Compilation of Factors Affecting Durability of Proton Exchange Membrane Fuel Cell (PEMFC)

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Abstract

Nowadays, the proton exchange membrane fuel cells (PEMFCs) underwent a huge development. But durability remains one of the most important issues in commercialization of polymer exchange membrane fuel cell PEMFC. PEMFC have the potential to replace the vehicle's internal combustion engine. Specifically, with fast-start capability and low temperature operation, the polymer electrolyte membrane (PEM) fuel cell is the current focus for light-duty vehicles. The durability of fuel cell systems, however, has not been established. To be commercially successful, fuel cell power systems must be as durable and reliable as current automotive engines, a 5000 hour lifespan and functionality over a range of vehicle operating conditions (-40° to +40° C ambient temperature). So this review paper aims at assessing the factors affecting durability of Proton Exchange Membrane Fuel Cell (PEMFC).

Index Terms: *Component, Degradation, Durability, Lifetime, Membranes.*

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1. INTRODUCTION

Proton exchange membrane fuel cells (PEMFCs) are considered as promising alternative power sources for transportation due to the high energy efficiency and clean utilization. Within the last decades, much progress has been made for PEMFCs in materials, system design, manufacturing and application. Up to now, durability is thought to be one of the major problems hindering the commercialization of PEMFCs besides cost, hydrogen storage, etc. While the lifetime for stationary used PEMFCs has already exceeded 3 years, the lifetime for transportation used PEMFCs is still below 3000 h, which is nearly half of the 2010/2015 target lifetime 5000 h set by U.S. DOE [1]. It is known that PEMFCs will gradually degrade in performance due to many factors including operation,

water-heat management, and materials deterioration, etc [2]. Membrane electrode assemble (MEA) is the most important component of PEMFCs. The deterioration of MEA directly leads to the degradation of PEMFCs. One of the most important causes for MEA deterioration is the degradation of catalyst in the cathode side due to the high voltage, low pH and oxidation environment [3]. For the most popular Pt/C catalyst, it is reported that the nano-sized platinum supported on carbon black will gradually grow larger during long-term operation, thus reduce the electrochemical active area (ECA) and resulted in an irreversible performance loss [4].

This degradation will be even severer when PEMFCs are operated under dynamic loadings. Borup investigated the

degradation behavior of single cell under cycling dynamic loading and found that the growth extent of cathode catalyst particles was greater than that observed during steady-state testing [5]. The half cell electrochemical potential cycling test also revealed that higher upper potential and scan frequency lead to a higher degradation rate [5]. Darling and Meyer calculated the Pt dissolution under potential cycling and found platinum will dissolve rapidly when transitioning from low to high potentials [6]. The proposed degradation mechanisms of Pt/C catalysts include coarsening of Pt particles, Pt dissolution, carbon corrosion [7, 8] and loss of contact of ionomer and Pt [9]. The purpose of this paper is to review issues associated with the durability of materials employed in the fabrication of the stack and, more specifically, to review the critical issues related to PEMFC stack life that results in voltage degradation.

2. FACTORS AFFECTING DURABILITY OF PEMFC

We would like to start with a brief review of the degradation of PEM fuel cells because the membrane degradation is closely related to the degradation of other PEM fuel cell components, such as electrocatalysts, gas diffusion media, etc. Degradation behavior of PEM fuel cells frequently manifests it self as a gradual, unrecoverable performance loss followed by a sudden failure of one or multiple cells in a stack, as shown schematically in Fig. This is indicative of multiple parallel degradation and failure mechanisms occurring inside PEM fuel cell stack. For a particular fuel cell design and an intended application, it is critical to identify and focus on the life-limiting degradation and failure mode as the basis for durability projection and life prediction.

Borup and al. published a comprehensive review on various aspects of durability and degradation issues of PEM fuel cells. Table 1 summarizes commonly observed degradation symptoms and the related degradation mechanisms for MEAs in PEM fuel cells. The gradual performance loss is typically associated with the reduction of catalyst activity by platinum catalyst sintering, dissolution, poisoning of electrocatalysts by various impurities (gaseous and ionic) and carbon corrosion, etc. Ionic contaminants can enter the electrolyte membrane and exchange with the proton associated with acid group in the ionomer membrane, reducing its effective proton conductivity.
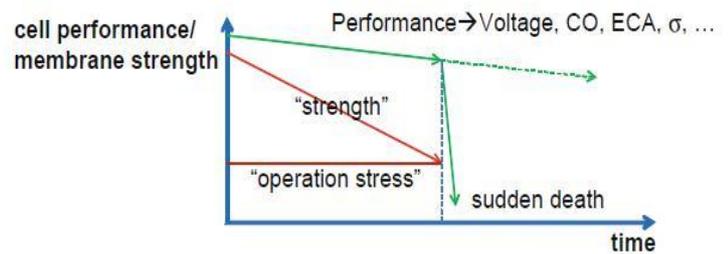
Fig-1: Competing degradation mechanisms in PEM fuel cells. (CO: hydrogen crossover rate; ECA: electrochemical area; σ: membrane ionic conductivity.)

TABLE I.   MEA Degradation and Failure Mechanisms

| Symptoms \ Mechanisms	Carbon corrosion (air-air start)	Gas impurities (e.g., CO, H2S, SO2 …)	Contaminants (e.g. some transition metal cations, anions)	Catalyst Instability (pt sintering, dissolution, re-rystallization)	GDL loss of wetproof (flooding)	Seal failure (gross leaking)	Membrane Failure (pinholing, and tear)
Activation loss (cathode Oxygen Reduction Reaction (ORR) & anode Hydrogen Oxidation Reation (HOR))	*	*	*	*			
Ohmic loss (ionic and electronic resistance)	*		*				
Mass transport loss	*				*		
Open circuit Voltage (OCV) drop (H2, O2 X-over)						*	*
Recoverable ?	No	Somewhat	Somewhat	No	No	Somewhat	No
Mitigation approach	Mitigation via. Potential control	air bleed, CO tolerant catalysts, …	avoid potential contaminants in conduits and in new alloy catalysts	new alloy catalysts, avoid OCV & potential-cycling	Surface modification	Alternative material, increase compression	avoid high T, dry & RH-cycling
Cross-cutting effects		oxygen into anode H2O2	membrane depolymerization via. Fenton reaction	Pt catalyzed membrane depolymerization	Flooding fuel or oxygen starvation	oxygen into anode H2O2	local eating via. Exothermal reactions

2.1 MEMBRANE DEGRADATION

The durability target for fuel cell components is closely related to the specific application, e.g. buses and passenger vehicles need to have a durability of 20 000 h and 5 000 h, respectively. In either case, the lifetime of the fuel cell is generally dependent on the lifetime of the membrane, as membrane failure not only implies a decrease in performance, but is equivalent to the end-of-life of the fuel cell. The three main degradation mechanisms affecting the membrane durability are: i) chemical degradation, ii) mechanical degradation and iii) thermal degradation Fig. 2, all three generally resulting in increased permeation rates and characterized by an increase in the hydrogen crossover.

Chemical degradation refers to the attack of the polymer chains and end-groups, e.g. carboxylic end-groups by hydroxyl (HOH) and peroxyl (HOOH) radicals [10]. These radicals are believed to be formed from hydrogen peroxide decomposition, which in turn is produced during the ORR [11] or during reactant gas crossover [12]. Chemical degradation leads to membrane thinning and can be evaluated in situ by measuring fluoride release from the fuel cell and increased gas crossover rates. Little work has been carried out to study the effect of high potential transients (>1.2 V) during Start-up and Shut-down (SU/SD) on membrane and ionomer durability. However, operating conditions such as extensive periods at OCV (~1 V) and low RH have been reported to have a large impact on the chemical degradation of the membrane [12]. Thus, it is highly probable that chemical degradation of the membrane is present during Start-ups and Shut-downs (SUs/SDs).

Mechanical degradation occurs when the membrane experiences creeping, cracking or formation of pinholes. Possible causes for this type of degradation are many, e.g. fabrication defects, improper MEA assembly by the seals, edges or by inhomogeneous compression of bi-polar plates (BPPs). Additionally, temperature and humidity cycling, differences in gas pressure between anode and cathode, as well as the spatial distribution along the flow channel of these conditions may also degrade the mechanical properties of the membrane [13]. These types of conditions, in particular temperature and humidity cycles, should definitely be expected to be present during SUs/SDs.

Thermal degradation occurs at high temperatures. Above 200 oC the loss of sulfonate groups begins to occur [14]. Consequently, during normal fuel cell operating temperatures (below 100 oC), thermal decomposition of membrane is often neglected. On the other hand, it has been proposed that membrane defects may allow the permeation of hydrogen to the cathode side, where it is likely to catalytically combust and produce local hotspots [13]. Yet another mechanism that may result in high enough temperatures to degrade the membrane may occur under transient conditions, e.g. when the fuel cell operates under gross fuel starvation [15] or under air starvation [16]. In the case of gross fuel starvation, it was shown that the high potentials generated at the anode resulted in local heat generation and holes in the membrane [15]. Therefore, it is reasonable to think that thermal degradation could be present during SUs/SDs operation as well.
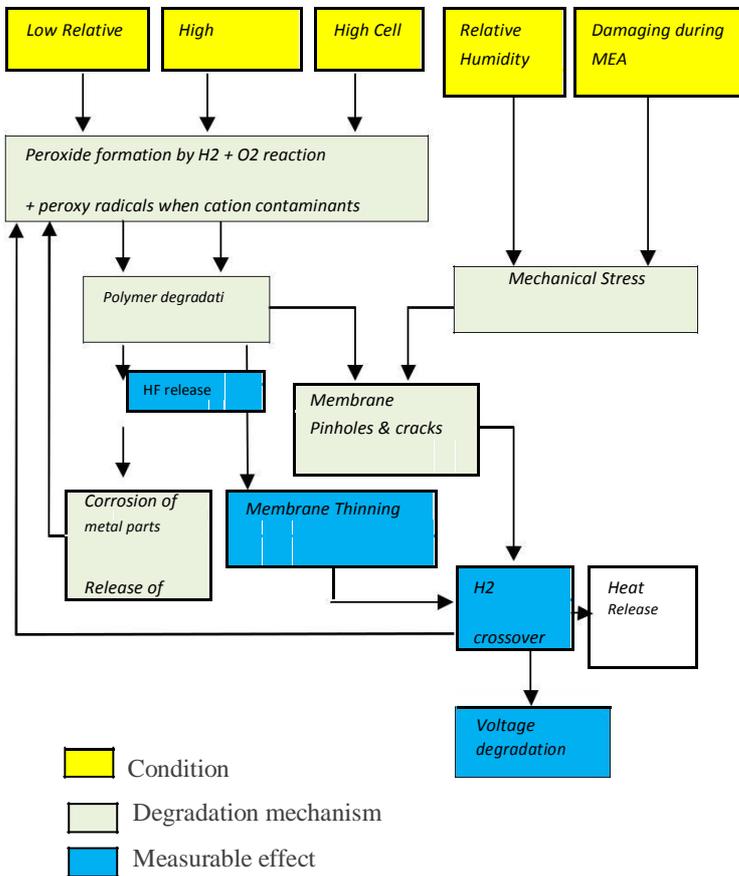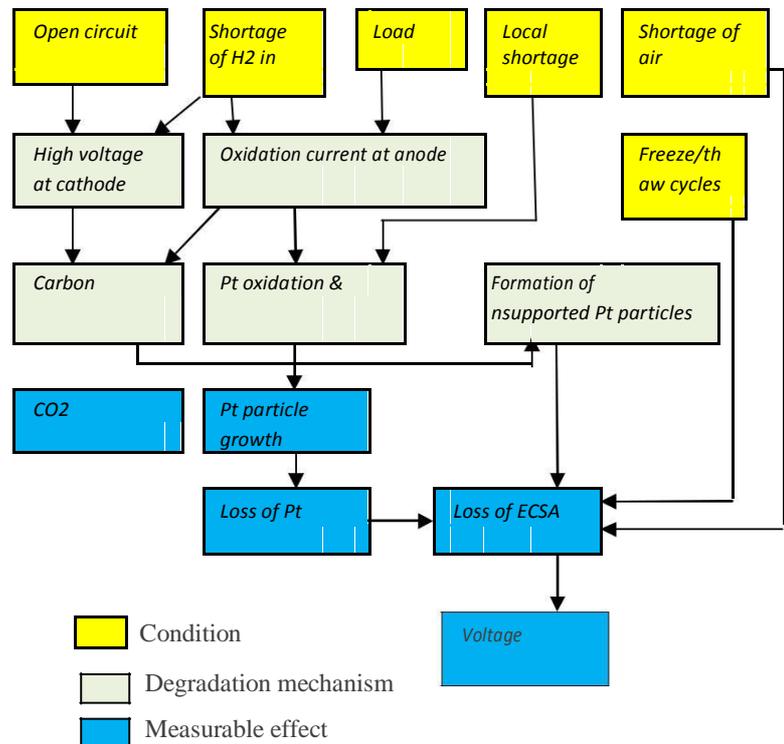
Fig- 2 : Factors affecting durability of membrane

2.2 IONOMER DEGRADATION

The ionomer in the catalyst layer is believed to be susceptible to many of the same degradation mechanisms as the membrane. Although pinhole formation and increased gas permeability are, for obvious reasons not of great concern, it is widely assumed that the dissolution of the ionomer in the catalyst layer occurs during long-term operation [17], causing loss of contact with the catalyst particles and decreased ionic conductivity. However, characterization of the ionomer degradation is very challenging due to difficulties in distinguishing the ionomer from the Pt/C phase using traditional morphology characterization methods, such as

scanning electron microscopy (SEM). Furthermore, measurements of fluoride release from effluent water do not discriminate between membrane and catalyst layer ionomer degradation. Zhang and al. [18] used X-ray photoelectron spectroscopy (XPS) to show that the catalyst layer ionomer dissolves and/or decomposes after 300 h of operation at high current densities. The degradation was characterized by a decreased concentration of CF3 and CF2 species. Wood III and al. [19] used neutron reflectometry (NR) to study the interaction between idealized layers of Nafion on either glassy carbon or Pt. They observed that the thickness of Nafion films increased with aging time, attributed to irreversible swelling. There are no studies on ionomer degradation related to fuel starvation conditions. However, Young [20] studied the performance of cathodes having different ionomer contents subjected to simulated SU/SD conditions. They found that the conductivity of the ionomer increased at low RH, but decreased at high RH after 30 h of degradation.

Fig-3 : Factors affecting durability of electrode

2.3 GAS DIFFUSION LAYERS (GDL) DEGRADATION

Although there is a vast amount of literature investigating the properties of the GDLs and how these properties may affect the performance of the fuel cell, degradation studies of GDLs are much fewer. Cindrella and al. [21] reviewed the key parameters affecting GDL performance, e.g. structure, porosity, hydrophobicity, hydrophilicity, gas permeability, transport properties, water management and surface morphology. Although GDL degradation studies are few, several degradation mechanisms that could influence the above-mentioned properties have been proposed. The loss of polytetrafluoroethylene (PTFE) is said to cause changes in the hydrophobic properties of the GDL. The modeling work of Seidenberge and al. [22] has shown that more and larger water clusters are formed in the GDL when the content of PTFE decreases over time. M. Schulze and al. [23] suggested that the degradation of the PTFE induced two times higher performance loss than Pt agglomeration after 1600 h of operation.

Oxidation or corrosion of the carbon in the GDL is another degradation mechanism of concern. Frisk and al. [24] used very aggressive ex-situ accelerated degradation tests (ADTs), i.e. using H2O2 to induce GDL oxidation. These were later used at the cathode of a PEMFC and had a large negative impact on the performance at high current densities. As the corrosion of the support during SUs/SDs became generally accepted, the corrosion of the micro porous layer (MPL) of the GDL also became an issue of concern. Owejan and al. [25] investigated the corrosion of the MPL using potentiostatic holds of 1.2 V and SU/SD simulation by air purging the anode. They suggest that carbon corrosion of the MPL at the GDL/electrode interface causes an increase of the mass-transport losses and that graphitized carbons in the MPL may be used to mitigate this type of degradation. Young and al. [26] simulated SUs/SDs while measuring carbon corrosion rates from the cathode exhaust. They found that the accumulative carbon loss was higher than the total carbon loading of the electrode, attributing the difference to corrosion of the MPL. On the other hand, others have found little or no degradation of the MPL upon high operating voltages of 1.5 V [27]. There are no reports on changes in MPL morphology or collapse of the MPL's porous structure and it is clearly an area of PEMFC research that needs more attention.

2.4 STAINLESS STEEL BI-POLAR PLATES (BPPs) DEGRADATION

There are two main issues concerning the durability of stainless steels as BPPs, these are: i) the corrosion of the stainless steels, releasing multivalent cations and ii) the increase in contact resistance between BPP and GDL due to surface oxides. Many research groups have reported the presence of metal ions either in the MEA or in the effluent water when using stainless steel BPPs [28-30]. It is suggested that metal ions diffuse into the membrane or catalyst layer ionomer, get trapped in the ion exchange sites decreasing the ionic conductivity of the ionic phase. It has also been suggested that metal ions, especially Fe ions, may react with peroxides formed during the ORR, producing hydroxyl radicals and increasing the chemical degradation of the membrane and ionomer [29]. Very little work has been carried out in the context of fuel starvation conditions and metallic BPPs. One possible reason for this is that the BPP is not expected to experience the same high potentials as the electrodes during these transients; since, the BPP is not in contact with the electrolyte. Yet, Eom and al. [31] have recently evaluated the durability of fuel cells using graphite, uncoated 316L and CrN-coated 316L BPPs under simulated SU/SD conditions at 1.4 V. As expected all three set-ups showed signs of carbon corrosion in the cathode catalyst layer. Interestingly, there were large differences in degradation of the fuel cell performance depending on the BPP material. Although the cell resistance was not measured in situ, the larger performance degradation exhibited by the fuel cell using uncoated 316 L was attributed to metal ion contamination of the membrane. This is also an area of PEMFC research that needs further attention.

2.5 CARBON CORROSION

The factors influencing the carbon corrosion are:

1) The structure of the carbon particle, conventional carbon blacks, e.g. Vulcan, have an amorphous core and a graphitic surface which could lead to different corrosion kinetics as function of time [32].

2) Carbon surface coverage by surface oxides, having the role of both intermediates and protective passive layer [33], also changing with time.

3) Changes in Pt oxide coverage or Pt surface in contact with the carbon; Pt has been shown to catalyze carbon corrosion [34].

4) The presence of water, which is directly involved in producing carbon surface oxides, as well as their further oxidation to CO_2 [35].

2.6. PT DEGRADATION

Voltage cycling clearly affects Pt dissolution to a higher extent than steady state operation. Unfortunately, voltage cycling is difficult to avoid in real applications, in particular in automotive applications. From a system point of view it is suggested that the loss of electrochemically active surface area (ECSA) caused by voltage cycling could be reduced by using the fuel cell stack in combination with a super capacitor or a battery. In such a configuration, the fuel cell could operate at more constant voltages, thus decreasing the impact of potential or load transients [36]. Another strategy for decreasing Pt degradation has been suggested by Mathias and al. [37]. They showed that the loss of ECSA could be minimized when the fuel cell was operated at lower RH and temperatures.

From a materials perspective, there is a vast amount of literature reporting alterative catalysts and combined catalyst/supports with enhanced catalyst durability compared to conventional Pt/C electrodes [38,39]. In general, Pt-alloys, e.g. PtCo, have been reported to show higher stability than Pt/C upon potential cycling [40,41]. However, dissolution of the alloy component has also been reported [42]. Antolini and al. [43] have reviewed Pt-alloy stability and concluded that the stability of the Pt-alloy catalyst may depend on the synthesis method, alloying degree and catalyst particle size. The particle size or grain size of the catalyst has a large effect on the degradation rate of the ECSA. Smaller catalyst particles are generally more unstable. This is one of the explanations given by Debe and al. [44] to explain the much improved stability of extended Pt-films, e.g. nanostructured thin films (NSTFs), compared to conventional Pt/C electrodes.

3. CONCLUSION

This study compiles available data from the literature on the factors that affecting the PEMFC durability. The causes of durability degradation in a PEMFC can be summarized as follows:

Degradation of the electrode/carbon paper material that can either change the mass transport properties, or release material that can contaminate the membrane

Contaminants from reactants (including dust)

Contaminants leached from fuel cell components

Degradation of electrode materials or polymer electrolyte Reactant shortage

Poor water management (flooding and dehydration) or simply the presence of liquid water

Loss of catalytic or electrolyte material

Catalyst migration or ripening; and uniformity of cell design and assembly

Temperature and humidity cycling

Differences in gas pressure between anode and cathode, as well as the spatial distribution along the flow channel .

These causes will ultimately lead to voltage decay in a PEMFC stack via the following failure modes:

Kinetic or activation loss in the anode or cathode catalyst

(due to physical or chemical changes in the catalyst); Loss of reformate tolerance of the catalyst;

Ohmic or resistive increases in the membrane or other components;

Delamination of the membrane from the electrode;

Loss of mass transport of the reactants in the flow channel Or electrode, or changes to the way water is transported;

Changes to electro osmotic drag properties or changes to the water diffusion characteristics of the membrane.

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