting method has been well known to industry for many years
22 and even commercialised by Ballard and Horizon, however there is a lack of understanding on
23 how it actually improves the performance. There have also been no studies on how to optimise
24 a short circuiting strategy for different fuel cell systems. This paper presents an experimental

1 study of the technique, and also the development of a methodology to establish the optimum
2 short-circuiting strategy for any system.

3 4 **Experimental Methods**

5
6 The short circuiting strategy was developed by testing on a 2 cell setup made from self-
7 humidifying cells provided by Arcola Energy. It is a closed cathode system with an active area
8 of 144 cm², 3 channel serpentine and parallel flow channels were used for the anode and
9 cathode respectively. Stoichiometry of 1.3 and 3 were used for H₂ and Air, respectively. None
10 of the gases were humidified and the relative humidity of H₂ and air were ~ 0 % and 10-15 %,
11 respectively. A simple block diagram for the test setup is given in Figure 1(a). The load bank
12 used was Scribner 850 e and Labview was used to control the short circuiting. The switching
13 between normal operation and short circuiting was done with the help of two switches shown
14 in block diagram as load switch (LS) and short circuit (SC). For normal operation of the fuel
15 cell, LS is closed while SC is open and to induce a short circuit first the LS switch is opened
16 and SC is closed. Cell voltage and current during short circuit was monitored using a National
17 Instruments (NI) compactRIO (cRIO) at 1 kHz with a 12 bit ± 10V analog input module. The
18 testing was done at 69.4 mA/cm² (25 °C), 138.9 mA/cm² (30 °C) and 277.8 mA/cm² (50 °C)
19 for different short circuiting times of 10 ms, 50 ms, 100 ms, 200 ms and 500 ms to find the
20 short circuiting interval which will give the maximum increase in the performance i.e. higher
21 cell voltage. A schematic of the short circuiting protocol is shown for 4 short circuits in Figure
22 1(b). However, the amount of short circuits were varied during the testing. During the
23 experiments, a load is applied to the fuel cell until it reached a steady state voltage. A short
24 circuit is applied to find the maximum power increase which was then set as the short circuiting
25 interval, this process was then repeated by doing 10 short circuits using the set short circuit
26 duration and interval to verify the improved performance.

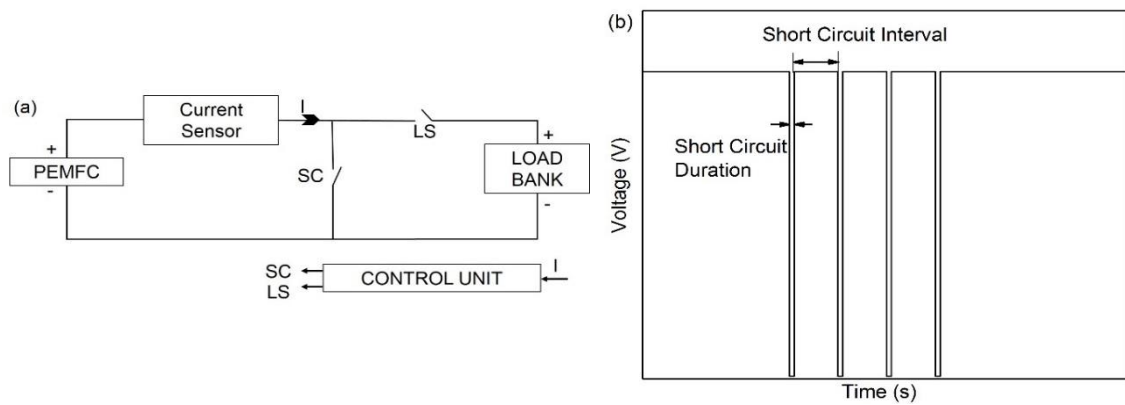


Figure 1: Schematic of the (a) short circuiting setup and (b) short circuiting protocol

After the formation of a short circuiting strategy using the 2 cell setup, this strategy was verified by application in a commercial Horizon H-100 fuel cell stack which is an open cathode system consisting of 20 cells in the stack each with an active area of 22.5 cm^2 . Two systems were used, a new H-100 system and a system that has been tested aggressively since 2014 and has degraded over time such that its maximum performance was around 40%. This was to test the hypothesis that a single optimisation strategy could be applied to stacks in different conditions.

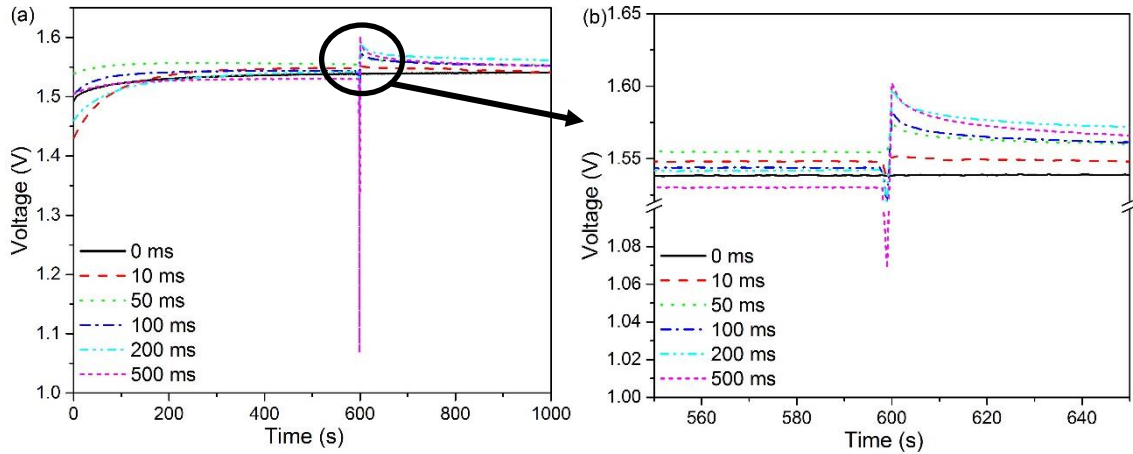
The testing was done at 3 different current values in the range of 0.6 V to 0.75 V which can be termed as the region where performance is thought to be dominated by the effect of PtOH and PtO. The testing was done at 44.4 mA/cm^2 ($22 \text{ }^\circ\text{C}$), 88.9 mA/cm^2 ($25 \text{ }^\circ\text{C}$) and 133.3 mA/cm^2 ($30 \text{ }^\circ\text{C}$) and the in-built controller was removed and replaced by the short circuiting controller used for the 2 cell system. The systems were conditioned by the procedure recommended by Horizon Fuel Cell systems to bring them to maximum efficiency. The testing was done at the same current values for both systems but the new H-100 system was also tested at 266.7 mA/cm^2 ($38 \text{ }^\circ\text{C}$) chosen so as to not exceed the 100 W power limit. The testing was done with hydrogen flowing through the system with no purging to exclude the effect of purging on the performance improvement. A fuel stoichiometry of 1.3 was used for H_2 and atmospheric air was used with operating the in-built fan at 5 V to maintain the temperature and

1 sufficient air supply for reaction. Dry hydrogen was used with a relative humidity of ~ 0 % and
2 the relative humidity of atmospheric air varied between 40- 45 % over the course of testing.
3 Electrochemical impedance spectroscopy was done at different current values to find the time
4 constant of the electrical double layer. The measurements were performed using an Autolab
5 potentiostat for H-100 stacks under load within a frequency range between 10 kHz and 0.1 Hz
6 with an amplitude of 5 % of the DC current. The spectra was recorded after the potential had
7 stabilised for the specified current value i.e. 1 A (44.4 mA/cm²), 2 A (88.9 mA/cm²), 3 A (133.3
8 mA/cm²), 6 A (266.7 mA/cm²) in the current study.

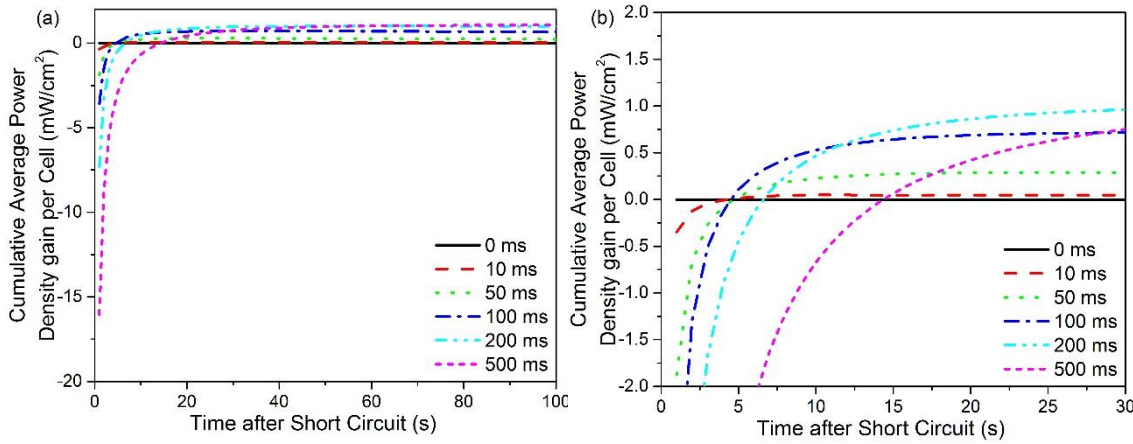
9 **Results and Discussion**

10 Figure 2 shows the voltage profile recorded at 69.4 mA/cm² for the 2-cell setup which
11 was subjected to a single short circuit from 10 ms to 500 ms. Results for other two current
12 values (138.9 mA/cm² and 277.8 mA/cm²) are reported in supporting information (Figure S1
13 and S2). First the 2-cell setup was held at 69.4 mA/cm² for 10 mins to reach a steady state as
14 can be seen by the plateau on the potential curve, then a short circuit (which appears as spikes
15 on the graph) was applied for different time periods i.e. 10 ms, 50 ms, 100 ms, 200 ms and 500
16 ms. It can be clearly seen from the curve that the short circuiting time of 10 ms was not enough
17 to induce any gain in the potential. All other short circuiting times showed a similar profile of
18 increase in the potential and then a gradual decline. This confirms that the short circuit duration
19 must exceed the time taken to discharge the double layer, and that for times greater than this a
20 potential increase can be induced which can result in the increase of power. However, the
21 maximum amount of short circuiting time should be kept to a minimum so as not to induce any
22 permanent damage to the fuel cell and to achieve maximum power gain. This improvement in
23 the potential is due to the removal of the Pt hydroxide and oxide layers formed on the Pt catalyst
24 and thus resulting in a larger effective surface area of Pt available for the electrochemical
25 reaction. If short circuiting is not performed Pt hydroxide and oxide will continuously form,

1 and with time the potential will decay gradually as can be seen in Figure 2. In order to find the
 2 amount of performance improvement from the short circuiting, cumulative average power
 3 density gain per cell was calculated taking into account the power decrease during the short
 4 circuit.



5
 6 Figure 2: Potential vs time comparison for short circuiting at 69.4 mA/cm^2 ($25 \text{ }^\circ\text{C}$) for Arcola
 7 2 cell setup at different short circuit time period
 8



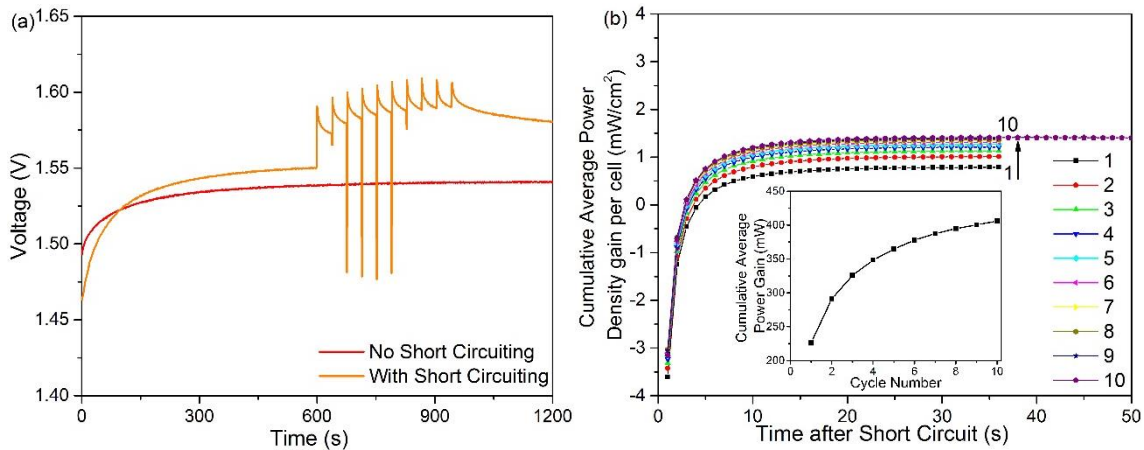
9
 10 Figure 3: Cumulative average power density gain per cell at different short circuiting time for
 11 short circuiting at 69.4 mA/cm^2 ($25 \text{ }^\circ\text{C}$) on Arcola 2 cell setup.
 12

13 Figure 3 shows a relationship between the cumulative average power density gain per
 14 cell with the increase of short circuiting time as a function of time after the short circuit. Sample
 15 calculation for cumulative average power density gain per cell is shown in supporting
 16 information S4. It can be clearly seen that the short circuiting time of 10 ms is a flat line very
 17 near to 0 and thus offers no significant improvement in performance. With the increase in short

1 circuiting time, the point at which the cumulative power density gain becomes a net positive
2 start shifting towards the right i.e. it takes longer to recover the energy loss incurred during
3 short circuiting. This is likely due a number of reasons. Firstly the double layer will become
4 increasingly discharged at longer short circuit durations and thus requiring additional time to
5 be recharged. Secondly, at these potentials the oxides are likely to be removed quickly and they
6 have a finite coverage. Therefore there is a fixed amount of time (or charge) needed to remove
7 them, and once this is exceeded any further time spent during short-circuit is unnecessary and
8 wastes energy. Figure 3 also shows the cumulative average power density increasing with
9 increase in short circuiting time. This reaches a maximum and then starts decreasing. This time
10 could therefore be around the optimum time needed to remove the oxides. So, in order to
11 develop an optimum strategy, the cumulative average power density gain should be maximised
12 by minimising the time taken to recover the energy loss during the short circuit. Based on these
13 two criteria, a short circuiting time between 100 and 200 ms is the optimum short circuiting
14 time for this system. To test this strategy, multiple short circuits were done at 69.4 mA/cm^2
15 with a short circuiting time of 100 ms and interval of 36 s (point of maximum cumulative
16 average power density gain).

17 Figure 4 shows the comparison between the 2 cell setup run without short circuiting
18 and with the short circuiting strategy. A 3% increase in the performance was observed for the
19 system run using short circuits. The low performance improvement is due to the only 2 cells in
20 the system and also running of the system at a very low current density. Thus even though the
21 potential increase is by 50 mV, the change in performance is only by 3 %. From Figure 4 (b),
22 it can be seen that with every cycle, there is an improvement in the performance and after
23 around 7-8 cycles it has reached a dynamic equilibrium. The results show that one short circuit
24 is not enough to achieve the maximum performance gain but requires multiple short circuits.
25 As discussed in figure 2 that the voltage improvement is due to the removal of Pt hydroxides

1 and oxides from the Pt catalyst. However, one cycle of short circuiting is not enough to recover
 2 the maximum performance and thus multiple short circuits are needed. This becomes clear
 3 from Figure 4 that with multiple short circuits, the voltage gain is increasing and becomes
 4 constant after ~ 7 cycles.



6
 7 Figure 4: (a) Potential profile at 69.4 mA/cm² without and with 10 SC cycles (SC time-100
 8 ms and interval of 36 s). (b) Cumulative average power density gain profile for each short
 9 circuiting cycle.
 10

11 The parametric short circuiting strategy developed above was then verified on a
 12 commercial Horizon H-100 system. A single short circuit was done at different current
 13 densities for different short circuiting times and short circuiting intervals, the optimum were
 14 found by the analysis of cumulative average power density gain per cell (Figure S3 for 44.4
 15 mA/cm²). This was the same procedure as that developed using the 2 cell Arcola Energy system.
 16 From the analysis of these results, the optimum short circuiting time was again between 100
 17 and 200 ms. As Horizon recommends using a short circuiting strategy of short circuiting time
 18 – 100 ms and interval – 10s, for a good comparison the short circuiting time of 100 ms was
 19 used with the short circuiting interval obtained from our strategy of obtaining maximum
 20 cumulative average power density gain.

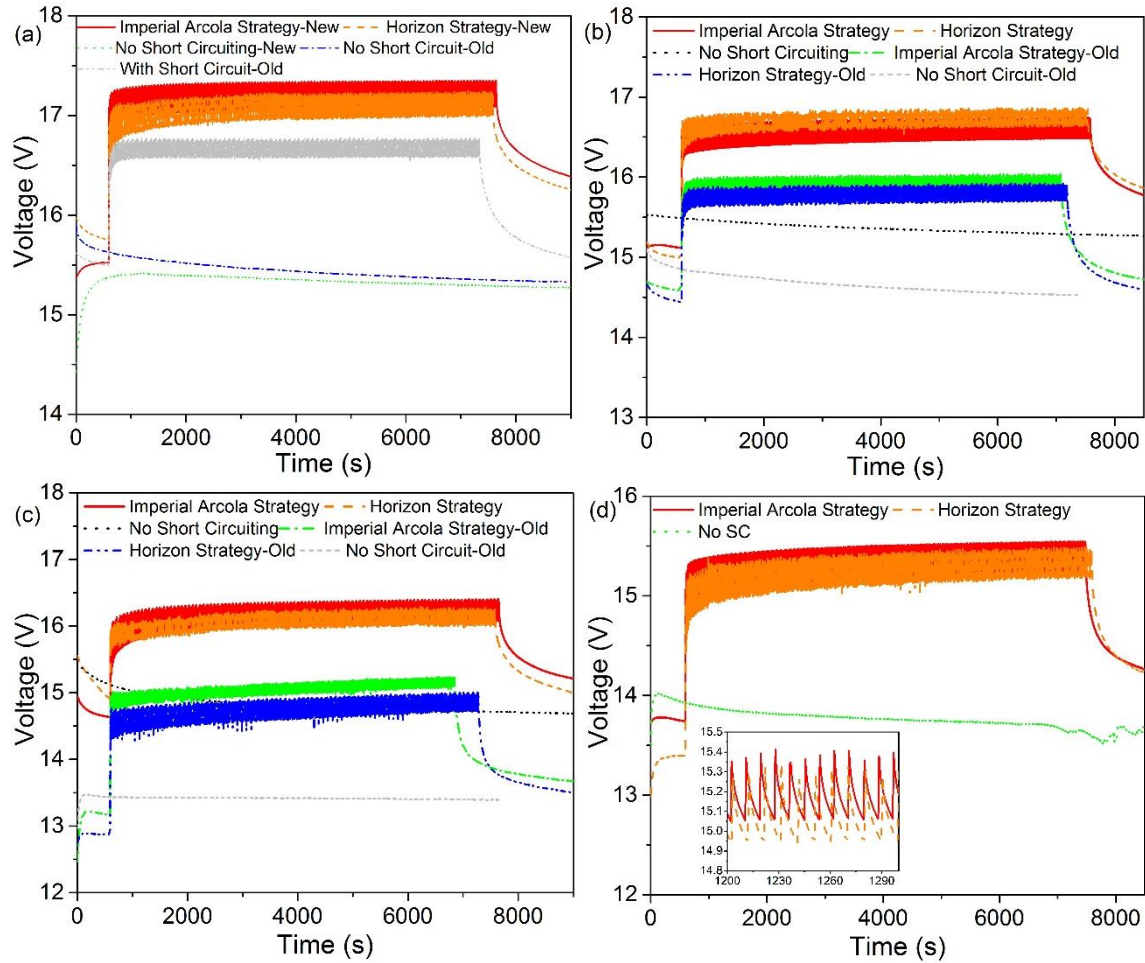


Figure 5: Potential comparison for short circuit testing using Imperial Arcola Strategy, Horizon Strategy and no short circuit at (a) 44.4 mA/cm², (b) 88.9mA/cm², (c) 133.3 mA/cm² and (d) 266.7 mA/cm².

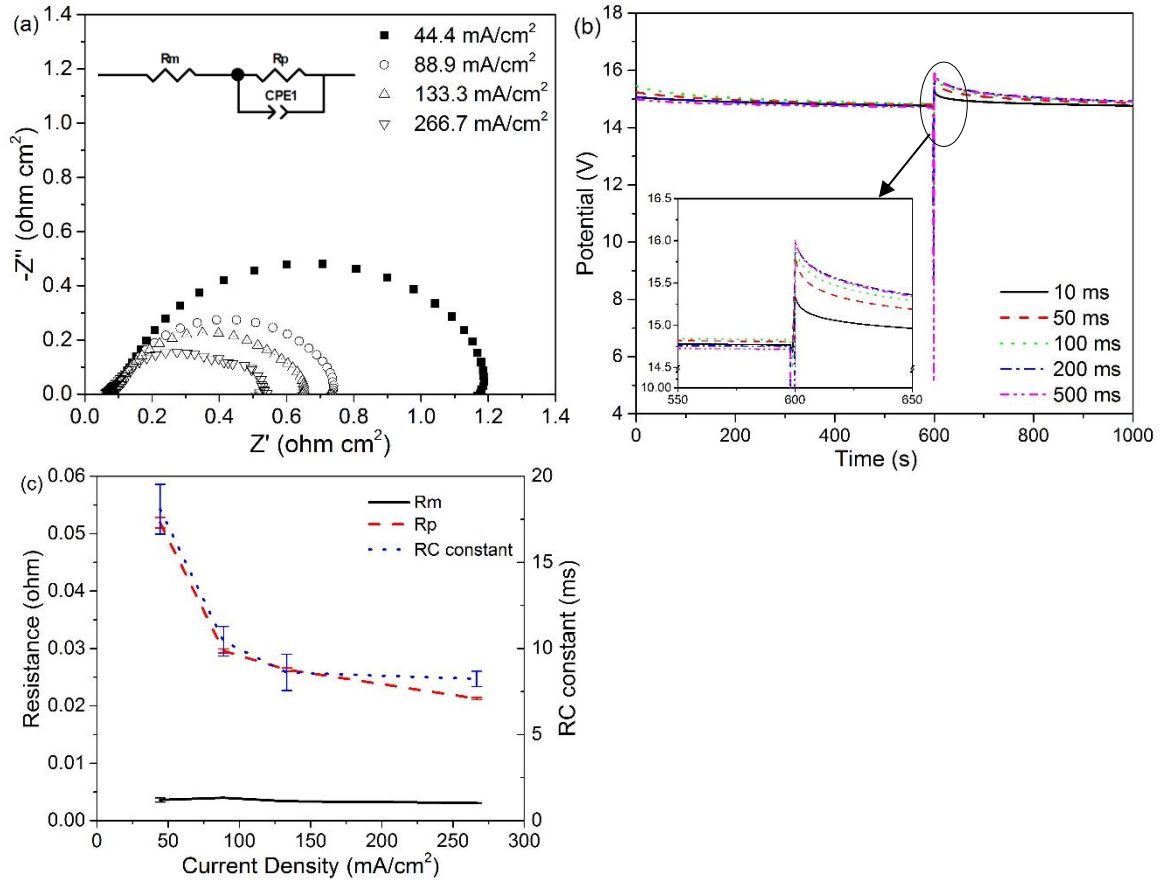
Figure 5 shows a comparison of the Horizon strategy, the developed strategy (Imperial Arcola strategy) and no short circuiting on the old and new H-100 systems at different current densities at a short circuiting time of 100 ms. As mentioned earlier, the old H-100 system was tested aggressively for 2-3 years and can only deliver 40-45% of its rated power, and thus higher current density testing was not possible. The results from this system are labelled as old. Figure 5 (a) shows the voltage profile of the testing done at 1 A (44.4 mA/cm²) and the sample with short circuiting shows an improvement of around 8 % in voltage value compared to the one without short circuit for the old system. There is only one curve for this sample as the short circuiting interval was the same for both the Horizon strategy and the optimised Imperial Arcola strategy. For the new stack, there is an improvement of 10 % (Horizon Strategy) and

1 12 % (Imperial Arcola Strategy) as compared to the testing without any short circuiting. As the
2 short circuiting was removed after 2 hours, the voltage starts to drop gradually which suggests
3 that the surface oxide coverage is beginning to return to its equilibrium value.

4 Figure 5 (b) shows the voltage profile for the testing done at 2 A (88.9 mA/cm^2) for
5 both H-100 stacks. The results demonstrate an improvement of 8 % (Horizon Strategy) and 10 %
6 (Imperial Arcola Strategy) for both the old and new stack in comparison to no short circuiting.
7 The system follows the same profile as seen in the testing done at 1 A (44.4 mA/cm^2) when the
8 short circuits is removed after 2 hours. Similar improvements in the range of 10-12 % are
9 observed for both stacks when tested at 3 A (133.3 mA/cm^2) with short circuiting in comparison
10 to no short circuiting as shown in figure 5 (c). As the old H-100 stack was not able to reach
11 higher currents, only the new stack was tested at 6 A (266.7 mA/cm^2) and the voltage profile
12 obtained is shown in figure 5(d) with an inset highlighting the profile for 100 s. Similar
13 improvement of 10-12 % are observed by the use of short circuiting as compared to no short
14 circuiting. The testing was not done at even higher current densities due to the limitation of the
15 stack as it is rated for 100 W and while doing short circuiting at 6 A (266.7 mA/cm^2), power
16 was around 92 W. Another point to be noted here is that we are able to exceed the performance
17 improvement using our Imperial Arcola strategy compared to the standard Horizon strategy at
18 all the current values. The improvement obtained by the Imperial Arcola strategy is almost 2 %
19 higher in all the cases which suggests that the short circuiting interval is an important factor
20 and it should vary with the operating condition of the stack like current density, temperature.
21 The other advantage using Imperial Arcola strategy is the validation and variation of the short
22 circuiting interval on the condition of stack i.e. new and old in the current study.

23 The relationship between short circuiting time and the electrochemical double layer
24 capacitance is also important. In order to investigate this, the double layer capacitance was
25 determined by electrochemical impedance spectroscopy. Electrochemical impedance

1 spectroscopy was done at the same current values which are used above for short circuiting in
2 the frequency range of 10 kHz and 0.1 Hz with an amplitude of 5 % of the DC current. Figure
3 6 (a) shows the impedance spectra obtained at different current values. All the spectra are fitted
4 using Z-View using an equivalent circuit consisting of R_m (CPE_1R_p) where R_m is the ohmic
5 resistance, CPE_1 is the constant phase element and R_p is the polarisation resistance. There is a
6 slight decrease in the ohmic resistance with the increase in the current which can be due to the
7 better humidification of the membrane as the current increases. Also the charge transfer
8 resistance decreases with the increase in the current as the driving force for oxygen reduction
9 reaction increases as can be seen at the low frequency values of impedance, as expected by the
10 Butler-Volmer equation. As the electrochemical double layer is modelled as a capacitor, the
11 RC constant was calculated from the parameter obtained by circuit fitting and the result is
12 shown in Table 1. The RC constant for a capacitor is termed as the time required to charge or
13 discharge 63 % of the capacitor and our theory is that the minimum short circuiting time should
14 be higher than the time needed to discharge the electrical double layer appreciably. It can be
15 seen that the RC constant decreases with the increase in current which is due to the improved
16 oxygen reduction reaction kinetics at the higher current and thus lower resistance. Figure 6 (b)
17 shows the voltage profile obtained at 3 A (133 mA/cm^2) by varying the short circuiting time
18 from 10 ms to 500 ms using a single short circuit. It can be seen that there is only a slight
19 improvement using the short circuiting time of 10 ms as the double layer is not completely
20 discharged and the effect of the short circuit is not prominent. As soon as the short circuiting
21 time is increased to 50 ms or above, the improvement is high as the double layer is discharged
22 to a significant amount during this time and the short circuit can induce other effects other than
23 just discharging the double layer.



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Conclusions

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A simple procedure to determine the optimum short circuiting strategy (the Imperial Arcola strategy) has been developed using a single short circuit at different current values. This strategy is based on maximising the cumulative average power density gain and minimising the time required to recover the energy loss during short circuits. This strategy was validated on a commercial H-100 stack using a degraded and new stack and the Imperial Arcola strategy showed an extra improvement of 2 % compared to the standard Horizon strategy recommended for use with H-100 systems. The optimum short circuiting duration and interval was shown to be dependent on the operating conditions of the stack (i.e. current density). An overall

1 improvement of 10-12 % in voltage for a given current density, and hence power, was
2 demonstrated using short circuiting as compared to no short circuit.

3 The optimum short circuit duration was also shown to be a function of the double layer
4 capacitance of the fuel cell. If the short circuit duration is too short the charge passed during
5 the short circuit mainly contributes to the discharge of the double layer capacitance rather than
6 stripping of the oxides from the Pt surfaces. We show that by using impedance spectroscopy
7 measurements, the double layer capacitance can be determined and used to ascertain the
8 minimum short circuit time.

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