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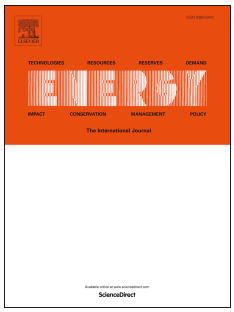

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Energy Efficiency Improvements by Investigating the Water Flooding Management on Proton Exchange Membrane Fuel Cell (PEMFC)

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Abstract

This paper presents a broad study of research work associated with the effect of water flooding and management in Proton Exchange Membrane Fuel Cells (PEMFC) which operates at relatively low temperatures at conditions that could allow the accumulation of water that degrade cell performance.

Recent studies confirm the importance of proper water balance during cell operation to avoid both dehydration and flooding. Condition to ensure the PEM remains hydrated while excessive water condensation is prevented are identified and analysed.

The work review current literature and examines the different mechanisms of water transport in PEMFCs and their relative importance and impact on cell operation. The work analyse the effect of water accumulation at both the anode and the cathode regions and discusses the impact on cell efficiency of each.

This work reviews recent development in this field and examines the approaches used such as improved flow field designs, improved membrane chemical formulation to increase hydrophilicity, manipulation of operating pressure, optimisation of operating temperature, the level of humidification, optimisation of gas flow rate and mechanical modification of the membrane structure among other techniques.

The work examines recent advances in the techniques for non-intrusive in-situ water detection, monitoring and characterisation and compares their effectiveness.

The work concludes by a critical review of recent studies that examined different strategies that could prevent water flooding and promote proper water management in PEM fuel cells. This includes water management control strategies designed to improve the voltage and current density at specific operating conditions.

Keywords: PEM fuel cells, Anode region, Cathode region, Membrane, Water flooding, Water management, Serpentine flow field channels, Open pore metal foam

1. Introduction

Concerns persist over the environmental impact of fossil fuels used in transport and electricity generation due to their harmful environmental and health impacts [1-7]. Also, the reliance on depleting resources such as fossil fuels, which are mainly produced by oil countries regions with political instability and security concerns, raises questions about the security of supply of commodities that are essential for our way of life and economies [8]. These are the major causes why there is urgent need for clean alternative energy sources. In recent years, the capacity of renewable energy technologies such as hydroelectricity, wind energy and solar power has increased considerably but still do not meet the energy demands of the world [9]. However, due to only heat and water being the by-products and for their high efficiencies, fuel cells are becoming credible candidates to provide energy for transport and other application but more research is still needed to improve the overall efficiency of fuel cells use cycle including the production of the fuel for the cells. In a PEM fuel cell an electrolyte membrane is sandwiched between two electrodes allowing protons to pass through it but at the same time it is impermeable to electrons [10-11]. The anode side is negatively charged electrode and the cathode side is positively charged electrode as shown in Fig. 1. Electrochemical reactions take place at the catalyst sites on both sides of the membrane. The hydrogen is split into protons and electrons at the anode catalyst layer, protons pass across the membrane while the electrons are transported through an external electrical circuit, which generates useful electric work and move across to the other side of the membrane (cathode region) where it meets with the protons to form hydrogen. The air or pure oxygen entering into the fuel cell from the cathode side will go through an electro-chemical process with

hydrogen at the cathode catalyst layer and the end products is water and heat [12-14]. The reactions that take place are shown in equation (1) to (3):

Anode region: $H_2 \rightarrow 2H^+ + 2e^-$ (1)

Cathode region: $\frac{1}{2}O_2 + 2H+ + 2e- \rightarrow H_2O$ (2)

Total reaction: $H_2 + \frac{1}{2}O_2 \rightarrow H_2O$ (3)

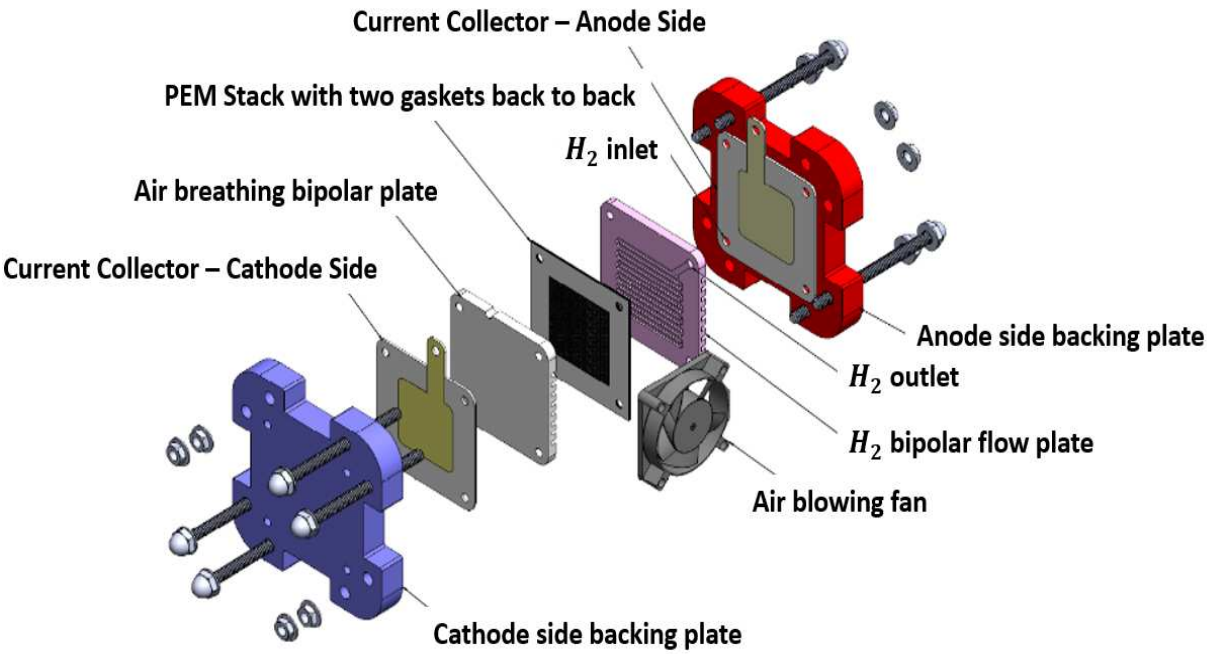


Fig. 1. Polymer electrolyte membrane fuel cell (PEM fuel cell) [13].

A major challenge that impacts the performance of the PEM fuel cell is the excess water in the cell which is produced by the oxygen reduction reaction as described by Eq. 2 at the cathode region or if protons are transported with water through the electrolyte membrane from the anode region to the cathode region through electro-osmotic drag [15].

Judith et al. [16] explain that if water generation level is higher than water removal level, there will be accumulation of water which leads to water flooding. Once the gas diffusion layer (GDL) pores are filled with water, transportation of reactants to the catalyst layer (CL) becomes impeded and the active catalyst sites will be masked with water. In addition, excess water in a PEM fuel cell will cause water bands and water columns forming inside the flow field channels whereby the flow of gas becomes clogged or blocked [17-18]. According to Alfredo and Pierre [19], water flows from the cathode region to the anode region in a process called back diffusion and the later part of the paper discusses this in more details. Chen et al. [20] studied the dynamics of water in the anode flow field channels at different operating

conditions. They found out that as liquid water droplets size increases, pressure drop also increases which reduces the time taken for the water to leave the channels.

Wolfgang and Ardalan [21] stated that when the rate of removal of water is higher than that of water production through reaction, there may not be enough water to keep the electrolyte membrane hydrated which will cause it to dry out resulting in a drop of voltage in the PEM fuel cell and its overall efficiency. Dong et al. [22] concluded that rib structure and capillary force have a tremendous effect on liquid water transport and distribution in the GDL which could enhance the removal of water from the GDL to avoid water flooding.

Carton et al. [23] modelled slug formation and droplet accumulation in the channels of a PEM fuel cell. They employed the Volume of Fluid (VOF) model in ANSYS Fluent to visualise the slug formation and water movement. Carton et al. [23] found out that coalescence of water droplets and excess water in the channels can lead to the formation of slugs in the cell and bends and steps in the flow channels could disrupt movement of slug and prevent channel blockage.

Erni et al. [24] investigated the impact of operating pressures and temperatures on water transport in a PEM fuel cell and reported that at the anode region relative humidity decrease was caused by higher pressures and temperatures. As the membrane water content reduced along the flow channel, the ionic conductivity of the membrane and electro osmotic drag coefficient decreased. Asif et al. [25] conducted a numerical study on the behaviour of liquid water rising from GDL in a dead end mode at anode channel. They concluded that when the current density increases the water production and flooding also increase in the channel.

Water management is essential in achieving durability and maximum performance of a PEM fuel cell. A minimum amount of water is necessary so as to improve the ionic conductivity in the proton electrolyte membrane but too much water causes flooding in the cell.

Yuehua et al. [26] argued that proper understanding of water management is important to find an equilibrium point between electrolyte membrane dehydration and flooding for the purpose of ensuring high performance of the cell.

2. Transport of water in PEM fuel cell

Water management plays an important role in overall fuel cell performance and it is one of the serious challenges facing fuel cells designers and manufacturers even when they implement widely used and studied designs. Nik et al. [27] stated that in order to achieve a proper water management in a fuel cell two requirements need to be fulfilled: preventing water flooding inside the fuel cell and keeping the membrane adequately hydrated.

The two main water transportation mechanisms in PEM fuel cell are electro-osmotic drag and back diffusion [28] and water transport determines the hydration level of an electrolyte membrane, which is essential in supporting proton conductivity to pass through it easily [29-30].

2.1. Mechanism of the transportation of water in PEM fuel cell

In addition to electro-osmotic drag and back diffusion, there are other methods through which water is being transported such as thermal-osmotic drag and hydraulic permeation [31]. Recent research has focused mainly on electro-osmotic drag and back diffusion because they are seen as the major causes of water flooding in fuel cells [32]. Good knowledge of the mechanisms of the transportation of water is key for improved performance of the fuel cell.

Fig.2 illustrates the mechanism of how water transportation occurs in a PEM fuel cell.

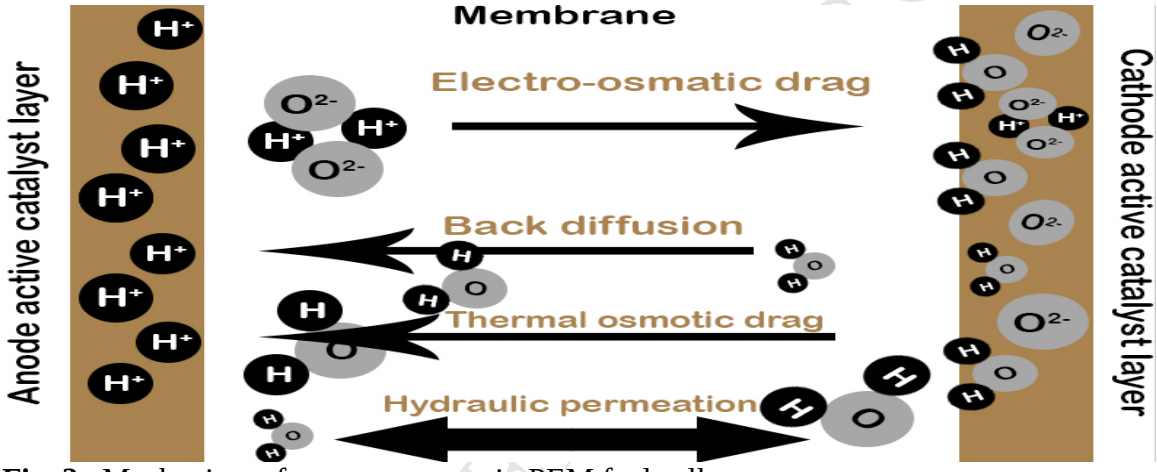


Fig. 2. Mechanism of water transport in PEM fuel cell.

2.1.1. Thermal-osmotic drag (TOD)

The effect of thermal osmotic drag on water management was not studied in details in the past and water transport as result of temperature effects started to attract attention recently. Thermal osmotic drag is caused by differences in temperature through the membrane in the fuel cell [33-35]. During observation of fuel cell operation, it was noted that water flow was from cold to hot regions [36]. During fuel cell operation a temperature gradient do occurs across the electrolyte membrane. In addition, it is considered that one of the main mechanisms of TOD is the effect of heat pipe. This heat pipe effect exists in a PEM fuel cell at the catalyst layer and is cause by the evaporation of water which moves as vapour then condenses down a path as a result of temperature decrease given the temperature gradient in the cell. This mode of transport is significant during start up and shut down of the fuel cell [37].

2.1.2. Electro-osmotic drag (EOD)

The protons travel along the membrane from the anode to the cathode region with water molecules being carried along through the membrane. The process of water being dragged by the protons across the membrane is known as electro-osmotic drag [38-40]. As explained earlier, water management affects the performance of the fuel cell and the hydration of the membrane increases the proton conductivity. When there is too much water in the membrane, the presence of EOD will lead to water flooding in the fuel cell which will impact the electrochemical reaction rate and lead to drop in the cell efficiency [41].

The coefficient of EOD is expressed as the number of moles of water associated with one mole of protons that is transported along the membrane in the absence of pressure and concentration gradients [42].

Feina et al. [43] studied water balance across a Nafion membrane by electrophoresis NMR (ENMR) method to determine electro osmotic drag coefficient. The results showed that the water electro osmotic drag coefficient within the Nafion membrane depended on the water content of the Nafion membrane and the kind of counter-cation used. Zhiping et al. [44] explained that electro-osmotic drag coefficient increased with increase in temperature between 20^{o}C and 90^{o}C.

2.1.3. Back diffusion (BD)

This type of water transport is a result of the excess water generated at the cathode region which diffuses back to the anode region. Water diffuses back from the cathode along the membrane empty space as result of water concentration gradients in the fuel cell [45-46] and research activity on BD from the cathode region to the anode region has attracted considerable interest recently. In addition to water concentration gradient, membrane thickness and pressure gradient through the membrane are among the other factors that determine the BD coefficient [47]. Experimental studies have also been conducted to measure the back diffusion coefficient of water [48]. The earlier research conducted showed huge differences between the measured values [48] and the discrepancies were attributed to the applied measurement techniques according to Majsztrik et al. [48] who concluded that the various methods used measured different rate processes. Majsztrik et al. also accounted for the swelling of the membrane and their work was further developed by Paul et.al [49] who emphasised the importance of the kinetics of sorption and desorption and their effect on water activity and temperature.

2.1.4. Hydraulic permeation (HP)

In a PEM fuel cell, hydraulic permeation takes place due to pressure gradient between the anode and the cathode regions. This results in water fluxes in the membrane because of the capillary pressure differential or gas phase pressure differential [51]. Amayol and Bahrami [52] stated that if the cells are pressurised separately in such a way that the fuel gas (anode side) is being supplied with a lower pressure than the oxidant gas (cathode side), this may help in reducing water at the cathode. It is important to note that before pressurising the anode and cathode region differently, membrane mechanical strength needs to be properly considered [53-54].

Water flux ($J_{nmw,\ hyd}$, kmol m^{-2} s^{-1}) can be related to hydraulic permeation resulting from the pressure gradient by eqn 4 [Need consistent reference to equations] [55]:

$$J_{nmw,hyd} = -c_{\mathrm{nmw}} \frac{\mathrm{K}_{nmw}}{\mu_{\mathrm{nmw}}} \nabla p_{\mathrm{nwm}} = -\lambda_{nf} \frac{\rho_{mem}}{EW} \frac{K_{nmw}}{\mu_{lq}} \nabla \mathrm{p} \qquad (4)$$

where C_{nmw} is concentration of non-frozen water in ionomer (kmol m^{-3}), K_{nmw} (m^2) is permeability, μ_{nmw} (kg m^{-1} s^{-1}) is the dynamic viscosity of non-frozen water in an ionomer (it is mainly replaced by liquid water properties), and p_{nmw} (Pa) is pressure of the non-frozen water in an ionomer [55]. The negative sign signifies the decrease of pressure decrease in the direction of water diffusion or increased path length.

Table 1 - Major mechanisms of water transport in PEM fuel cell

Mechanisms	Descriptions	References
Thermal-osmotic drag(TOD)	Caused by differences in temperature across the membrane.	[33-35]
Electro-osmotic drag (EOD)	The process where water molecules are dragged by protons through the membrane.	[38-40]
Back diffusion (BD)	Water transport as result of the excess water generated at the cathode region which diffuses back to the anode region.	[45-46]
Hydraulic permeation (HP)	Water transport due to the pressure gradient between the anode and cathode regions.	[51]

3. Impact of water management (flooding) on PEM fuel cell (Water issues within PEM fuel cell)

As the system operation continues and water builds up over time, severe water flooding will occur especially when operating at low temperature and this could temporarily block the gas flow paths [56-57].

In most of the investigations conducted so far, the membranes of most fuel cells where the electrochemical reactions occur are made up of Nafion. To increase the protonic conductivity of most Nafion membranes, the membrane must be fully hydrated and by doing this the proton conductivity is increased by an order of magnitude of six [58]. Water often accumulates at the cathode region at low operating temperatures. This makes the fuel cell prone to flooding especially when reacting hydrogen and oxygen electrochemically together at temperatures less than 100^{o}C at atmospheric pressure. It is also possible to channel water from the cathode to the anode region via the membrane through local pressure and concentration gradients [59].

Datong et al. [60] concluded that occurrence of liquid water flooding in the GDL hinders the oxidant transportation and flooding of the cathode catalyst layer does not only block oxygen transport in the cell but covers the active catalyst area and degrades the performance of the cell.

Ugur and Wang [61] explain that flooding reduces the mass transport functionality of catalyst layer and gas diffusion layer predominantly in the cathode region. This reduces the magnitude of the current and voltage being generated hence degrades the overall performance characteristics of the fuel cell. Once the fuel cell begins to get flooded, water needs to flow from the electrodes regions through the GDL into the channels and flow out of the fuel cell to reduce flooding. Very often nearly one third of the membrane is not utilised in instances where the fuel cell is flooded [60].

Two hypothesises explain the movement of the water through the GDL with the first being the converging passageway of water transport mechanism and the second being channelling liquid water transport mechanisms and the two theories suggested the possibilities of water flooding in the GDL [62]. The water formed often starts as droplets that then coalesce and start building up in the GDL before flowing into the flow field channels or the bipolar plates. This phenomenon occurs because of capillary effects in the GDL [60-62].

According to the work of Turkmen et al. [48], when the gases flow through a thin GDL with small pores a good electrical conductivity is observed but for effective mass transport the pores need to be larger. Cao et al. [64] study stressed the fact that these conditions normally occur in the fuel cell depending on its operational conditions such as the inlet temperature, humidification level and gas flow rates.

In addition to impeding gas movement, flooding in the fuel cell causes water to cover the catalyst sites thus creating a barrier between the catalyst and the reactive substances. To avoid the impact of flooding on the performance of the fuel cell, understanding the mechanisms of water transport in the fuel cell is necessary. Although measures need to be taken to avoid flooding, it is important to recognise that keeping the membrane well hydrated increases its life span and reduces the cost of the cell [65-67].

3.1. Impact at the Cathode

At the cathode region, oxygen is being supplied to the PEM fuel cell by diffusion via the GDL to the cathode catalyst layer and it interacts with the protons that move along the membrane and electrons that travel across the external load to form water and produce heat [68-70].

Water accumulation at the cathode may be due to the formation of electro-chemical reaction products at the cathode on its own or coupled with other factors such excess humidification of the hydrogen feed and electro-osmotic drag [71]. McCain et el. [72] explains that when there is flooding in the cell the water may accumulate at the GDL which will block the surface of the catalyst layer that is supposed to facilitate the electro-chemical reaction thus preventing oxygen reduction reaction (as shown in Fig. 3). This lead to a situation where the current and voltage will not be predictable and the performance of the cell will deteriorate. However, excess accumulated water could be removed by capillary transport of cathode channels, by means of water-vapour diffusion or evaporation and BD to anode region. The evaporation and water-vapour diffusion usually happen at high cell operating temperature [73-74].

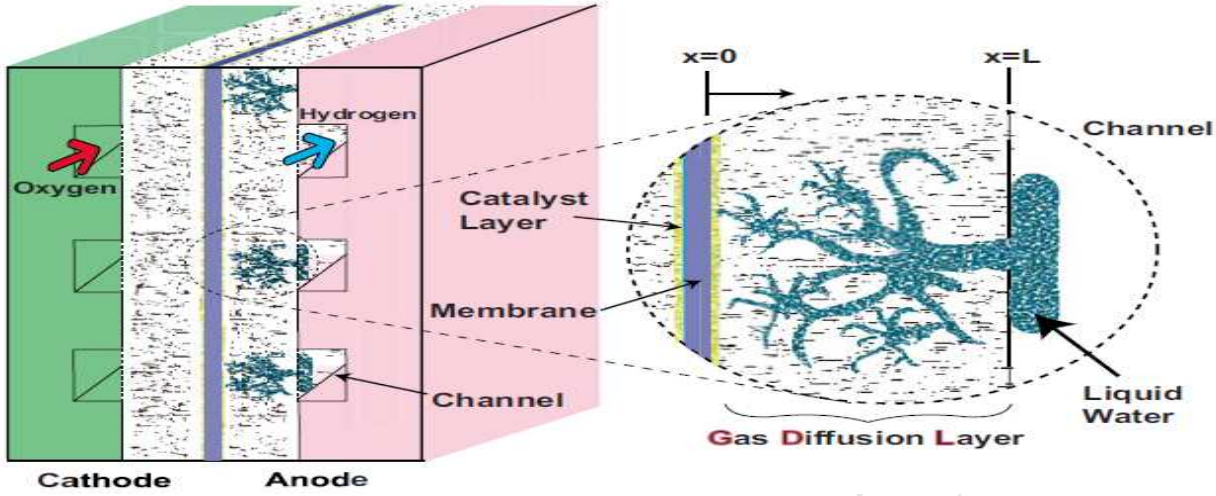


Fig. 3. A conceptual schematic describing liquid water accumulation in the GDL and later flow to the field channel to build a blockage reactant film [72].

Jamekhorshid et al. [75] investigated the performance of cathode flooding environments of a PEM fuel cell. Two-dimensional GDL models of partially flooded cells based on electrochemical relations and laws of mass conservation were proposed for the study. Jamekhorshid et al. [75] observed that the cell performed well when it was in operation at a higher temperature which prevent water flooding as more water joins the vapour phase. Fig. 4 shows the polarization curves for different levels of water flooding as well as a flooding free PEM fuel cell. The three curves for the flooded cells show drastic fall in the cell performance as the water flooding increases in the cell. This is due to the fact that as water accumulation increases, more of the catalysts sites are covered by water and made inaccessible to the reacting gases.

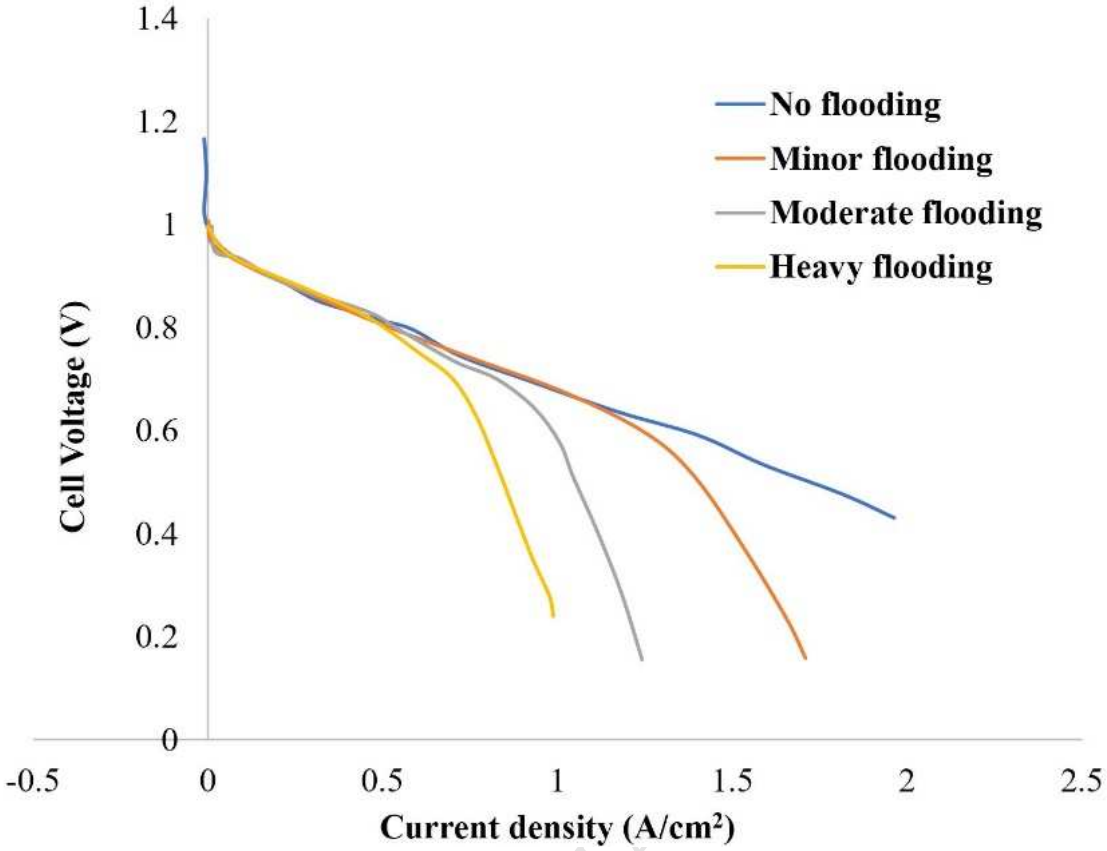


Fig. 4. Polarization curves of a PEM fuel cell showing no flooding and the effect of water flooding on cell performance [76].

Eikerlinga [77] studied water management of cathode catalyst layers (CCL) in PEMFC. The results show that CCL behaves similar to watershed in the fuel cell which helps to balance the opposite water flux across the membrane and cathode region outlet. Najjari et al. [78] conducted experiments to observe the effects of water flooding on GDL because of the accumulation of water in the fuel cell and their results confirmed that excess liquid water affects fuel cell performance drastically. Najjari et al. [78] concluded that a thinner GDL will result in a lower resistance for oxygen transport and liquid water is more likely to block the pores easily than vice-versa. Das et al. [79] investigated the effect of flooding on cell performance and they concluded that if the cathode catalyst layer active reaction area reduces due to liquid water, an increase in activation over-potential will occur.

3.2. Impact at the anode

At the anode region hydrogen is dissociated into protons and electrons on the catalyst layer. The protons pass across the membrane to the cathode region while the electrons are transported through the external circuit to the cathode region [80-81]. The membrane needs to be hydrated to avoid dryness that leads to ionic resistance. However, water can flow to the

anode region from the cathode region through back diffusion which will result in flooding and drop in produced voltage [82-83]. Fig. 5 shows voltage against time for a single cell at a stable current density of 1 A cm^{-2}. As the voltage starts dropping over time, water flooding increases [84]. Lee and Bae [85] experiments showed that the major cause of anode flooding is low gas flow rate in the channel. However, in the fuel cell stack when water blocks a channel, the fuel gas is diverted to the unblocked channel of another cell which causes fluctuations and voltage drop. Lee and Bae [85] concluded that this occurs as a result of water flooding at the anode or cathode regions. Zhang et al. [86] investigated the water vapour transportation from the cathode region to the anode region during operation at high temperatures within the fuel cell. The polarization curve, transient ohmic resistance and the voltage were used in analysing the results and they concluded that in flow through mode of operation water vapour has little effect. On the other hand under dead end mode of operation it brings about stability. Zhang et al. [86] further argued that water cross over to the anode hinders the diffusion of hydrogen and causes reduced cell performance.

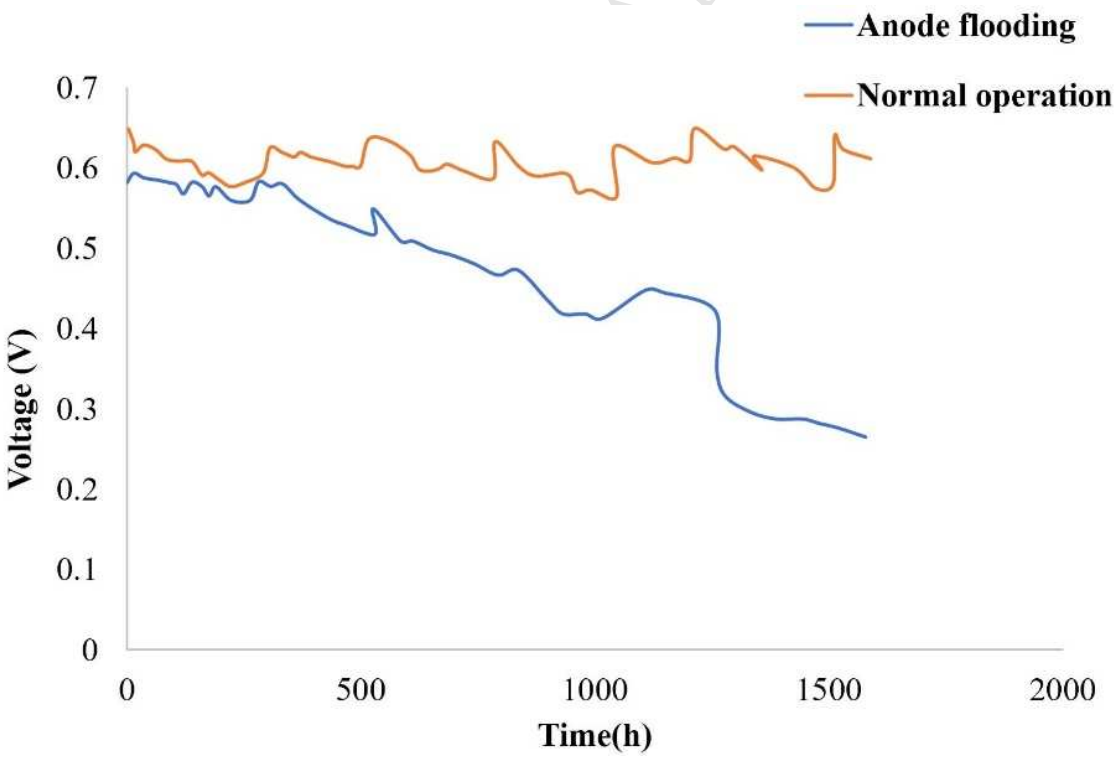


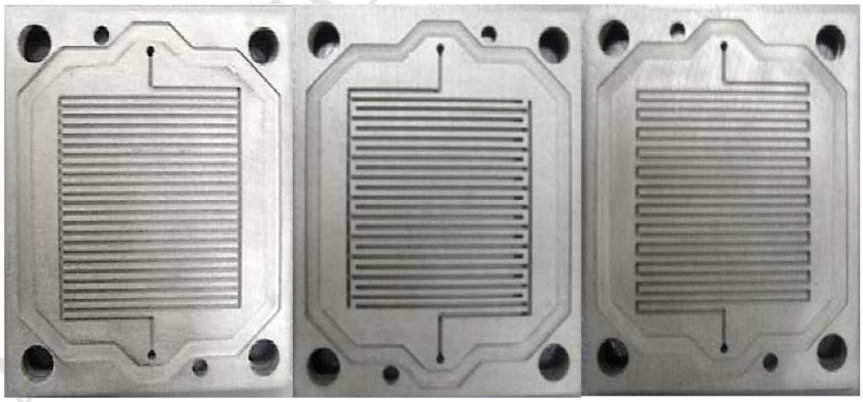
Fig. 5. A profiles of voltage decline profiles of a single cells for 1600 h at constant current density of 1A cm^{-2} [84].

3.3. Impact on flow field channels

Water flooding also occurs in the gas flow field channels and this depends on the design of the flow channels, characteristics of the material of the components of the fuel cell and the operating conditions. Flooding in the flow field will stop the reactant gas from reaching the catalyst layer and prevent the electro-chemical reaction from taking place [87].

As could be seen from the discussion so far, the design of flow field channel contributes to water flooding and most common types of flow field designs are “Parallel”, “Interdigitated” and “Serpentine” as shown in Fig. 6. Researchers have proved over the years that parallel flow field design is liable to flooding and unacceptable amount of non-uniformity in the air streams flow because the gases flow inside the parallel channels through the surface of the GDL which causes water accumulation [88] but parallel flow design is appropriate for application that requires low pressure drop and high gas flow rates [89].

The interdigitated flow field has dead end in both the inlet and outlet of the flow field channel. Consequently, both the reactant and product transport to and from catalyst layer along the GDL is by convection rather than by diffusion [90-91]. The serpentine flow field is mostly used in the industry as a result of its high cell performance, good reliability and durability resulting from its layout [92]. Serpentine flow channel is also better because it is not prone to flooding like the other conventional designs but it has some limitations like major power loss due to high pressure drop and drop in the reactant concentration along the flow field path [93].



(a) (b) (c)

Fig. 6. (a) parallel flow field channel (b) interdigitated flow field channel (c) serpentine flow field design [94].

Liu et al. [94] carried out an experimental study on different types of flow field channels in order to determine their effect on cell performance using the geometries shown in Fig. 6 and concluded that the serpentine flow channel performed better than the other counterpart

because it help to prevent water flooding in the channel. It also keeps good humidity in the channels of the cathode which will avoid membrane dehydration and has acceptable pressure drop. A similar conclusion was made by Arvay et al. [95] who investigated the effect of water on interdigitated and serpentine flow field channels.

Arvay et al. [95] stated that changing the active area had a less effect on the interdigitated flow fields when compared to serpentine flow field. Lim et al. [96] stated that the parallel flow field shows a poor rate of water removal when compared to other flow field designs. In a similar way Shimpalee et al. [97] compared parallel, interdigitated and serpentine flow fields and they reported that interdigitated and serpentine flow fields are better than parallel flow field in terms of water management due to the dead end channel of the parallel design.

3.4. Water transport in membrane

The transport of water in Nafion membranes is based on water absorption at membrane and gas diffusion layer boundary and water desorption [98]. Several models were developed for water transport via the membrane based on its structure. Most membranes material constitute improved polyethylene with hydrogen as a replacement for fluorine. The electrolyte is supported by sulfonic acid group (HSO_3), merged ionically, enabling the acid group to attract the H^+ ions. The acid group (HSO_3) can be found in the chain end and this structure is referred to as an ionomer [99]. It is feasible to vary the equivalent weight but it will have immense influence on the transport and mechanical properties. An effective way of improving the mechanical properties of fuel cells is by increasing the electric weight but this might decrease the protonic conductivity and sulfonic acid group concentration. Membranes for PEM fuel cells application are currently made up of 1100 EW Nafion because there is a good balance of protonic conductivity and mechanical properties [100]. Increase in water activity of the fuel cell often leads to additional water being absorbed by the membrane which causes the hydrophilic area of the PEM fuel cell to swell up and this phenomenon reduces protonic transport resistance in the membrane [101-103].

There are three possible forms in which water could be generated through electrochemical reaction. This includes water vapour, liquid water and water contained within the membrane as shown in Fig 7.

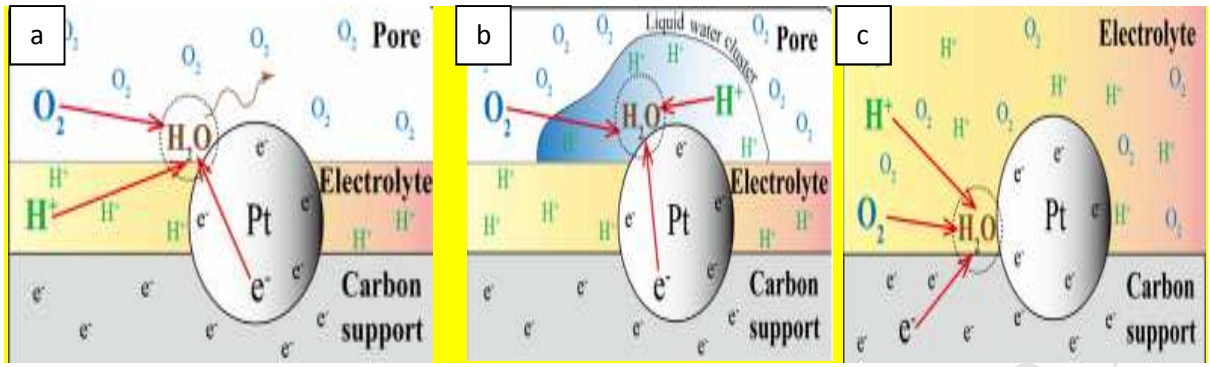


Fig. 7. Schematics of different mechanisms of cathode catalyst water generation. (a) water vapor generation, (b) liquid water generation, (c) membrane water generation [104].

The water flow of the net membrane $\dot{n}_{H2O,m}$ in the cell is assumed to be the arithemetic sum of two elements:

$$\dot{n}_{H2O,m} = \dot{n}_{H20,m,backdiff} - \dot{n}_{H20,m,osmotic} \quad (5)$$

When the net flux is from cathode to anode ($\dot{n}_{H20,m,backdiff} > \dot{n}_{H20,m,osmotic}$)then, $\dot{n}_{H2O,m}$ will be positive and negative if it is vice versa. In the latter instance, the effect of osmotic drag is greater than water diffusion($\dot{n}_{H20,m,backdiff} < \dot{n}_{H20,m,osmotic}$). The terms of the equation (5) on the right side is explain below.

3.4.1. Diffusion of water

Water back diffusion is caused due to the concentration gradient from cathode to anode regions. Using Fick's law, mass transfer diffusion rate is expressed as [1]:

$$\dot{n}_{H20,m,backdiff} = D_w \frac{dc_w}{dy} = \frac{1}{t_m} \int_{c_w^{anode}}^{c_w^{cathode}} D_w dy \quad (6)$$

The water content variation in cross direction can be approximated by averaging. In equation (6) the diffusion coefficient of water in the membrane is D_w [cm^2s^{-1}]; water concentration in the membrane at anode and cathode region is c_w (mol cm^{-3}); perpendicular distance to the membrane is t_m [cm]. $c_w^{cathode}$ and $c_w^{cathode}$are water concentration in the membrane which contribute to the conversion of membrane water content λ to mass are establish by:

$$c_w = \frac{\rho_{m,dry}}{M_{m,dry}} \lambda \quad (7)$$

where $\rho_{m,dry}$ is the dry membrane density[g/cm3] and $M_{m,dry}$ is the dry membrane equivalent weight[g/mol].

If no current passes across the membrane, then the ionomer phase water content (λ) can be defined as the number of water molecules per equivalent of the polymer in membrane with water vapour local activity or water liquid present in the pores of catalyst layer. Zawodzinski

et al. [105] measured water content in a membrane, which is found from the following equation:

$$\lambda_{\text{equi,i}} = \begin{cases} 0.043 + 17.81a_i - 39.85a_i^2 + 36.0a_i^3 & ,0 \le a_i \le 1 \\ 14 + 1.4(a_i - 1) & ,0 < a_i \le 1 \end{cases} \quad \text{i} = \text{anode; cathode} \quad (8)$$

Water activity of the membrane is found to be:

$$\text{a}_\text{i} = \frac{y_{w,i}p_i}{P_{sat,i}} = \frac{P_{w,i}}{P_{sat,i}} \quad \text{i} = \text{anode; cathode} \qquad (9)$$

a_m is the water activity of the membrane mean value between the water activity of the anode and the cathode.

The diffusion coefficient of water is obtained using a revised Arrhenius equation derived from experimental measurements and depends on temperature and water content as:

$$\text{D}_\text{w} = \text{D}_\lambda \cdot \exp\left(2416 \cdot \left(\frac{1}{303} - \frac{1}{\text{T}_\text{fuel cell}}\right)\right) \qquad (10)$$

Equation (6) was originally correlated using experimental measurement between 30^{o}C and 80^{o}C using the nafion electrolyte. D_λ is the diffusion coefficient and depends on membrane hydration.

$$D_\lambda = \begin{cases} 10^{-6} & ,\lambda_\text{m} < 2 \\ 10^{-6}\left(1 + 2(\lambda_\text{m} - 2)\right) & ,2 \le \lambda_\text{m} \le 3 \\ 10^{-6}\left(3 + 1.67(\lambda_\text{m} - 3)\right) & ,3 \le \lambda_\text{m} \le 4.5 \\ 1.25 \times 10^{-6} & ,\lambda_\text{m} \ge 4.5 \end{cases} \qquad (11)$$

Dutta et al. [106] reported that water contents in the membrane (Nafion) and water diffusion coefficients vary from one another significantly.

The water transport from the anode to cathode side (EOD) can be defined as:

$$\dot{\text{n}}_{\text{H20,m,osmotic}} = \text{n}_\text{d}\frac{\text{i}}{\text{F}} \qquad (12)$$

which is defined as the amount of water molecules conveyed by one of the proton [mol sec^{-1} cm^{-2}], F [C mol^{-1}], is Faraday constant and i [Acm^{-2}] is current density of the fuel cell.

It is noticed consistently that if liquid water is in equilibrium with membranes, then EOD will be larger but if membrane are in equilibrium with water vapour, EOD will be smaller. EOD coefficient is likely to increase with temperature.

3.4.2. **Membrane water transport with sorption/desorption**

From the previous model, it is assumed that water in the contacting fluid phase and water in membrane are in equilibrium. Studies have shown that water activity in the contacting fluid and membrane phase are likely not to be in equilibrium. Combined with other transport resistances, this will limit water transport across the membrane and this depends on the operating conditions [107]. A non-equilibrium model for water transport can be expressed as [108]:

$$\dot{n}_{H20,m} = \dot{n}_{H20,m,osmotic} - \dot{n}_{H20,m,backdiff} = \frac{\rho_m}{EW_m}\phi k\left(\lambda_{equi,m} - \lambda_m\right) \qquad (13)$$

Where EWm [grams of polymer mole of sulphonic acid groups] is ionomer equivalent weight and ρ_m [kgm-3] is density of dry ionomer. Table 2 also captures other investigations on water management in fuel cells.

3.4.3. Water sorption/desorption

Water sorption and desorption at the GDL/ membrane interface is function of the equilibrium offset, where $\lambda_{eq}^{ads.side} - \lambda^{des.side}$ are the real water content in the ionomer phase and the eqillirium is the number at equilibrium as defined by [109]

$$\dot{n}_{H20,m} = \frac{\rho_m}{EW_m}\phi K_a\left(\lambda_{eq}^{abs.side} - \lambda^{abs.side}\right) \qquad (14a)$$

$$\dot{n}_{H20,m} = \frac{\rho_m}{EW_m}\phi K_d\left(\lambda^{des.side} - \lambda_{eq}^{des.side}\right) \qquad (14b)$$

A side of the membrane is either in absorption or desorption mode and this depends on the direction of net membrane water flux [$\dot{n}_{H20,m}$]. The set of equations is also applied for anode and cathode regions. K_a and K_d account for the mass transfer coefficients for absorption and desorption. ϕ is the surface roughness factor which is introduced in equation 10a and 10b to account for the difference in a particular surface area, if various types of ionomer loading is used in the catalyst layer. $\lambda_{eq}^{ads.side}$ and $\lambda_{eq}^{des.side}$ are the water content values at equilibrium. K_a and K_d [1/S] are the kinetic absorption and desorption coefficients describing the operating conditions of non-equilibrium membrane water uptake.

$$k_{d/a} = a_{d/a} \cdot f_{v,An/Ca} \cdot \exp\left[2416\left(\frac{1}{T_o} - \frac{1}{T_{stk}}\right)\right] \qquad (15)$$

T_o is the reference temperature. [110]

Kinetic absorption and desorption water coefficients as well as chemical diffusion coefficient are proportional to water volume fraction in the membrane f_v. (Transport in polymer-

electrolyte membranes.) and could be given with Arrhenius temperature difference depending on the following equation:

$$f_{v,An/Ca} = \lambda_{An/Ca}\left[\frac{\overline{V}_w}{\overline{V}_{mem}+\lambda_{An/Ca}\cdot\overline{V}_w}\right] \quad (16)$$

where $\overline{V}_w$ is the partial molar volume of water [18cm^3/mol] and partial molar volume of Nafion is $\overline{V}_{mem}$ [cm^3/mol] which is defined as: $\overline{V}_{mem} = \frac{EW_{mem}}{\rho_{mem}}$ (17)

Table 2 - Some Studies on different approaches conducted to understand water flooding and management in PEM fuel cell

Author	Study	Methods	Results	Conclusions	Reference
P.K. Bhattacharya	Water flooding in PEMFC.	Water uptake in the membrane was investigated experimentally and a model was used to validate the result using COSMOL multiphysics.	The result show from the polarisation curves that as the current densities is increased, more liquid water was produced The simulation results follow almost the same trend.	Water management strategies needs to be considered when designing a fuel cell in order to obtain an optimum cell performance.	111
Li et al.	A flow channel design procedure for PEM fuel cells with effective water removal.	The channel cross section of serpentine design was modified and characterised.	The reactant forced water out of the system when the pressure drop was increased.	The modification of the serpentine channel and proper pressure drop helped in preventing flooding in the channels.	[112]
Qi and Kaufman	Improvement of water management by a microporous sublayer for PEM fuel cells.	Different material of sublayers of micro- porous layer containing 24, 35 and 45 per cent micro-porous layer of various thickness between carbon paper and catalyst layer were assessed	The 35% perform better than the others, which helps in enhancing gas diffusion layer to manage water effectively.	Micro-porous layer with the right percentage of PTFE contributes to a proper water management.	[113]
Liu et al.	Water flooding and pressure drop characteristics in flow	Different conventional flow field	The interdigitated and cascade performed better in liquid water	At low temperatures, liquid water	[114]

	channels of proton exchange membrane fuel cells.	channels used including interdigitated, cascade and parallel flow field	removal than parallel channels during cell operation which helps to prevent flooding.	concentration is high in flow channels than at high temperatures. If there is liquid water in channels, increasing flow rate may force water out of the cell.	
Hickner et al.	Real-Time Imaging of Liquid Water in an Operating Proton Exchange Membrane Fuel Cell.	Neutron imaging techniques were used to measure the extent of water content in the cell when varying temperature.	They observed that as cell temperature increases from 40^{o}C to 80^{o}C, the liquid water amount reduced over time.	They concluded that cell temperature and heat do affect liquid water content in the cell.	[115]
Liu et al.	Experimental Study and Comparison of Various Designs of Gas Flow Fields to PEM Fuel Cells and Cell Stack Performance.	A graphite plate PEM fuel cell stack was fabricated. Different flow field channels were used to conduct the experiments.	The results showed that serpentine flow channels performed better because of a reasonable pressure drop and prevented water flooding better than the other designs.	Serpentine flow field channel do optimise water management in the cell better than the other flow channels.	[94]
Jithesh et al.	The effect of flow distributors on the liquid water distribution and performance of a PEM fuel cell.	Parallel, serpentine and mixed flow field channels were modelled and stimulated numerically.	The mixed flow field channel tends to perform better in terms of water removal, effective water distribution and good membrane hydration.	Flow field channels design have effect on water management in PEM fuel cell.	[116]
Su et al.	Studies on flooding in PEM fuel cell cathode channels.	An experiment was conducted using serpentine and serpentine-interdigitated flow field channels to study flooding occurrence and the results were compared.	They observed that serpentine-interdigitated flow field performs better for water removal than serpentine because the pressure in upstream helps to push the water down to the downstream channels.	The serpentine-interdigitated flow field design Is more effective for water removal than serpentine alone.	[117]
Bozorgnezhad	The experimental	A transparent	It was seen that liquid	They concluded	[118]

et al.	study of water management in the cathode channel of single-serpentine transparent proton exchange membrane fuel cell by direct visualization.	stack was used for direct visualisation and experiments were performed with several operating parameters.	water was accumulated at the elbow of the channels.	that the elbows of the channel are important for a water management.	
Lee and Bae	Visualization of flooding in a single cell and stacks by using a newly-designed transparent PEMFC.	A transparent thin gold plate and polycarbonate plate is (one plate or two different plates?) substitute with graphite bi-polar plate.	They observed water droplets at the ribs and elbows in both anode and cathode side of channels which later turned to water slugs in the anode side of flow channels but not at cathode side due to flow rate effects.	Water droplets and slugs were formed from condensation in the cell.	[119]
Wang and Zhou	Liquid water flooding process in proton exchange membrane fuel cell cathode with straight parallel channels and porous layer.	A computational fluid dynamics (CFD) was used to numerically simulate the process.	Liquid water was observed in the channels, which comes from the porous layer.	It was concluded that the channels do influence water flooding in cathode side and water accumulation begins from the zones in porous layer	[120]

4. Visualizing liquid water in PEM fuel cells

There is no established reliable predictive process to know when and where flooding is taking place because there are various factors such as feed stream humidity, temperature and pressure of the fuel cell that affect water transportation in the various components of the PEM fuel cell [121]. However, there has been extensive research on diagnostic and detection tools for water flooding including direct visualisation, neutron imaging, nuclear magnetic resonance and X-ray imaging.

4.1. Direct Visualization

These methods involve the use of a transparent cell plate that permits visual access to the channels by optical devices such as high-speed cameras, digital camcorder, CCD camera and infrared detection devices. It is used directly in observing the operational conditions effects on water droplets growth, formation and movement [122].

Ge and Wang [123] employed direct visualization to investigate water drops formation at anode flow channels to examine hydrophobic and hydrophilic GDL. It was observe that at low current density in hydrophobic GDL water tends to condense on the channel while in hydrophilic GDL condensed water stores up and blocks the channels. Furthermore, Weng et al. [124] studied the various effects of stoichiometry of the gas concentration and humidification at the cathode. They observed that as the stoichiometry of humidified oxygen increases, the cell performances increases. Spernjak et al. [125] experimentally examined water formation and transport by directly visualising it on various GDL materials for the removal of water from PEMFC and they concluded from their experimental results that the untreated GDL did not push water to the membrane and pores.

Aslam et al. [126] used thermal and digital cameras to view the relationship between temperature and liquid water within the cell at the cathode side by using a transparent PEM fuel cell as shown in Fig 8. Aslam et al. [126] observed that as the air flow rate increases, the temperature distribution through the electrolyte of the cathode region becomes non uniform, as a result of the liquid water seen in the flow field channels (Fig. 9) causing a reduced efficiency of heat dissipation.

Fig. 8. A transparent PEM fuel cell [126]

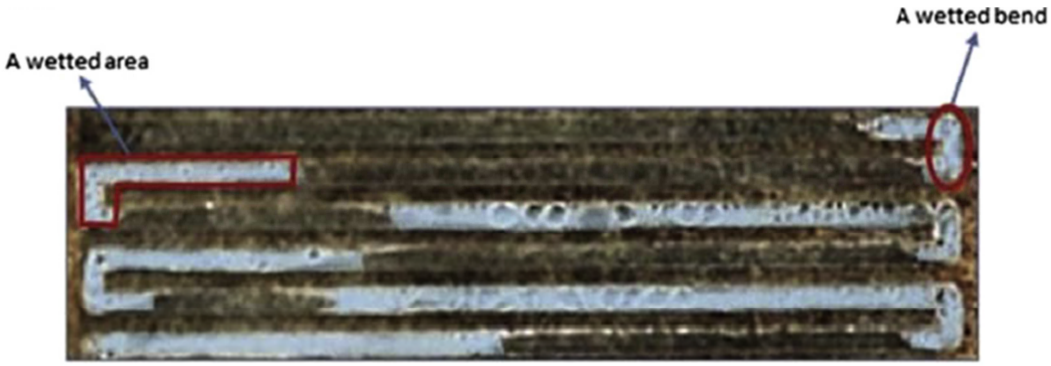


Fig. 9. Images of the wetted area in the flow field channel [126]

Hussaini and Wang [127] conducted a visualisation study of liquid water flooding in the flow field channels of the cathode side using different range of current densities, humidification and stoichiometry that are similar to the one experienced in an automotive environment. Some of the conclusions from the study are that in operating conditions with a low stoichiometry and low current densities at any humidity level, the fuel cells are more vulnerable to severe flooding occurring in any of the operating conditions they considered. In addition, they noticed that the actual water distribution was different from one experiment to another and from one channel to another, but at specific operating conditions liquid water was spotted in the channels. Daino et al. [128] conducted an experiment observing GDL cross section by visualising it using a digital microscope at a higher magnification in which the small droplets were seen to be condensing. Water droplets were observed to have developed at the GDL across the section at a current density of 0.4 Acm^{-2}. The video recorded was processed to detect the locations and condensate quantity. Fig. 10 shows the actual image), water detection locations image and water detection with GDL cross section. The results shows that water droplets and transportation on GDL cross-section can be detected with the use of digital microscope.

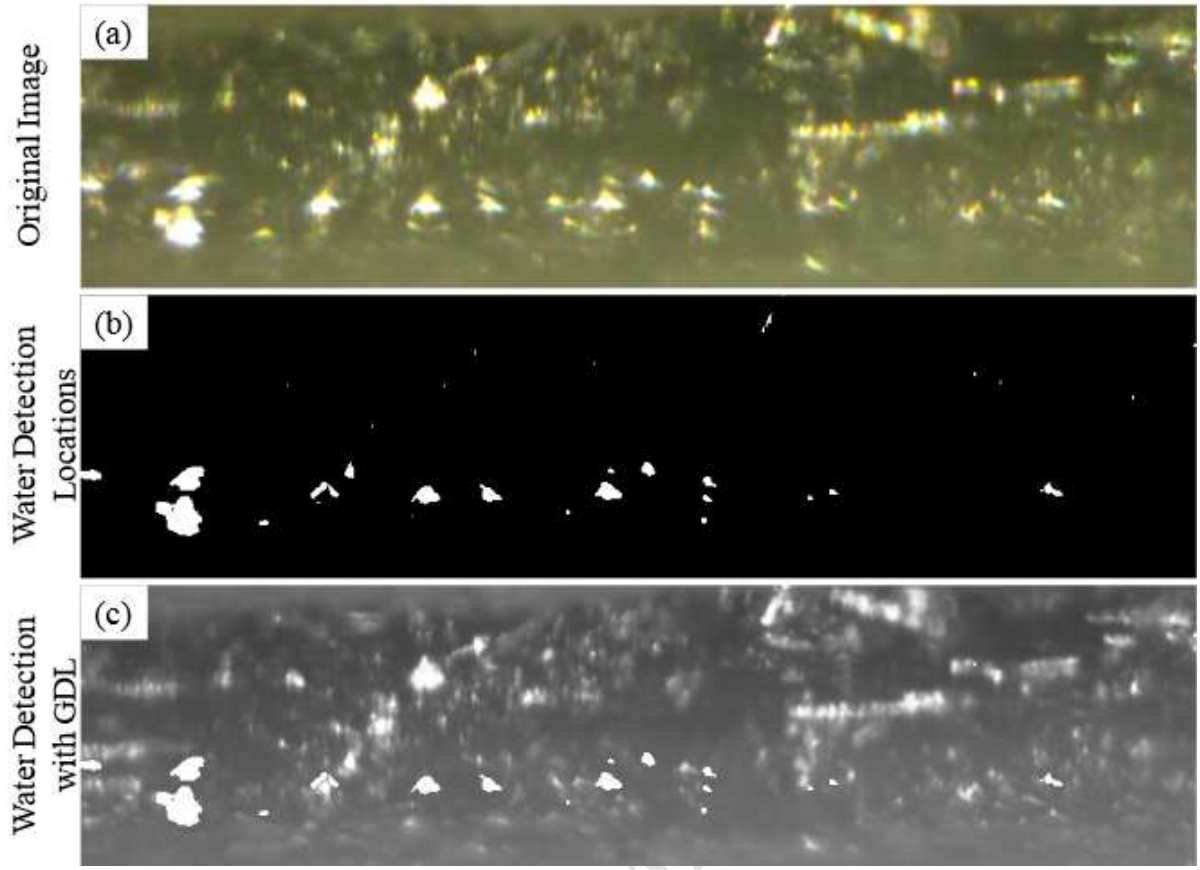


Fig. 10. Direct visualization of water on the cathode GDL cross-section (a) original image (b) water detection locations and (c) water detection with GDL [128].

Ous and Arcoumanis [129] investigated water build up at the anode and cathode regions of serpentine flow field channels at different operating conditions using direct visualisation techniques to examine the water droplets and slugs made in the flow channels. Hydrogen stoichiometry, air stoichiometry, electric load and cell temperature were studied in order to assess the formation and extraction of water out of the flow channels. The outcome shows that both hydrogen and oxygen stoichiometry do contribute to water accumulation at cathode flow field channels.

4.2. Neutron imaging

The neutron imaging technique was the only diagnostic tool recognized by Ballard to give all the three requirements (minimal invasiveness, in-situ applicability and ability to give local information) in water management [130]. The concepts of using neutron imaging techniques in the fuel cell is based on hydrogen ability to scatter neutrons creating specific finger prints for hydrogen consisting components like water [131]. Researchers have been using these

techniques to carry out experiments in-situ for the non-destructive analysis of PEM fuel cells [132-133]. Hickner et al. [115] used neutron imagining experiment to study water content at different operation conditions. They recorded that as temperature increases, this result into decrease in water content because of the evaporation of liquid water at a higher temperature.

Fuel cell industry is showing keen interest in neutron imaging techniques for visualisation of fuel cells due to the usefulness of the technique in investigating water distribution of an operating PEM fuel cell [134]. The technique is able to visualise water location in different flow fields during fuel cell operation. Neutron imaging techniques are also considered as useful tool to optimise the operating conditions and flow field in order to increase the efficiency of the fuel cell. Owejan et al. [135] investigated interdigitated flow field channel within a PEM fuel cell using neutron imaging techniques to observe water accumulation at the cathode region and the reported liquid water accumulation at the GDL. The neutron imaging techniques could be used to check the distribution of water which is helpful for proper water management in PEM fuel cell [136-137].

4.3. Nuclear magnetic resonance imaging (NMRI)

NMRI, also known as magnetic resonance imaging, is able to visualise water in opaque structures and there have been successful experiments carried out using this technique for the in-situ measurement of the distribution of liquid water in an operating fuel cells in which water could be spotted in the gas channels and land areas [138]. NMRI is both non-invasive and non-destructive technique which is useful in the observation of the properties of water transport inside the membrane of PEMFC [139]. Feindel et al. [140] investigated co-flow and counter-flow configuration using NMR microscopy and their result shows that co-flow configuration dehydrated at the PEM fuel cell inlets while counter flow distributed water uniformly in the fuel cell. Dunbar and Masel [141] used NMRI techniques to measure quantitative 3D water distributions in a fuel cell that is in operation. Dunbar and Masel [141] observed that at the cathode water was generated because of oxygen reduction reaction (ORR) and is first transported across the GDL before forming a big drop at the surface of the GDL.

Tsushima et al. [142] developed NMRI techniques to study liquid water that is supplied directly to the membrane of the fuel cell. They noticed that direct water supply to the membrane increased the cell voltage due to the low membrane resistance

4.4. X-ray Imaging

The X-ray imaging techniques have demonstrated that they can be used for the study of water management in PEM fuel cell [143-144]. Mankel et al. [145] investigated the behaviour of

water transport in a PEM fuel cell during its operation along with some plane resolution using x-ray imaging techniques. The researchers looked at the viability of using synchrotron radiation in observing the accumulation of liquid water across the cross section of the fuel cell. Mankel et al. [145] detected that at higher current, there was build-up of water at both anode and cathode regions close to the MPL and channel ribs. Lee et al. [146] used this technique to determine the thickness of water by attenuation of x-ray conventional source using x-ray camera and tube.

It was stated [Lee et al. [146]] that coupled device (CCD) camera is useful in acquiring direct X-ray images. A picture of X-ray imaging system is shown in Fig. 11.

Lee et al. [146] observed the rays attenuation is linear with respect to the thickness. Markotter et al. [147] investigated water distribution in PEM fuel cell in order to visualise water transport by means of x-ray imaging technique which has the ability of achieving a spatial resolution of 3-7 μm. Markotter et al. [147] observed that water was accumulated and discharged from the pores. The water distribution was uniform which indicates that the water was in continuous flow and the eruptive transport adds water droplets from the GDL to the channels. To obtain clearer images using this technique, a compact imaging system was developed using medical X-ray tube which functions as the source of light.

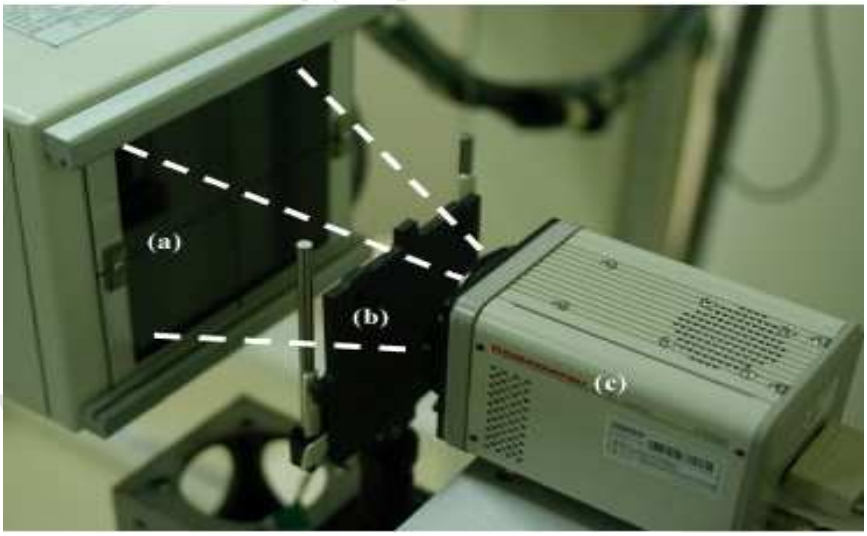


Fig. 11. An X-ray imaging system for a fuel cell experiments: (a) X-ray tube (b) fuel cell components and (c) X-ray CCD camera [146].

Kuhn et al. [117] also studied high temperature PEMFC applications using synchroton x-ray imaging techniques to give an insight of electrolyte concentration and distribution. Their research showed that important data such as humidification degree, mediation utilisation and temperature can be obtained from the electrolyte concentration and distribution. This technique has several potentials and can be employed in investigating water movement within a PEM fuel cell. However, there are challenges in achieving progress in using high temporal and spatial resolution in capturing liquid water droplets in the GDL [148].

Table 3 - Common methods of visualizing liquid water in PEM fuel cells

Methods	Enlightenments	References
Direct visualization	It is used directly in observing the operational conditions and their effects on water droplet growth, formation and movements.	[122]
Neutron imaging	This method in fuel cell was established on the sensitive reaction of neutron to hydrogen consisting of compounds like water.	[131]
Nuclear magnetic resonance imaging	This method is useful in measurement of in-situ for the distribution of water in operating fuel cells.	[138]
X-ray	This method is attenuation of synchrotron and non- synchrotron which have demonstrated that they could be used for the studies of water management in PEM fuel cell.	[143-144]

5. Ways that could prevent water flooding in PEM fuel cell

5.1. Operating temperature

In PEM fuel cell, mainly temperature and pressure determine if the water present is liquid or vapour and at higher temperatures vaporisation will intensify. Due to surface tension of liquid water being strong, water vapour is much easier to be removed from the cell than liquid water. Liu et al. [149] investigated water flooding and two phase flow of reactants and products in cathode flow field channels using a transparent PEM fuel cells and they concluded that when the cell is operating at low temperature there is a higher tendency of liquid water to accumulate in flow field channels than at high temperature. Formation of water columns is inevitable at low temperature due to water accumulation in the flow field channels. This water accumulation can hinder mass transport and reduce the catalyst area available for the electrochemical reaction. High temperature operation results in lower water

accumulation in the flow channels and water vapour is removed from the cell before condensation occurs.

M.Perez and V. Perez [150] carried out experiments with various fuel cell temperatures in a humidified 300W fuel cell stack operated on dead end mode andexperiments were performed in the 20^{o}C to 60^{o}C temperature range. M.Perez and V. Perez by studying the polarization curves they observed that the cell performance increases as temperature increased from 20^{o}C to 40^{o}C then became constant between 40^{o}C and 50^{o}C before decresing as the temperature increased further. In terms of voltages, the increase in the cell performance between 20^{o}C to 40^{o}C was due to the increase in gas diffusivity and conductivity of membrane. Thus, as the gas diffusivity increases, the cell performance increases. The drop in cell performance above 50^{o}C is due to the increased rate of water evaporation as temperature is increased. As there is more water evaporation than water production by the electrochemical reaction, the membrane starts to dehydrate and dry out leading to poor cell performance. However, with increased the humidification of the cell temperature increase from 20^{o}C to 60^{o}C improves the overall performance of the cell [150].

Wang et al. reported that if the cell operating temperature is higher than the temperature necessary for the fuel to be properly humidified, the cell will dehydrate and the cell performance will degrade [151]. Ozen et al. [152] considered the effects of operating temperature on cell performance and kept the humidification level for anode and cathode regions equivalent to those desired at at 70^{o}C then varied the cell temperature from 50^{o}C to 80^{o}C. The cell performance increases up to 70^{o}C which was attributed to reduction of activation losses. The cell performance dropped at 80^{o}C, which was higher than the maintained humidification temperature due to dryness in the membrane.

Natarajan and Nguyen [153] concluded that in order to avoid loss of cell performance, as a result of membrane dryness, anode humidification temperature must be increased if cell temperature increases.

5.2. Operating pressure

A fuel cell performance can be improved by increasing the pressure. If there is any water accumulation in the cell, increase in pressure will cause the reactant to force the water to flow. Kerkoub et al. [154] conducted experiments on the effect of pressure gradients between the electrodes and investigated their effect using various parameters on water management within a PEM fuel cell. Kerkoub et al. [154] noticed that when the pressure was increased the cell performance increased. They also noted that at the anode side, and due to the increase in pressure gradient, the membrane begins to dry out and as a result of this the protonic

membrane conductivity begin to reduce overtime and causes decrease in cell performance. Santarelli and Torchio [155] investigated the effect of operating pressure on water distribution of a PEM fuel cell and used backpressure to adjusting the operating pressure. Santarelli and Torchio [155] varied the reactant inlet pressure from 1.0 bar to 3.1 bar and they observed that when the pressure increases the cell performance increases and this was due to the increased flow rate of reactants.

Amirinejad et al. [156] experimentally studied the effect of operating pressure between 1 atm to 3 atm within a PEM fuel cell of an open circuit voltage and they noticed an increase in cell performance as the pressure increased according to Nernst equation. The increment was due to the increase of reactant gases flow and diffusivity which helped improve water management of the cell and reduced mass transport resistance.

5.3. Replacing conventional flow field channels with open pore metal foam

In conventional flow field channels the presence of ribs, channels and dead end mode where liquid water could accumulate may result in a non-uniform distribution of reactants and water flooding is distinct possibility. Open pore metal foam is being seen as a substitute to conventional gas flow fields of fuel cells due to the absence of ribs, channels and dead ends which could reduce or solve the issue of water flooding in the fuel cell. Open cell metal foam, as shown in Fig. 12, possess a high efficient thermal conductivity, porosity up to 99%, high specific surface area of almost 10,000 m^2 m^{-3} and a random flow path which helps the reactant reach the catalyst surface to enable the electro-chemical reaction [157].

Fig. 12 - Open pore metal foam

Su et al. [158] conducted an experiment using different conventional flow fields channels in PEM fuel cell such as parallel, interdigitated, serpentine flow fields. In the parallel flow field flooding starts to happen once liquid water appears in the central channels. Water will continue to accumulate until the channels eventually become blocked. For the interdigitated flow field, flooding occurs mostly in the downstream channels than the upstream channels. While in the serpentine flow fields it was notice that the corners of the channels are more prone to flooding than the upstream and downstream channels. Yang et al. [159] studied the characteristics of fuel cell performance at dead ended anode using three various flow fields by measuring the local current densities and fuel cell voltages operating modes and they concluded that the parallel flow and interdigitated flow field water accumulation is very high which leads to water flooding but in the serpentine flow field water accumulation is less.

Tseng et al. [160] replaced the conventional flow field channels with open porous metal foam in PEMFC and the results obtained by them showed that using metal foam as flow distributor offers exceptional mass transport properties and better convective gas flow with minimum flow resistance than the conventional flow channels.

Baroutaji et al. [161] conducted a Computational Fluid Dynamic (CFD) analysis between a serpentine flow plates and open pore cellular metal foam flow field. They observed that there was a large pressure drops in serpentine flow field plates because of the velocity disturbances at the ribs which affect the reactant flow, and there were low velocity flows in the channel edges and dead end mode. In contrast, there was low-pressure drop in the open pore cellular metal foam flow field and velocity disturbances were at minimal due to its high porosity.

Tabbi et al. [29] carried out a computational fluid dynamic modelling study to investigate the characteristics of flow field channels and compared serpentine flow field channels and open cellular metal foam flow. They concluded from their results that liquid water accumulation is likely to occur in the serpentine flow channels due to the dead end zone which will prevent some reactants from reaching the MEA, while this can be avoided using open cellular metal foam flow which will improve the general fuel cell performance.

Carton and Olabi [162] conducted an experiment for double flow field channel and developed a 3D computational fluid flow dynamic model for metal foam and compared the results for a PEM fuel cells. The results point out that there was water accumulation in the ribs of the double flow channels which has the potential to lead to local water flooding. In the metal

foam water is distributed uniformly and water accumulation is minimal which will prevent water flooding.

5.4. Micro porous layer (MPL) within the cell

Microporous layer help in reducing liquid water saturation from CCL to the flow channel across the GDL of the cathode [163-164]. MPL can hinder the water condensate from moving from the channels or GDL back to the catalyst layer. Again, they provide electrical contact between catalyst layer and GDL, and reduces the quick dry out of the membrane at low humidity [165-166]. The importance of MPL is obvious mostly at higher current state which shows that it improves mass transfer. MPL have a way of distributing liquid water favourably for gas phase transport in the cell [167]

Kim et al. [168] investigated micro-porous layer (MPL) assembly in PEM fuel cell as a function of elctrochemical losses and investigated the use of MPL at cathode only, both sides of the electrodes and without MPL. It was seen that the cells with MPL on cathode side and at both sides performed better because it helps force the liquid water out of the cell, aid back diffusion and reduces gas diffusion layer liquid saturation at high current densities. The EIS response comparison shows that the addition of MPL in the cells reduces charge transfer resistance, mass transport resistance and ohmic resistance when compared without MPL in the cell. They concluded that MPL helps manage water in a PEM fuel cell.

Chen et al. [169] conducted similar experiment as mentioned earlier and they concluded that the use of MPL reduces water loss to flow field channels and complement back diffusion which help in membrane hydration.

Blanco and Wilkinson [170] studied the effect of microporous layer on water management using novel diagnostic method in PEM fuel cell. They conducted an experiment with the use of cathode MPL and without MPL in the cell. The test conducted indicated that the MPL ameliorate the cell performance as a result of water saturation in the CCL which leads to improved oxygen diffusion and increases back diffusion as shown in Fig. 13 . It was also seen that anode pressure drop increases with the use of MPL than without MPL.

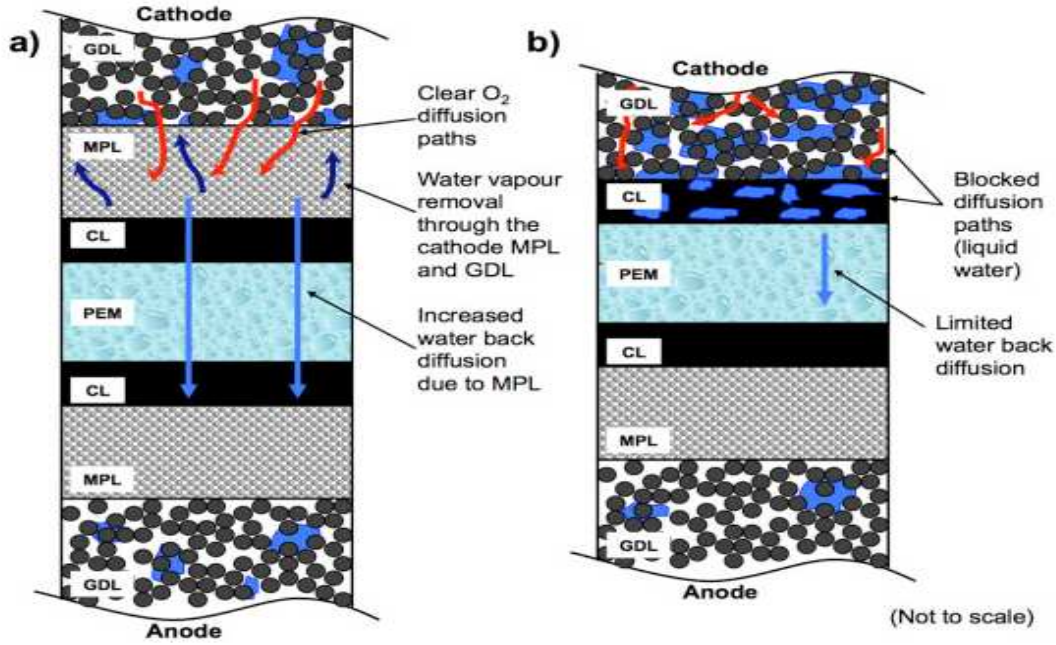


Fig. 13: Schematic water transport of (a) with MPL (b) without MPL[170]

Deevanhxay et al. [171] studied the effect of water in gas diffusion media (GDM) with and without MPL on cell performance. Deevanhxay et al. [171] concluded that liquid water was found on the CL and GDM of the cell without MPL which had a critical effect on the overall cell performance. However, there was reduced water accumulation on the CL and GDM surface of the cell with MPL which result in a better cell performace. Fig. 14 shows GDM without MPL and with MPL. Pasaogullari and Wang [172] experimental results also confirm that using MPL in the cell enhances the removal of liquid water and prevent the water from covering the active area of the catalyst surface for electrochemical reaction. Tseng and Lo [173] investigated the effects of MPL on water management and cell performance by using a commercial 25cm^2 catalyst coated membrane along with a MPL and GDL in the single fuel cell assembly. Tseng and Lo [173] concluded from their experiment that the use of MPL increases the cell performance, especially at high current density because the mass transfer limitation was eliminated.

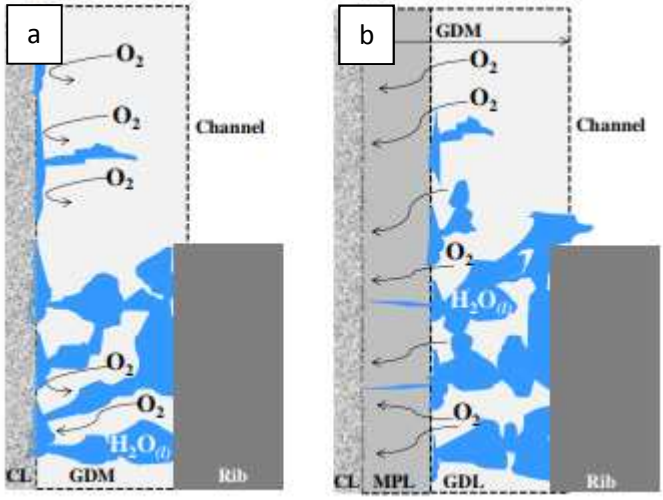
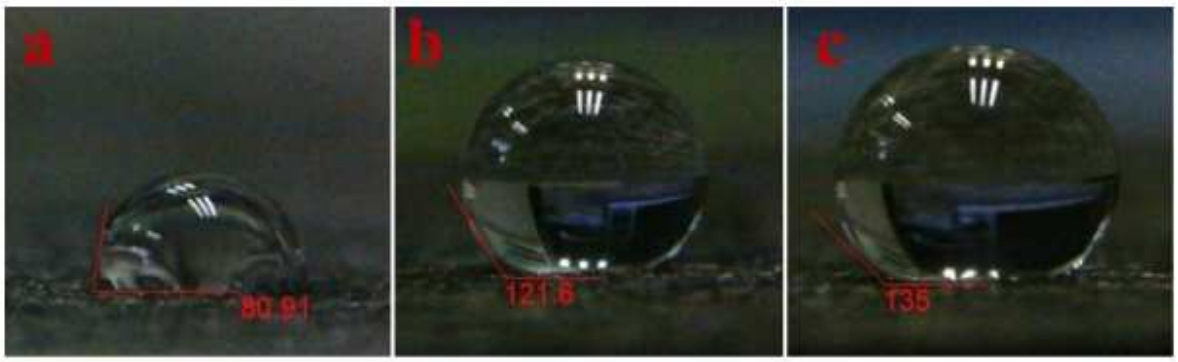


Fig. 14: (a) GDM without MPL (b) GDM with MPL [171]

5.5. Enhancing the hydrophobicity of Gas diffusion layer (GDL)

Several research findings shows that the coating of GDL with a hydrophobic agent like fluorinated ethylene propylene (FEP) or polytetrafluoroethylene (PTFE) help in facilitating the removal of liquid water from the cell [174]. The untreated GDL hold more water due to its hydrophilic nature when compared to coated GDL and this result in more susceptibility to water flooding in the GDL or membrane hydration, and increased mass transport resistance [175-176]. Chen et al. [177] studied various PTFE coatings on GDL to see tthe effect on cell performance. The contact angle measurement for the hydrophobic GDL material of different PTFE content was made by using a digital microscope of high resolution. It was seen that the higher the PTFE content in GDL material, the higher is the liquid water droplet contact angle as shown in Fig 15. This means that the hydrophobicity of GDL material increases with increase in the contents of PTFE. The hydrophobicity of the GDL material is good for water management because it helps remove liquid water from the cell.



(a-0wt% PTFE, b-10wt% PTFE, c-20wt% PTFE)

Fig. 15 Liquid water droplet contact angle (a- PTFE 0wt%, b-PTFE 10 10 wt%, c- PTFE 20 wt%) [177].

A similar experiment was performed by Kakaee et al. [178]. Their results show that PTFE coated GDL helped in the removal of liquid water which avoid water flooding of the GDL. Chen et al. [179] employed PTFE to treat the surface of GDL so as to study the water behaviour in a PEM fuel cell. Their findings show that uncoated GDL get wet easily than the treated GDL especially at low operating conditions.

Chan and Wang [180] investigated the effect of different volume of fluorinated ethylene propylene (FEP) coating varying from 10 to 40 wt. % on the GDL. It was noticed that the 10 wt. % FEP treated GDL performed better due to a better hydrophobic surface and it facilitated water removal, while the others FEP content over 10 wt.% blocked the GDL surface pores which caused mass transport limitations. In another experiment Hasanpour et al. [181] compared a woven GDL and a coated woven GDL with FEP. Their results show that liquid water flow is higher in FEP coated GDL than the uncoated.

5.4 Effect of Water management on characteristics performance of PEM Fuel cell

An investigation conducted by a group of researchers in Republic of Korea showed that the performance of a fuel cell was subject to the moisture content in the membrane at varying operating conditions [182]. The outcome of their investigation captured in Fig. 16 and Fig. 17 explains the performance of the fuel cell at 100 percent relative humidity and 50% relative humidity.

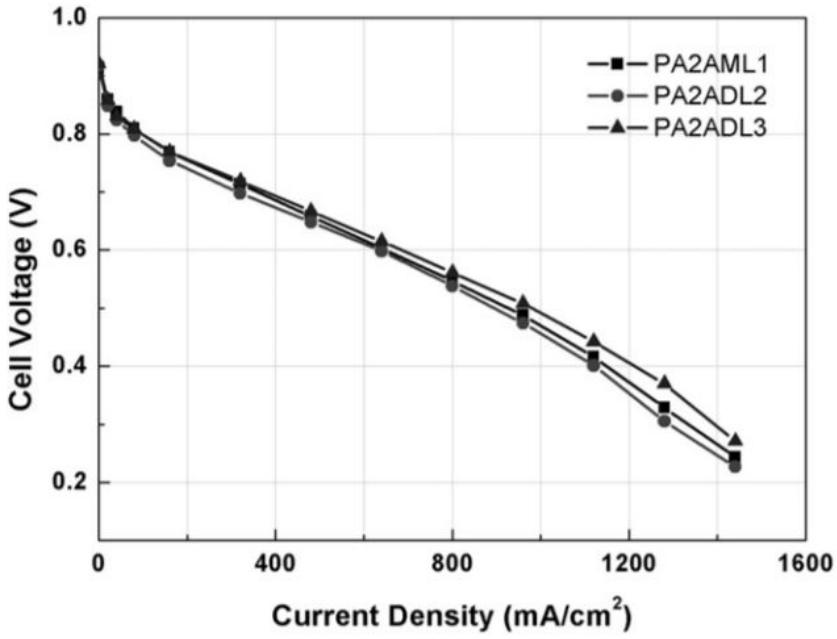


Fig. 16. Polarization curves of fuel cell performance at 100% humidification conditions [182].

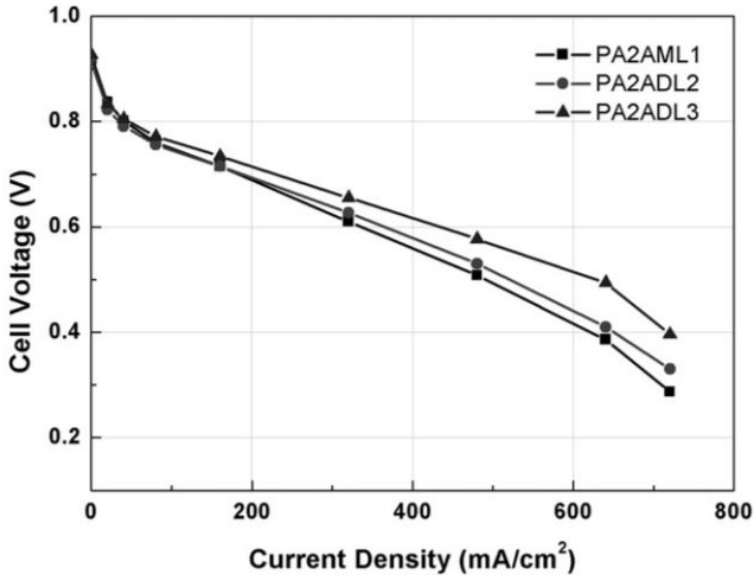


Fig. 17. Polarization curves of fuel cell performance at 50% humidification conditions [182].

The findings indicate the key role water management plays in terms of the overall characteristic performance of the fuel cell. It is imperative that the water produced at the cathode catalyst layer is eliminated particularly at high humidification conditions to prevent the fuel cell becoming flooded. Stagnation of water in the fuel cell implies that a portion of the membrane electrode assembly will not participate in reaction hence reducing the current density being generated from the fuel cell [183]. Subin et al [184] also conducted several experiments on fuel cell performance at varying operational conditions. From Fig. 18, it is observed that the fuel cell performance increases appreciably between cell operating temperatures of 45^{o}C – 50^{o}C. When the fuel cell is operated at cell operating temperature of 45^{o}C, the performance is reduced at low current densities. This is because the electrochemical kinetics at this cell operating temperature is poor. The performance of the fuel cell at higher current density is observed to be better at cell operating temperature of 45^{o}C compared to that of 50^{o}C. At 50^{o}C, there is decrease in fuel cell performance at high current density due to many factors. The researchers argued that flooding, dehydration of the membrane electrode assembly and reactant starvation, all directly related to water management in the fuel cell, were some of the reason for this fuel cell characteristics.

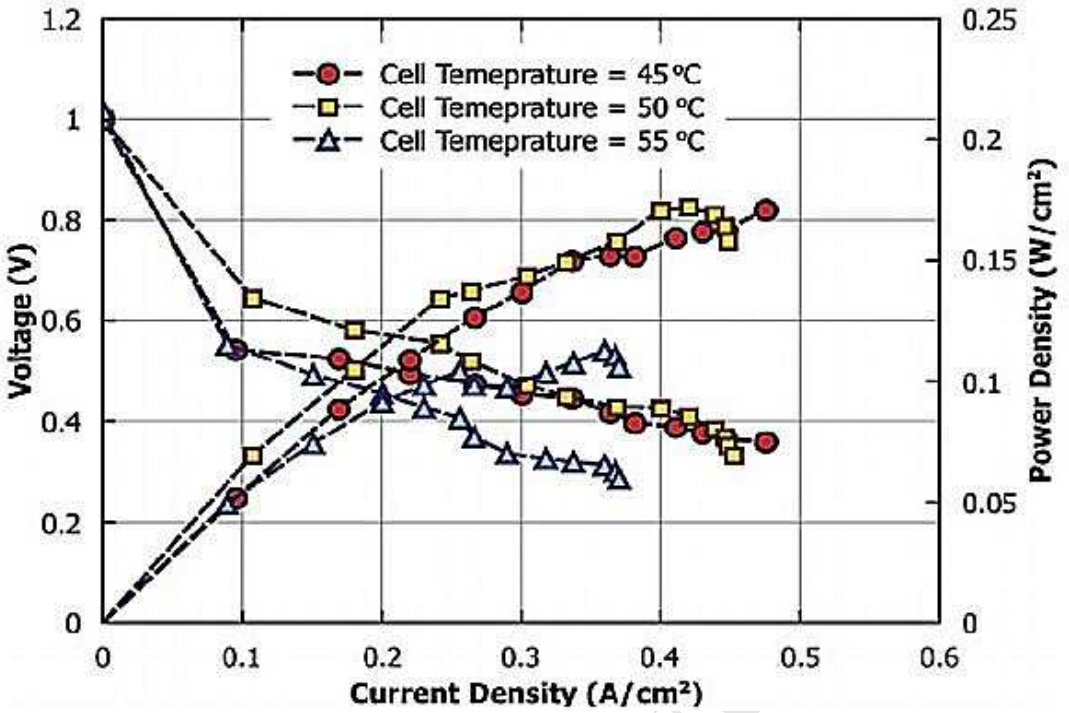


Fig. 18. Performance of a fuel cell at varying cell temperature [184].

The voltage generated from the fuel cell is significantly affected by membrane dehydration especially when the cell operating temperature keeps increasing [185]. Atlower cell operating temperature, there is condensation of water which eventually accumulates leading to flooding in the fuel cell. When the current density is kept constant at varying cell operating temperature (in this case 45 ^{o}C – 50^{o}C), the water produced will be same for all operating conditions. Again, the rate of reactant that will go into reaction will also be high for both conditions since rate of reaction increases with temperature. Water accumulation in the cell also increased between 45^{o}C and 50^{o}C at high current densities. This indicates that there is an optimum cell operating temperature which can reduce condensation of water and rapid evaporation of water. This will help reduce water flooding and membrane dehydration which will reduce the overall performance of the fuel cell. Li et al [26] used cathodic pressure drop of PEM fuel cell to determine the water management characteristics in a fuel cell. They explained that regulating the fuel cell temperature, inlet pressure and the inlet relative humidity were strategies to effectively regulate the water management characteristics in a fuel cell. Electrochemical impedance spectroscopy (EIS) was used to determine the amount of water in the cell. There was no flooding after the fuel cell was regulated using the three (3) strategies presented by Li et al [26]. Increasing the pressure at the inlet by 25 kPa helped the voltage to recover 25 mV and this stabilized the performance of the fuel cell. Increasing the

temperature by 2 K also caused an 8 mV to be recovered as well, maintaining the performance of the fuel cell. The range of recovery reduced when the inlet relative humidity was used as a strategy to maintain the water management characteristics in the fuel cell. For most commercialized fuel cell facilities, a change in the inlet pressure is one acceptable mean of reducing flooding in the fuel cell. Fig. 19 shows the fuel cell characteristics at varying pressure drop and voltage during flooding conditions in the cell.

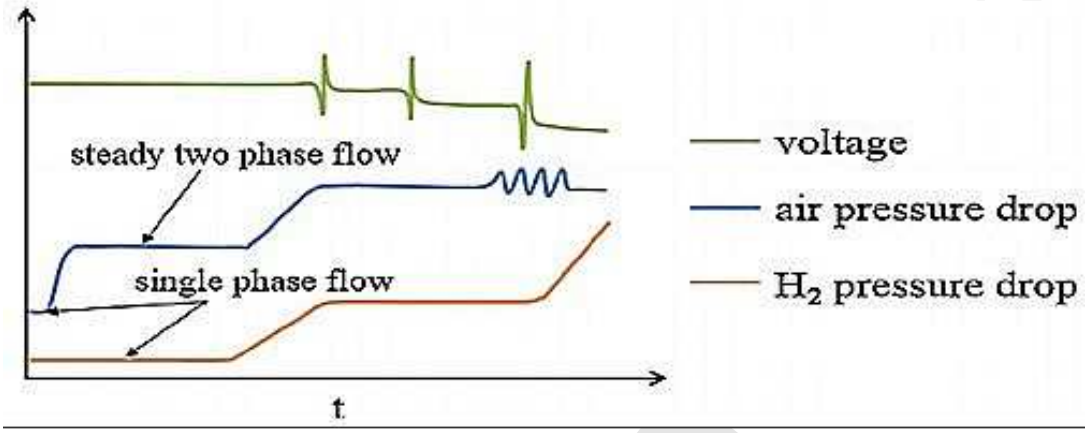


Fig 19. Characteristics of a fuel cell pressure drop and voltage during flooding in the cell [186].

Li et al [26] presented Fig. 20 to demonstrate water management using the cathodic pressure drop strategy through the regulation of the fuel cell temperature. They argued that there is a steady water state in the fuel cell where the pressure drop for two phase flow and voltage becomes stable for a longer period. The pressure drop can therefore be used as a reference point for controlling the water management in the cell [135]. Li et al [26] deduced further that should the real pressure drop exceeds the theoretical line, the water in the fuel cell will increase and the fuel cell is likely to become flooded. The operating temperature of the fuel cell at these operating conditions is increased to help evaporate excess water from the cell. The pressure drop from Fig. 20a shows that the pressure drop has exceeded the theoretical line by 3 times hence the temperature increased by three. The temperature was increased by 2 K for the first two scenarios and 4 K for the third case. The pressure drop reduced as the temperature increased.

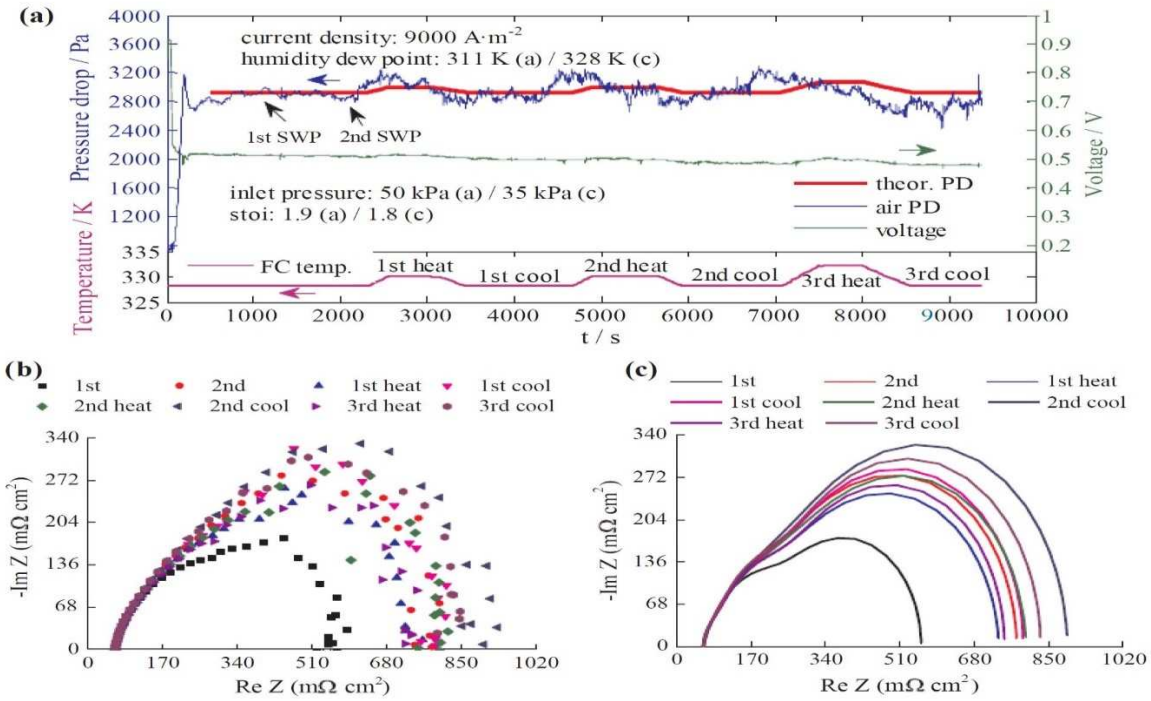


Fig. 20. Regulation of fuel cell temperature to enhance the water management a) Varying pressure drop, voltage and cell operating temperature during water management [134].

From Fig. 20c, it is observed that there is apparently no direct link between high frequency resistance and flooding. It can be observed that the low frequency resistance is directly affected by the amount of water. The low frequency resistance increases from EIS sweep 1 to EIS sweep 2. The low frequency resistance then decreases afterwards when the fuel cell heats up. It implies that there was change in the amount of water as the temperature was being regulated.

Fig. 20 shows a marginal increase in voltage as the temperature increased. If this control strategy was not applied, the fuel cell was likely to become flooded reducing the voltage during the three hours that the experiment was conducted. Fig. 20 also shows voltage obtained at various water management conditions.

The redline in fig. 20 also indicates the theoretical pressure drop, blue line shows the actual pressure drop, the green line shows the fuel cell voltage and the purple line shows the cell operating temperature b) EIS results obtained at specific cell operating times c) Curve fit for results generated in b [26]. Fig. 21 also explains the change of voltage with respect to time at varying cell operating conditions.

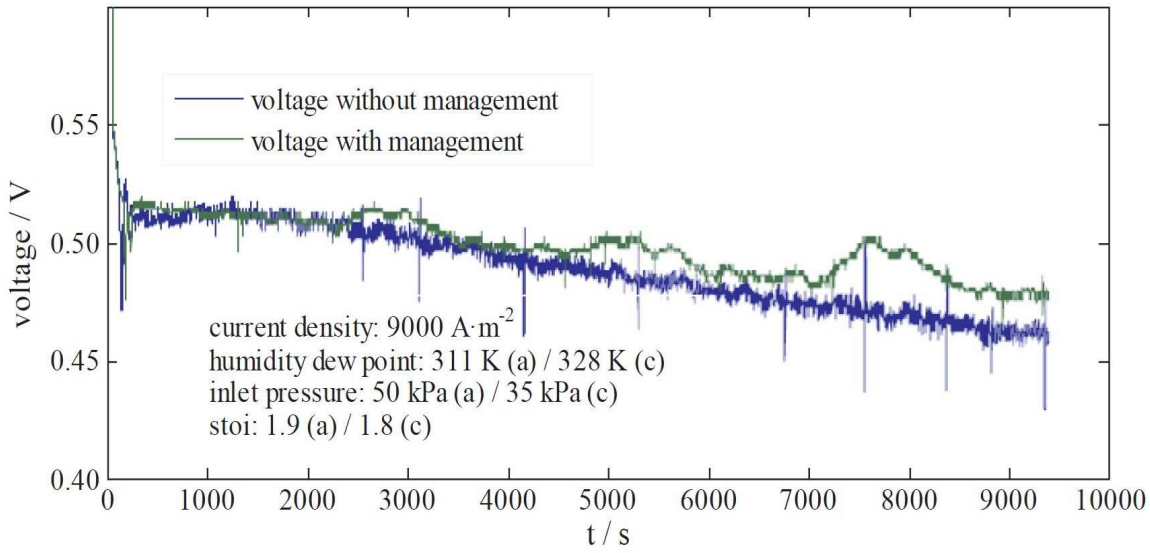


Fig. 21. Comparing the voltage obtained for regulating the cell temperature with respect to water management [26].

Changing the inlet pressure and relative humidity is also captured in Fig. 22 and Fig. 23 in a manner similar to changing the cell temperature. There is a significant improvement in the voltage value for all these conditions because voltages did not exhibit frequent variations and less drop in performance was experienced compared to the flooded operation mode. Increasing the inlet pressure caused the three step downs in the graph in Fig. 23. The impedance also decreases whenever the inlet pressure goes up. Fig. 24 shows the voltages generated varying with the water management conditions mentioned earlier. In comparison with a fuel cell operated under normal conditions, the results obtained using the three strategies did not indicated any sign of voltage drop. The three strategies can help curb flooding in a fuel cell and increase the running time for a fuel cell at varying operating condition.

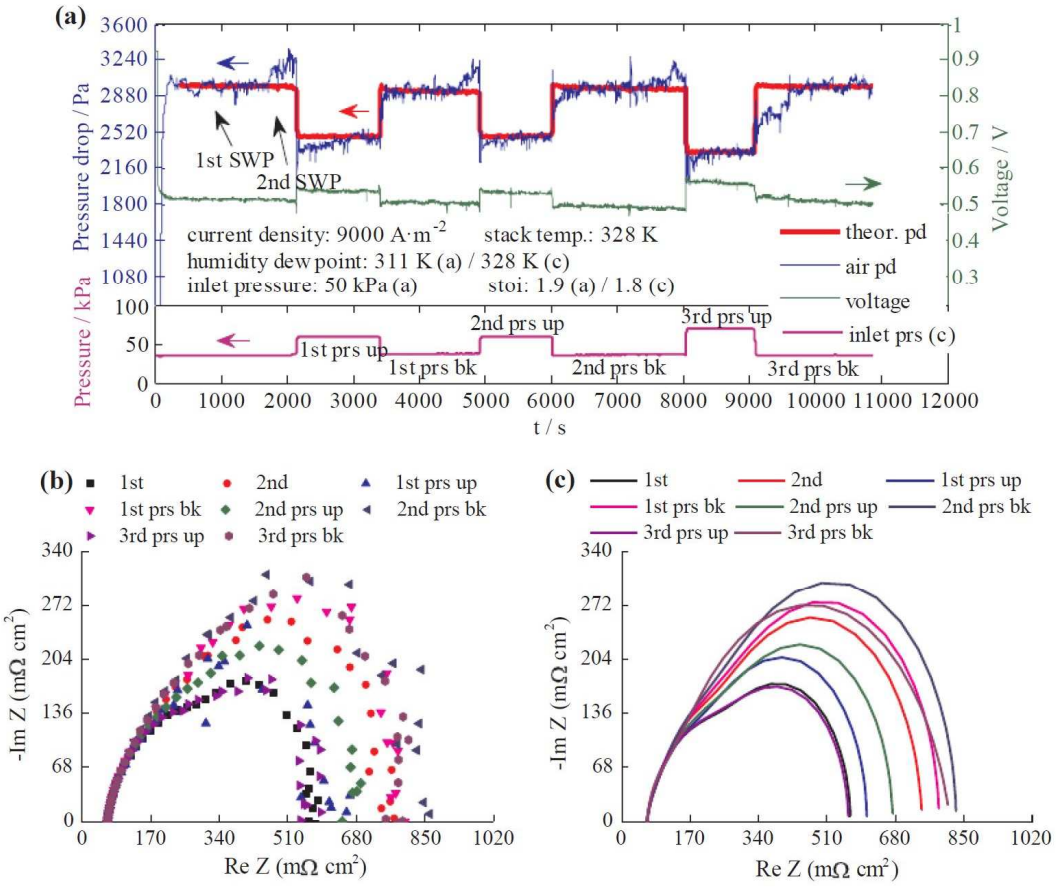


Fig. 22. Water management control strategy using cathodic pressure drop a) Varying pressure drop, voltage and cell inlet pressure during water management b) EIS results obtained at specific cell operating times c) Curve fit for results generated in b [26]

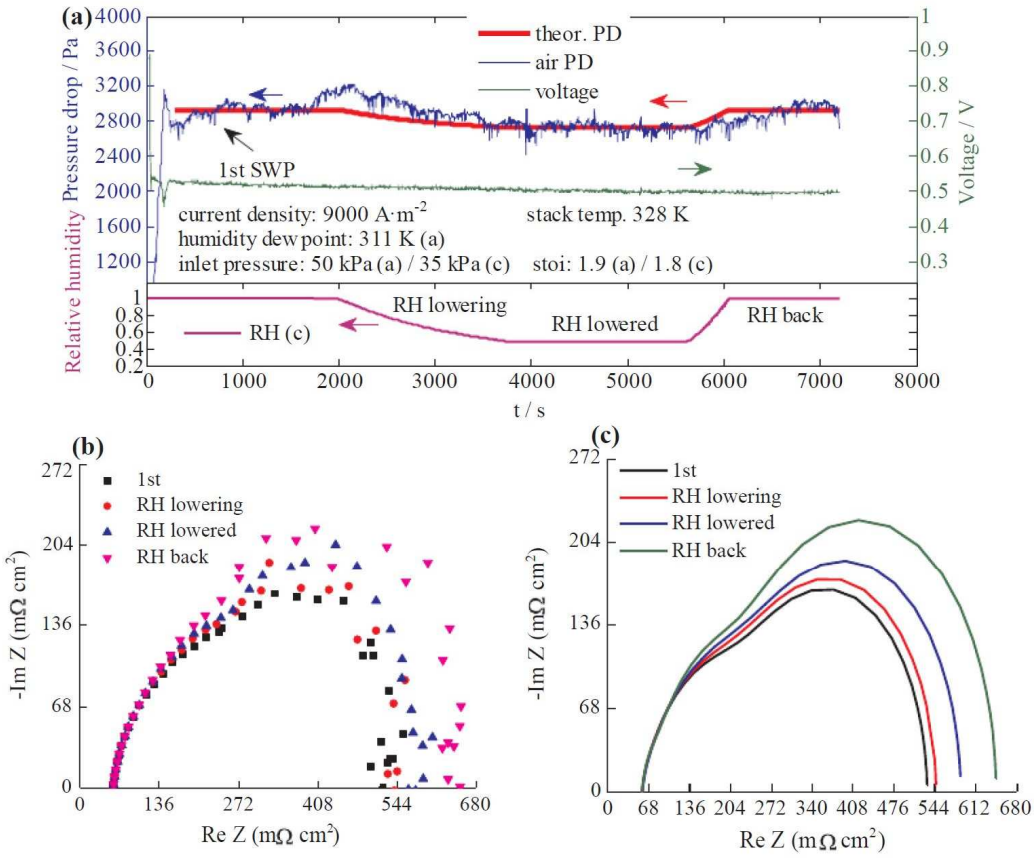


Fig. 23. Water management control strategy using cathodic pressure drop a) Varying pressure drop, voltage and cell inlet relative humidity during water management b) EIS results obtained at specific cell operating times c) Curve fit for results generated in b [26]

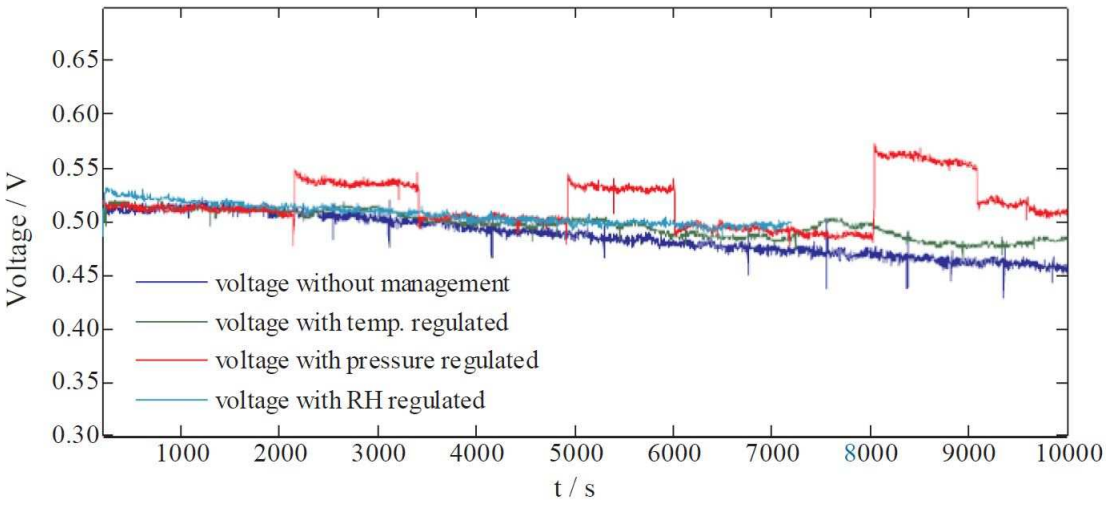


Fig. 24. Voltage comparison for different water regulation strategy [26]

6. Conclusions

The commercialisation and the wide use of fuel cells technology is not only dependent on cost, and their efficiency and longevity will be deciding factors in their successful adoption.

With water being the major by-product of electro-chemical reactions in fuel cells, water accumulation and subsequent fuel cell flooding remain important factors in the operation of PEM fuel cells and the degradation of their efficiencies.

This work provided and expose of the phenomena and discussed the different mechanisms for water transport in PEMFCs and the effect on water flooding of the different regions of the fuel cell. Research reviewed by this work highlights the importance of development of good knowledge of all water transport mechanism and consider their relative importance when optimising fuel cells operations and not only concentrate on electro-osmotic drag and back diffusion. When temperature gradients are experienced within the fuel cell it is important to consider the effect of thermal osmotic drag. Similarly, when large pressure drops exist hydraulic permeation becomes a factor in cell operation and performance.

Recent research also stresses the importance of the formulation of the PEM material as well as its mechanical configuration. Increasing hydrophobicity plays an important role in preventing water accumulation at membrane surfaces and masking of catalyst sites. The use of other mechanical barriers is also conducive to the cell operation.

The design of the flow channels is of paramount importance in improving the efficiency of the fuel cell and the improvement of its efficiency. Cells operated at low pressure drops do not always provide good water management and they are prone to blockages of their flow channels. Other designs that ensure water is clearer from the fuel path need to be operated at higher pressure drops and also may not be efficient in fuel use due to the requirement of large flow rates. Interest in the use of porous foams for the flow plate is increasing given their superior ability to distribute gasses, eliminate water accumulation, and their operation at low pressure drops.

The adoption of analytical techniques from other disciplines such as neutron imaging and magnetic resonance imaging are providing reliable tools to study water behaviour in fuel cells

and to develop better fundamental understanding that can aid in the development of better designs and optimised operation strategies.

Further research is required to help produce better designs for fuel cells flow plates and also to develop better understanding of the effects of the combination of the different operating parameters that influence the overall efficiency of fuel cells.

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Paper Highlights

1. Effect of water flooding in fuel cells is critically investigated.
2. Effective water management procedures to increase fuel cell performance is discussed.
3. Techniques for the visualization of liquid water in fuel cell is presented
4. Effect of porous material in terms of water management in fuel cells is presented.
5. Overall energy efficiency improvement has been introduced.