PROTON EXCHANGE MEMBRANE FUEL CELLS: TECHNOLOGY, PROSPECTS AND CHALLENGES

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Abstract*--Proton exchange membrane fuel cells (PEMFCs) are considered to be a promising technology for clean and efficient power generation in the twenty-first century. This interest is because of their high efficiency, high power density and zero environmental pollution. Polymer electrolyte membrane fuel cells (PEMFC) are the most appropriate type of fuel cells for use in vehicles due to their low performance temperature and high power density. Proton exchange membrane is the key component of fuel cell system. In spite of the currently promising achievements and the credible perspectives of PEMFCs, there are many challenges remaining that need to be overcome before PEMFCs can successfully and economically substitute for the various traditional energy systems. During the last couple of decades or so, numerous efforts have been made to advance the PEM fuel cell technology and fundamental research. Factors such as safe storage of hydrogen, durability and cost still remain as the major barriers to fuel cell commercialization and so have been the major concern among scientists.

The objective of this review paper is: (1) to provide an overview of the technology of polymer electrolyte membrane fuel cells, their characteristics and (2) applications to real systems such as transportation, residential power generation and portable computers; (3) to outline the major commercialization challenges of these fuel cells and the needs for fundamental research for the near future.

***Index Terms*-- Proton exchange membrane (PEM) fuel cells, Polymer electrolyte membrane, applications, durability, cost.**

CONTENTS

1. INTRODUCTION

Proton exchange membrane fuel cells, also known as polymer electrolyte membrane (PEM) fuel cells, are a type of fuel cell being developed for transport applications, stationary application and portable fuel cell applications. By definition, it is an electrochemical apparatus which converts chemical energy stored in a fuel such as hydrogen, directly and economically to electrical energy by giving water as the only by-product. It has a potential to shrink our energy use, pollutant emissions and dependence on fossil fuels. Five kinds of fuel cells which have received major efforts in the current research: (1) polymer electrolyte membrane (PEM) fuel cells (PEMFCs), (2) solid oxide fuel cells (SOFCs), (3) alkaline fuel cells (AFCs), (4) phosphoric acid fuel cells (PAFCs) and (5) molten carbonate fuel cells (MCFCs). Among all these types of fuel cells, PEMFCs have the highest power density of 350 mW/cm^2 and hence have the maximum prospects. They have the potential to reach approx. 60% in electrical energy conversion with more than 90% reduction in major pollutants [1]. Table 1 shows the comparison of these various types of fuel cells relating their types of electrolyte used, operating temperatures, efficiency etc. A PEMFC is constructed using Nafion (Polymer electrolyte membrane) as a proton conductor and platinum (Pt)-base material used as a catalyst [1, 2]. Their notable features being low operating temperature, sustained operation at a high current density, low weight, compactness, potential for low cost and volume, long stack life and easy scale up. The practical efficiency of a PEM fuel cell is in the range of 40-60%. Activation losses, Ohmic losses and Mass transport losses are the main factors that create losses.

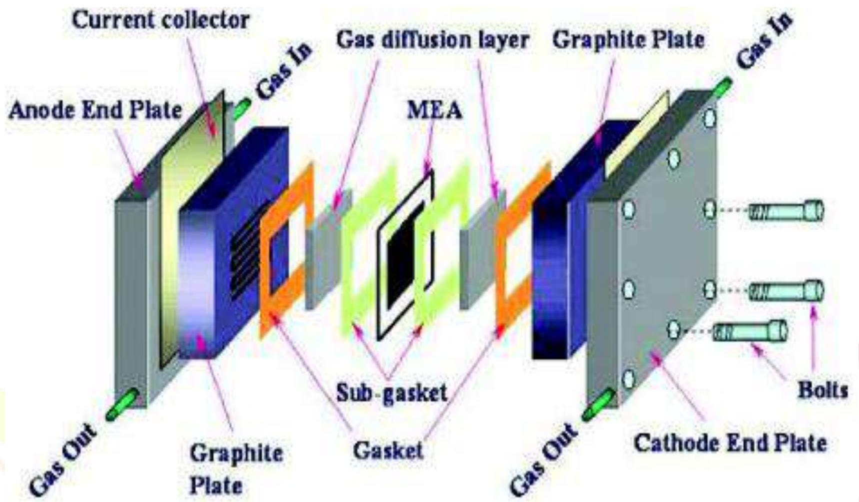

Fig. 1: Various components of a PEM fuel cell.

1.1. Reactions

In a fuel cell system, the chemical energy of the fuel directly changes into electricity, water and heat by carrying out certain electrochemical reactions using certain oxidants. Hydrogen, methanol and ethanol etc. have been usually used in fuel cells. Hydrogen injected as a stream is delivered to the anode side of the Membrane Electrode Assembly (MEA). At the anode, hydrogen splits catalytically into protons and electrons. These electrons move through external circuit to the cathode to conduct electricity while newly generated protons permeate through the polymer electrolyte membrane to the cathode side. The electrons from external load and protons permeated from membrane to the cathode side creating the current output of the fuel cell. Meanwhile, a stream of oxygen is delivered to the cathode side of MEA. At the cathode side, oxygen molecules react with the protons permeating through the PEM and the electrons arriving through the external circuit to form water molecules. These two reactions (i) oxidation half- cell reactions or hydrogen oxidation reaction (HOR) and (ii) reduction half-cell reaction or oxygen reduction reaction (ORR) [3] are represented by:

At the anode: $H_2 \rightarrow 2H^+ + 2e^-$ (1)

At the cathode: $1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O$ (2)

Overall reaction: $H_2 + 1/2O_2 \rightarrow H_2O$ (3)

The most important part of the fuel cell is the Membrane Electrode Assembly (MEA) having two main parts namely electro-catalyst and membrane. In this region, electrochemical reaction takes place. The main components of PEM fuel cells are shown in the Fig.1 above.

1.2. Proton exchange membrane (PEM) and Gas diffusion layer (GDL)

The proton exchange membrane is the heart of PEM fuel cell. It is a semipermeable membrane generally made from Ionomers and designed in such a way that it is permeable to protons while acting as an electronic insulator and reactant barrier, e.g. to oxygen and hydrogen gas. This is their main function when incorporated into a membrane electrode assembly (MEA) of a proton exchange membrane fuel cell i.e. separation of reactants and transport of protons while blocking a direct electronic pathway through the membrane [2].

The most widely used is a Perfluorosulfonic acid membrane. The Gas Diffusion Layer (GDL) is a central supporting material in a Membrane Electrode Assembly (MEA) that plays an important role of electronic connection between the bipolar plate with channel-land structure and the electrode. In addition, the GDL also performs the following essential functions: passage for reactant transport and heat/water removal, mechanical support to the membrane electrode assembly (MEA), and protection of the catalyst layer from corrosion or erosion caused by flows or other factors [2]. The table shows different type of fuel cells along with their features.

Table 1 [62]

FUELL CELL TYPE	COMMON ELECTROLYTE	OPERATING TEMPERATURE	TYPICAL STACK SIZE	EFFICIENCY	APPLICATIONS
POLYMER ELECTROLYTE MEMBRANE (PEM)	Perfluorosulfonic acid	50-100°C 122-212°F Typically 80°C	<1 kW-100 kW	60% transportation 35% stationary	• Backup power • Portable power • Distributed generation • Transportation • Specialty vehicles
ALKALINE (AFC)	Aqueous solution of potassium hydroxide soaked in a matrix	90-100°C 194-212°F	10-100 kW	60%	• Military • Space
PHOSPHORICN ACID (PAFC)	Phosphoric acid soaked in a matrix	150-200°C 302-392°F	400 kW 100 kW module	40%	• Distributed generation
MOLTEN CARBONATE (MCFC)	Solution of lithium, sodium and/or potassium carbonates, soaked in a matrix	600-700°C 1112-1292°F	300 kW-3 MW 300 kW module	45-50%	• Electric utility • Distributed generation
SOLID OXIDE (SOFC)	Yttrium stabilized zirconia	700-1000°C 1202-1832°F	1 kW-2 MW	60%	• Auxiliary power • Electric utility • Distributed generation

The Remaining portion of this review paper discusses the applications of PEM fuel cells in various fields, commercialization barriers and challenges in the field of PEMFC's.

2. APPLICATIONS OF PEMFCs

Proton exchange membrane fuel cells (PEMFCs) have their main applications in the area of transportation, residential power generation and portable computers. As a result of the increasing environmental hazards by emissions of petroleum based energy resources, whole scientific community is concerned to find their alternative and fuel cells due to their high efficiencies and low emissions proposed a strong candidature to become an alternative. Fuel cells and PEMFCs in particular have attracted the scientist ever since their discovery due to their light weight and eco-friendly power generation. Different sections have different power requirements, the power of electric passenger car, utility vehicles, and bus ranges from 20 kW to 250 kW. The stationary power by general fuel cells has a wide range of 1–50 MW, and for the remote telecommunication application it is 100–1 kW [4]. Fig 2 shows the proportion of fuel cells consumed in different categories in previous years.

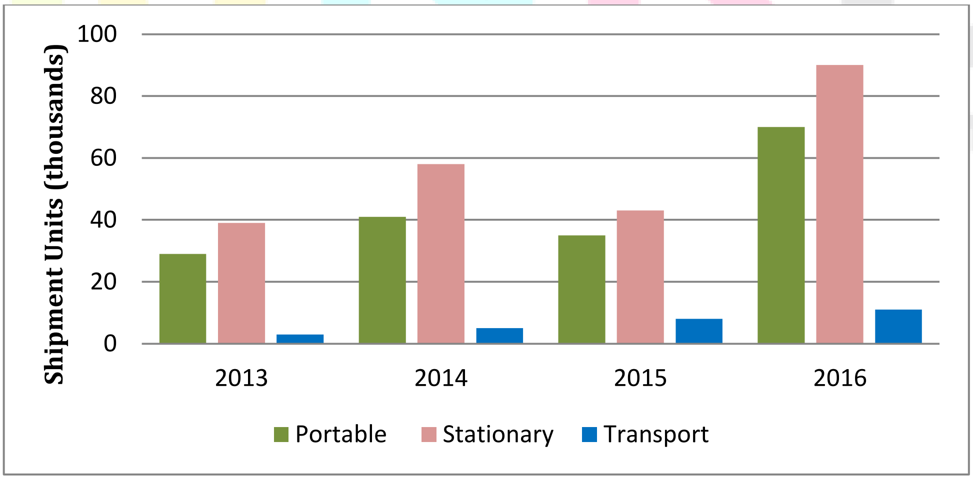

Fig. 2: Fuel cell systems shipped by sector: 2013-2016 [5, 6, 7]

2.1. Transportation applications

There is a growing concern over the environmental degradation, by conventional internal combustion engines (ICEs), around the globe; moreover constant depletion of petroleum fuel reserves has made the situation even worse. Therefore in transportation, there lies the most important application of PEMFCs. PEMFCs due to their fewer emissions and high efficiency have the potential to replace ICEs. As reported by McNicol et al. [8] fuel cell vehicle (FCV) systems are better than ICEs in every aspect except cost. The recent 2016 draft reports on '*Hydrogen energy and Fuel cells in India'* [9] recommends the allocation of Rs2765 crore till 2022 for the adoption of hydrogen as fuel in transportation sector. The national hydrogen energy roadmap (NHERM) has initiated several programs like The Green Initiative for Future Transport (GIFT) which aims to develop and demonstrate hydrogen powered ICEs and fuel cell based vehicles [10]. NHERM envisioned a goal of one million hydrogen fuelled vehicles on Indian roads by 2020 [10]. Ballard power systems have published a technology roadmap to provide future specifications of PEMFCs for FCVs [11]. This roadmap lays down that the durability; cost, freeze start and volumetric power density are the critical challenges for commercial adaptation of fuel cell vehicles.

2.1.1. Light weight vehicles

During the past two decades there is a constant rise and fall in the usage of fuel celled vehicles. The fuel cell light weight vehicles market in past few years is led by Honda & General Motors globally. General Motors have launched '*Project Driveway*' program in 2007 through which company delivered 100 units of Chevrolet Equinox FCVs to different states of USA which by 2009 had accumulated over 1,000,000 miles of driving [12]. Honda had also launched its sedan class FCV: FCX clarity even though the vehicle is only available to the costumers of southern California, that too on lease basis, this achievement presents a bright future of FCVs [13]. Other automobile makers have also launched/announced their FCVs such as Kia Borrego by Hyundai-Kia, B-class by Daimler, Passat Lingyu by Volkswagen and FCHV-adv by Toyota Fig. 3 [14]. 2-wheelers and 3-wheelers comprise the major part of the light weight vehicles in India. Therefore there is a need of hydrogen-based technologies particularly in this vehicle segment in Indian market. Domestic automobile maker Mahiondra & Mahindra (M&M) have developed a hydrogen based 3-wheeler 'HyAlpha' Fig. 4 [15]. Recently Bajaj auto and the Conversion Devices of the US jointly have also developed a hydrogen fuel-run auto Fig. 4 [16]. Also Swedish electric vehicle maker launches its Zbee 3-wheeler Fig. 4 in Indonesian market [17].


Toyota FCHV-adv


Daimler B-class


Hyundai Kia Borrego


Volkswagen passet Lingyu


Chevrolet Equinox


Honda FCX Clarity

Fig. 3: Fuel cell vehicles (FCVs) by various car manufacturers [18]

Fig. 4: 3-wheeler FCVs by M&M, Zbee and Bajaj respectively [19]

2.1.2. Heavy weight vehicles

Heavy weight vehicle such as trucks and buses comprise major proportion of automobile sector. Hence buses provide a promising future for the adoption of PEMFC technology; particularly in India where there is an enormous public transport sector comprising mainly buses. Tata Motors and Ashok Leyland, two biggest heavy weight vehicle makers in India, have started developing fuel cell based buses for public and private transport [20]. Tata Motors in collaboration with Indian Space Research Organization (ISRO) have developed a PEMFC powered bus named *Starbus* which was displayed at AutoExpo 2010 in Delhi [21]. Recently during the 2016 AutoExpo in Delhi Ashok Leyland presented their hybrid bus *Hybus* [22]. Several government initiatives around the world such as CUTE (Clean Urban Transport for Europe) and ECTOS (Ecological City Transport System) in Europe and STEP (Sustainable Transport Energy Project) program in Australia [23] have enlightened the future of powered buses and has encouraged other countries around the world to switch to eco-friendly transportation.

Fig. 5: Fuel cell powered buses by Ashok Leyland and Tata Motors respectively [24]

2.2. Stationary power generation

Stationary power generation sector is the most growing fuel cell market in India. Indian economy is growing day to day and to keep this growth on a constant pace, power shortage problem has to be tackle down. India is facing a huge power shortage which hinders the growth of economy. Adoption of PEMFC technology for power generation can tackle this shortage problem. Key markets for fuel cell technology in India, as per the study conducted by the TERI- Delhi, are telecommunication sector, chlor-alkali industry, luxury hotels, dairy industry and paper pulp industry [25]. The conventional centralized large-scale power plants have many benefits such as high efficiency, yet there are power loses due to long distance transmission which affects the efficiency. The way to undertake this drawback is a decentralized power generation which generates heat and energy for local usage. The back-up power generation is also a potential area for adoption of PEM fuel cells as power generating unit. Institutions such as banks, hospitals and telecom sector require uninterrupted power supply (UPS) and avoid unexpected power breakdowns and thus are potential customers of back-up power supply by PEMFCs. Recently RelianceJio has ordered fuel cells from Ballard power systems for their back up power deployment [26]. The Automatic Weather Stations (AWS) installed by ISRO at Shillong and Thiruvananthapuram Fig.6 uses fuel power systems for weather monitoring [27].

Fig.6: Fuel cell AWS installed by ISRO at Shillong [27]

2.3. Portable power generation

During the past few decades, there is a boom in market of portable electronic devices such as laptops and mobile phones. As the technology is growing day by day there is a high demand of power for use in portable devices. Conventional batteries used for this purpose doesn't produce satisfactory results due to their low power outputs and long charging durations. Micro PEMFCs can tackle down these problems efficiently due to their high power efficiency and short charging duration. Various fuel cell fabrication models have been proposed by technology developers worldwide. Hayase et al. proposed a model in which fuel cell and gas diffusion layer (GDL) are fabricated in Silicon wafers [28]. Other developers have also utilized different techniques to fabricate micro PEMFCs like Hahn et al. are using reactive ion etching (RIE) for production of micro channels in stainless steel plate [29], LIGA technology i.e. X-ray lithography is used to fabricate flow channels in metallic bipolar plates [30], Cha et al. uses lithography, physical vapor deposition (PVD) and focused ion beam (FIB) to formulate flow field plates [31] etc. Area of portable power in today's world is not only confined to laptops and mobile phones but also used in robot and power toys, boats, radio control cars and emergency lighting is also encouraged these days. In addition to this, fuel celled portable radios and other electronic devices are being used for military expeditions [32]. Recently launched fuel celled portable chargers, have gained worldwide appreciation. Fig.7 shows some portable devices running on fuel cell technology.

Lilliputian-fuel-cell-charger

Sony fuel cell speaker cum charger

Horizon fuel celled charger *myFC JAQ™ fuel celled charger*

Fig. 7: Some fuel celled electronic devices [33]

3. REMAINING CHALLENGES: COMMERCIALIZATION BARRIERS OF PEM FUEL CELLS

Irrespective of the worldwide considerable features of PEM fuel cells, there are some problems as well that need to be solved before full commercialization of these fuel cells. There are three main challenges that are common to each application viz. stable supply of high-purity hydrogen, cost reduction of the PEM fuel cell system and durability [34]. There are some other technological problems e.g. fuel cell components such as the MEA, suffer degradation during long-term operations [35]. Many problems of cost, durability or reliability, could be fixed with other material, catalyst or sealing. Thus, R & D of fuel cell commercialization has been guided to solve issues of materials, chemistry, water and hotspot [36] and many measures have been suggested in industries, such as related systems for water and heat management, high temperature PEMFC, and cheaper catalyst [37-41].

3.1. Cost reduction of PEM fuel cell system

The most critical challenge for PEM fuel cells today still remains in their cost. The cost of a typical PEM fuel cell is the makeup of the cost of the membranes, platinum electrodes, bipolar plates, peripherals and the assembly process. About 80% of the total cost is the cost of the bipolar plates and the electrodes including platinum [42]. A typical PEM fuel cell system costs USD 53/kW as per 2016 stats [43]. But competitively in order to sound more economical, fuel cell system must cost USD 30/kW [43]. Especially the platinum content in these fuel cells contributes more to the overall cost, so in one way or another, this platinum content should be reduced or totally removed. Various researches on platinum loading matter have focused on improving the electro-catalytic activity of platinum based catalysts. Another approach is to eliminate the use of Platinum entirely by developing a Non-platinum group metal cathode catalyst whose performance rivals that of platinum based technologies [44].

3.1.1. Reductions accomplished so far

Catalytic modifications have greatly helped in reducing the prices and thus approaching the cost target. Simplifications in the air humidification and coolant systems have gifted significant savings. For example, some years back water spray injection was used for air humidification, and now 2016 model does not have air humidification system at all [45]. Platinum loading of PEM fuel cells has been reduced by two orders of magnitude in the last decade [45].

Key accomplishments obtained from fuel cell technologies office FY 2016 budget data:

- Reduced the cost of automotive fuel cell systems to USD 55/kW in 2014, which is a reduction of more than 50% since 2006 and is well on the way to achieving the 2020 target of $30/kW.
- Reduced platinum content of fuel cells by more than doubling catalyst specific power from the 2008 baseline of 2.8 kW/g of platinum group metal (PGM) to 6.3 kW/g in 2014 that resulted in the reduction of PGM content by 80% since 2005.
- Reduced the capital cost of electrolyzer stacks by 80% since 2002, which will help to achieve a cost of less than USD 4/gge for renewable hydrogen by 2020.

A clustered column chart below displays the progress made in the reduction of cost of PEMFC System transportation in the previous years:

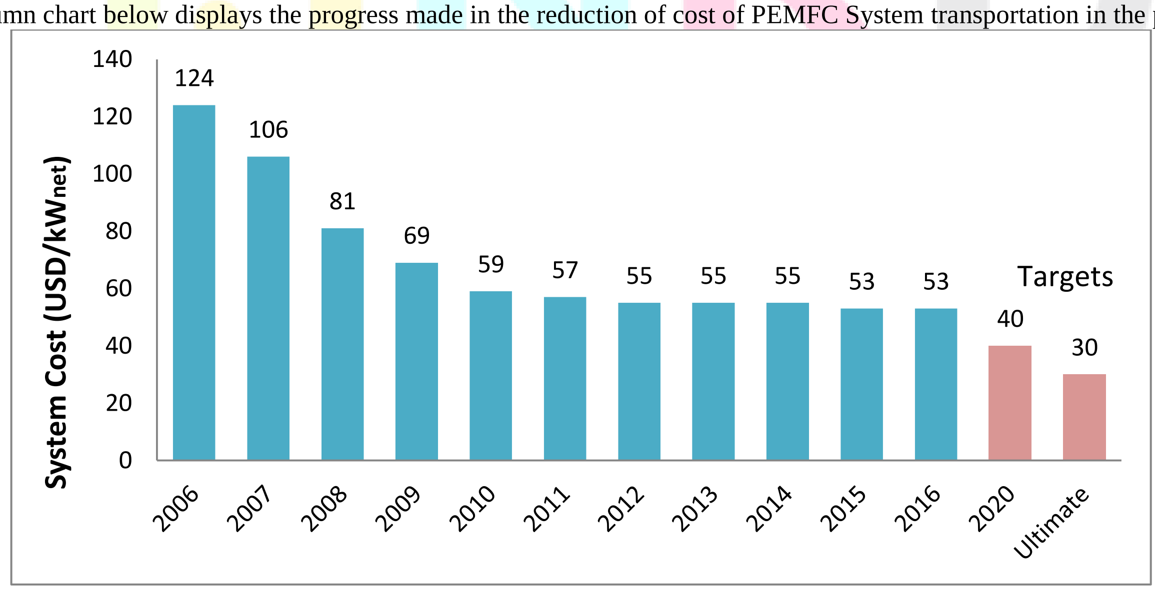

Fig.8: Modeled cost of an 80-kWnet PEM fuel cell system (500,000 units/year) [63]

Apart from the reductions attained so far, more reductions are required to make the technology economically possible for commercialization.

3.2. Stable supply of hydrogen with high purity

Traditional methods of hydrogen production like the steam reforming of natural gas or by coal gasification, causes the unavoidable CO_2 emission, which can lead to greenhouse effect [47]. In addition the production of CO can cause serious poisoning of the anode electro-catalysts in the fuel cell. Hydrogen can be stored as compressed gas or as a cooled liquid [47, 48]. Despite these methods, storing hydrogen to power a car requires a large tank. If these issues not overcame, there is probability that PEMFC's will lose the various application fields to other types of the fuel cell systems. Several studies have been carried out on high-purity hydrogen producing technologies including the water electrolysis, using the electricity from wind turbines and solar cells. For example, Lee et al [49] carried out a study of producing hydrogen using the excess electricity from wind turbines. The price of the hydrogen produced by this program was more than twice that produced by methane reforming [49]. So it can be said that further studies on areas such as the electrolyzer design and energy flow will be needed to develop a more efficient system. A compact on-site hydrogen generator produces very much pure hydrogen from methanol water mixture [50]. This system consists of a methanol steam reformer to get hydrogen rich reformed gas and a metal membrane purification part to recover high purity hydrogen from the reformed gas [50].

3.3. Durability

To commercialize PEM fuel cell technology, another important criterion is to achieve long lifetime of the overall system. Various factors are responsible for limiting the longevity of PEM fuel cells for example electro-catalysts and GDL carbon corrosion [51, 52]. Mechanical, thermal and chemical mechanism occurring over time, results in membrane degradation in fuel cell. The membrane degradation is mainly classified as electrochemical degradation and physical degradation [53, 54]. As for the chemical degradation, hydrogen peroxide and its decomposition intermediate products HO_ and HO2_ with strong oxidative characteristics generated during the fuel cell operation have been considered as one of the important factors resulting in the membrane degradation [55]. There are two different pathways for the H_2O_2 generation and the free radical species: (1) generating at the cathode due to the electrochemical two-electron reduction of oxygen (mechanisms 1-3) and (2) generating at the anode due to the chemical combination of crossover oxygen and hydrogen at the anode (mechanisms 4-7) [55].

$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2$ (4)

$H_2O_2 + M^2+ \rightarrow M^{3+} + HO^* + HO^-$ (5)

$HO^* + H_2O_2 \rightarrow H_2O + H0_2^*$ (Hydrogen peroxide radical attacks PEM) (6)

$H_2 + O_2 \rightarrow [0] + H$ (on Pt surface) (7)

$H_2 \rightarrow 2H^*$ (via Pt catalyst) (8)

$H^* + O_2$ (diffused through PEM to anode) $\rightarrow HO_2^*$ (9)

$HO_2^* + H^* \rightarrow H_2O_2$ (diffused into PEM) (10)

Effective strategies should be taken to stop such membrane degradation. The active approach is to suppress the free radicals attack, such as avoiding H_2O_2 formation, destroying H_2O_2 or killing the free radicals [56]. Trogadas and Ramani [57] prepared Pt/C/MnO_2 hybrid catalyst to minimize the effect of reactive oxygen species at fuel cell operation condition. However the hybrid catalyst can lessen the generation of hydrogen peroxide, the activity of the catalysts is poor at the same time [58]. So if the materials and technical levels can be satisfied, fuel cell durability will be improved.

3.4. Other technical issues

In addition to the previously discussed challenges, there are some other technical issues as well in a PEMFC system. They include water and thermal management, scale-up from single cells to cell stacks, fuel processing, CO poisoning of the platinum anode electro-catalysts and the over potential of cathode electro-catalysts [59]. Sometimes due to improper thermal management, comes various complications. Among them the electrolyte dehydration and cathode flooding impose the most critical challenges to PEMFC operation. Liquid water accumulation in the pores of the cathode electrode (including catalyst layer and GDL) causes the cathode flooding [60]. Also the delaying oxygen transport to the catalyst site is one of the most common issues in water and thermal management [61].

4. CONCLUSION

The excellent applications of PEM fuel cells have been look over. The major challenges to world-wide commercialization were explained viz. stable supply of high purity hydrogen, durability and cost reduction. Perfluorosulfonic acid membranes are the most widely used membranes but proton conductivity of these membranes is very small at high temperatures and they are very expensive. Therefore studies have been also focused on the investigation of alternative membranes. Ultimately, there is a serious need on fundamental research and associated challenges. Concentrating on materials development, cheap alternatives, attainment of fundamental knowledge and experimental tools are required. Hydrogen fuel research and development is also essential to reduce the cost of producing hydrogen from renewable resources and to economically advance hydrogen storage systems.

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