Fault Tree Analysis of Polymer Electrolyte Fuel Cells to predict degradation phenomenon

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Abstract:

Hydrogen Fuel Cells are an electro-chemical, zero-emission energy conversion and power generation device. Their only products are heat and electrical energy, and water vapour. One of the major hurdles to the uptake of this technology is the reliability of the fuel cell system.

This hurdle can be overcome through in depth reliability analysis including Failure Mode and Effect Analysis (FMEA) and Fault Tree analysis (FTA) amongst others. Research has found that the reliability research area regarding hydrogen fuel cells is still in its infancy, and needs development. This paper looks at the current state of the art in reliability analysis regarding Polymer Electrolyte Fuel Cells (PEMFC). A recent fault tree (FT) from the literature is qualitatively analysed to ascertain its practicality in relation to PEMFC degradation analysis.

The fault tree was found to harbour certain aspects that could be improved upon. There was no FMEA undertaken to precede the FT which would have given a greater understanding of the possible failure modes in a PEMFC system and their relationships. The FT was found to be lacking dependant relationships which are apparent in a PEMFC system. The data from the literature was also analysed to check its relevance in today's fast moving PEMFC research. Conclusions are given to the way forward for future reliability evaluation of PEMFCs.

1 Introduction

Climate change issues and sustainability concerns have increased in interest and awareness in recent years, since anthropogenic activities have been found to impact considerably upon the environment [1]. The way in which manmade activities contribute to climate change is mainly due to greenhouse gas (GHG) emissions. These include, among others, carbon dioxide (CO2), methane (CH4) and nitrous oxide (N2O) that contribute to the greenhouse effect. Additionally, energy prices are set to continue to rise by alarming rates[2] which will disrupt the UK's energy system due to a rise in oil prices.

The UK emitted 549.3 Million tonnes of Carbon Dioxide equivalent ($MtCO_2e$) in 2011[3] and 122.2 $MtCO_2e$ was due to the transport industry, with 74% of this figure due to cars, taxis and busses[4]. Due to the aforementioned negative environmental impacts of emissions from fossil fuel energy sources, this figure needs to be dramatically reduced not only to meet government targets, but for the health of the biosphere.

There are some technologies that can be used as alternatives to the fossil fuel dependent transport industry and alleviate our negative impact upon the environment. Battery electric vehicles (BEV)s have increased in popularity in recent times due to their potential to be zero emissions (when charged with renewable power sources). However they have not been very popular due to their small ranges and long recharging time requirements[5]. These negative attributes have affected their uptake with the general public customer base, and stinted their growth and commercialization.

Hydrogen fuel cells (HFC) negate the above issues as they are an electro-chemical, zero-emission energy conversion and power generation device. Their only exhaust emissions is water that is so pure, it was used by the Apollo astronauts as drinking water on the lunar missions[6]. They can be re-fuelled in a similar time to conventional Internal Combustion Engine (ICE) vehicles, and can operate to a similar range. These positive attributes have put the HFC in the limelight as an attractive alternative to the fossil fuel dependant ICE.

At present, degradation and lifetime analysis of Polymer Electrolyte Membrane Fuel Cell (PEMFC) is sparse and still undeveloped. Although overall reliability analysis of PEMFCs is lacking in development, component level degradation data is somewhat abundant in the Fuel Cell (FC) arena. There are many useful review papers[7] [8] [9] [10] that identify the possible failure modes of a PEMFC and links to the experiments that suggest an associated degradation rate for the component.

There are certain examples of research that have aimed at addressing the holistic modelling of a PEMFC system. Rama, et al. [11] constructed a qualitative Fault Tree (FT) of a PEMFC which was structured in such a way as to segregate the top failure events into the main loss pathways in a fuel cell; Activation losses, Mass Transportation losses, Ohmic losses, Fuel Efficiency losses and Catastrophic cell failure. This split the overall analysis into five separate trees to represent the ways in which a FC can fail. There is no quantitative data used in this model, thus the tree can be used to gain a greater understanding of the failure characteristics of a FC.

Placca and Kouta[12] constructed a quantitative FT of a PEMFC using aggregated data from the literature to be used to predict the lifetime of a PEMFC. They split the top event of 'degradation of the cell' into physical components associated with a PEMFC; Membrane, Gas Diffusion Layer and Catalyst Layer. After constructing the tree, they inputted failure and degradation data from numerous sources to predict the lifetime of a cell. A potential limitation of this method of data gathering, is that experimental results seldom use the same materials, operating conditions and parameters.

FT analysis (FTA) was most recently used in relation to PEMFC by Yousfi Steiner, et al.[13] Previously, the authors had looked into using FTA for the water management issues related to a PEMFC[14], however the recent work took a more systemic level approach. The authors used FTA to model a FC stack and its auxiliary components such as air blowers and piping. However the stack tree was somewhat basic and did not split the FC down into basic events such as in the trees of Rama and Placca & Kouta.

Even though the latest work was more systemic, it overlooked the basic events that could cause a reduction in stack voltage output which is an area that needs to be developed.

Wieland, et al. [15] used Petri-net modelling techniques to try to accurately predict the lifetime of a PEMFC stack or fleet of cars, incorporating Monte Carlo simulation techniques. The model can take into account reversible events, spontaneous events and repairable items. As with the aforementioned FT work, failure and degradation data was taken from the literature to input into the model. The model presented can be quickly and easily adapted to new situations during operation, where new transitions can be freely added. However the authors state in their concluding remarks that a lot of simplifications had to be undertaken to achieve this model. A step forward would be to address the simplifications and to map operating time realistically, which would allow for an availability analysis to be undertaken.

The current level of research relating to reliability analysis and lifetime prediction is underdeveloped and requires advancement. FTA is known for being most suited to simple systems with no dependencies between failure modes and interlinked relationships. Petri-Net analysis can take dependencies into account, however it is a lot more complex than FTA and involves a lot more computing time in comparison. Even though there is a multitude of FC degradation experimental data available, it is seldom uniform and directly comparable to other data sets.

This paper looks into the most recently proposed FTA research and analyses the techniques used and methods adhered to, to achieve the said research. The goal of this paper is to understand how to advance the PEMFC durability and degradation research area through in-depth analysis of the current research available.

2. Hydrogen Fuel Cell Overview

There are five main types of HFC that have been developed over the years, these are;

- Polymer Electrolyte Membrane Fuel Cell (PEMFC)
- Alkaline Fuel Cell (AFC)
- Phosphoric Acid Fuel Cell (PAFC)
- Molten Carbonate Fuel Cell (MCFC)
- Solid Oxide Fuel Cell (SOFC).

The main way in which they are segregated is by their constituent materials for the electrolyte based upon the operating temperature. PEMFC, AFC and PAFC have relatively low operating temperatures (<200^{o}C), and can thus utilise aqueous electrolytes, whereas MCFC and SOFC operate at temperatures from 600^{o}C and 1000^{o}C respectively, and thus cannot use aqueous electrolytes due to vapour pressure.[16]

Out of the many classifications of hydrogen fuel cell, the PEMFC is commonly singled out as the most appropriate to be implemented into an automotive application. This is due to its relatively low operating temperature of around 50-80^{o}C, its ability to use air as the cathode reactant and its rapid start-up time. The PEMFC will therefore be the focus of this paper.

2.1 Operation of a PEMFC

A fuel cell is an electro-chemical energy generation device that directly uses H_2 and O_2 to create electrical and heat energy, with the only by-product of the reaction being water. The overall reaction that takes place in a PEMFC is shown in **Error! Reference source not found.**:

$$H_{2(gas)} + \frac{1}{2}O_{2(gas)} \rightarrow H_2O_{(liquid)} + \text{heat} + \text{electricity} \qquad (1)$$

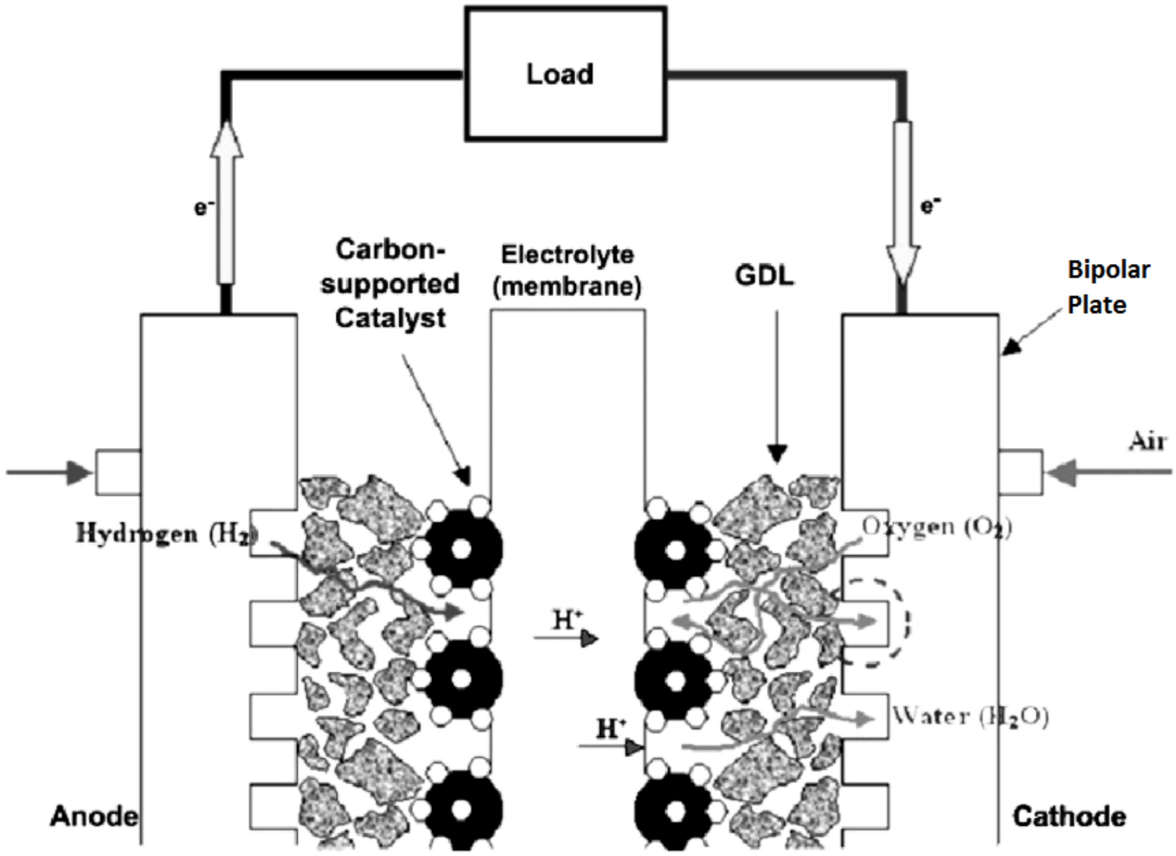

Figure 1 - PEMFC components, adapted from Kandlikar & Lu, 2009.[17]

A PEMFC is made up of four main components;

- Polymer Electrolyte Membrane
- Catalyst layer
- Gas Diffusion Layer (GDL)
- Biploar Plate

As can be seen in Figure 1, the catalyst layer, GDL layer and bipolar plates are repeated either side of the central membrane creating a ‘sandwich’ called a cell. The cells are then layered to create a ‘stack’ which forms the power plant in a PEMFC vehicle. Hydrogen gas is supplied to the anode side of the cell, and Oxygen is supplied to the cathode either in pure form, or as part of ambient air intake.

The voltage of a PEMFC can be expressed as in **Error! Reference source not found.**

$$V_{cell} = E_{nernst} + \eta_{act,a} + \eta_{act,c} + \eta_{ohmic} + \eta_{concentration} \quad (2)$$

Where $E_{nernst}$is the open circuit voltage potential of the fuel cell, $\eta_{act,a}$and $\eta_{act,c}$are the activation losses at respective electrodes, $\eta_{ohmic}$is the loss due to electrode, connections and polymer proton resistance, and $\eta_{concentration}$relates to the losses due to concentration of fuel.

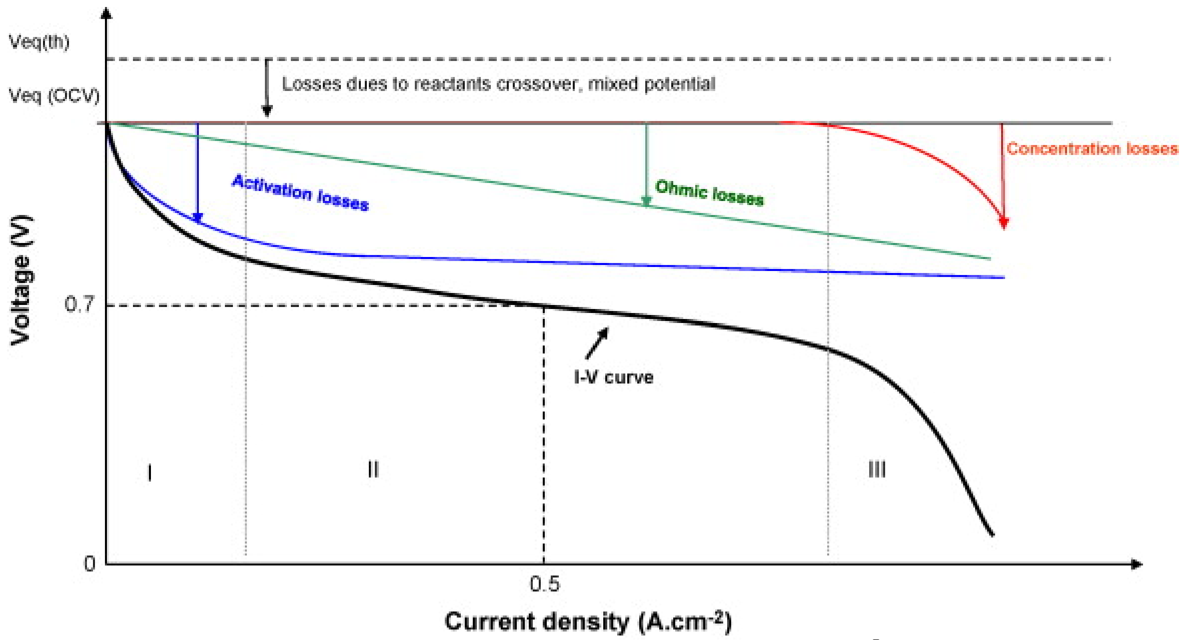

Figure 2 - Standard polarisation curve[8]

Figure 2 presents the standard polarisation curve which is used throughout FC science. It indicates the evolution of the PEMFC voltage in relation to the applied current, and the losses associated with certain fuel cell phenomenon. V_{eq}(th) shown by the dotted line at the top of the graph is associated with the fuel cell's theoretical maximum potential or Open Circuit Voltage (OCV), this is commonly known as 1.229V.

2.2 PEMFC Degradation

Currently there are three main hurdles to the commercialisation of PEM fuel cells and their competition with the ICE, these are; infrastructure, cost and durability[9]. PEMFC durability issues can be mitigated against through reliability analysis techniques. Degradation of a PEMFC is therefore a prominent area for extensive research to aid in the goal of commercialising the PEMFC car.

Degradation in a PEMFC is measureable via a reduction in output voltage.[10] Current lifetime goals accepted by the fuel cell community state that a fuel cell in an automotive application must operate for 5000 hours with a reduction in output voltage of no more than 5% over the 5000 hours period. This gives a solid indicator and timeframe for the measurement of undesirable degradation in a PEMFC.

3 PEMFC Fault Tree Analysis

3.1 Current state of analysis

Fault trees have been used by some researchers to try to gain a greater understanding of the failure modes of PEMFC's. The following sections look at a recent paper regarding a FT analysis of a PEMFC, commenting on possible improvements and future work to develop this field further. The following work was chosen to be analysed due to the fact that it is the most up-to-date FT of a PEMFC. Previous attempts had been developed qualitatively[11] which have been built upon since, and as such, the latest work is chosen to be evaluated and developed further.

Placca and Kouta[12] recently used FT analysis to investigate PEM fuel cell degradation. They constructed a FT of a single cell PEMFC with the overall concerning factor being the top event of ‘Degradation of the cell’ to an extent that was detrimental to the functioning of the PEMFC system. They used reliability data compiled from many sources within the literature, and extrapolated the data to acquire a formatted degradation measure. From various literary sources, they came to the conclusion that there are 37 individual basic events to be considered when analysing the degradation of a PEMFC. The FT presented is a ‘physical’ analysis of a single cell PEMFC, splitting the top-event of the ‘Degradation of the Cell’ down through an OR gate into three physical components of a PEMFC:

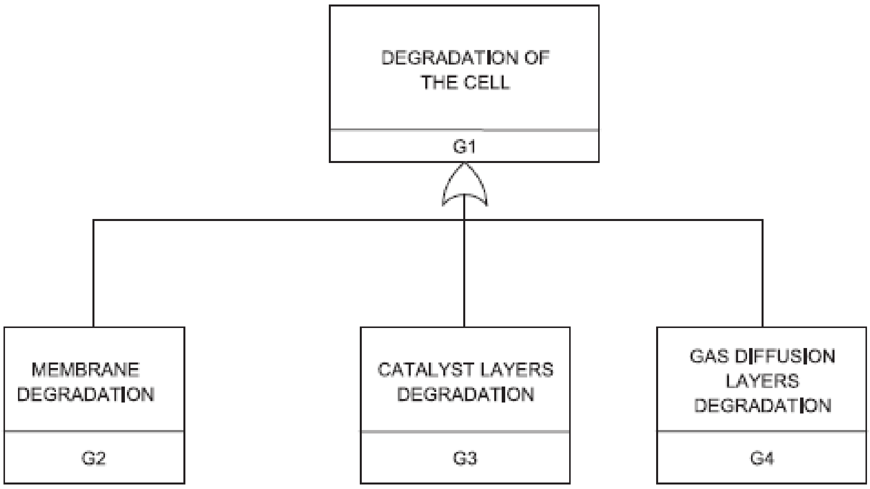

Figure 3 - 'Global' Fault Tree presented by Placca and Kouta.

These are three of the four main physical components of a PEMFC with only the bipolar plate being omitted. G2, G3 and G4 each had 12, 12 and 6 intermediate events respectively, which further branched down through OR gates to the basic events. As all of the gates in the presented FT were of the OR variety, the minimum cut sets are simply all order one’s, representing the basic events of the tree.

3.2 Analysis review – Limitations Determined

3.2.1 Top Event: The top event for the tree developed by Placca and Kouta can be interpreted as vague; ‘Degradation of the cell’ does not directly inform the reader of what ‘degradation’ is classed as, and what drop in voltage output is considered to be degraded. A limitation with using fault tree analysis for this type of application is the fact that the events in the tree need to be binary in nature. A failure or a success of a component or process is an ideal scenario for using FT analysis, however the top event of ‘Degradation of a cell’ does not fit these criteria. A more prudent way of defining the top event would be to suggest a rate of degradation over a time period that is unacceptable.

3.2.2 Bipolar Plate Omission: The omission of the bipolar plate component of a PEMFC is an issue that needs to be addressed. There are many studies in the literature that document and analyse the degradation and failures of bipolar plate materials in PEMFC. Failure modes affecting the bipolar plates include, but are not limited to; corrosion of the metal bipolar plate when in contact with the aqueous and acidic environment of the PEMFC, mechanical fatigue caused by repeated thermal cycles and silicone sealant used as a gasket on the bipolar plate can degrade and enter the membrane.

Corrosion and mechanical failure have been documented occurrences in a number of studies.[18,19,20] Y.C. Chen et al. showed in a 2012 study[21] that cell performance can be dramatically reduced through bipolar plate corrosion and the formation of passive oxides creating an oxide film. The corrosion of the plate material and the formation of the oxide film reduce the electrical conductivity of the plate. They showed that the film gradually increases in thickness with age, and as such the resistance increases with the thickness of the film. This phenomenon is only present on the cathode side where O_2 is the fuel and hence having the opportunity to form the oxide layer. At the anode side, H_2 is the fuel, and thus this issue is not apparent.

The bipolar plate can also affect other parts of the PEMFC, for example steel bipolar plates can release Fe^+ and Cu^+ ions that can have a detrimental affect elsewhere. The presence of foreign cations is included in the FT. For example in an intermediate event in the membrane branch named ‘Contamination by trace metal ion – G10’.

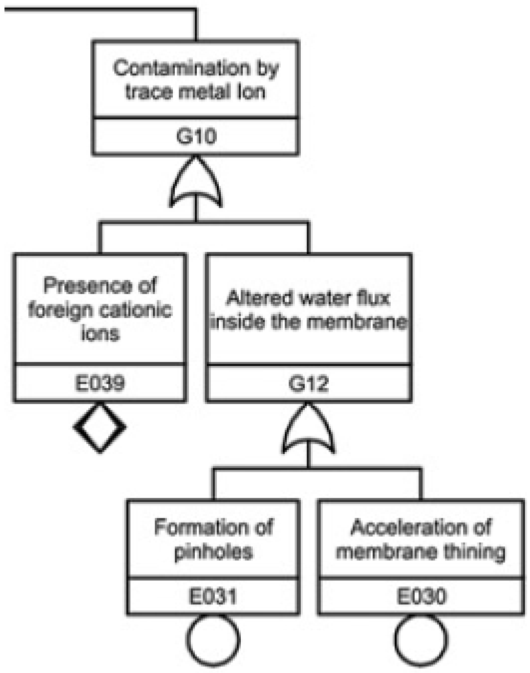

Figure 4 - Metal Ion Intermediate Event

As can be seen in Figure 4, G10 is split into the presence of foreign cations and altered water flux. The FT presented puts accelerated membrane thinning and pinholes as the cause of altered water flux, whereas review papers[7,9] suggest that the presence of the foreign cations displace the H+ ions in the membrane, and would lead to membrane thinning and possibly pinhole formation. This indicates that the logic of this section of the membrane branch needs re-evaluating.

As mentioned earlier, foreign cations can stem from the bipolar plate material as well as inlet piping or humidifier materials. This fact, alongside the other bipolar plate corrosion issues mentioned previously, would indicate that the overall structure of the tree would need further evaluation.

3.2.3 Relationships between failure modes: One of the main limitations with current fault tree analysis of PEM fuel cells is the lack of forged links between different failure modes, where upon there exists key relationships between certain failure modes in a PEM fuel cell system.

In order to understand the links between the different basic events in the current FT, the entirety of the basic events contained in the intermediate events branches were plotted out with any potential links and relationships highlighted, the relationships are shown in Figure 5 for G2, ‘membrane degradation’.

In the figure, if one event can cause another, it is linked by a solid arrow. Additionally if one event can increase the impact or occurrence of another event, it is linked by a dotted and dashed arrow. A dashed line indicates where ‘basic events’ are the same, and as such shouldn’t be classed as different basic events. A dashed box indicates how a basic event in another branch can affect the membrane’s degradation rates.

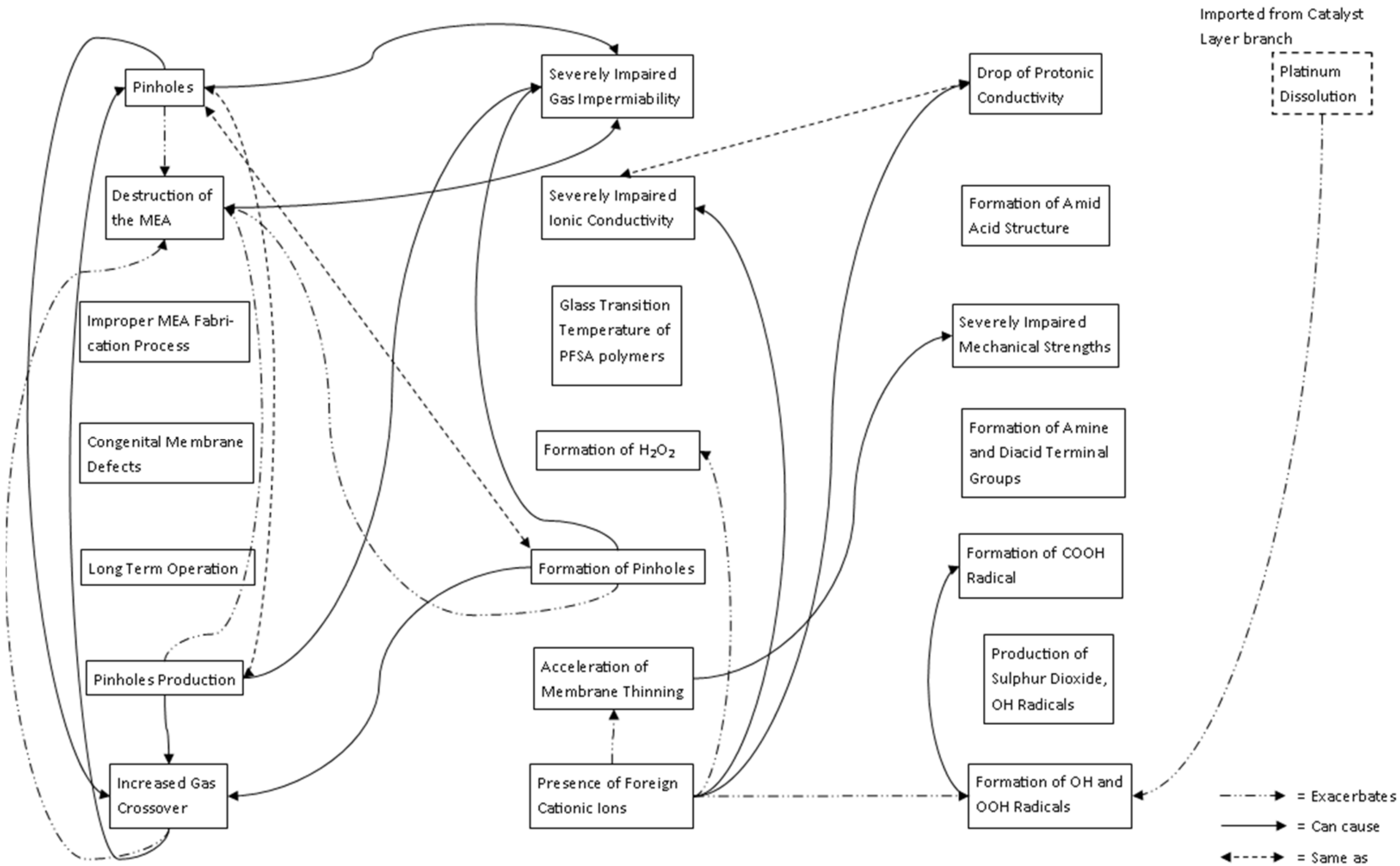

Figure 5 - Membrane Basic Event Links

Figure 6 shows how basic events in the membrane branch of a PEMFC FT are intrinsically linked. Some events can lead on to other events occurring, additionally some events can exacerbate other issues. To discuss this further, under the membrane degradation branch of the tree, G2 in Figure 3, gas crossover has been listed as a basic event ('Increasing gas crossover' – E020). H_2O_2 formation is listed as a basic event under the peroxide/radical degradation and is placed under the chemical degradation branch. In the 'Electrocatalysts and catalyst layers degradation' branch, a basic event of 'Platinum dissolution' highlights the issue of Pt nanoparticles separating from the CL and migrating to other areas of the cell. Bruijn, et al.[7] suggest that radicals are formed at either; the cathode through H_2O_2 which is formed as part of the oxygen reduction reaction, or through the decomposition of H_2O_2 at the anode through the crossover of O_2 from the cathode to anode. Additionally, they state that recent work shows how radicals can be formed through a more direct pathway as opposed to the H_2O_2 intermediary pathway, in the presence of Pt. This is where favourable conditions for degradation can be provided by a reaction between molecular H_2 and O_2 in the presence of Pt particles that have separated from the CL through electrode degradation. This shows that gas crossover, H_2O_2 formation and platinum dissolution are interlinked and therefore should not be listed as segregated basic events. Gas crossover creates H_2O_2, which causes radical introduction, and Pt nanoparticles from dissolution create radicals that attack the membrane. The radical attack causes thinning of membrane which can create pinholes and increased gas crossover. Operating conditions have been proven to exacerbate the above relationships, namely; high temperature, low humidification and high gas pressure.

3.2.4 Ambiguity of Intermediate Events: As with the top event, many of the intermediate events are not as well defined. The events in G1 – G4 are all degradation events and as such can be a small, large or complete drop of voltage. It is not clear what level of degradation is being modelled. For a more reliable FT of a single cell PEMFC, one would need to amend the current G1 – G4 events to intermediate events with binary outcomes such as 'G1 – Failure of PEMFC' and 'G3 – Failure of catalyst layer'. Degradation levels could be specified to make use of the existing failure modes, such as '60% loss of electrochemically active surface area' for the catalyst layer (G3). Basic events leading to this could include, but are not limited to: CO contamination taking up catalytic active sites, or Pt agglomeration reducing surface area of Pt catalyst for the redox reaction.

As with the previous issues found with the top events and first intermediate events, the majority of the basic events are also equivocal and are not necessarily binary in nature. E002 'long-term functioning' is ambiguous in description, not informing the reader whether it means that the cell is completely failed or is degraded to a lower output state due to long-term operation. There is no explanation of what this pertains to, such as a time frame, or what the failure mode is. During long term operation, many components can fail by any number of failure modes.

The membrane branch contains three basic events that need to be further considered. 'Pinholes', 'Pinhole Production' and 'Formation of Pinholes' are all listed as basic events, and are explained as follows; 'Pinholes' are stated as occurring 'due to exothermal combustion between H_2 and O_2'. 'Pinhole Production' is listed as not due to, but related to 'mechanical degradation'. Finally 'Formation of Pinholes' is considered to be 'due to contamination by trace metal ion'. The above would suggest that the three ‘basic’ events could be further broken down to fundamental basic events.

4 Possible ways of advancing the reliability study of a PEMFC

The initial FT developed by Placca and Kouta is a good first step in addressing the durability issues in PEMFC systems. However there are limitations in the study as shown by this research, which need to be addressed. The areas suggested are; Top Event, Bipolar Plate Omission, Relationships Between Events, Ambiguity of Events and Lack of Standardised Data.

4.1 Top Event

The standardly agreed criteria for PEMFC is to have a reduction in output voltage of no more than 5% over the 5000 hours period[9]. A top event reflecting this standard would alleviate the uncertainty with the current top event. It is suggested that a new top event is used to emulate the commonly accepted lifetime requirements of a PEMFC for portable applications as mentioned in section 2.2. A top event of; ‘5000h cell lifetime with less than 5% drop of output voltage’ would clearly define the cut-off point for any values that exceed this level.

4.2 Bipolar plate omission

The bipolar plate can degrade in two main ways; corrosion which releases metal ions, and oxide film formation.

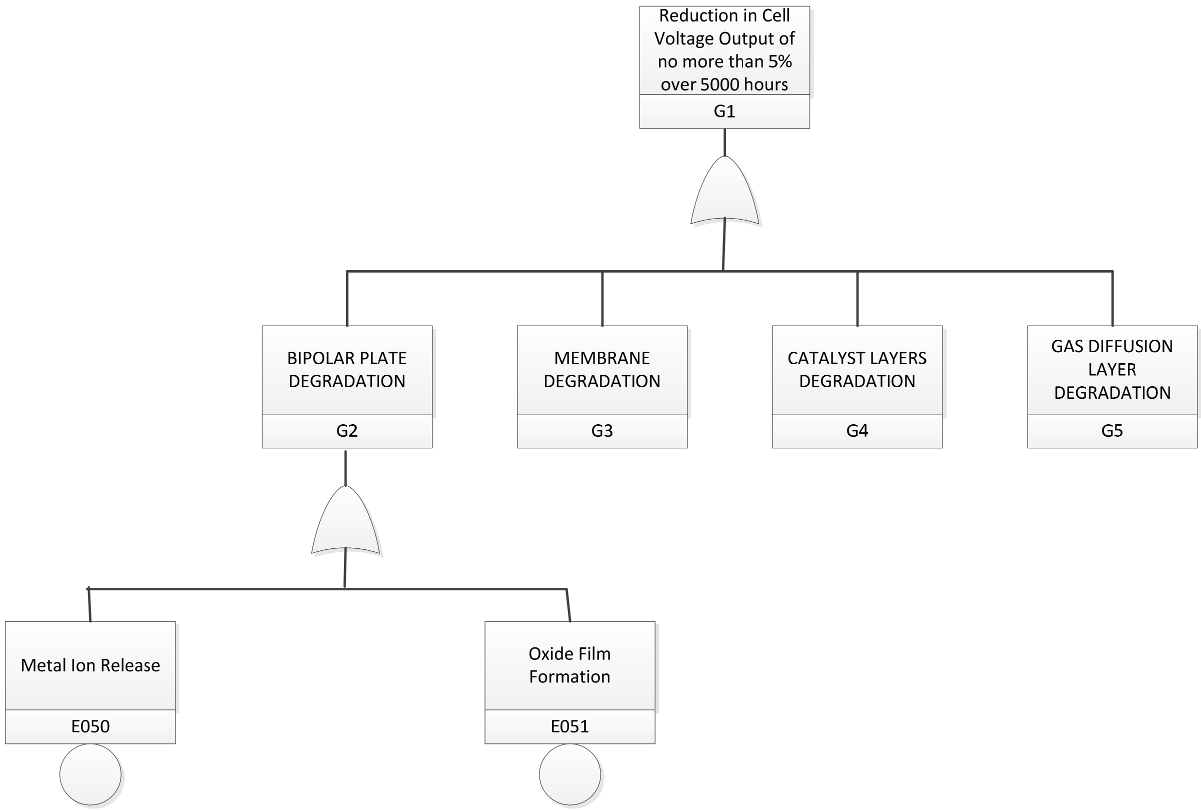

Figure 6 - Propsed change to 'global' tree

Trace metal ions such as Fe^{2+} from the bipolar plate can have adverse effects throughout the cell. It is known that these metal ions can contaminate the membrane and poison the electrode catalyst. As mentioned in section 3.2.2, the current logic regarding the trace metal ions in the membrane degradation section needs re-evaluating. The presence of foreign metal ions leads to accelerated membrane thinning and possibly pinhole generation, therefore the tree should reflect this. This would require the replication of the above basic event 'E050 – Metal Ion Release' in the newly proposed G2 and G3 intermediate events.

It has been stated that oxide film formation can increase the contact resistance of the bipolar plate by 'many orders of magnitude'[19]. This basic event would need to be included in the overall model due to its effect on the reduction of output voltage, which is the quantifier for degradation.

4.3 Relationships between events

It has been highlighted that the basic events presented by Placca and Kouta have certain relationships that make it difficult to make equivocal statements regarding the FT logic.

Pinholes were found to be three events listed as basic, but were all caused by certain conditions or phenomena. Therefore they need to be broken down further to the basic events that cause the pinholes. One way of representing pinholes in the 'mechanical degradation' branch is proposed below in figure 8.

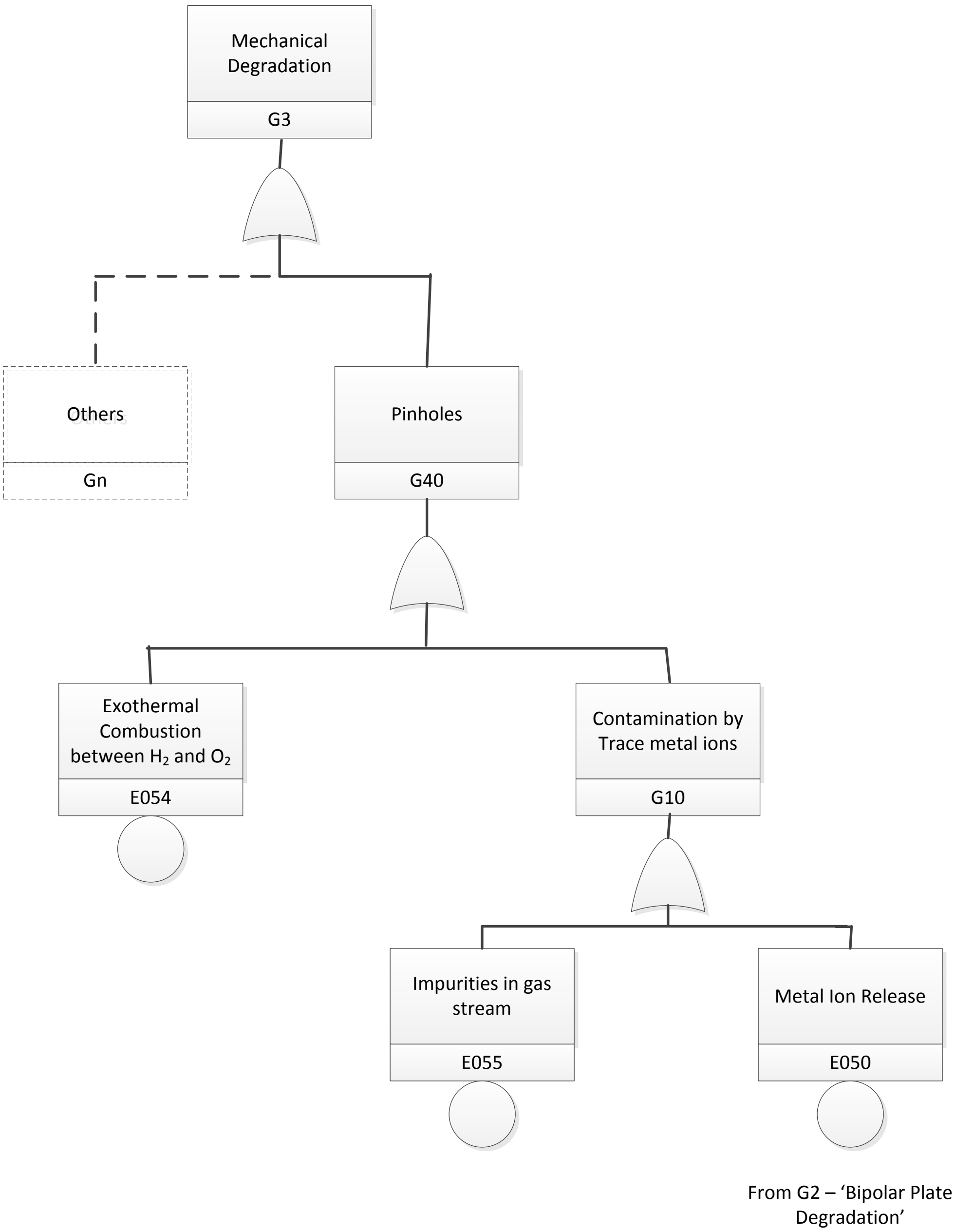

Figure 7 - Proposed pinhole logic

Figure 5 showed how certain basic events can lead on to others, and how they can also make other events worse. In particular, this instance of making others worse, questions the assumptions of the FTA techniques and its suitability for modelling these relationships with the PEMFC. It is therefore suggested that further research should look at re-evaluating the logic and structure of the presented FT. This would identify if FTA can be used for PEMFC degradation analysis, and failure forecasting.

4.4 Ambiguity of events

As with the top event, it is recommended that for a more comprehensive FT, one would need to modify all ambiguous events with definitive labels. All labels would need to be explained thoroughly or linked back to an FMEA for a clear understanding of the event.

4.5 Lack of standardised data

The lack of homogenised data for a PEMFC is a pitfall that can only be overcome by an increase in experimental analysis of certain failure modes in a fuel cell. An ideal scenario would incorporate sets of standardised experiments to homogenate degradation data, aiding with the validity of failure analysis. These would use the same cell materials, size and construction to make sure that degradation data is as reliable as possible.

5. Conclusions

Although hydrogen fuel cells have been praised as an alternative to the internal combustion engine with their ‘no moving parts’ slogan being used, the PEMFC of today is a highly complex machine and harbours extensively intricate relationships between components and operating conditions.

It is apparent that there is a need to aggregate the abundance of component level degradation data, into a comprehensive systemic model to forecast degradation of a PEMFC stack. This analysis will be the catalyst for a definitive way forward for modelling the lifetime prediction of a PEMFC.

A recently presented quantitative FT by Placca and Kouta has proven to be a good first step in degradation analysis and failure forecasting. Some areas that need to be addressed have been identified in this current study, in particular critical component omission, basic event logic & structure, ambiguity of events and lack of standardised data sets. It is envisaged that if these issues can be addressed, the overall degradation analysis of PEMFCs will become increasingly more accurate.

Future work will entail a full re-evaluation of the current FT logic and structure, in an attempt to account for all relationships and links. To justify the new logic and structure of the new tree, a full FMEA will be completed which will aid in the understanding of all possible links between basic and intermediate events. Close attention will be paid to potential dependencies that exist within the system whereupon a technique more suited to this analysis, such as Markov Modelling, may be incorporated within the FT.

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