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Electrode for proton exchange membrane fuel cells: A review

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ABSTRACT

The electrode is the key component of the membrane electrode assembly (MEA) of proton exchange membrane fuel cells (PEMFCs). The electrochemical reaction of hydrogen (fuel) and oxygen that transform into water and electrical energy occurs at the catalyst site. Attempts to improve the performance and durability of electrodes have sought to overcome the challenges arising from utilizing PEMFCs as an efficient and competitive energy source. To accomplish this goal and to solve the problems related to using PEMFC electrodes, the structure and function of each component and the manufacturing method must be comprehensively understood, and the electrode performance and durability of the cell must be characterized. Therefore, in this paper, we discuss the components, preparation, functions and performance of the electrodes used in PEMFCs. This review aims to provide comprehensive information regarding PEMFC electrodes.

1. Introduction

The proton exchange membrane fuel cell (PEMFC) is the most promising candidate for renewable and sustainable energy conversion devices due to its zero CO₂ emissions. This technology is expected to be an efficient energy source that is free of pollutants, and it has a high energy density compared to conventional energy sources. Considering the global energy demand from the human population, the threat of fossil fuel shortage is a major concern. Therefore, extensive research and development have been focused on renewable energy sources as well as reducing of CO₂ emissions. PEMFCs are widely used as clean energy conversion devices, especially in vehicles and in stationary and portable power generation systems. Because of their power density, energy efficiency, lack of pollutant emissions and low operating temperature [1–7]. However, the commercialization success of PEMFCs depends on their ability to demonstrate optimal fuel to electricity conversion with a high current density [8].

In PEMFCs, hydrogen (H₂) gas feed at the anode is oxidized to

release protons and electrons. The electrons generate electricity at the external circuit connected to the load. The hydrogen ions (protons) migrate through the polymer electrolyte (proton exchange membrane) to recombine with electrons and oxygen to produce water at the cathode. There are two important key factors that slow the development and commercialization of PEMFCs: cost and durability. The high cost of PEMFCs is largely due to the use of noble metals (platinum) as catalysts, which accounts for 55% of all PEMFC manufacturing costs [9]. While researchers are currently seeking alternative catalysts, platinum is still the most commonly used catalyst because it is very effective and has a high chemical stability, exchange current density and work function [10]; however, platinum is expensive and easily poisoned [11]. Therefore, the development of fuel cells is directed toward developing new electrodes and reducing the use of platinum catalysts [11,12], perhaps even by replacing platinum with a non-noble metal, such as Co, in the anode [9]. Although their performance is not as high as that of a platinum catalyst, Co catalysts in PEMFCs may decrease the cost of PEMFC manufacturing. The durability of a PEMFC varies

Abbreviations: AB, acetylene black; AST, accelerate stress test; BL, backing layer; CCM, catalyst coated membrane; CFGDL, carbon-filled gas diffusion layer; CL, catalyst layer; CV, cyclic voltammetry; CNF, carbon nanofibers; CNT, carbon nanotubes; DOE, department of energy; DSO, dimethyl silicon oil; ECSA, electrochemical active surface area; EIS, electrochemical impedance spectroscopy; EDX, energy dispersive X-ray; FE, four electrode; FPE, four point electrode; FEP, fluorinated ethylene propylene; FCTESTNET, fuel cell testing and standardization thematic network; GDB, gas diffusion backing; GDL, gas diffusion layer; GDM, gas diffusion media; HALS, hindered amine light stabilizers; HDP, homogeneous deposition precipitation; IBAD, ion beam assisted deposition; LSV, linear sweep voltammetry; MPL, macroporous layer; MEA, membrane electrode assembly; NIOSH, national institute of occupational safety and health; OCV, open circuit voltage; ORR, oxygen reduction reaction; PBI, polybenzimidazole; PFSA, perfluorosulfonic acid; PTCA, perylenetetracarboxylic dianhydride; PPA, polyphosphoric acid; PTFE, polytetrafluoroethylene; PVDF, polyvinylidene fluoride; PED, preferential pulsed electrodeposition; PEMFC, proton exchange membrane fuel cells; SEM, scanning electron microscope; TEM, transmission electron microscopy; XPS, X-ray photoelectron microscopy; XRD, X-ray powder diffraction

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according to its application mode. In 2015, the DOE (Department of Energy, USA) declared a lifetime target of 5000 h for vehicle energy systems and 40,000 h for stationary power systems. However, the actual PEMFC technology achievements to date are 1700 and 10,000 h, respectively [13]. Some durability test standards have been developed by the DOE and the Fuel Cell Testing and Standardization Thematic Network (FCTESTNET). In addition to endurance tests, accelerated tests are also used for conditioning or fuel cell incubation to achieve optimum conditions. Depending on the membrane electrode assembly (MEA) type, conditioning can require several hours or even days. Some theoretical aspects and considerations for conditioning include activating and cleaning the catalyst from impurities, cleaning the remaining catalysts and membranes from the dry and wet membrane electrode structures and activating the Nafion ionomer as a proton conductor [14]. A commonly used method involves heating the cell without a gas supply followed by applying a shortcut connection between the anode and cathode for a few minutes. After the hydrogen and oxygen gas flow and the voltage reach 1.0 V OCV (open circuit voltage), the fuel cell is operated under a load for 6 h. The conditioning process is considered complete when the voltage changes by < 1 mV.

Discussing the structure, components, manufacturing, and characterization methods of electrodes is very important for highlighting and solving the problems related to PEMFC electrodes. Researchers have attempted to investigate electrodes to improve their performance and durability, but a limited number of review papers exist regarding all aspects of the electrode. For example, Litster et al. [12], focused on the catalyst layer (CL) in a very constructive manner but presented a limited discussion on the gas diffusion layer (GDL). In fact, the current research has focused on the durability of electrode materials, but the degradation of the PEMFC cannot be avoided. However, the degradation can be minimized by understanding the degradation mechanism and cell components [14].

This paper intends to review the prior studies on electrodes regarding the function of the catalyst components, manufacturing and characterization methods, electrode performance and cell durability to provide a reference for future electrode researchers. Moreover, the aim of this work is to provide a critical review as well as a comprehensive discussion on the electrode synthesis process, the function of electrode components in terms of cell performances, and electrode system management that could be effective for PEMFC researchers.

2. Electrode structure and components

Generally, electrodes are constructed with three layers: the backing layer (BL), gas diffusion layer (GDL) and catalyst layer (CL) [15]. Some researchers consider that an electrode is composed of two layers, the GDL and CL, while the GDL consists of two layers, namely, a macroporous layer (MPL) of carbon powder and hydrophobic / hydrophilic agents and a backing layer composed of carbon paper or carbon cloth. The electrode structure according to Park [16,17] is presented in Fig. 1.

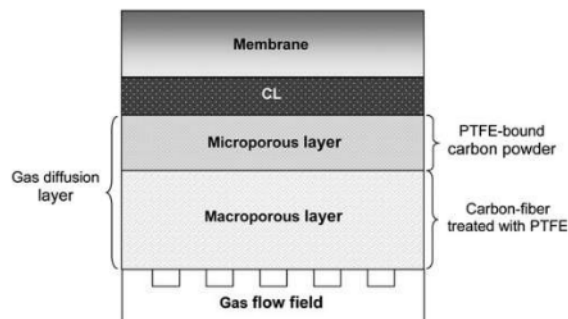


Fig. 1. Basic structure of PEMFC electrode (Reprint permission from Ref. [16]).

During electrode preparation, the GDL (GDL = BL + MPL) and CL should be carefully considered. The MPL that supports the catalyst layer should have the following features: high electrical conductivity, good interactions as a catalyst support, a high surface area, the ability to repel water and prevent flooding, corrosion resistance and the ability to easily restore catalyst functions [10]. The GDL and CL can be prepared using several methods, including casting, painting and brushing [4,19]; injection molding [5,20]; impregnation [11,21]; spraying [17,22]; thin layer deposition; using a catalyst coated membrane (CCM) [23,24]; and electro deposition [25,26].

In addition to serving as electrochemical reaction centers, electrodes must continuously allow electrons to flow from anode to the cathode side. In accordance with this function, an electrode must fulfill three aspects. First, the electrode must have appropriate pores for the reactants. Second, the electrode should contain a chemical catalyst to break the bonds of the fuel to form more reactive ions. Third, the electrode must conduct electrons to the external circuit. A proper PEMFC electrode structure has three phases: the gas phase reactant, catalyst particles and ionic conductors. The catalyst particles must directly contact the electron conductor. Electrons will flow through the carbon on which the catalyst particles are immobilized [27].

In MEA, the electrodes included an anode and a cathode. The anode is the site where the hydrogen gas (feed) is oxidized and split into protons (H^+) and electrons. Protons can pass through the proton exchange membrane to the cathode site. In contrast, electrons flow through the outer circuit to reach the cathode, thus generating electrical energy. Meanwhile, oxygen from the air is reduced at the cathode to produce water (H_2O). The electrochemical reaction that occurs at the cathode is $4H^+ + O_2 + 4e^- \rightarrow 2H_2O$ as shown in Fig. 2.

Many electrode studies have focused on the electrochemical reactions that occur on the cathode [21,28–34]. For example, the oxygen reduction reaction (ORR) is slower than the hydrogen oxidation reaction [35], requires a high catalyst content to maintain a tolerable response speed [32], requires a large over potential and causes the loss of 80% of the cell voltage [36]. In addition, PEMFC applications (especially for vehicles) require more oxygen from the air at the cathode. Since the oxygen content in air is approximately 20%, more oxidant intake is required, and the catalyst must have a high affinity for oxygen. Due to the increased oxidant flow, more impurities are introduced that can cause catalyst poisoning. Yu et al. [36,37] comprehensively discussed the catalyst activity and durability of Pt/C catalysts for PEMFC cathodes. Moreover, many researchers [38–42] have focused on catalyst poisoning by CO and H_2S gases at the anode. Meland et al. [43]

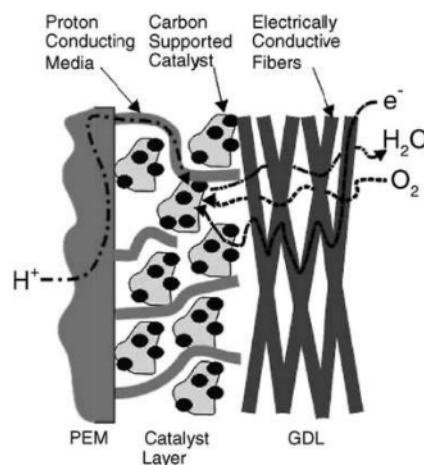


Fig. 2. Electrochemical processes forming water at the cathode (Reprint permission from Ref. [12]).

suggested that CO poisoning of the anode involves three stages; adsorption / diffusion, the degree of charge transfer and proton hydration. CO covers the active sites of the catalyst, adsorbs onto the carbon matrix and then reduces the speed of hydrogen adsorption on the electrode surface.

2.1. Gas diffusion layer (GDL)

2.1.1. GDL structure and functions

In general, the GDL is a mixture of carbon, water, alcohol and polytetrafluoroethylene (PTFE) or other hydrophobic substances. The purpose of PTFE is to facilitate the transport of gas and water during fuel cell operation under flooding conditions [12,44]. The GDL connects bipolar plates with the CL and consists of a macroporous layer as a backing layer that is composed of carbon paper or carbon cloth and a MPL that is composed of carbon powder and hydrophobic/hydrophilic substances [16,17,45,46]. Han et al. [47] reported that a GDL needs to only consist of a BL to be a monolayer GDL and should contain an additional MPL to be a double-layered GDL. A few researchers [48,49] have noted that a GDL can consist of a macroporous layer only (without a MPL) or contain an MPL, and other researchers [50–53] have indicated that the GDL is the BL on its own.

Carbon powder is one of the most important components in the GDL. Comparison studies of carbon substrates as components of MPLs or CLs have been conducted by a few researchers [10,47,54]. Carbon black, Vulcan XC-72R, and acetylene black are commonly used as carbon substrates, but progress has been made in using nanosized carbon sources, such as carbon nanotubes (CNTs) and carbon nanofibers (CNFs); substrates of MPL fillers and catalyst layers based on titanium, tin and silicon have also been widely used [10]. Liu et al. [55] investigated the influence of the carbon black content (0–10 wt%) on the performance of a PEMFC and found that the density of the carbon paper increased with the increasing carbon black content. However, this increase in carbon black content did not significantly increase the thickness and resistivity of the carbon paper surface. The use of various types of carbon and carbon-based compounds is essentially an attempt to improve the performance and durability of PEMFCs.

Moreover, the BL has been used by several researchers [56–59] as a supporting layer for PEMFC electrodes. In addition to the BL, the term “supporting layer” has also been applied to gas diffusion backing (GDB) [60,61] or gas diffusion media (GDM) [62–65]. The BL is a layer that serves as a retainer and keeps moisture away from the electrodes [66]. It is also a gas diffuser and provides electron and water routes outside of the electrode. Carbon-based materials are commonly used as BLs due to their acid resistance, improved gas permeability, good electrical conductivity, elastic characteristics under compression, and ability to maintain porosity [16]. Thus, the BL must meet several requirements: high electron conductivity, hydrophobicity for draining water and the ability to facilitate gas transport to the catalyst layer for the electrochemical reaction [67]. In addition, the BL should be sufficiently strong to support the overall electrode layer. Typical BL thicknesses are in range from 0.2 to 0.5 mm. The carbon substrate in the BL forms macropores with 60–90% porosity, a 100–400 μm (0.1–0.4 mm) thickness and a 20–50 μm pore diameter. The hydrophobic polymer depth merges into the carbon substrate in the range of 5–40 μm [27].

The macropore substrate in the BL is the largest contributor to the porosity for gas flow, and it has a high electrical conductivity. The MPL (composed of carbon powder and PTFE and located between the BL and CL) serves to reduce the resistance current, arrange the hydrophobicity level and water traffic control [7,68–70] and prevent the CL from seeping into the BL [47]. Nam et al. [7] showed the influence of the MPL on water regulation. On the CL side, the MPL enhanced the catalytic activity by lowering the degree and level of water saturation on the MPL-CL interface, whereas on the GDL side, the MPL reduced the level of saturation on the GDL. The addition of an MPL improves the performance of the GDL. Han et al. [45] introduced a specific GDL

called a CFGDL (carbon-filled gas diffusion layer) that pervaded both sides of the carbon cloth of the MPL, and they found that the CFGDL increased the electrical contact between the GDL and bipolar plates as well as between the GDL and the catalyst layer. Additionally, the CFGDL was a good support substrate for the catalyst layer. Han et al. also showed that the best composition of the PTFE and carbon content in the CFGDL was 40 wt% and 6 mg cm^{-2} , respectively.

The GDL of the PEMFC performs several functions, such as a gas spreader, current collector, and water guide [8,17,23]. The porous was shown to GDL act as a gas spreader in an additional study [47], and it also transfers heat from the catalyst layer to the current collector [46]. As a current collector, the GDL should be highly conductive. Moreover, since the GDL contributes to removing water from the electrode layer, it should be sufficiently hydrophobic, especially when the fuel cell is working at a high current density and using air as the oxidant [46,60]. Meanwhile, Lin et al. [71] stated that the GDL has three important functions: controlling the water balance in the MEA, providing mechanical support for the MEA and facilitating the electrical contact between the electrode and current collector. Further, studies based on the hydrophobic and hydrophilic properties, gas diffusion, water management and surface design of GDLs have been conducted. According to Arvay et al. [46], the GDL has four main functions: (1) transmitting electrons to or from the CL with a resistivity range of 0–0.08 $\Omega\text{ cm}$; (2) transporting the reactants and products to and from the CL with a typical porosity of 0.7–0.8, (3) transporting heat from the CL to the current collector, and (4) providing mechanical support for the electrolyte. On the CL side, the MPL enhances the catalytic activity by decreasing the size and level of saturation of the water spots at the MPL-CL interface, while on the GDL side, the MPL reduces the level of saturation by focusing the density level towards the GDL.

Cindrella et al. [8] reviewed the features of the GDL (hydrophobicity, porosity, permeability, transport properties, compaction and structure) and its influence on the fuel cell performance and manufacturing and coating process. Meanwhile, Park et al. [16] reviewed studies on GDL materials and design, and Arvay et al. [46] discussed GDL characterization techniques. Furthermore, Zamel et al. [52] studied the effective transport properties of PEMFCs and focused on the GDL (structure, diffusion coefficient and thermal and electrical conductivity). Therefore, the important features of GDLs were determined to be the structural design, porosity, hydrophobicity, hydrophilicity, gas permeability, water management and surface morphology. Although, the manufacturing cost of GDLs is not very high, these layers greatly affect the PEMFC performance.

2.1.2. GDL preparation

The GDL is very important because its function directly affects the PEMFC performance. Therefore, GDL research has grown to encompass composition, manufacturing and characterization methods. As a part of the GDL, BL research has received increasing interest, especially the manufacturing methods and composition of BLs. BLs are usually composed of carbon paper or carbon cloth that is coated with a Teflon emulsion to increase the hydrophobic nature of the layer.

To improve the performance of the BL and MPL, the functions of the PTFE content and other hydrophobic substances and their relationship with the conductivity and hydrophobicity have been widely studied [62,72–76]. Hydrophobicity is important for the GDL so that it can repel water to avoid flooding. Therefore, PTFE is used to increase the hydrophobicity to avoid the occurrence of flooding and closure of the electrode pores [74]. In addition, PTFE provides some mechanical stability in the GDL. Lin et al. [44] investigated the influence of PTFE on GDLs and noted that the addition of PTFE can increase the gas and water transport when fuel cells operate during flooding. Many other researchers have also investigated the influence of PTFE [62,77,78], and they found different results for the optimal content of each component depending on the material used. Moreover, other polymeric substances are commonly used, such as polyvinylidene fluoride (PVDF),

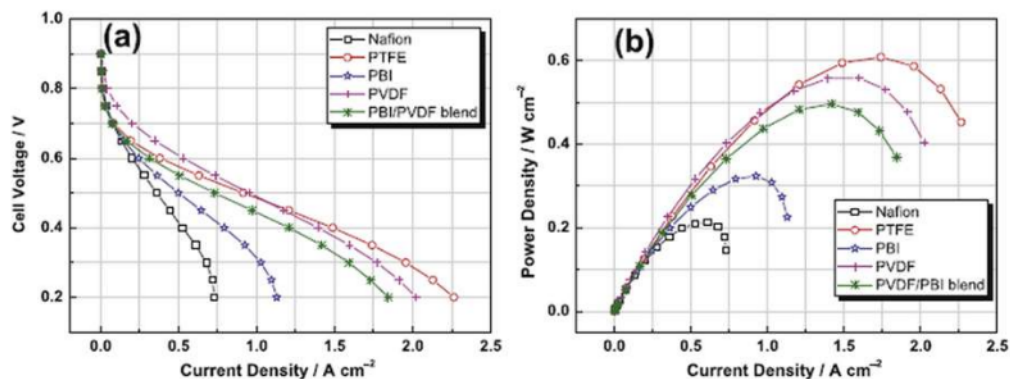


Fig. 3. The cell performance has recorded under varying polymer binders at 160 °C: (a) polarization curve and (b) power density (Reprint permission from Ref. [83]).

sulfonated polymer (Nafion) [79], polybenzimidazole (PBI), fluorinated ethylene propylene (FEP), and polybenzimidazole-blended polyvinylidene difluoride (PBI/PVDF) [80–82]. Nafion is the conventional polymer binder for low-temperature PEMFCs. However, at high operating temperatures, Nafion cannot function properly due to the water dependency, and it may tend to block the catalyst reactive sites. The PVDF binder results in a better performance than Nafion or PBI binder under this condition. Su et al. [83] fabricated five types of GDEs using the optimum binder content and the same catalyst loading (0.5 mg cm⁻²) and active area (5 cm²) for the Nafion, PTFE, PBI, PVDF and PBI/PVDF blend separately. In a single cell, the PTFE and PVDF based GDEs exhibited the best performance compared with the others as shown in Fig. 3. The power density was recorded as 0.61 W cm⁻² at 0.6 V and 160 °C. The current density of PVDF-GDEs was 0.52 A cm⁻² under the same conditions. This result is 121% higher than the PBI and Nafion based GDEs (0.24 A cm⁻²), which is very promising for commercial applications [83,84]. Lim et al. [85] studied the influence of FEP as a hydrophobic substance in the GDL on the performance of PEM fuel cells using a Toray 090 backing layer. In addition, Liu et al. [51] used carbon cloth as a BL and PEF as a binder and measured their thickness, contact angle, horizontal gas permeability and resistivity. Li et al. [75] used dimethyl silicone oil (DSO) on the cathode as a hydrophobic substance to prevent flooding. Wang et al. [50] added sucrose on carbon paper for further carbonization before adding PTFE, which increased the BL hydrophobicity by reducing the PTFE content. Chun et al. [86] utilized two microporous layers, one had internal hydrophilic properties and surface hydrophobic properties, and the other hydrophilic layer served to increase the internal humidity, which improved the performance of a single-cell PEMFC under low humidity conditions, as shown in Fig. 4.

Chiu et al. [74] introduced a sputtering method to add PTFE onto carbon cloth to prepare BL electrodes. These authors suggested that this technique is faster and cleaner. In the conventional method, macropores are often blocked by large PTFE molecules which reduce the gas permeability and electron transfer efficiency from the electrode. In addition, the placement of hydrophobic substances can be accomplished via various methods, such as dipping, spraying and brushing [16]. The effect of PTFE on carbon cloth and carbon paper are shown in Figs. 5 and 6, respectively. Park et al. [17] explained the influence of a GDL composed of carbon paper and carbon cloth with or without MPL and analyzed properties such as the porosity, surface structure, hydrophobicity, water absorption, polarization and resistivity. They found that carbon paper with 5 wt% PTFE in the MPL exhibited better performance than the commercial carbon cloth Elat-LT-1400 W and demonstrated that the total volume and average pore diameter of the carbon paper were larger than those of the carbon cloth with the same PTFE content. Meanwhile, Wang [87] analyzed and compared the performance of carbon paper and carbon cloth in PEMFCs and noted

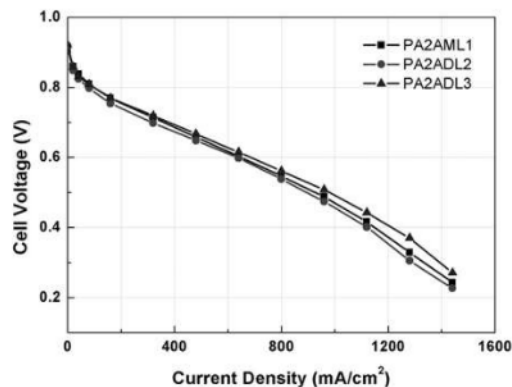


Fig. 4. GDL polarization curves under different conditions: PA2AML1 for a single MPL, PA2ADL2 for a double MPL with the hydrophilic layer above and PA2ADL3 hydrophilic layer below (Reprint permission from Ref. [86]).

that carbon cloth is a better choice under high humidity conditions. However, under dry conditions, carbon paper is better due to its pore structure and ability to retain water to increase the moisture level of the membrane and ionic conductor. The structures of several BLs are presented in Fig. 7.

Other studies have investigated and characterized the influence of the hydrophobic substance used on various BLs. However, the role of the BL in the mechanism of the mass transport of the reactants and products has not been widely discussed. In addition, the sintering time and temperature have also not been reviewed. The addition of carbon substrates and different types of BLs during the preparation of the GDL is correlated to the overall thickness. The influence of the carbon content on the performance of a PEMFC substrate was studied by Liu [55], while the influence of the BL thickness on the MEA performance was also studied [71]; the results are shown in Fig. 8.

Lin et al. [71] suggested that larger pore radii are responsible for high current densities compared to using thinner layers, and they recorded a 330 μm GDL thickness with 3.0 mg cm⁻² carbon content. In addition, Lee [88] studied the influence of the thickness and porosity of the GDL on PEMFC performance. This author manufactured the GDL after rolling, spraying and screen printing as shown in Fig. 9, which resulted in different thicknesses and porosities. When the GDL thickness was above 300 μm, better performance was observed. Moreover, the spraying and screen-printing methods were shown to produce better results than the rolling method. GDL manufacturing processes, especially the process of MPL implantation in BLs, have been performed using coating [70,85,89], screen printing, [88] and spraying methods

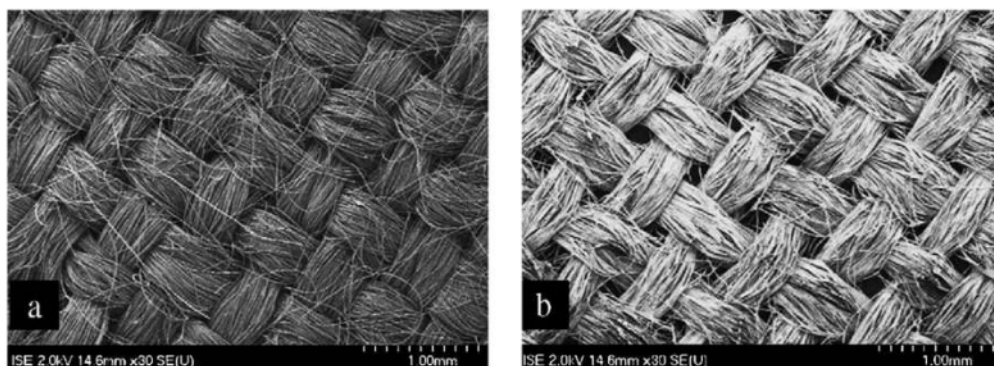


Fig. 5. SEM images of carbon cloth a) without and b) with 15 wt% Teflon (Reprint permission from Ref. [67]).

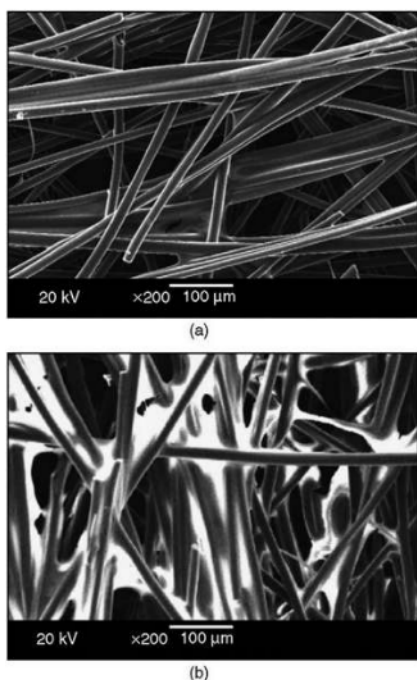


Fig. 6. SEM micrographs of carbon SGL704E paper (a) before and (b) after hydrophobic treatment (PTFE content: 40%) (Reprint permission from Ref. [45]).

[15,18]. One spray method has also been studied using a robotic spraying technique [90]. Celebi [91] fabricated GDLs by the pasting of CNFs on carbon paper using nickel through a homogeneous deposition precipitation (HDP) method. The use of nickel with the HDP method enabled successful CNF deposition on one side of the layer while controlling the GDL thickness.

Chen et al. [92] comprehensively compared the manufacturing processes of MPLs for GDLs using wet and dry methods. They found that MPLs fabricated using the dry process exhibited better performance, especially when the fuel cell operated with high oxygen consumption at high humidity levels. Eventually, the mass transport improved. The dry method produced more mesopores, resulting in higher electrical conductivity and more stable hydrophobicity as shown in Figs. 10 and 11. The dry method was conducted by mixing Vulcan XC-72 carbon and PTFE with a powder content of 30 wt%; the mixture was placed on carbon paper, heated at 240 °C for 30 min and then sintered at 340 °C

for 30 min.

2.1.3. GDL characterization

In general, GDL characterization is divided into two methods: ex situ (GDL separately) and in situ (in a fuel cell system). Ex situ methods are used to characterize the electrical and thermal conductivity, mechanical properties (tensile strength, compressibility and bending properties), porosity and pore size distribution, gas permeability, surface morphology, cross-sectional morphology, contact angle and surface energy, and for cyclic voltammetry (CV) for simulations, and modeling; in situ methods are used to measure the impedance and water transfer and for modeling and simulations [46]. Studies have been performed to characterize the gas permeability and anisotropy of several types of GDLs in various directions [93]; the in-plane direction exhibits larger gas permeability than the through-plane direction. In addition, GDLs with uniform fibers have a greater degree of anisotropy.

Radhakrishnan [94] analyzed the stress reduction, performed compression tests, and measured the channel intrusion and electrical resistivity of carbon paper and carbon cloth to compare their performances. This author showed that the through-plane resistivity of carbon paper is higher than the resistivity of carbon cloth. In contrast, the in-plane resistivity of carbon paper is lower than that of carbon cloth. Radhakrishnan also suggested that the intrusion of carbon paper is smaller than that of carbon cloth. However, according to Yang [95], carbon cloth is more practical as a BL than carbon paper because of its compressibility, elasticity, and flexibility. One of the advantages of carbon paper is its more even distribution and more delicate structure, but it is easily broken and must be carefully handled. Overall, both carbon paper and carbon cloth perform well when their thermal and chemical stability is tested for use as BLs in a PEMFC. The influence of the BL and GDL on fuel cell performance has been widely studied by Ismail [78], who investigated the influence of PTFE addition on GDL electrical conductivity and found that GDLs with a 50 wt% PTFE content performed better than GDLs with a 25 wt% PTFE content in the case of in-plane conductivity. Ismail [78] and Zhou [32] showed that in-plane conductivity is more useful for determining the conductivity of BLs than through-plane conductivity.

Zhou [32] studied the influence of GDL conductivity on fuel cell performance and found that the performance improved with a higher conductivity. In addition, the through-plane conductivity influenced the ohmic loss, and the in-plane conductivity affected the over potential and local current density. Zhou also investigated the influence of the GDL thickness on the current density and found that the relative characteristics of the current density did not change with an increased thickness. However, if the thickness of the GDL was reduced, then the current density decreased drastically in the shoulder and returned to normal at the channel. Meanwhile, Ismail [78] explained the effect of adding PTFE on the GDL conductivity using a carbon paper BL; this

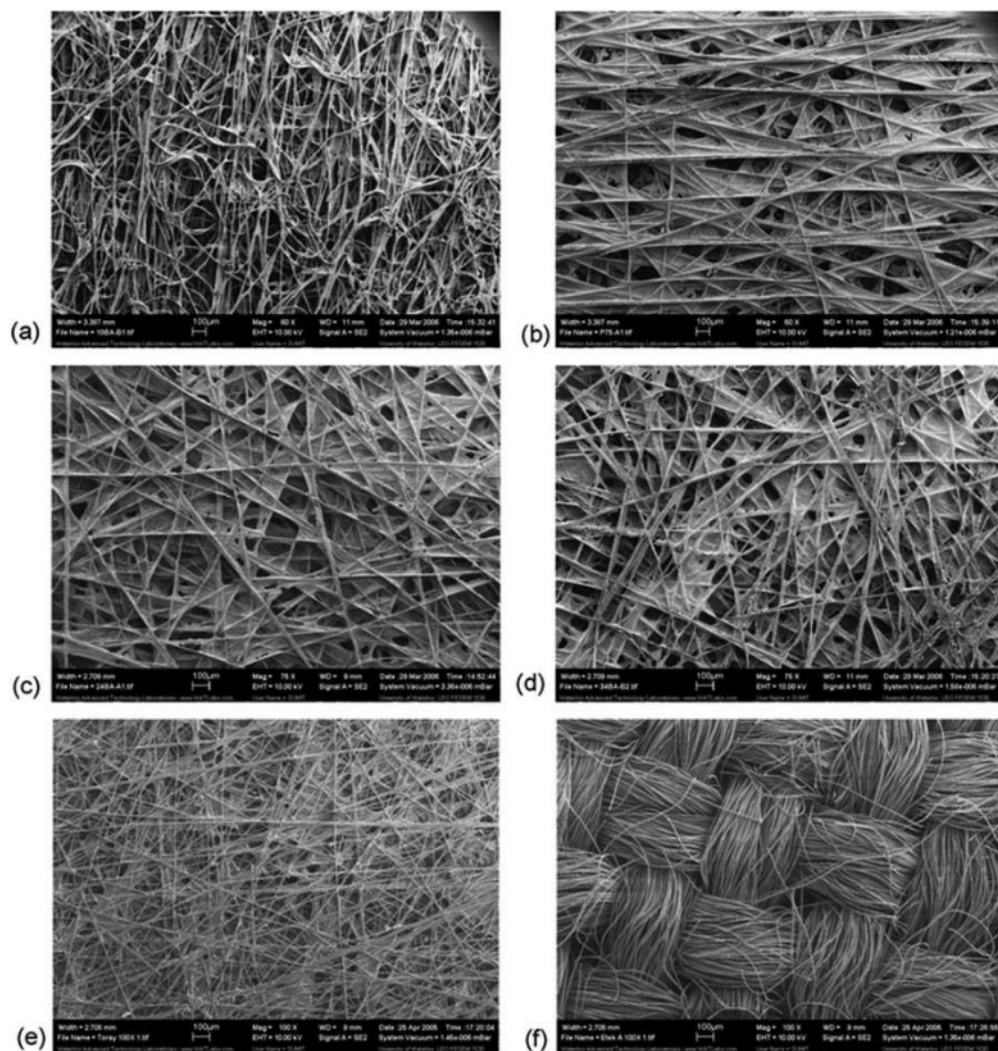


Fig. 7. Several types of BLs: (a) SGL 10BA, (b) P75 Ballard, (c) SGL 24BA, (d) SGL 34BA, (e) 090 Toray, (f) E-Tek Cloth 'A' (Reprint permission from Ref. [57]).

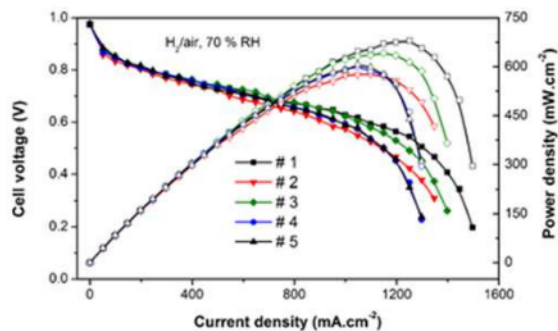


Fig. 8. Influence of GDL thickness on the performance of the MEA. #1. 330 μm, #2. 250 μm, #3. 210 μm, #4. 170 μm, #5. 130 μm (Reprint permission from Ref. [71]).

author found that the PTFE content influenced the in-plane conductivity of the GDL. GDLs with in-plane conductivity along different perpendicular directions exhibited double conductivity. Increasing the PTFE content in the MPL did not affect the conductivity. The MPL conductivity with a PTFE content of 50 wt% was greater than that of the MPL with 25 wt% PTFE content. Under different conditions, Park [62] investigated the influence of PTFE content in the BL (defined as the GDM) on the gas permeability, pore diameter, and I-V performance in a single cell; he found that the gas permeability and pore diameter increased up to a certain rate and then decreased with increasing PTFE content. Park also found that increasing the PTFE content did not have a positive impact on the I-V performance when high water condensation levels were present on the boundary surface of the GDM with the CL. The influence of temperature on the electrical resistivity and tensile strength of the BL was studied by Zhang [96], who observed that the electrical resistivity and tensile strength decreased with increasing temperature, according to SEM (scanning electron microscope) and XRD (X-ray powder diffraction) analyses.

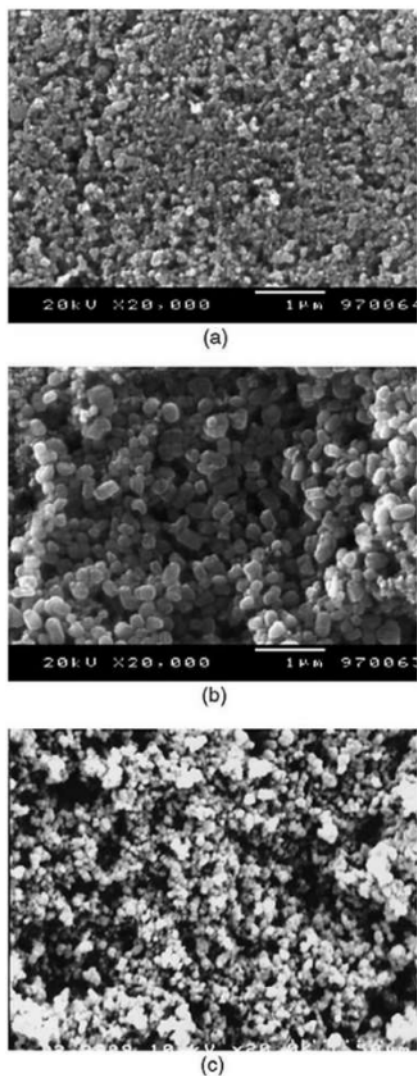


Fig. 9. SEM images of GDL produced by (a) rolling, (b) spraying and (c) screen printing methods (Reprint permission from Ref. [88]).

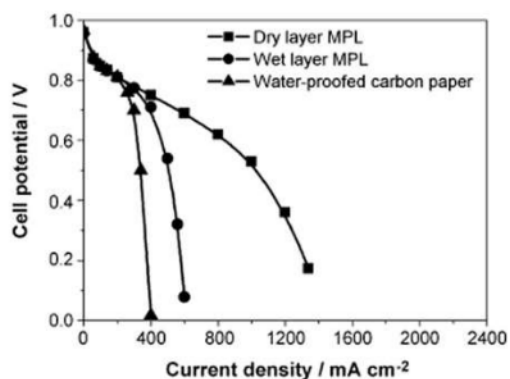


Fig. 10. MPL performance comparison between the dry method and wet method at the speed of 70 wt% oxygen consumption (Reprint permission from Ref. [92]).

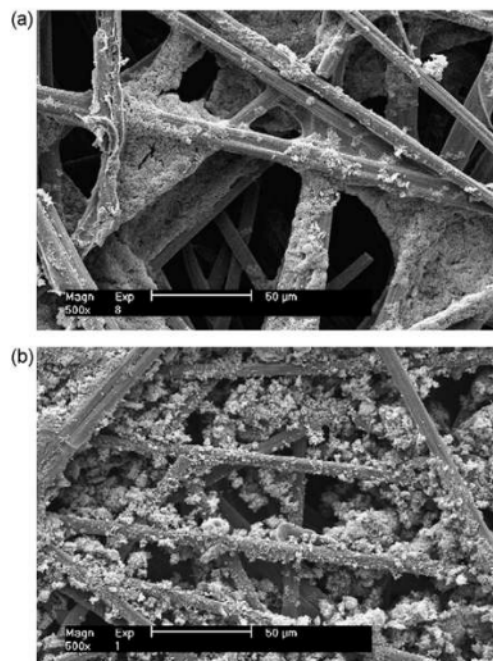


Fig. 11. Comparison between MPL produced with (a) wet and (b) dry methods with carbon content of 0.1 mg cm^{-2} (Reprint permission from Ref. [92]).

2.2. Catalyst layer (CL)

2.2.1. CL components and functions

In the CL, the electrochemical reaction occurs, which converts the hydrogen gas feed and oxygen (air) to water and electricity. Generally, the CL consists of a catalyst, a carbon substrate, a hydrophobic substance, and an ionomer from an electrolyte, such as Nafion. The catalyst plays an important role in accelerating the oxidation of the hydrogen gas and reducing the oxygen gas to water, while the Nafion ionomer provides access for H^+ ions to pass through from the anode to the cathode side [97]. The CL thickness is typically between 5 and 100 μm , with a porosity of 40–70%, and the catalyst should be well dispersed with a particle size of 1–10 nm [27]. Carbon-based materials are used as catalyst substrates because of their high conductivity; examples include carbon powder, graphite, and active carbon. Marie [98] introduced the use of carbon aerogels as CL substrates with larger porosities and surface areas compared to using carbon black. The catalyst loading on the catalyst layer is between 0.01 and 5 mg cm^{-2} depending on the thickness of the electrode layer.

Hydrophobic substances in the CL serve as a catalyst binder and maintain the hydrophobicity of the CL. To bind the catalyst and increase the hydrophobicity, PTFE is generally used [99]. Thus, the reactant gas and hydrophobic state should be optimized carefully to achieve high absorption so that a CL is produced with a high level of catalyst. In addition, the durability of the catalyst is an important design constraint [12]. The CL should be able to facilitate the effective flow of electrons generated or absorbed by the electrochemical reaction in a given situation and should be able to drain H^+ ions from the anode to the membrane. Therefore, the Nafion ionomer is typically added to the CL. Several properties of the catalyst layer should be optimized to produce the desired performance of the catalyst material, including the level of adsorption of the reactants, the hydrophobicity level, and transmission of ions and electricity [12].

2.2.2. Catalysts in the CL

The electrocatalyst accelerates the electrochemical reaction of the H_2 gas fuel reactant, which is transformed into chemical energy. The catalyst must have several features, including high internal activity (especially for the cathode), high electrical conductivity, and environmental friendliness.

The PEMFC catalysts can be grouped into three categories: platinum based catalysts, platinum modified based catalysts (which include other metals such as Cr, Cu, Co or Ru) and non-platinum-based catalysts, such as non-noble metal or organometallic catalysts [100]. Platinum is still the most effective catalyst used in PEMFCs. To date, platinum-based catalysts in PEMFCs have the dominated research, and typically, the influence of the Pt content and particle size has been studied [11,29]. Moreover, many researchers have focused on the type of carbon substrate used [101], the CL placement or manufacturing method [21,97,102–106], the influence of the solvent [6,107,108], the influence of hydrophobic substances [99,109], catalyst and CL characterization [110], the effect of porosity [111], the effect of various surface structures and layers [30,31], modeling and optimization [35,112], the degradation and durability of Pt [3,100,113–115], or the expansion of Pt catalysts on carbon nanotubes [116].

Platinum alloys are used to reduce the content of Pt catalysts without significantly decreasing the catalytic performance. Examples of Pt alloy catalysts that include other metals are Pt-Ru [117], Pt-Co [118,119], Pt-Pd [120,121], Pt-Ru-Co [122], Pt-Co-Cr [123] and combinations of Pt with Fe, Co, Cu and Ni [124]. It has been reported that the use of metal alloys can improve the catalyst performance [36] because the presence of other metal alloys with Pt can increase the size of the active catalyst site. However, the durability of the catalyst remains to be proven. The use of non-platinum catalysts remains a challenge for researchers and is a barrier to the development of fuel cell catalysts. Moreover, platinum is an expensive metal catalyst and is easily poisoned. The use of non-platinum catalysts has been attempted, including 5,10,15,20-tetrakis-(4-methoxy-phenyl)-porphine cobalt (II) [125], Ir-V alloys [126], cobalt-based catalysts [9], nitrogen-based catalysts [127], metal carbide and other metal oxides [39], and catalytic iron acetate [128]. Faubert et al. [128] reported that 0.2 wt% Fe absorbed in perylenetetracarboxylic dianhydride (PTCDA) on the cathode exhibited a similar performance to that using a platinum catalyst with 2 wt% content in an MEA (1 cm^2).

2.2.3. Polymer ion content in the CL

Conductive polymer ionomers, such as Nafion, contribute to improving the performance of the electrode. Nafion ionomers that are added to the CL help H^+ ions flow from the anode to the polymer membrane and then through the cathode [129]. In addition to serving as proton transmitters, ionomer polymers also function as CL binders and provide hydrophilic groups to retain moisture and ionic conductivity [24,130]. Increasing the amount of Nafion in the CL provides two effects: improving the electrochemically active surface and enhancing the ion conductivity. However, there is an optimal threshold amount of Nafion; an amount that is too low will limit the proton delivery to the membrane, an amount that is too high will negatively affect the absorption and flow of gas [12,21]. Passos et al. implanted catalyst layers on membranes using Nafion solutions with different content levels, from 10% to 40% dry weight, calculated from the total weight of the catalyst layer. They observed that the current of hydrogen desorption and the platinum active surface area increased with as the percentage of Nafion increased. Several researchers have observed the effects of Nafion ionomer addition [24,73,131]. Lee et al. [131] studied the effect of adding Nafion ionomers on the surface and compared the results with those from adding Nafion in the CL; these authors found that the distribution of Nafion on the surface resulted in better performance than distributing Nafion inside the CL, as shown in Fig. 12. Meanwhile, Zhang et al. [30] characterized the influence of using Nafion on dual-bonded PEMFC cathode structures, and proved that the

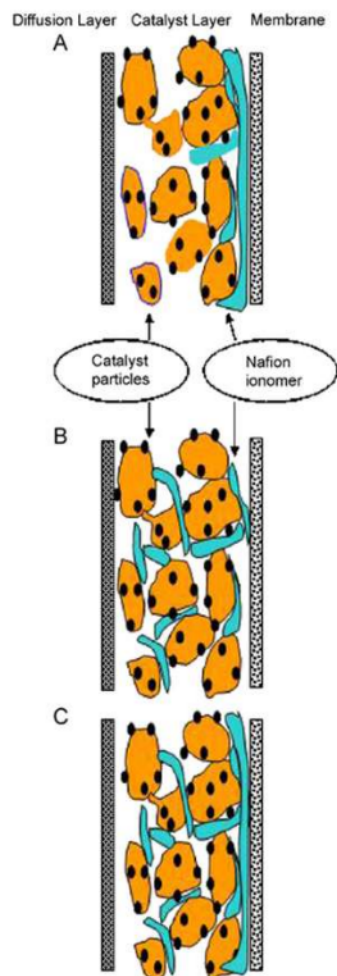


Fig. 12. Schematic planar representation of the catalyst layer. The Nafion ionomer was incorporated within the catalyst layer on its surface (A), inside (B) and both together (C) (Reprint permission from Ref. [131]).

reduction rate of Nafion in the CL hydrophobic layer improved the electrode performance. Chaparro et al. [73] discovered that the optimal Nafion ionomer content for the cathode using the electric spraying method was 15 wt%. The influence of the Nafion content on the catalyst activity was characterized by Lai [132], who concluded that the optimum Nafion content based on I-V tests and electrochemical impedance spectroscopy (EIS) was 1.0 mg cm^{-2} , and the interface resistance was very closely linked to the three-phase region (nutrient response, electrolytes and catalysts). In addition, Sasikumar [133] attempted to investigate the dependence of the optimum Nafion content in the catalyst and found that a low catalyst content required more Nafion. Furthermore, the Nafion content was not related to the membrane thickness. Ahn [130] investigated the influence of the Nafion ionomer equivalent weight and showed that ionomer polymers with a low equivalent weight are ideal under low humidity conditions because they maintain the moisture and ionic conductivity due to the greater number of sulfuric acid functional groups present.

2.2.4. Manufacturing of CL

Attempts have been made to improve the CL performance by selecting the catalyst type and content, the manufacturing method, or the

ionomer type and content. CL manufacturing has grown based on recent research developments and viewpoints. Some methods for manufacturing electrocatalysts, including their advantages and disadvantages, were described by Park [134]. According to Park, the thin layer method for CL manufacturing is effective using Pt catalysts, but for micro PEMFC applications, this method has been proven to be effective because of the greater Pt content required. In the electrodeposition method, the required amount of Pt is lower, but the method is not ready for actual utilization. However, the sputtering method can directly deposit Pt for various types of MEAs with ultralow Pt loading. Moreover, low consumption and low Pt function remain the restrictions for CL manufacturing. To overcome these problems, the sputtering technique may be a promising approach in the near future. In contrast, the dual ion beam assisted deposition (IBAD) method combines the electro spray technique and sol Pt with the ultralow Pt loading method for optimal use of Pt. Meanwhile, Lee [88] noted that the spray technique is more advantageous than the brush technique to reduce the charge transfer resistance and extend the three-phase region. Hwang [135] also discovered that the screen printing technique is more satisfactory than the spray method and other techniques. Therefore, the screen printing technique can be used for the large-scale production of MEAs.

Regarding casting methods, Hwang [135] adhered polyphosphoric acid (PPA) on the membrane media of PEMFCs at high operating temperatures and found that the slot die extrusion method was advantageous due to its cost effectiveness, continuity, and uniformity. This method is generally chosen because of its simplicity, ability to produce various shapes and sizes, and relatively low cost.

The spraying method has been used by many researchers [73,90,136,137] due to its advantages in terms of convenience (especially for multilayers), uniformity, and high performance. Furthermore, the electrocatalyst manufacturing method with galvanostatic pulses [138] and preferential pulsed electrodeposition (PED) [104] have already been explored. Although the performance results have been relatively low, the idea of optimizing the Pt catalyst is still being considered.

Abaoud [139] introduced a hybrid or mixed technique (a combination of spray and screen printing methods) for manufacturing CLs. The performance of electrodes fabricated using the hybrid method was better than that using electrodes fabricated with either the screen printing or spray method alone. A diagram for electrode manufacturing using the hybrid method is presented in Fig. 13. A performance comparison among the spray, printing and hybrid methods is presented in Table 1.

Table 1
Electrochemical report of electrodes prepared by different fabrication techniques [131].

Fabrication techniques	Pt-loading anode/cathode (mg cm^{-2})	Cell resistance at 0.5 V ($\text{m}\Omega$)	Current density at 0.5 V (A cm^{-2})	Power density (W cm^{-2})
Spraying	0.15/0.15	10.0	1.6	0.80
Screen-printing	0.10/0.15	6.5	1.4	0.75
Hybrid	0.10/0.15	5.8	2.1	1.00

Song et al. examined the influence of PTFE in ink on the CL using a new approach called the CCM-modified method [99]. In this method, a platinum and PTFE containing catalyst ink is sprayed on a layer of aluminum foil and then sintered at 340°C before being pasted on a Nafion 212 membrane and pressed with a heater. Song conducted a study on the influence of PTFE content on electrochemical activity, hydrophobicity, and ohmic resistance; the optimal reported PTFE content was 5 wt%. A large PTFE content increased not only the hydrophobicity but also the ohmic resistance. Therdtianwong et al. [109] focused on the cathode while investigating the influence of various PTFE and CL patterns and showed that a chess pattern resulted in a higher performance than a striped pattern at a catalyst content level of 0.5 mg cm^{-2} . One study sought to improve the performance of the catalyst layer by using the electro spray deposition method with different platinum and ionomer content levels. Chaparro et al. [73] found that the optimum Nafion ionomer content was 15%, as shown in Fig. 14.

Chaparro et al. showed that the electro spray method is not significantly different from the conventional method (brushing) in terms of fuel cell performance at the same catalyst content level but requires a slightly higher ionomer content. Antolini [140] discussed the types of fuel cell carbon backing catalysts and their activities. Contrary to the results obtained by other researchers, Antolini stated that carbon mesopores (pore size of 2–50 nm) yielded better results for the placement of Pt particles than micro (< 2 nm) or macropores (> 50 nm). The mesoporous structure facilitated mass transfer. This discussion led to efforts for improving the performance of the catalyst layer. Meanwhile, Wee [103] compiled research on the various available methods of manufacturing catalyst layers with low Pt catalyst content. In addition, this author discussed the modified thin layer, electrodeposition, sputtering deposition, dual ion-beam deposition, and electrodeless deposition methods. According to Wee, the use of catalysts with a low Pt content is intended to reduce MEA manufacturing costs and cell stack weight.

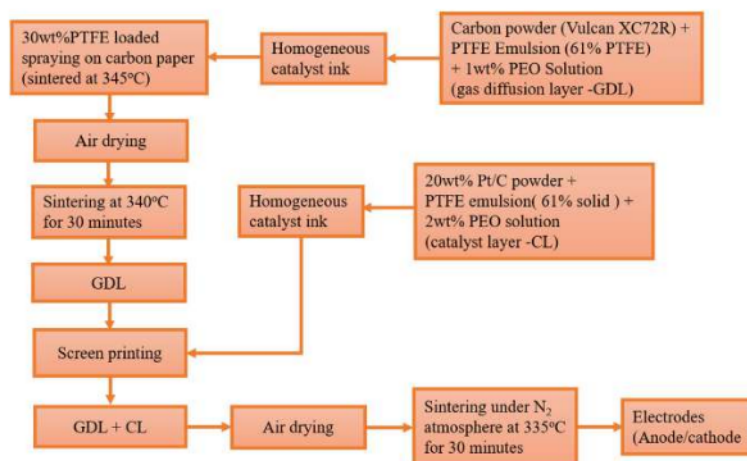


Fig. 13. CL manufacturing diagram using the hybrid method (Reprint permission from Ref. [139]).

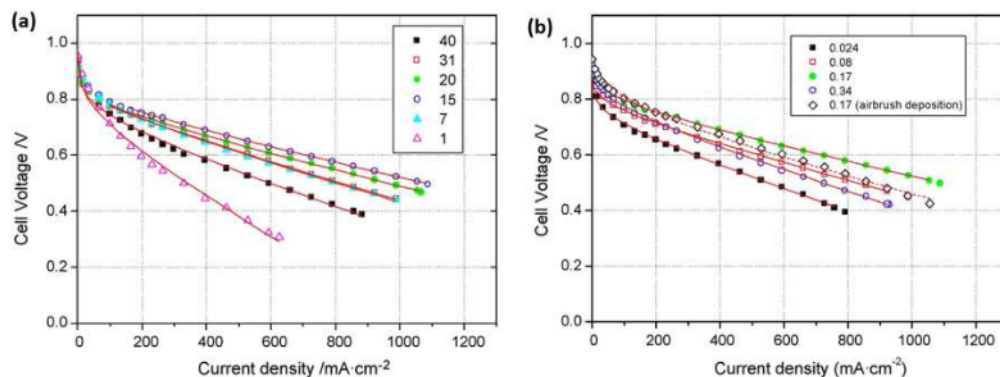


Fig. 14. Polarization curves corresponding to MEAs with cathodes deposited by electrospray with different indicated amounts of ionomer in the catalyst layer (values are given as weight percentage within the catalyst layer). Pt cathode load: 0.20 mg cm^{-2} (Reprint permission from Ref. [73]).

Actually, the reduction of the Pt content depends on the manufacturing method, substrate content and electrode structure.

In addition to research on the influence of the material content and operation conditions, various CL layer structures have been studied. Generally, the CL layer is composed of a catalyst component, hydrophobic substances, and electrolyte ionomer components. Double layer type CLs have been studied [141,142] to increase the performance over that of single layer type CLs. Moreover, triple layer type CLs have also been manufactured to reduce the interface resistance between the CL and GDL in the first layer and between the CL and membrane electrolyte in the second and third layers [137].

3. Electrode degradation and durability

Degradation is a highly concerning issue for PEMFCs because it can decrease their lifetime. Degradation processes can be divided into three categories: baseline degradation that occurs because of long-term application, which is irreversible and cannot be avoided; degradation that occurs due to repetition or operating conditions; and degradation that occurs when fuel cells experience adverse operating conditions, such as a lack of fuel [143]. Many researchers have sought to understand the degradation mechanisms of each component to reduce the degradation speed of the fuel cell as a whole. Based on the results from studies, degradation often occurs in the GDL, platinum catalyst and the CL, carbon supporter, membranes and bipolar plates [144,145]. In addition to the above factors, degradation can occur due to inefficient water management, which leads to flooding or even dehydration, poisoning the fuel and oxidant [144]. GDL degradation occurs because of the oxidation of carbon, PTFE decomposition and mechanical degradation [145,146].

Furthermore, the degradation of the CL includes catalyst maturation, the loss or transfer of catalysts, the corrosion of carbon, electrolyte and interface degradation [100], and a decrease in the catalyst active surface area due to catalyst agglomeration that primarily occurs in the cathode [147]. The degradation of PEMFC as a system was previously studied by Wu [145], who investigating degradation in terms of the mechanisms and strategies to overcome it. Other topics of study have included the degradation of the membrane, catalyst and catalyst layer, GDL and bipolar plate. The degradation of the catalyst and catalyst layer includes catalyst contamination by impurities, losses in activity due to the sintering process, catalyst migration to the carbon substrate and catalyst dissolution to the membrane layer.

When the catalyst or the CL is degraded, the change in the catalyst particle size generally cannot be observed during fuel cell operation conducted at a constant voltage and current when the operation is running. However, the degradation of the anode appears when the fuel cell is used for long periods, while at the cathode, the particle size

grows with the increasing temperature, length of the test, potential and moisture content [147]. Zhang et al. [100] comprehensively studied platinum-based CL degradation, covering the causes of CL degradation and its components, equipment, and diagnostic methods and strategies to reduce or overcome this degradation.

The general degradation mechanism in an MEA, especially in a multi-stack, is due to polarity reversal during fuel cell operation, which causes permanent damage [148]. The main cause of polarity reversal is a lack of reactants and fuel at the anode while the fuel cell receives a load. Taniguchi has tested the influence of air and fuel shortages on the degradation mechanism. One factor that causes a decline in the PEMFC life for vehicle applications is the rapid change of vehicle operating conditions, such as changes in the loading cycle, high power conditions, and idling conditions and the cycle of turning the vehicle on and off. Yu [13] investigated these causes, their consequences and methods to overcome the performance degradation of PEMFCs due to the influence of turning the fuel cell on and off. Another matter that requires attention is increasing the durability of PEMFC equipment. Wu [145] explained various PEMFC degradation mechanisms and mitigation strategies. The worst-performing components observed by Wu were the membrane, CL, GDL and bipolar plate.

To increase the endurance and durability of MEAs, Accelerated Stress Tests (AST) were performed by Zhang [149]. There are three clusters of research regarding MEA durability: 1) investigation and experimental validation, 2) mathematical modeling, and 3) performance degradation handling strategies. Tanuma and Terazono [150] used hindered amine light stabilizers (HALS) to increase the lasting power of MEAs and proved that it increased drastically. The HALS amino groups reacted with the carboxyl groups at the BL, forming complex compounds that could slow the formation of hydrogen peroxide during fuel cell operation.

4. Electrode characterization and performance tests

Electrode characterization and performance tests are very important stages for assessing the suitability level of the electrode to be used. Common electrode characterization and performance tests are described below.

4.1. Conductivity

The conductivity of PEMFC electrodes is divided into two categories: electrical and ionic. Electrical conductivity is a measure of the ability of an electrode to conduct electrons from the CL to the GDL and then to the bipolar plate. The measured electrical conductivity consists of through-plane and in-plane conductivity. Unlike through-plane conductivity, whose data are usually provided by the GDL

manufacturer (depicting the conductivity level of the GDL conducting the electrons), in-plane conductivity is highly important to properly describe the ability of the GDL to conduct electrons on every surface to the bipolar plate. In contrast, ionic conductivity describes the ability of an electrolyte membrane to conduct H^+ ions from the anode to the cathode. These two conductivity parameters contribute significantly to the performance of the PEMFC. Ismail [78] investigated the influence of the PTFE content on the through-plane and in-plane electrical conductivity. Furthermore, Mironov [151] compared two methods of electrical conductivity measurements: four electrode (FE) and four-point electrode (FPE) methods. The influence of conductivity on the conduit and floor section of the GDE was measured [152]. Zhou [32] modeled the electrical resistivity influence on GDL performance and demonstrated that the electrical resistivity of GDL could be ignored. Meanwhile, Bai [153] characterized the ionic conductivity of sulfonated polyarylenethioether sulfones as the electrolyte in a membrane fuel cell, and the data revealed that the ionic conductivity was dependent on the temperature and humidity. To improve the electrical conductivity of PEMFCs, the pressure pins between the components can be increased. However, the increase in pressure should be maintained so that component damage does not occur and reactant traffic in the GDL is not reduced [154].

4.2. Hydrophobicity

Many researchers are interested in hydrophobicity because it affects the water rejection rate of electrode components. Water molecules need to continue to the outer layer and should not be retained in the pores of the BL, GDL or CL because they inhibit the reactant gas flow and cause flooding for that reason, the electrodes must be sufficiently hydrophobic. In addition, the use of PTFE and the influence of PEF hydrophobic substances in the GDL on PEMFC performance has been studied [51,85]. Li et al. [75] investigated the effect of adding the hydrophobic substance dimethyl silicon oil (DSO) on PEMFC cathode performance and found that a DSO content level of 0.5 mg cm^{-2} was optimal. Several researchers have also focused on the influence of CL hydrophobicity on PEMFC performance; Yu et al. [155] investigated the hydrophobicity of CLs and their influence on PEMFC performance. Interestingly, these authors characterized the effect of time against hydrophobicity, by conducting a morphological study using FESEM; the observations were confirmed by measuring the contact angle. In addition, Chun [86] analyzed the contact angle of both a GDL double layer consisting of hydrophobic and hydrophilic layers and a GDL single layer in the order shown in Fig. 15; the contact angle observations are also shown in Fig. 16. Hydrophobicity is measured using a contact angle meter, which measures the contact angle of water molecules on the substrate surface. Greater contact angles imply higher hydrophobicity.

4.3. Porosity, surface and particle analyses

The porosity of the electrode (including the BL, GDL and CL) significantly affects the transport of the reactants and products at the electrodes [47]. The porosity also regulates the traffic between the oxygen gas entering the electrode and the water leaving the electrode.

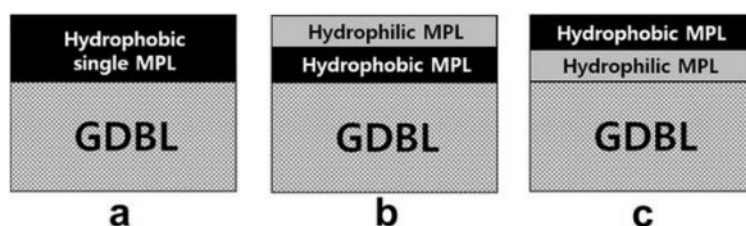


Fig. 15. GDL structures with single- and double-layer MPL: (a) PA2AML1, (b) PA2ADL2, (c) PA2ADL3 (Reprint permission from Ref. [86]).

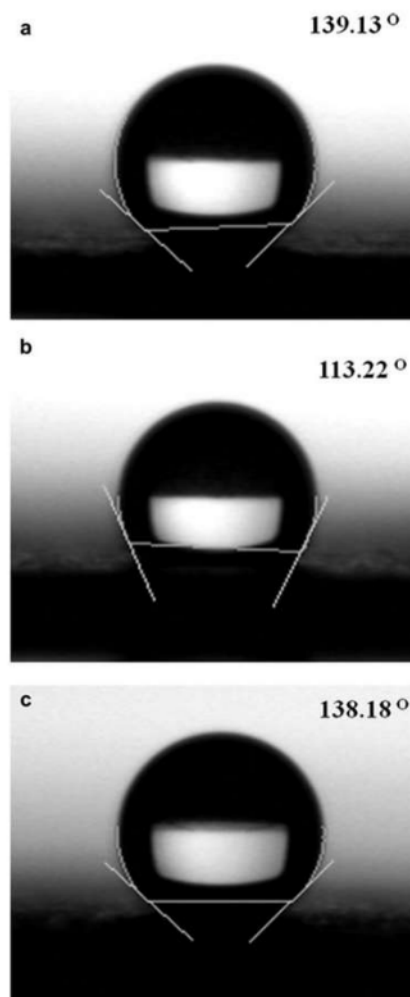


Fig. 16. MPL contact angles: (a) PA2AML1, (b) PA2ADL2, (c) PA2ADL3 (Reprint permission from Ref. [86]).

When, the porosity is balanced with adequate levels of hydrophobicity, oxygen is able to pass through the pores, even while water is exiting [8]. The influence of the pore structure on PEMFC performance gained the attention of Yoon [111]. An even pore structure facilitates the flow of oxygen to the reaction zone and the release of water. Some researchers have performed in depth studies of the influence of porosity on electrode performance based on the size or distribution of the pores. Jordan [54] compared the influence of the use of acetylene black (AB) carbon powder and Vulcan carbon black XC-72 by examining the

performance of electrodes based on pore sizes produced by the two types of carbon and found that the use of AB was superior because of the more effective transport mechanism of the reactants. Huang et al. [156] modeled the effect of porosity gradients on the performance of electrodes and observed that the porosity gradients helped to transfer condensate water, increased oxygen consumption and reduced the level of stress.

The porosity and surface structure were analyzed using SEM which can determine the pore structure, thickness and cross section structure of the electrode. SEM is typically combined with energy dispersive X-ray (EDX) measurements, which clarify the elemental composition of the electrode surface. EDX is a valuable technique that analyses changes in the elemental composition of a fuel cell layer either through particle migration, growth and catalyst particle accumulation or through fuel cell component degradation due to operation [157].

Transmission electron microscopy (TEM) analysis is very useful for analyzing the structure of the fuel cell because it can reveal the spatial distribution of each component and analyze the composition of the local area at the nano or atomic scale [158]. TEM has three important advantages as a material characterization technique. First, TEM captures images of material structures with atomic-level resolution; second, TEM provides a steady theoretical basis for image analysis; and finally, TEM is capable of microanalysis when combined with spectroscopic methods. The weakness of TEM is the difficulty of preparing samples and the sample damage caused by high-energy electrons [101,122,126,138,159,160].

X-ray photoelectron microscopy (XPS) is an internal characterization tool that uses monochromatic X-ray radiation. The full energy of the X-ray is transferred to an electron in an inner energy layer, which causes electrons to leave the surface. Photoelectron energy comes from the X-ray and provides the energy required to leave the surface. The energy required to escape from the surface depends on the type of atom present [101,122,123,136].

XRD is the most widely used technique for materials characterization. It is a nondestructive technique that reveals detailed information about the chemical composition and crystallographic structure of natural materials and synthetic substances [161]. This technique is a rapid and important method for determining the stability of a catalyst by calculating the size of the metal particles. Catalytic activity is highly dependent on the shape, size and distribution of the metal particles. In addition, catalyst supports also play a role in improving the catalytic function. X-ray diffraction and electron microscopy can both provide information about particles smaller than 10 nm. However, XRD analysis provides information regarding the size of fine crystals instead of the actual particle size, while TEM yields local information generated from the results by calculating the average volume [159]. The use of XRD analysis for the development of fuel cells includes Pt/C catalyst characterization using various types of carbon [101,117], non-platinum catalysts characterization [126,160], catalytic activity and durability tests in liquid electrolytes [147,159], characterization of the resultant catalyst [122], the influence of the catalyst content on thin layers of the electrode [162], and electro-kinetic oxygen reduction [163].

4.4. Electrochemical performance

Electrochemical characterization and electrode performance tests, especially in the CL, are accomplished using various methods, namely, CV, linear sweep voltammetry (LSV), and EIS.

CV is an important technique that is used often for electrochemical analyses because it offers a wide range of experimental information and insight regarding the kinetic and thermodynamic aspects of chemical systems. CV methods are used in fuel cell electroanalyses to calculate the electrochemical active area in the catalyst layer, estimate the limit of catalyst oxidation, and analyze the catalyst activity. CV analyses can be performed using in situ or ex situ methods. The in situ method uses a two-electrode configuration in which the analyzed electrode is the

working electrode, and the other electrode is used as the reference and counter electrode. The in situ method has been widely used to determine the electrochemical active area and analyze the use of catalysts in PEM fuel cells. In addition, the ex situ method uses three electrodes: the analyzed electrode is a working electrode, the counter electrode is a second electrode, and a calomel electrode is the reference electrode. The ex situ method can simply and quickly screen electrocatalysts, but it is not suitable for the assessment of fuel cell electrodes under different operating conditions [157]. Many researchers have tested the performance of electrodes using CV [110,126,159,163–165].

LSV is the most frequently used technique for estimating the gas crossover events that pass through the membrane in a PEM fuel cell, characterizing the membrane deterioration, and detecting short circuits. LSV can be used directly under various fuel cell operating conditions [138]. During the test, the linear potential of the fuel cell electrode is scanned to obtain the limited current, which is useful for calculating the rate of cross-hydrogenation. This test can be performed online without disassembling the cell system. Thus, the gas cross rates provided by LSV reflect the realistic membrane situation under actual working conditions. Due to its convenience and suitability, LSV has become a basic test method for determining the suitability of different materials when evaluating a new membrane. During deterioration tests in the worst scenarios or under long-term fuel cell operation, LSV can be easily used before and after surgery or during any period of operation to evaluate the evolution of gas crossover. Lee [166] conducted a galvanostatic analysis of single and multicells in situ without needing to disassemble the MEA.

EIS is widely used by researchers for characterizing and testing PEMFCs [167]. EIS can be operated without damaging the sample, providing detailed diagnostic information on the electrochemical phenomena that occur including the load transfer reactions at the electrode and electrolyte interface, the reaction mechanisms and the electrode material properties. EIS has been used for PEM fuel cell studies with the following goals: (1) to provide microscopic information about the fuel cell system, which in turn can aid in structure optimization and the selection of the most suitable operating conditions; (2) to equip the system model with an appropriate equivalent circuit and obtain electrochemical system parameters; (3) to distinguish the individual contribution of each component, which can help in identifying problem fuel cell components; and (4) to identify individual contributions to the total impedance of the PEM fuel cell from the different electrode processes, such as interface charge transfer and mass transport in both the catalyst and supporting layer [168].

At least one review of the EIS method has been published, which can be used to determine the capacity resulting from the kinetic process, ohmic loss, and mass transfer, and this review has also successfully distinguished the influence from various processes [168]. Several researchers have used EIS for PEMFC characterization to accomplish various purposes: to perform a PEMFC multistack analysis operated under various conditions and loadings [169], to diagnose PEM fuel cells [170,171], to correlate the capacitance of the catalyst active area and ion transportation properties with ionic conductivity [172], to analyze high-temperature PEM fuel cells [173,174], to characterize the anti-flood mechanisms in the CL [175], to test the CL using different parameters [171], to characterize the PEMFC failure [176], to evaluate the MPL [177], to investigate GDL compression in ongoing fuel cells [178], and to determine the post force of the membrane protons at frequencies between 1 and 300 kHz using a swing voltage [179].

5. Electrode management system

System management is the comprehensive work that generates efficient energy by controlling few a few parameters in the process to obtain an expected outcome based on the mode of application. The most common variables of electrode system management are discussed below.

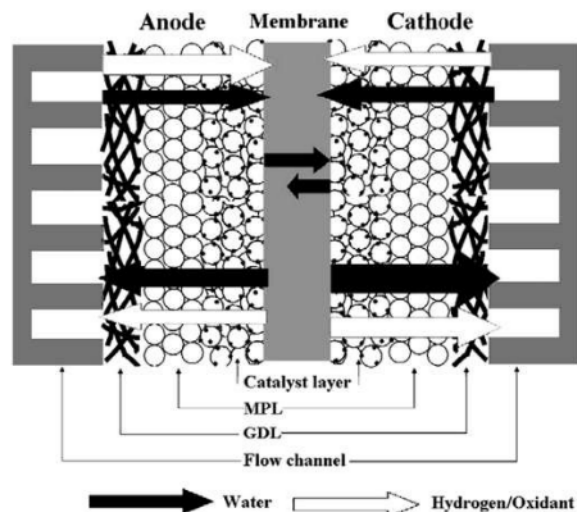


Fig. 17. Mass flow diagram in PEMFC electrodes (Reprint permission from Ref. [181]).

5.1. Water management

Water management within the electrode cannot be ignored. Water is needed to hydrate the electrolyte membrane and accelerate H^+ circulation in the CL. For these conditions, adequate amounts of water are necessary. However, the presence of excess of water causes flooding that can cover the GDL and CL pores. Thus, water management is one of the most important issues for the commercialization of PEMFCs to ensure high proton conductivity in the electrolyte membrane and to supply sufficient amounts of reactants to the reaction center [180,181].

During the electrochemical reactions of a PEMFC, electricity, water and heat are generated. The generated heat evaporates the water in the reaction system causing drying, particularly on the membrane. Perfluorosulfonic acid (PFSA) membranes, such as Nafion, exhibit their protonic post force in the hydrated state. The increased reaction heat accelerates the membrane drying process, thus decreasing the PEMFC performance and increasing the resistance of the membrane. For this reason, adequate membrane hydration without causing flooding is an important aspect for water management [180]. Therefore, the GDL must regulate the traffic, quantity of reactants, and water management. The mass transport mechanism in PEMFC electrodes is illustrated in Fig. 17. The role of the MPL in regulating water can be seen in Figs. 18 and 19.

A few studies have specifically discussed water management within electrodes by using X-ray imaging to evaluate the PEMFC system [183], characterizing the water produced at different operating temperatures [184], removing water in the anode as a diagnostic tool to check flooding in PEMFC cathodes [49], characterizing the water transport in the GDM [63,185], transferring the water from the GDL by reactant flow [186], and studying water in the liquid form in PEMFCs [187].

Some water management considerations from studies conducted by researchers are summarized below.

- The MPL should have a high level of hydrophobicity and small GDL pore size (in the case of using a BL) to prevent flooding at the electrodes. The small pore size (especially at the CL-MPL interface) prevents the growth of water droplets and reduces the saturation level at the CL-MPL interface [7].
- The water balance is critical for not only the performance but also the durability of the PEMFC [182].
- During water condensation, the water flows due to capillary pressure, which is determined by the pore radius and hydrophobic level.

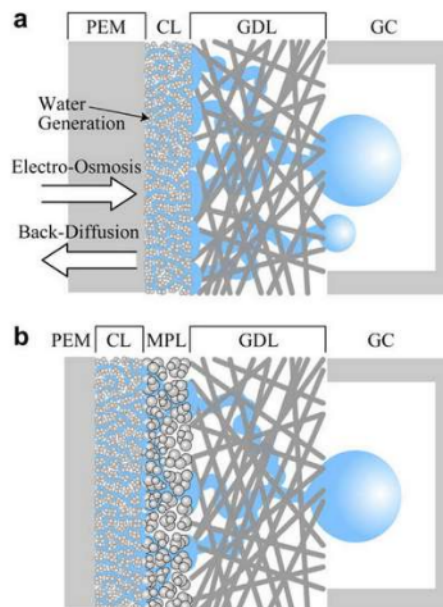


Fig. 18. Cathode structure a) without and b) with the MPL (Reprint permission from Ref. [7]).

- Hydrophobic materials are one of the criteria that facilitate water management [51,74,85,86,175].

5.2. Reactant management

Reactant management has similar functions in both high and low temperature PEMFC systems and is important when the PEMFC is applied in loading because uniform feed loading is required for linear PEMFC performances. In addition, an insufficient reactant supplies at either the anode or cathode directly affects the current density and voltage in cells. At the cathode, the reactant management depends on three factors: the optimum flow rate, pressure, and stoichiometric ratio [188]. Regarding the electrochemical reactions, for the fuel oxidation on the anode side, both pure hydrogen and reformat gas containing up to 3% carbon monoxide are acceptable for use, and oxygen or air can be used for the oxidant reduction on the cathode side [189]. The electrochemical reaction is favorable when a high supply of hydrogen gas feed is used, possibly leading to a high power density but leading to an inefficient net power output or a tendency to damage the MEA [190]. However, the reactant supply is too low, then feed starvation at the electrode may occur. This issue is related to a few factors, such as rapid changes in the load, improper gas supply, start up or start down process, failure of the bipolar gas supply channel in a single cell or stack, and system failure. Therefore, the reactant supply should be at an optimum flow rate to obtain a uniform current density [143,191–194]. Orfanidi et al. [195] reported that hydrogen starvation occurred due to catalyst flooding by the acid leaching from the polybenzimidazole electrolyte membrane. The electrochemical active surface area (ECSA) of an electrode is gradually reduced due to the corrosion of carbon based catalyst support materials [148,196–198].

5.3. Heat management

In PEMFCs, heat energy is generated during the electrochemical reactions. Therefore, PEMFCs are exothermic reaction systems when the combustion of hydrogen and oxygen reactant gases, particularly for the reduction of oxygen on cathode side. The generated heat is

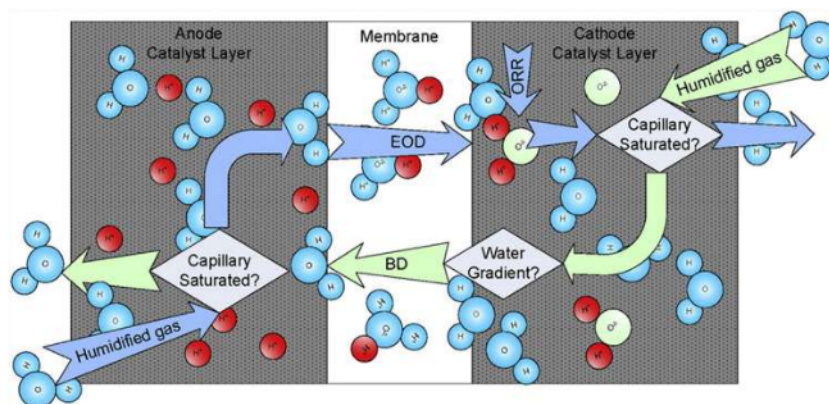


Fig. 19. Mechanism of water transport in the CL and membrane (Reprint permission from Ref. [182]).

transported over the cooling cell (which is inside the bipolar plate) and through the flow channels containing the reactant gases inside the anode and cathode. Heat management is another important parameter for cell performance. The even distribution of a metal (usually Pt) catalyst in the electrode enables even electrochemical reactions to proceed and heat energy to be produced. In contrast, an uneven catalyst distribution causes uneven reactions and variable amounts of heat energy; moreover, an uneven distribution may generate a local hot spot inside the MEA, leading to drying out of the electrolyte membrane especially in low temperature PEMFCs. Therefore, the ionic resistance of the membrane may be increased, thus lowering the proton conductivity from the anode to the cathode side [52]. Cathode flooding primarily causes water accumulation in the cathode pores, which prevents oxygen transport to the electrode reactive sites. This issue is caused by both improper heat and water management. The heat distribution is related to the condensation or evaporation processes of the cathode, which in turn cause cathode flooding [199].

High temperature PEMFCs have potential advantages over low temperature PEMFCs, such as high electrochemical reactions, low ionic resistance of the membrane, high ORR rate, high tolerance of fuel contamination and simple system design. However, these cells have an accelerated degradation rate of the cell components, thus reducing the life time of the cell. Therefore, the temperature should be precisely maintained at the optimum level to stabilize the cell performance. In addition, heat transport from the cell stack chamber can be difficult. Zhang et al. [200] suggested that the heat can be removed quickly by using the cathode air flow, thus simplifying the design of the stack system, and directly reducing the cost while improving the power density. Furthermore, PEMFCs can be stored at ambient temperature with the cell components at a different temperature. In this manner, heat is rejected rapidly due to the large temperature difference between the cell stack and the surrounding environment [189].

6. Post-processing of PEMFC

Post-processing of PEMFC is a little bit complicated term that refers to the take care of cell stack, repair or replacement of single/multiple cells in the stack, disposal of the cell stack, input variables of stack etc. Therefore, it is closely related to the overall cell durability, performances, and environmental viability. The PEMFC stack or single cell is needed the replacement for one or two more times of entire vehicles life. During the replacement or disposal of the cell, it is necessary to consider the environment and health security. The NIOSH (national institute of occupational safety and health, USA) was research conducted and suggested that the nanoscale particles such as carbon nanotube, graphene, carbon powder, carbon nanofibers etc. have a little

bit health risk compare than the bulk materials [201]. In addition, a metal catalyst such as Pt, Pd, Ni, Ru etc. have also toxicity characteristic in some cases that are commonly used as supporting materials in PEMFC electrodes [202,203]. Moreover, the most common input variables of the cell are namely as back pressure, anode stoichiometry, cathode stoichiometry, relative humidity, inlet gas temperature and so on that are closely connected with the overall cell performance as well as cell longevity. Zhang et al. [204] were suggested that the poor management of those operating parameters can badly have effected on the stack durability. Kanani et al. [204] also found that the high and low stoichiometry of cathode and anode respectively can attribute lower power density. In contrary, at the medium level of gas feed and oxidant stoichiometry regards the higher power density.

7. Applications of PEMFCs

Over the last decade, the convincing worldwide research efforts have caused PEMFCs to successfully pass the demonstration and commercialization. However, these fuel cells must overcome challenges, including infrastructure development, cost reduction, cell durability, H₂ fuel refilling and storage system issues. However, PEMFC technology is one of the most promising green technologies and represents a realistic energy conversion system for the future. Currently, PEMFCs have been applied in various applications, such as transportation (public and private sectors), stationary power, portable devices, aircrafts, military submarines, and toys. [205].

PEMFCs based vehicles have over potential advantages over battery and electric based vehicles regarding CO₂ emissions and a much greater well-to-wheel efficiency. PEMFC-based vehicles also have a long driving capacity, short start-up time, dynamic load demand and shorter refueling time than battery and electric-based vehicles. The transportation mode is predominantly focused on buses, cars, trains, golf cars, trams, motorcycles, and bicycles, as well as material handling transport applications. America, Japan, Canada, Germany, and Australia, along with many other countries have been launching fuel cell-based vehicles for public and private sectors in particular areas to raise the public's awareness. Moreover, many well-known bus and car manufactures, such as Honda, BMW, Toyota, Hyundai, Ford, Mercedes-Benz, Mitsubishi, Nissan, Kia, Suzuki, have been commercializing PEMFCs-based vehicles. Since 2004, this technology has been successfully demonstrated for use as passenger cars. During the initial stages, problems such as start-stop operation and steep transient load cycling (leading to water management and gas transport problems) primarily affected the lifetime of these FC systems [206]. Many problems have already been solved over the last few years, but methods of cost reduction are still being considered. In 2014, the DOE calculated the large-scale

production costs (500,000 units/annum) as \$55/kW for 80 kW PEMFC systems. The ultimate target is \$30/kW for 30,000 units, and the cost is expected to be \$40/kW in 2020. Some car manufacturing companies are namely, Daimler, Nissan, and Ford are forming an alliance to allow for a flexible cost burden, and they will produce PEMFC-based vehicles together from 2017 onwards [205].

For portable applications, the power requirement is approximately 25 W to 5 kW, but in reality, portable PEMFC devices are not very promising. In this case, the focus has been on the reducing the emissions and noise and on optimizing device operation conditions rather than energy efficiency. These devices are also popular for military purposes, and Hitachi, Toshiba, NEC, Samsung, Panasonic and use PEMFC and DMFC power supplies for notebooks and mobile phones in the range of ~ 5–75W [207,208].

PEMFCs can also be applied for stationary power supply purposes, such as residential combined heat and power (resCHP), large CHP, and uninterrupted power supply (UPS). Tri-generation systems are under development for power, cooling and heat (via an added absorption chiller), mainly for areas in which the thermal demand during the cold season is balanced by an almost equal cooling demand during the hot season [209]. Moreover, PEMFCs can be used for fire prevention by reducing the oxygen from the air in areas where an accident occurred. In 2014, 70% of global fuel cell revenues was from the stationary fuel cell sector, and this revenue level is expected to continue in the coming years. Navigant research teams have reported that the annual shipments of stationary FCs will grow from nearly 40,000 in 2014–1.25 million in 2022 [205].

8. Future developments and conclusions

The electrode is the heart of the MEA of PEMFCs and it plays a vital role with important functions. The electrode converts chemical energy to electrical energy during electrochemical reactions that are directly related to the cell performance. However, the electrochemical reaction pathway is not simple because it involves the mass transfer of materials and reaction products as well as electrons. Therefore, understanding the structure and function of each component, the manufacturing methods, and the characterization and testing methods for the electrode performance and durability is critical. Regarding these points, electrode research is necessary for improving electrode performance.

The preparation of an electrode is a comprehensive process that includes the GDL, CL and MPL layers. Dry preparation processes for MPLs are better than wet processes during fuel cell operation with high oxygen consumption under high humidity conditions. Moreover, dry processes produce more mesopores, higher electrical conductivity, more stable hydrophobicity and higher mass transport. The PTFE content in the MPL should be maintained at the optimal level while increasing the hydrophobicity which has an appreciable effect on the GDL conductivity. Furthermore, the GDL thickness needs to be considered during the GDL preparation process. In addition, the Pt content in CLs can be reduced without significantly affecting the performance of the fuel cell by using Pt alloy (Pt-Ru, Pt-Co, Pt-Pd, Pt-Ru-Co and Pt-Co-Cr) or bimetal (Pt with other metals such as Fe, Co, Cu and Ni) catalysts. Moreover, using the optimum ionomer content in the CL improves the electrochemical active surface area, ensures good compatibility and enhances the ionic conductivity. The catalyst ink distribution, Pt utilization and agglomeration of catalyst particles should be considered during electrode fabrication. Furthermore, durability is still a major challenge for the commercialization of PEMFCs. To maintain the degradation rate at minimal level, the material compatibility should be considered during the electrode fabrication process.

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