

# Morphological features (defects) in fuel cell membrane electrode assemblies

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## Abstract

Reliability and durability issues in fuel cells are becoming more important as the technology and the industry matures. Although research in this area has increased, systematic failure analysis, such as a failure modes and effects analysis (FMEA), are very limited in the literature. This paper presents a categorization scheme of causes, modes, and effects related to fuel cell degradation and failure, with particular focus on the role of component quality, that can be used in FMEAs for polymer electrolyte membrane (PEM) fuel cells. The work also identifies component defects imparted on catalyst-coated membranes (CCM) by manufacturing and proposes mechanisms by which they can influence overall degradation and reliability. Six major defects have been identified on fresh CCM materials, i.e., cracks, orientation, delamination, electrolyte clusters, platinum clusters, and thickness variations.

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## 1. Introduction

There are still many challenges to fuel cell commercialization; issues include the development of hydrogen infrastructure and storage technologies, and the durability, reliability and cost of the fuel cells themselves. Durability and reliability is of particular importance because these are main factors that will determine the ultimate uses of fuel cells. Currently, durability targets for polymer electrolyte membrane (PEM) fuel cells exceed 5000 h for automotive applications [1] and over 40 000 h for stationary applications [2]. The durability and reliability challenges encompass many factors not traditionally studied in published fuel cell research such as the initial quality and stability of fuel cell materials.

Research on membrane electrode assemblies (MEAs) often focuses on beginning-of-life (BOL) performance with little regard for end-of-life (EOL) operation. Recently, however, many longer-term degradation studies have appeared in the literature

[1,3–5]. These include examination of many different degradation modes that include radical attack [6–9], cell reversal and catalyst migration [1,10–13], and contamination [7,14,15]. Some modelling work has also been undertaken to predict the effects of ageing [16] or to explain the dynamics of degradation [17]. Nevertheless, systematic studies on both ex situ and in situ degradation modes have not been fully explored. Prior to attempting to understand degradation and reliability issues with fuel cell systems, it is necessary to categorize the different causes, mechanisms and effects. Included in this is the development of an understanding of the quality issues that face MEA manufacturing. In essence, a failure modes and effects analysis (FMEA) is required for a delivered manufactured component. Such analysis will allow successful de-coupling of causes and effects and will provide better data to improve fuel cell components.

The work reported here begins to categorise the different aspects that lead to degradation of fuel cell materials. Particular attention is paid to the factors that influence operational reliability. More detail and discussion of one of the factors, namely quality, is undertaken by examining the role of defects in catalyst-coated membranes (CCM) that are created during the manufacturing process.

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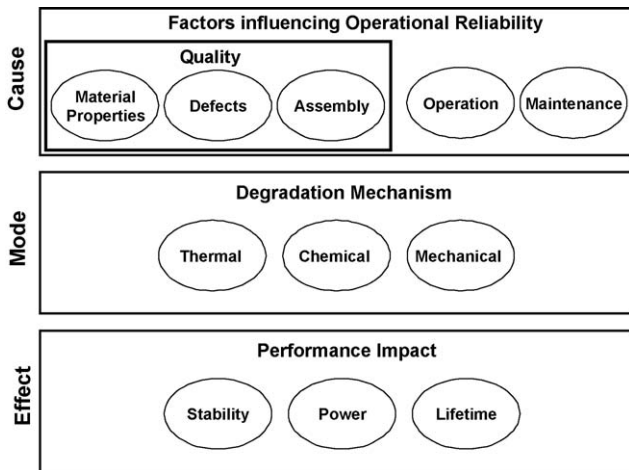


Fig. 1. Schematic representation of fuel cell degradation.

### 1.1. Degradation and Reliability Categorization

The reliability of any device can be broadly defined as the probability of the device to perform its purpose. When applied to fuel cells, the failure path can be divided into three segments, as shown in Fig. 1. The first segment is the cause and includes factors that influence the operational reliability of the fuel cell, e.g., the quality of fuel cell components, the operational conditions, and the maintenance procedures. Quality refers to the degree of conformance to set specifications and workmanship standards [18]. Quality is determined by the inherent properties of the materials used in fuel cell construction, defects in the materials, and the assembly process of the fuel cell. These factors determine the degradation and failure mechanism in the second stage, i.e., ‘the mode’. Finally, the effect of degradation that occurs in the fuel cell will have a certain performance and lifetime impacts, i.e., ‘the effect’.

The factors influencing product quality are variables that can either be controlled, introduced or are at least known to the user prior to fuel cell operation. These variables have been categorized into three different segments: (i) material properties; (ii) material defects; (iii) assembly. Within each of these categories are a number of different factors which, in combination with maintenance activities and operational conditions, lead to the degradation and ultimate failure of the fuel cell system.

To clarify some of the terminology used in this paper, the component properties encompass the physical properties of the different materials in the fuel cell system, as well as the physical properties including dimensions. The component properties partially govern the performance and dynamics of the performance of the fuel cell, e.g., the current density, the amount of water retained in the system, and the heat generated and rejected by the system. The component properties, which are dependent on factors such as the chemical structure, the manufacturing process, and the operating conditions of the cell, include: (i) electronic or ionic conductivity; (ii) mechanical strength, transition temperatures, flexural modulus, creep characteristics; (iii)

transport properties for mass, momentum, and heat (including flow path effects); (iv) compatibility with other materials; (v) composition; (vi) hydrophobicity; (vii) thickness; (viii) gas permeability; (ix) pore-size distribution; (x) flow path and coolant path design.

Although the influence of component defects on bulk component properties is included as part of the component properties, the defects also exert an impact on the quality of the fuel cell. Component defects are undesirable, inhomogeneous anomalies or features in the larger matrix of a component. On a scale consistent with their characteristic size the defects will tend to have different properties than the surrounding matrix. It is these non-uniform properties in the component that can give rise to an adverse effect on fuel cell lifetime. Whereas the origin and type of the features in the component are typically a function of the method of manufacture, features can also be introduced during the assembly process (as is the case with plate cracking). Component defects include, but are not limited to: (i) cracking in the catalyst layer; (ii) electrolyte inclusions and bubbles; (iii) delamination; (iv) scratches; (v) thickness variations; (vi) macroscopic orientation.

The third factor that contributes to the quality of the fuel cell is the method of assembly. Assembly includes the steps involved in building the cell and stack, as well as in integrating the overall system. The work reported here focuses on the assembly of the cell/stack since this is of importance to the lifetime of the fuel cell. The assembly operations (and material selection) for the balance-of-plant components have implications to the overall reliability of the system, but do fall within the scope of this study. Variables in the stack and system assembly operations are: (i) compression pressure; (ii) compression method (press, bolts, other); (iii) orientation; (iv) number of cells in a stack.

Once a fuel cell has been assembled, the only area in which the operator has some measure of control is the operation environment. Nevertheless, whereas this may be the case with single cells and stacks in research operations, it is not the case with end products that must face the un-predictability of the outside world. In the latter situation, there may be some understanding of how operational conditions will change in different applications. Since the operational environment exerts the greatest influence over the degradation mechanism in a fuel cell, it is of primary interest in this investigation. The important variables are: (i) current, temperature, humidity, stoichiometry; (ii) cyclic changes in the variables in (i); (iii) start–stop characteristics; (iv) shock and vibration; (v) feed gases; (vi) acidity.

Finally, maintenance includes any activity on the fuel cell after commissioning that does not include environmental changes. This can consist of: (i) re-builds; (ii) replacement parts (membranes, plates, etc.); (iii) re-sealing; (iv) cell removal; (v) balance-of-plant replacement.

The purpose of this work is to catalogue and document some of the manufacturing features and defects that impact the quality of the fuel cell at the CCM level. The study also proposes mechanisms by which the features and defects may influence the durability of a fuel cell.

## 2. Experimental

Samples of CCM were obtained from Ion Power Inc. and used Nafion 112 as the electrolyte and carbon-supported platinum as the catalyst. Scanning electron microscope (SEM) analysis was performed with a LEO SEM that was fitted with a field emission Gemini Column. X-ray compositional analysis was undertaken with an electron dispersive (EDS) collector manufactured by EDAX. The detection limit of the EDAX system is atoms with atomic weights equal to or larger than carbon.

Samples of membrane electrode assemblies and gas diffusion layers (GDLs) were cut into squares of approximately  $0.5\text{ cm} \times 0.5\text{ cm}$  and fixed to an aluminium stub with double-sided tape. It was possible to place two or three samples on each stub. The presence of conductive paths from the material of interest to the aluminium stub is important to prevent charging of the sample. In order to improve the conductivity of the samples, conductive tape was placed on a corner of most samples and attached to the aluminium stub.

Cross-sections were made by means of two different methods, namely, cutting and freeze fracture. Cutting involved taking a  $0.5\text{ cm} \times 1\text{ cm}$  sample and cutting it in to two  $0.5\text{ cm} \times 0.5\text{ cm}$  halves with a blade. One half was then mounted upright for study. The second method involved taking a strip of sample and submerging it in liquid nitrogen. Once frozen, the sample was broken in half while still submerged. At that point, one of the halves was chosen for mounting. For the work presented here, freeze fracture was the preferred method as it was shown to have minimal impact on the surface to be imaged.

When examining the resistance of the membrane to swelling effects, both deionized water and propylene glycol were used. The propylene glycol caused the membrane to swell more than with water and hence magnified the mechanical stresses associated with swelling.

## 3. Results and discussion

Several morphological anomalies have been catalogued with new CCMs, namely, cracks, orientation, delamination, electrolyte clusters, catalyst clusters and thickness variations. The following is a discussion of each of these manufacturing features and their potential impact on durability.

### 3.1. Cracking

A very common feature among all of the analyzed samples is the development of cracking on the surface of the catalyst layer. As shown in Fig. 2, cracking involves breaking of the catalyst layer without breaking of the electrolyte membrane. Thus, although the fuel cell can still operate in the presence of cracking, it is possible that performance or durability may be affected.

Cracking generally occurs over the entire catalyst layer surface. There are several possible causes for cracking, one of which the MEA manufacturing process. The catalyst layer is made by mixing catalyst powder with ionomer and an appropriate solvent. The resulting slurry is then applied to the electrolyte surface or

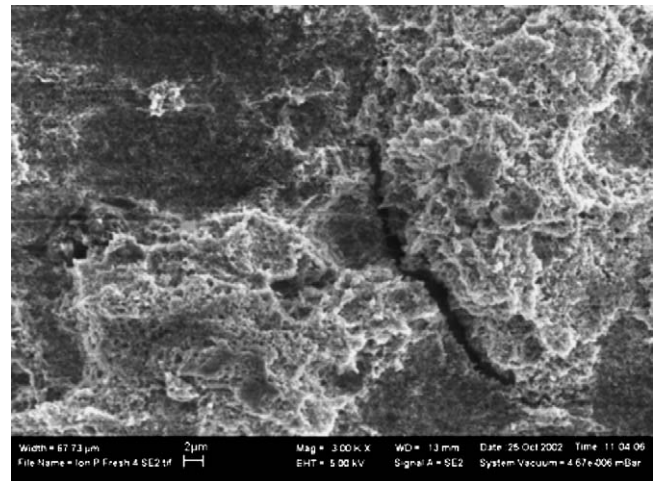


Fig. 2. Scanning electron micrograph of cracking in catalyst layer of Ion Power 522B.

made into a decal to be applied later. The layer is dried to remove solvent and simultaneously create the pore structure of the catalyst layer. The top of the layer dries first and then the solvent vapours from the lower regions break through the top, to cause cracking.

Another cause for cracking is simply through poor handling of the membrane and therefore which may also be related to the manufacturing method. Since the catalyst layer is less flexible than the electrolyte membrane, bending or stretching of the MEA can result in cracking. Bending can take place during preparation and assembly of the fuel cell. Also, as the electrolyte (in particular perfluorosulfonic acid membranes such as Nafion™) swells during hydrated fuel cell operation, the MEA will tend to wrinkle. Again, this may cause cracking.

Some possible impacts of cracking in the CCM layer on the performance of a fuel cell include the following.

#### 3.1.1. Defect propagation to a pinhole

A crack may represent an area at which there are local stresses. During the hydrated and heated operating conditions of the fuel cell, these areas may be prone to stretching and, hence, to pinhole/tear formation. Further, cracks may increase the resistance in the catalyst layer to electron and proton flow. Cracked areas will then have higher losses to heat that, in turn, may increase the risk of pinhole formation, which is a principal failure mode of a fuel-cell stack.

#### 3.1.2. Increased resistance of catalyst layer

As described, cracks break up the continuity of the catalyst layer. Thus, they will increase the total resistance of the catalyst layer. Also, if the cracked areas leave parts of the ionomer exposed, they will also increase the contact resistance between the catalyst layer and the electrolyte.

#### 3.1.3. Flooded areas

Cracks provide suitable areas to hold pools of water and thus can increase the water residency time in the MEA and

surrounding areas. Flooding in these areas will reduce the rate of reaction at the surrounding catalyst sites by preventing reactant gases from reaching catalyst sites. Flooding of the crack will also create a direct path for dissolved contaminants to react with the electrolyte ionomer.

#### 3.1.4. Areas for catalyst erosion

An important impact is that cracking exposes catalyst surfaces that are already weak. With the flow of water and gases at fuel cell temperatures, the gradual loss of material from erosion is a concern. The result of erosion is a gradual loss of catalyst activity over the service life of the catalyst layer.

#### 3.1.5. Areas with higher radical concentration

Radicals that may be produced at the edges of the crack and then move into the bulk of the crack (assuming that it is filled with water) will have unfettered access to the electrolyte membrane and therefore increased chemical degradation may occur at crack locations.

### 3.2. Orientation and roughness

Orientation of material gives features on the catalyst layer of the order of 10–100  $\mu\text{m}$  that are arranged or angled in a particular direction. The orientation in an MEA is illustrated in Fig. 3.

The most possible cause for orientation in the catalyst layer is from the processing equipment of the catalyst layer. Application of the catalyst slurry on to the electrolyte or decal may involve a spreading procedure such as tape casting as is the case with the MEA shown in Fig. 3. When spreading slurries in this manner, large agglomerates of un-dispersed catalyst powder can create drag marks on the surface. Other factors that will impact the degree of orientation on the surface are the speed and temperature of the caster. Imperfections on the knife edge, inhomogeneous slurry mixture, as well as build up of slurry behind the knife will also impact the surface characteristics.

Given the scale of the macroscopic variations, orientation may have very minimal impact. Further, in fabrication processes where GDLs are hot-pressed on to the catalyst layer, the pressing

step may flatten out the catalyst layer surface. The potential impacts of orientation may be as follows.

#### 3.2.1. Variable contact resistance

A very rough surface with randomly distributed peaks and valleys may have inconsistent contact with the electrolyte or, more likely, with the GDL. This can create increased contact resistance over the entire MEA area with localized areas of high and low conductivity (which can lead to localized heating).

#### 3.2.2. Mechanical stress variations

Upon compression, the variation in surface topography can cause variations in the pressure on the MEA. These, even small ones, can create a number of problems, such as parts of the catalyst layer being pushed into the electrolyte, bulging of the MEA into the channel, pitching of the electrolyte.

#### 3.2.3. Less control over morphology

Orientation in this case is indicative of a process where morphology is not completely controlled. Defects such as this undermine efforts to impart the desired morphology into the catalyst layer. This may adversely impact the porosity or proton conductivity.

### 3.3. Delamination

Delamination describes an MEA feature whereby the catalyst layer has separated from the polymer membrane electrolyte, as shown in Fig. 4.

It is unlikely that the freeze-fracturing process produced the observed delamination, since it was rarely observed in CCMs that were prepared in this way (and was much more prevented in aged samples). Therefore, possible causes may be through the lamination conditions used during manufacturing, such as catalyst casting speed, pressure, temperature, or even the amount of solvent used in the slurry. For example, higher temperatures during the catalyst drying stage may cause vapour to form at the interface of the ionomer and catalyst slurry. If the top of the catalyst layer dries too quickly, this vapour may become trapped

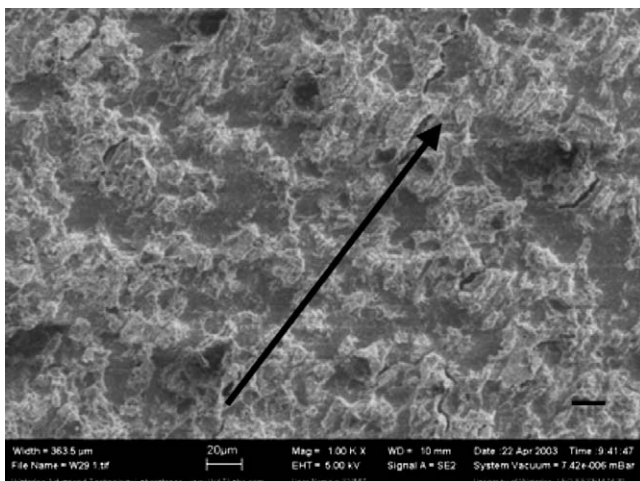


Fig. 3. Orientation of catalyst layer in CCM.

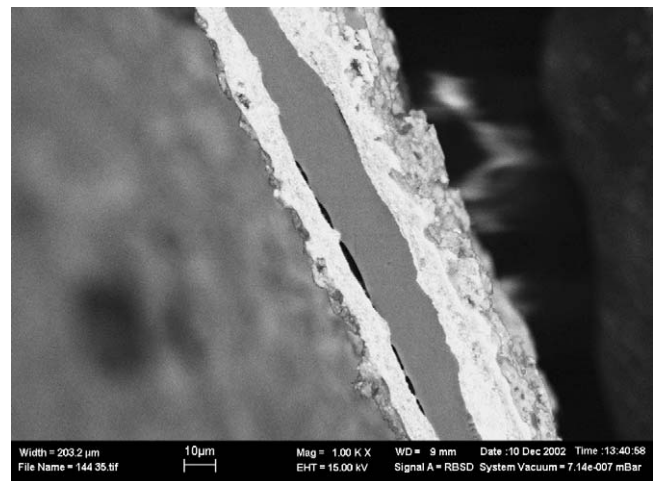


Fig. 4. Delamination between electrolyte and catalyst layer in a CCM.

and thereby create an area where the adhesion between the two layers is poor. This can then result in further delamination under the environmental stresses of the cell. Differences in thermal and hydrated expansion properties of the different materials may also be responsible for delamination over time.

Since the catalyst slurry contains electrolyte, it is generally assumed that there is a perfect melding between the catalyst layer and the electrolyte as the polymers are expected to bind together during the lamination process. This may not be the case, however, even if the temperature is above a glass transition temperature. Also, when using a decaling method to apply the catalyst, any wrinkles in the decal can result in the formation of voids between the layers.

Swelling studies with propylene glycol have also shown that layers within the CCM can be easily delaminated by exploiting the differences in swelling in each of the electrolyte and catalyst layers, as well as the manner by which they were laminated. It is also found that different CCMs manufactured via different methods delaminate to different degrees.

Some of the potential impacts of delamination on fuel cell performance are as follows.

### 3.3.1. Development of flooded areas

Pockets created by delamination can become filled with water and cause flooding of the pores in the surrounding catalyst area. This will increase the resistance to reactant transport to the catalyst sites.

### 3.3.2. Increased resistance in MEA

Separation of the catalyst layer from the electrolyte reduces the total contact area between the two layers. Hence, the contact resistance between the materials will increase, the protons will have a longer path to travel to catalyst sites, and the water barrier will be more resistive to proton conduction than pure electrolyte.

### 3.3.3. Development of pinhole areas

With more ionic current being redirected away from delaminated zones to neighbouring regions, there will be higher resistive heat generation in the latter. The increased heat can degrade the electrolyte and cause a pinhole. This mode may be further exacerbated if the void provides an area for hydroxyl radicals to concentrate.

### 3.3.4. Loss of apparent catalytic activity

Delamination can also lead to dead zones where no reaction takes place due to a lack of reactants.

### 3.3.5. Development of areas susceptible to erosion

When not attached to the electrolyte, the catalyst layer is very weak and hence may easily disintegrate. Parts of the catalyst layer that become delaminated from the electrolyte are at risk of flaking away.

## 3.4. Electrolyte clusters

The mixing process in the creation of the catalyst ink is very important for the dispersion of the catalyst powder with



Fig. 5. Scanning electron micrograph of Nafion<sup>TM</sup> cluster (highlighted circle); resolution  $\times 100,000$ .

the electrolyte. Another feature related to the mixing process is the presence of electrolyte clusters in the catalyst layer. An electrolyte cluster is essentially an area where there is significantly more electrolyte than in the surrounding areas, see Fig. 5. Electrolyte clusters are possibly formed if carbon agglomerates are not well dispersed during the mixing stage or if too much electrolyte is used in the ink.

The following impacts may be associated with electrolyte clusters.

### 3.4.1. Increased resistance

Excess electrolyte and less carbon black agglomerate creates a region with higher electrical resistance for electrons that penetrate and react at catalyst sites within the electrolyte cluster. This will lead to localized heating.

### 3.4.2. Lower active catalytic area

In areas occupied by the electrolyte clusters, there is little catalyst to contribute to reactions. The excess electrolyte presents a barrier to gases from reaching the catalyst sites. Further, carbon agglomerates will be completely covered in electrolyte so that the electrical conductivity will be reduced and the active area of the catalyst will be decreased.

## 3.5. Catalyst clusters

By using SEM in the backscattering mode, it is possible to identify areas that contain atoms with higher atomic weights than their surroundings. Atoms with higher atomic weights appear brighter than atoms with lower atomic weights. This technique has been used to determine the extent of catalyst clustering in the catalyst layer, see Fig. 6(a). X-ray microanalysis data for a cluster are presented in Table 1. Catalyst clusters can be formed through insufficient mixing or ball milling. It is also possible that the catalyst powder itself may have contributed to the presence of larger catalyst particles. At greater magnification (see Fig. 6(b)), the difference in morphology between the cluster and the surrounding area is evident. The cluster clearly lacks the 20-nm carbon spheres and necklace-like

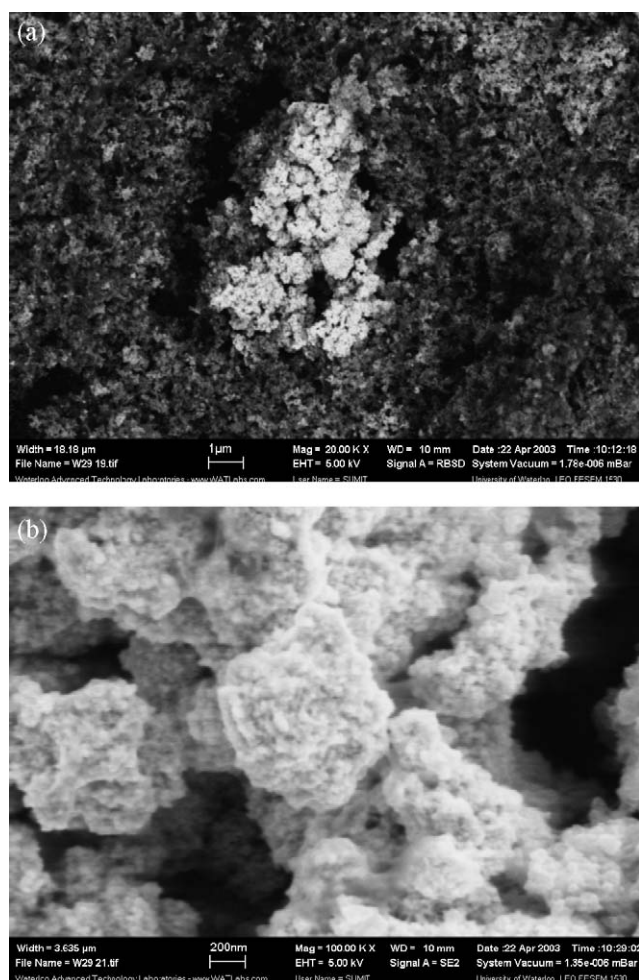


Fig. 6. Catalyst cluster in a CCM: (a)  $\times 20\,000$ ; (b)  $\times 100\,000$ .

agglomerate chains. In addition, the cluster appears to be less porous.

The impact of catalyst clusters on fuel cell performance and durability are as follows.

### 3.5.1. Reduced activity over entire active area

The dispersion of the catalyst on the carbon support is crucial to creating a high catalytic surface area. A catalyst cluster will obviously have a lower surface area than that of the same mass of catalyst well dispersed throughout the matrix. Further, as shown in SEM micrographs, the structure of the catalyst cluster is significantly different from that the surrounding cat-

alyst region. Qualitatively, the catalyst cluster has a very fine microstructure with very small pores. Thus, it can be expected that gasses will not be able to penetrate as deeply into the catalyst cluster, and that water can more easily flood these areas. This further emphasizes the reduced active area. As a result, the catalytic activity over the entire geometric area of the CCM would decrease.

### 3.5.2. Increased point activity

On a smaller scale, a catalyst cluster represents a region where there is a higher weight of catalyst and hence high catalytic activity. Though the overall catalytic activity is diminished by the loss of catalytic surface area, on a small scale the reaction rate in regions of high catalyst concentration will be faster than the surrounding lower concentration areas. Thus, catalyst clusters introduce the risk of hot-spots on the cathode side where the reaction is exothermic. The increased temperature at a hot-spot may melt the surrounding electrolyte and cause a pin hole.

### 3.6. Thickness Variations

The manufacturing method of the ionomer or catalyst decals can give rise to variation in the thickness of a CCM as seen in Fig. 7. There are many possible causes for such variation. For instance, if using a decaling method, the catalyst layer is first made on a film of PTFE or glass and thickness variations in the catalyst layer can occur if there are large clusters of agglomerates in the catalyst ink or if there are micro-variations in the membrane as a result of the casting or extrusion process. Thickness variations can also occur if the casting process creates flaws in the electrolyte.

The issues associated with thickness variations are as follows.

#### 3.6.1. Variable resistance through MEAs

Thicker areas of the catalyst layer will have higher electronic resistance, while thicker electrolyte segments will have

Table 1  
Microanalysis of catalyst layer of membrane electrode assemblies

Atom	Catalyst cluster		Homogeneous catalyst	
	(wt.%)	(at.%)	(wt.%)	(at.%)
C	19.01	76.89	61.07	92.50
O	0.58	1.76	1.28	1.45
F	0.70	1.76	2.94	2.81
Pt	79.51	19.59	34.71	3.24

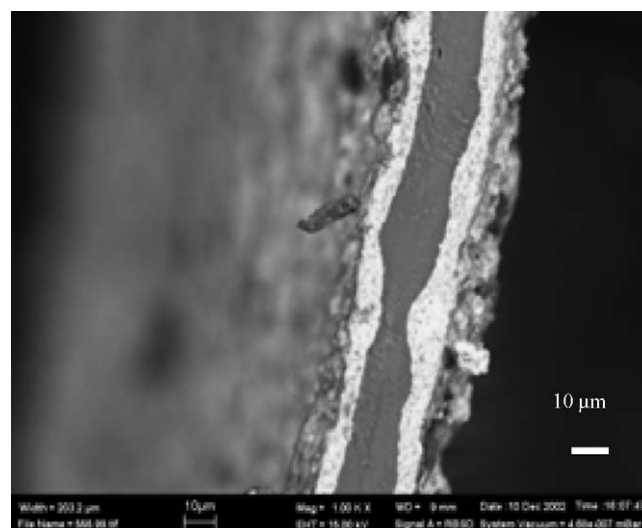


Fig. 7. Thickness variations in a CCM.

increased ionic resistance. For thinner areas, the opposite will be true.

### 3.6.2. Areas susceptible to pinhole formation

Thinner areas of the ionomer coupled with a thicker catalyst layer (which may promote more reaction and hence more heat generation) will be more susceptible to degradation by heat.

### 3.6.3. Mechanical weakness

Since the mechanical strength of the MEA comes partially from the electrolyte membrane, areas where the electrolyte is 'pinched' by the catalyst layer may be mechanically weak and thus may tear easily with tension. This may cause problems with pressure differentials and mechanical stress during thermal and hydration cycles.

## 4. Conclusions

In order to understand degradation in fuel cell materials, it is important to categorize and differentiate between failure modes and resulting effects. There are three main categories that can be used in failure modes and effects analysis. The first concerns quality factors (i.e., component properties, component defects, assembly), operational environment and maintenance that influence system reliability. The second category is the mechanism of the failure mode, which is completely determined by the selection of the variables in the first category. Finally, the mechanism of degradation leads to a performance impact or effect, which is the third category.

Controlling the manufacturing process of catalyst-coated membranes (CCM) is one way in which quality can be improved. The manufacturing process can impart features such as cracks, surface roughness and orientation, delamination, electrolyte and catalyst clusters, and thickness variations. In this work, a number of such features has been observed, and mechanisms have been proposed by which these features can lead to CCM degradation. The latter include increased radical attack, hot-spots, and low mechanical strength. These are very important issues when attempting to improve the durability and reliability of PEM fuel cells.

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