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Recent Advances in Proton Exchange Membrane Fuel Cells: A Review

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Abstract: A fuel cell is an electrochemical energy converter that converts the chemical energy of fuel into electrical energy. Among the various types of fuel cells, the present review focusses on Proton exchange membrane fuel cells (PEMFCs). High energy conversion efficiencies, quick start-up and zero by-product emissions are the key advantages of PEMFCs. Main components of PEMFCs are Membrane Electrode Assembly (MEA), Gas Diffusion Layer (GDL), bipolar plates and so on. This paper mainly reviews and summarizes research carried out on proton exchange membrane fuel cells in the past six years (2010 to 2016). For ease and clarity of presentation, the present review is divided into experimental, numerical, combined experimental and numerical, and studies on some of the reviews on fuel cell. The paper also lists conclusions and suggestions for future work.

Keywords: Fuel cells, proton exchange membrane fuel cells, gas diffusion layer, bipolar plate.

Nomenclature

GDL-Gas diffusion layer
MEA-Membrane electrode assembly
MPL-Micro porous layer
PEMFC-Proton exchange membrane fuel cell
PTFE-Polytetrafluoroethylene

I. INTRODUCTION

Energy demand is increasing day by day, but its production is decreasing. Mainly traditional fossil fuels are used but pollutions and its effects, depletion of fossil fuels so on leads to the development of renewable sources and other alternatives. However, existing issues including high cost, intermittency of energy supply, grid connection and storage are the main concerns with renewable energy sources. Among the various techniques to mitigate this problem, fuel cells plays a vital role. Fuel cells using hydrogen as fuel is collectively known as hydrogen fuel cells. Most of the fuel cells uses atmospheric air as the oxidizer.

Different types of fuel cells are Proton exchange membrane fuel cells,Alkaline fuel cells, Direct methanol fuel cells, Solid oxide fuel cells, Phosphoric acid fuel cells,Molten carbonate fuel cells. Of these Proton exchange membrane fuel cells are very promising because of its low operating temperature(in the range of 100°C),Polymeric material(Nafion) is used as the electrolyte which avoids concentration problems when compared with electrolytes in other fuel cells, high fuel utilization etc. The working of PEMFC is defined by the reactions 1.1 to1.3:

At the anode $H_2 \longrightarrow 2H^+ + 2e^-$……………………………………….1.1

At the cathode $(½) O_2 + 2H^+ + 2e^- \longrightarrow H_2O$…………………………………………....1.2

Cell reaction $H_2 + (½) O_2 \longrightarrow H_2O$…………………………………………….1.3

Hydrogen is given to the anode side and oxygen to the cathode side, hydrogen is split into its ions and electrons by oxidation reaction at anode and at cathode reduction reaction takes place. Ions produced at the anode side is transferred to the cathode side through the electrolyte and electrons flows to the cathode side through an external circuit, load can be connected to the external circuit to take the electric current output. At cathode side water is produced which can be taken with the help of proper water trap system.

The main parts of a PEMFC are shown in Fig. 1.

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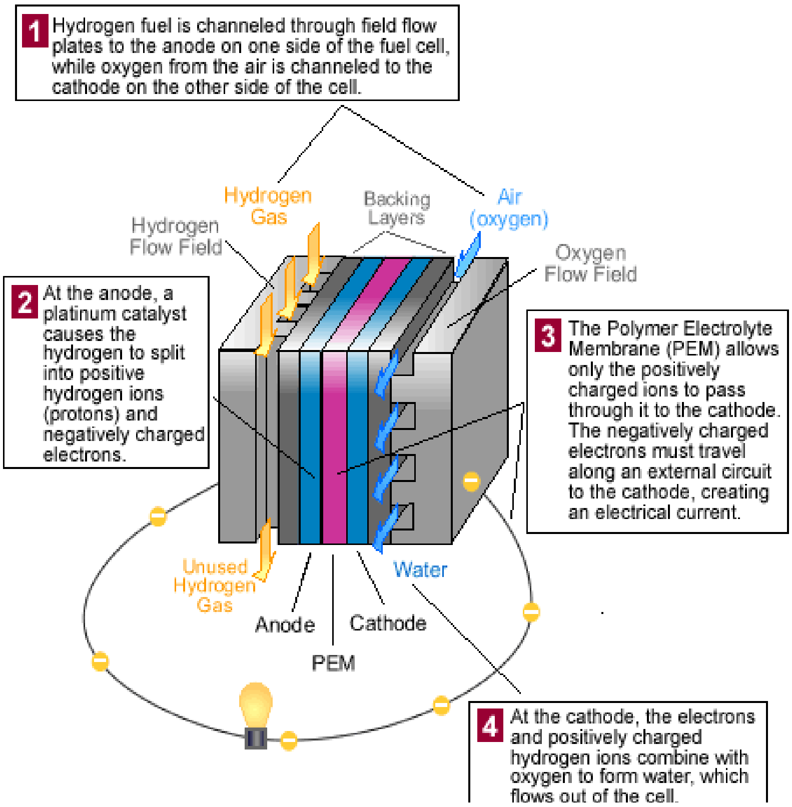

Fig. 1. Components-Proton Exchange Membrane Fuel Cells[27]

A. Membrane Electrode Assembly

The polymer electrolyte membrane, catalyst layers, and Gas Diffusion Layer (GDL) together form the Membrane Electrode Assembly (MEA) of a PEMFCs. Nafion is used as the polymer material which helps in the transportation of ions from anode to cathode side and it block the passage of electrons. Catalyst layers attached to the anode and cathode side. Usually platinum is used as the catalyst which helps in the oxidation reaction in the anode side and reduction reaction in the cathode side. Gas diffusion layer is made up of carbon paper and it is coated with polytetrafluoroethylene (PTFE). GDL helps to the transport of reactants into the catalyst layer, as well as removal of product water.The micro porous layer can help to adjust the balance between water holding and water release capacity.

B. Bipolar plates

Each membrane electrode assembly produces voltage which is very small (in the range of 1 V), as a result in practical applications multiple MEAs are used which is connected in series and forms a sandwich structure. Bipolar plates are used to hold the MEA structure in series.

II. REVIEW OF LITERATURE

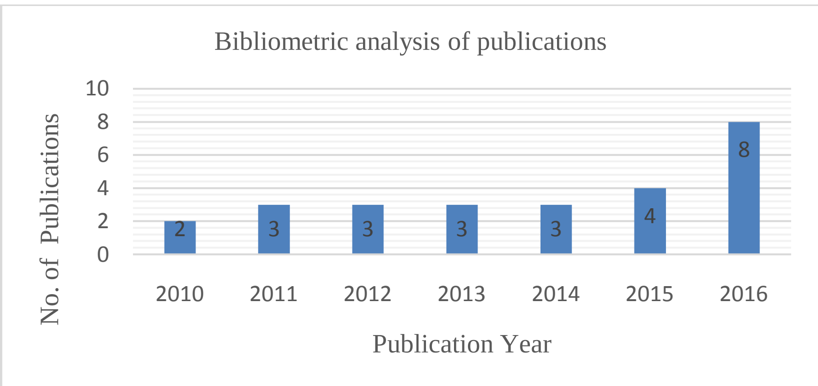

Fig. 2. Bibliometric analysis of publications used in the present study

Many review papers are available on fuel cells [24, 25, 26] but majority of them were not considering the general trends in PEMFCs. This paper is an attempt to review the papers mainly PEMFCs from 2010 to 2016 detailing the various components considered, important parameters analysed and also list key results.Bibliometric analysis of publications are shown in Fig. 2.

A pie-chart of apportioning the literature in the present review is shown in Fig. 3. A total of 26 papers are taken for this study. Out of these, thirteen papers focusses on experimental, five on numerical, five on combined experimental and numerical investigations and three explainsvarious reviews of fuel cell.

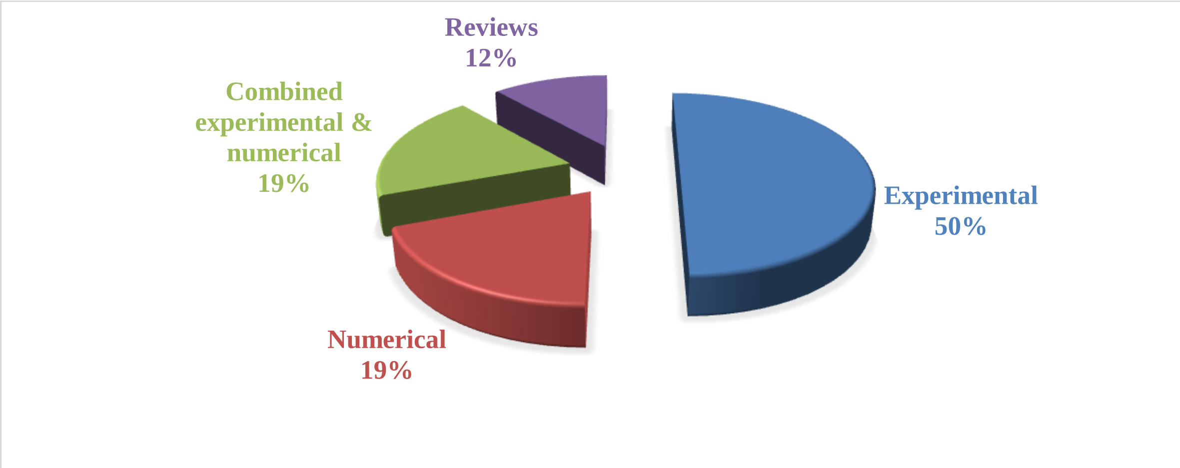

Fig. 3. Chart of apportioning of the literature

A. Experimental Investigations

Many researchers have experimentally studied about the proton exchange membrane fuel cells and its performance level. Yusuke, et al. [1] analysed the effect of humidity in PEMFC. High humidity level in PEMFC helps to achieve high ion conductivity and thus cell performance but this causes the water flooding. Effective water management is required for the effective running of PEMFC. Humidity factor and water flooding mainly depend on the pore structure of GDL. Simple carbon paper is used to prepare a high resistant GDL. Experimentally it was observed that flooding was prevented by a GDL with a smaller pore structure. An investigation carried out by Giovanni, et al. [2] analysed the GDL compression by Electrochemical Impedance Spectroscopy. Uses two GDLs based on same carbon cloth with and without MPL. It was observed that ohmic and mass transfer resistance were sensibly affected by increase in compression of the GDL. Chien, et al. [4] reported the design of a ultra-thin GDL. Carbon fibre paper is used as the GDL. Polyacrylonitrile (PAN) carbon fibre of length 3, 6 and 12mm was selected. Thickness of GDL in the range of 100 µm. Results shows that 3 and 6mm length carbon fibre have more strength than 12mm because, in long carbon fibres bonding is not stronger. Ultra-thin GDL(thickness 48µm) is best suited to reduce the fuel cell stack volume. Schweiss, et al. [5] conducted the design of MPL with multiwall carbon nanotubes using wet chemical method. Multiwall nanotubes helps to improve mass transfer, electronic conductivity. Multiwall nanotubes shows excellent electric and heat conductivity.

Yuka, et al. [6] focussed their attention to the cell voltage and life time. Long lifetime with high cell voltage is the one of the basic requirement that is expecting from the fuel cells. Cell voltage decreases as time increases because of the depletion of the phosphoric acid. Chemically cross linked poly membranes were used. Test results shows that cell with ordinary PEMFC shows 10% decrease in cell voltage with given period where new cross linked poly membranes assisted cells shows only 4.4% decrease in cell voltage for the same time period. Jason et al. [7] address the challenges of PEMFCs and how to overcome these challenges. Parameters which are specifically related to GDL and Micro Porous Layer(MPL) were analysed in this experimental study. Lifetime of a fuel cell mainly depends on catalyst and membrane durability. Main parameters of GDL which affects the cell performance are reactant permeability, product permeability, electrical conductivity, thermal conductivity, mechanical support. GDL thickness is an important factor. Thicker GDL will provide better protection for the membrane, gives mechanical strength, have longer diffusion paths lengths, gives more thermal and electrical resistance. Around 10% difference in cost for a 50% increase in thickness, but it is very difficult to handle very thin materials (<100 microns). It was observed that thick GDL is best suited for low current density or dry applications and thin GDL for high current density or wet applications. Satoki, et al. [8] introduces a hydrophilic layer in the GDL. Hydrophilic layer is placed between MPL and the carbon paper. Experiment was conducted at normal operating temperature and sub-freezing temperature conditions. GDL with hydrophilic layer shows high performance when compared with conventional GDL.

Kong, et al. [9] focused their studies in self-humidification effect in PEMFCs. Uniform structured GDL and heterogeneous structured GDL was used for the experiment purpose. Heterogeneous structured GDL shows better electrical performance. Durability of fuel cells and related investigations was conducted by Zhang et al. [10]. Durability is the one of the important challenges for the commercialization of PEMFCs. Experiment aims to recover losses due to kinetic and mass transport after accelerated stress tests. Platinum oxide reduction and platinum reattachment is the main reason for the kinetic recovery, where removal of oxide groups causes mass transport. Results shows that performance degradation can be recovered by performance recovery methods. Recovery methods help to improve the lifetime. Kong, et al. [11] reported start up characteristics of fuel cells. Start-up characteristics was analysed at different flow arrangements, mainly co-flow and counter flow, counter flow arrangement has good start up characteristics. It was observed that reactant flow arrangement has significant influence in the stable operation of fuel cells. Start-up cell temperature or initial temperature has also significant influence in the cell performance.

Heterogeneous porosity and its effects was analysed by Banarjee, et al. [12]. Analysed the bilayer structure of GDL. X-Ray computed tomography is used to study the structure of the bilayer GDL. This is a NDT method. Many other techniques commonly used for the study of structure of GDLs are mercury intrusion porosimetry, atomic force microscopy, optical profilometry etc. Results shows that porosity distribution has a strong basis for the calculation of thermal and electrical conductivities, permeability and diffusivity. Iryna, et al. [13] mainly focused their attention towards the compression and porosity. GDL porosity measurement is very challenging because of their thin, compressible and inhomogeneous geometry. Result shows that porosity decreases with compression of GDL. Porous structure and mass transfer effects wereanalysedby Takahiro Suzuki, et al. [14].In this study porous structure formation of catalyst layer was analysed. Porous structure has significant effect on the mass transfer in the electrode. Experiment was conducted at different hot pressing pressure varies from 0.5 to 10MPa. Results shows that pore size has more importance than porosity with respect to over potential in the polarization curve. A summary of experimental investigation is given in table 1.

B. Numerical Investigations

Huang, et al. [15], reported the interaction between heat and water transport in PEMFCs. Performance mainly depend on the platinum metal, but cost of platinum metal is very high. Results shows that porosity gradient enhances the gas transport which in turn improve cell performance. Fanglong [16] developed the design of fractal model, which is used to predict the liquid water permeability of the GDL. Here GDL having parallel and perpendicular combination of channels to the fluid flow direction is used. Fractal model is more suitable for the prediction of permeability of GDL. Fractal model gives a strong foundation for the design of GDL with enhanced fuel cell performance.

Dahua et al. [17] developed 1 dimensional, 2 dimensional and 3 dimensional model which is used to analyse the diffusivity. Influences of porosity, fibre distribution, fibre orientation were analysed, Results shows that randomly distributed fibres have lower diffusivity when compared with orderly distributed fibres. Diffusivity can increased by using less chemical binders and prevention of chemical binders. Degradation rate was reported by Taegon,et al. [18]. Degradation rate was analysed in high temperature PEMFCs. Effects of degradation with respect to temperature was considered. Degradation factors mainly affects the activation, ohmic and concentration potentials. Results gives an idea about how temperature changes affects life time and average performance of fuel cell and also it was observed that high temperature PEMFCs degradation rate at initial time is low and then increases rapidly later.

Straubhaar, et al. [19] developed Pore Network Model. This is used to find out the liquid water formation by vapour condensation in the GDL at an operating temperature of 80°C. Pore Network Model is very essential and useful to find out the transfer phenomena in GDL. This model is used to stimulate the condensation in GDL. Water transfer is connected with heat transfer because condensation in GDL is due to the temperature difference in GDL.

TABLE I SUMMARY OF EXPERIMENTAL INVESTIGATIONS REPORTED IN THE PRESENT PAPER

SL. No.	Authors	Year	Component considered	Parameter analysed	Key results
1	Yusuke, et al. [1]	2010	GDL	Pore diameter, humidity level, water flooding	GDL having low pore diameter (24 μm) shows 0.7 V cell voltage at 0.5 Acm^{-2} (current density) and GDL of 45 μm pore diameter shows only cell voltage < 0.2 V at 0.5 Acm^{-2}
2	Giovanni, et al.[2]	2011	GDL	Compression of GDL with and without MPL	Without MPL power density is < 0.30 Wcm^{-2} at a current density of 0.7 ACm^{-2} but MPL coated GDL shows power density of 0.35 Wcm^{-2} at the same current density. Presence of MPL improves

					the power density.
3	Chien, et al. [4]	2012	GDL	Thickness, length	6 mm length carbon fibre shows better mechanical strength when compared with 3 mm length carbon fibre. Ultra-thin GDL produced was 48µm thickness gives a tensile strength of 32~39 N/cm and current density of 815 mAcm^{-2}
4	Schweiss, et al. [5]	2012	MPL	MPL having multiwall carbon nanotubes	Observed 50 % reduction in the resistance by the addition of multiwall carbon nanotubes in the MPL.
5	Yuka, et al. [6]	2013	High Temperature PEMFC	Temperature (150 ° C), cross linked poly membranes	Conventional systemshows 10% decrease in cell voltage with given period where new cross linked poly membranes assisted cells shows only 4.4% decrease in cell voltage for the same time period.
6	Jason et al. [7]	2014	GDL	Thickness, Relative Humidity (RH)	At dry operating conditions (<30% RH) thick GDL(55µm) shows maximum current density(1.5 Acm^{-2}) at 0.4 V but thin GDL(5µm) shows only cell voltage less than 0.2V. At wet operating conditions (>70% RH) thin GDL shows maximum current density (> 1.5 Acm^{-2}) at 0.4 V but thick GDL maximum current density is 1.2 Acm^{-2} and cell voltage is less than 0.1 V.
7	Satoki, et al.[8]	2014	GDL	Hydrophilic layer between MPL and carbon paper	With hydrophilic layer cell voltage is in the range of 0.75 V (at 1.5 Acm^{-2} current density & 100 % RH). Without hydrophilic layer cell voltage is in the range of 0.70 V (at 1.5 Acm^{-2} current density & 100 % RH).Introduction of hydrophilic layer improves cell voltage by only ~7%.
8	Kong, et al. [9]	2015	GDL	Self-humidification, uniform GDL, heterogeneous GDL	Heterogeneous GDL shows current density > 1.2 Acm^{-2} at 0.4 V(at self-humidification condition), but under these conditions uniform GDL shows current density in between 1 and 1.2 Acm^{-2}. Similarly heterogeneous GDL shows 0.5 Wcm^{-2} power density but uniform GDL power density is less than 0.4 Wcm^{-2} (at self-humidification conditions)
9	Zhang et al. [10]	2015	GDL	Recovery mechanism, Accelerated Stress Tests(AST), degradation percentage	With recovery mechanism maximum current density obtained was 700 mAcm^{-2} without recovery mechanism only 100 mAcm^{-2} was obtained(under 15 hours after AST & 0.2 V). 30% reduction in the percent of degradation of performance was obtained by recovery mechanism(15 hours after AST)
10	Kong, et al. [11]	2015	Anode and cathode	Flow arrangements in anode and cathode side	Counter flow arrangements shows a current density in the range of 0.8 Acm^{-2} and co-flow arrangements current density was less than 0.2 Acm^{-2}. Hydrogen crossover and membrane hydration is responsible for the changes in current density in both arrangements.
11	Banarjee, et al. [12]	2016	GDL	Porosity distribution with and without MPL	Porosity distribution is more uniform in GDL without MPL, but it is complex in GDL having MPL.
12	Iryna, et al. [13]	2016	GDL	Compression and porosity	Porosity decreases with compression. GDL sample (SGL 24 BA) shows reduction in porosity from 0.77 to 0. 57 for strains 0.02 and 0.4

13	Takahiro Suzuki, et al. [14]	2016	Catalyst layer	Porous size distribution in catalyst layer	Polarization curve shows drastic drop in voltage when pore size distribution changes from 70 to 30 nm. Compression of catalyst layers results in decrease in pore size distribution, so more compression will results in more reduction in voltage.

C. Combined Experimental and Numerical investigations

The combined experimental and numerical investigation on fuel cells conducted by Enju, et al. [20], mainly focused on the role of micro porous layer and how it affects the water transport system. If water level is low which will increase ohmic resistance, if water level is high it will leads to water flooding. So water management is an important aspect. Capillary pressure of MPL and GDL helps in the water transport system. Pore Size Distribution model was used to find out the capillary pressure. Study mainly focused to establish relationship between capillary pressure and liquid saturation. It was found that analysis of capillary pressure of MPL has significant role in the cell operation especially in high humidity conditions.

Tamayol, et al. [3] mainly deals with PTFE content and mechanical compression of PEMFCs. Permeability was calculated with the help of PTFE content and mechanical compression. Experimental analysis shows a reverse relationship between the permeability and PTFE content. Also the authors address the main challenges associated with PEMFCs, main challenges are reliability, cost, power density, performance and membrane lifetime. Mass transport loss which is related to GDL permeability results in the reduction of maximum power density. Cell components were compressed together to avoid gas leakage from the system. More power density will affect the proper water management system.

Zhang, et al. [21] analysed the effects of variation in local current density. Numerical model was used to analyse the optimum porosity distribution. Experimental study was used to analyse the local current density and porosity distribution. Main challenges addressed by the authorsis the low reliability in service life because of MEA degradation. Variation in local current density is due to the reduced reactant concentrations. Variations in local current density increases temperature and stress gradients. Optimum porosity was found to be decrease with increase in temperature and increase with increase in cell voltage.

Reduction in the size of fuel cells was conducted by Kong, et al. [22]. External humidifier is the one of the important component which will helps to maintain proper humidification in the cell. Self-humidification is achieved and avoid external humidifier, which will helps to reduce the size of PEMFCs. This study gives support to the design of self-humidified PEMFCs. Shiro, et al. [23] suggests the usage of metal sheet GDL. Perforated metal sheet was used. Stainless steel 316L was the material selected. Numerical model conducted with the help of simulation software STAR-CCM+. Results shows that metallic GDL gives sufficient strength and thickness to the GDL structure. But one of the drawback of this metallic sheet GDL was the chances of corrosion which will reduce the lifetime of the cell.

D. Various Reviews on Fuel Cell Designs.

Yun et al. [24] conducted a review about the application of fuel cells, research fundamentals and major challenges in the field of PEMFCs.Linfa, et al. [25] focussed their attention towards the bipolar plates of fuel cells. Usually graphite plates were used as the bipolar plates, but in this paper authors reported the usage of Stainless steel bipolar plates and its significance. Flow field design, micro forming process, joining process and coating process are the steps in stainless steel bipolar plates formation. Bipolar plates should have good electrical conductivity, high resistance to corrosion, mechanical stability, light weight, cheap etc. Graphite and its composites having these characteristics but its high production cost, brittleness etc. are some of the drawbacks. Authors suggested that stainless steel bipolar plates are more effective than graphite bipolar plates. Wang, et al. [26] reported about unitized regenerative fuel cells (URFC). URFC consist of fuel cell part and electrolyser part and these two parts are unitized in a single electrochemical cell. Advantages of URFC are lower capital cost, simpler structure, higher specific energy, no need for auxiliary heating. URFCs have wide application in aviation, aerospace, power supply and transportation etc.

III.CONCLUSIONS

From the literature survey, it is seen that the GDL and its porous structure has the significant effects on the performance of PEMFCs. The following lists conclusions and the future scope.

1. The analysis of the recent literature on PEMFCs shows the dominance of experimental techniques over numerical techniques.
2. From the review of the literature, it is seen that porous and porosity factors of GDL were greatly influence PEMFCs.

3. Majority of literature focused their attention to improve the electrical performance of PEMFCs. Thermal analysis were scarce in the literature. More studies are required to highlight the thermal performance of PEMFCs.

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