A review on prognostics and health monitoring of proton exchange membrane fuel cell

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Abstract

Fuel cell technology can be traced back to 1839 when British scientist Sir William Grove discovered that it was possible to generate electricity by the reaction between hydrogen and oxygen gases. However, fuel cell still cannot compete with internal combustion engines although they have many advantages including zero carbon emissions. Fossil fuels are cheaper and present very high volumetric energy densities compared with the hydrogen gas. Furthermore, hydrogen storage as a liquid is still a huge challenge. Another important disadvantage is the lifespan of the fuel cell because of their durability, reliability and maintainability. Prognostics is an emerging technology in sustainability of engineering systems through failure prevention, reliability assessment and remaining useful lifetime estimation. Prognostics and health monitoring can play a critical role in enhancing the durability, reliability and maintainability of the fuel cell system. This paper presents a review on the current state-of-the-art in prognostics and health monitoring of Proton Exchange Membrane Fuel Cell (PEMFC), aiming at identifying research and development opportunities in these fields. This paper also highlights the importance of incorporating prognostics and failure modes, mechanisms and effects analysis (FMMEA) in PEMFC to give them sustainable

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competitive advantage when compared with other non-clean energy solutions.

1. Introduction

A fuel cell is simple electro-chemical device as shown in Figure 1, which converts chemical energy into electrical energy from a hydrogen fuel or hydrogen-rich fuels [1]. A fuel cell basically consists of three main components: anode, cathode and electrolyte. The electrolyte, which is made of non-conductive materials, allows charges to pass through and is sandwiched between catalytic electrodes, i.e., the anode and the cathode. Electricity is produced from the cathode to the anode, i.e., electrons flow from the anode to the cathode through an external circuit [2, 3]. Based on the materials used for the electrolyte, the anode and the cathode, there are many different types of fuel cells. Particularly based on the non-conductive materials used for the electrolyte, fuel cells can be classified into alkaline fuel cell (AFC), proton exchange membrane fuel cell (PEMFC), direct methanol fuel cell (DMFC), phosphoric acid fuel cell (PAFC), molten carbonate fuel cell (MCFC) and solid oxide fuel cell (SOFC) [4, 5].

An individual fuel cell typically delivers low voltages and high currents. Typical voltage and current ranges are from 0.4 to $0.9V$ and from 0.5 to $1A/cm^2$ respectively [6, 7]. For example, the fuel cell developed at the Sustainable Energy Technology Centre, University of Hertfordshire, is reported to produce about $0.7V$ (after losses) and $0.6A/cm^2$ [8]. In order to achieve a higher power output, the fuel cells need to be stacked together as shown in Figure 2. Depending on the power output and the applications, fuel cells come in various shapes and sizes [9]. Fuel cell has demonstrated to be an attractive alternative energy generation technology from hydrogen-rich fuels [2, 3, 5]. A fuel cell does not have any mechanical moving parts, but have high energy efficiency and zero emissions i.e., to deliver no pollution to the environment during operation. Hence, there is a good potential for fuel cell to gradually replace internal combustion

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(IC) engines in the future [10, 11]. Fuel cells are used in many applications, such as in aerospace and automotive vehicles, in small and large scale power generation plants, in portable power generators, in combined heat and power (CHP) generation, and in backup power applications [12].

PEMFC is the most suitable type of fuel cells for many applications because of their operating temperature range (between 20°C and 100°C) and quick response time compared with other types of fuel cells. Compared with high temperature fuel cells, PEMFC can be operated very quickly in lower temperature. PEMFC materials cost is lower than that of a high temperature fuel cell. If hydrogen is used as a fuel, lower temperature fuel cells are safer to use [5]. Hence hydrogen PEMFC is most suitable for portable power generators such as those used in spaceships or automotive vehicles [13, 14]. However, main drawbacks are the cost and the lifespan of fuel cell because of durability, reliability and maintainability issues associated with them [13, 15, 16]. So far, general life expectancy of a fuel cell is not up to the expectation in industries. For example, a typical life expectancy of the PEMFC is around 2500 hours, whereas transportation applications require at least 5000 hours and stationary applications require at least 40000 hours [17, 18].

Health monitoring has been used in engineering systems for many years to ensure performance, safety, availability and reliability [19, 20, 21, 22]. Generally speaking, sensors are used to monitor the operating conditions, performance and loading cycles. Anomalies and faults can be detected on time, hence avoiding otherwise unpredicted incidents, down time and fault propagation [23]. Typically, small faults in a part of the system may develop as a major fault, so that ultimately the system may fail adversely. If the fault can be detected or predicted at an early stage, then the system can be scheduled to maintenance on time before the fault develops into something more serious [24]. Therefore, applying health monitoring techniques will be a definite advantage, not only for the safety reasons, but also for a considerable reduction of unscheduled maintenance costs. [25].

Prognostics is an engineering process of diagnosing, predicting the remaining
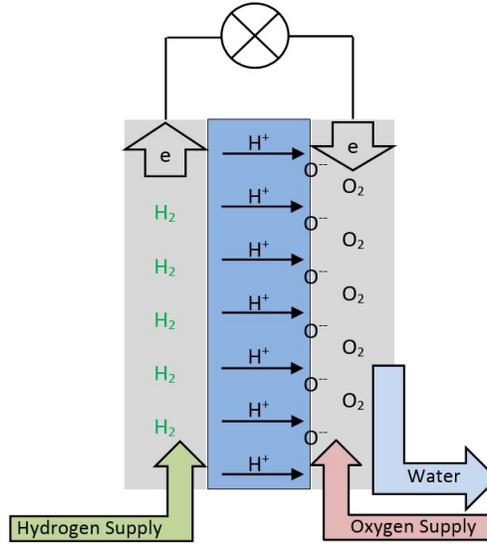
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Figure 1: Schematics of the principle of operation of a fuel cell

useful lifetime (RUL) and estimating the reliability of a system [26, 27, 28, 29]. It has emerged in the last decade as one of the most efficient approaches in failure prevention, reliability estimation, RUL prediction of various engineering systems and products [30, 31]. There are three different approaches to prognostics, namely (1) data driven approach, (2) model driven approach, and (3) fusion approach [26]. As prognostics can provide state-of-health (SOH) and RUL information of the fuel cell, the operation of the fuel cell can be optimised using an appropriate control strategy. Maintenance tasks can be scheduled, thus reducing down time. Although prognostics was used in safety critical systems in early days, it is nowadays an integral part in many engineering systems, products and applications [26]. Hence application of Prognostics, along with health monitoring to PEMFC, can be used to improve the reliability, sustainability and maintainability (through evidence based decision making), reduce the life cycle cost, and can also provide feedback to the design and validation process [32, 33, 34]. Health monitoring sensors can be used to monitor the important parameters such as precursor parameters and loading conditions [35]. Prognos-
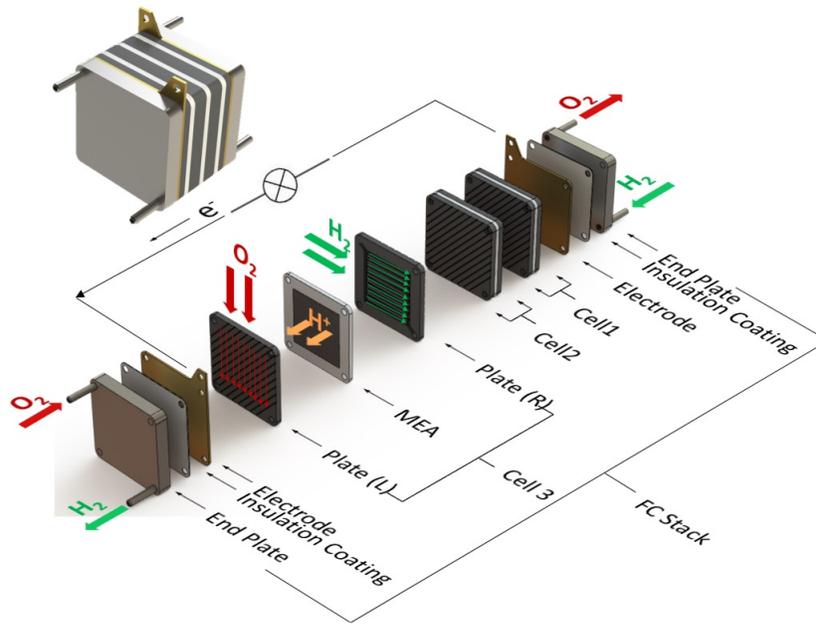
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Figure 2: Schematics of a fuel cell stack operation and components

tics can use sensor information to predict the remaining useful life time (RUL),
to diagnose failures well before they develop into a serious problems, and to
provide information to control systems to automate contingency management
[36, 37, 38]. Hence both health monitoring and prognostics can play a vital role
in improving the durability, reliability, and maintainability of PEMFC system.
This will help to overcome the main challenges faced by the PEMFC industry
today.

Although some developments have been reported in prognostics for PEMFC,
more research needs to be done in the field. Development of prognostics for
PEMFC has become a hot topic in the recent years as PEMFC has the po-
tential of replacing the internal combustion engine in the future [39]. PEMFC
is a very sensitive electrochemical device which involves heat transfer, charge
transport, electrochemical reaction and multi-phase flows, hence developing a
prognostics methodology has become a complex and complicated process [17].
Although a fuel cell has no mechanical moving parts, membrane electrode as-

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sembly (MEA) undergoes degradation processes similar to those occurring in mechanical systems because of the electrochemical reaction and multi-phase flows. These degradation processes might be natural in some cases but most of these processes of degradation could be accelerated by loading cycles, operating conditions, etc. Furthermore, failures in MEA are very difficult to measure or observe directly as MEA is placed between the bipolar plates. Failure modes, mechanisms and effects of MEA are not very well researched and understood. Therefore, a detail study of FMMEA is necessary for two main reasons: (1) to apply prognostics at the deployment stage of the fuel cell systems and (2) to understand the underlying physical processes of degradation and improve the design of MEA and other components using novel materials which have high resistance to degradation. FMMEA studies can also help to identify the precursor of failures in the MEA to start the process of prognostics and health monitoring.

This paper presents a critical review in prognostics approaches and health monitoring techniques to improve the lifespan of the PEMFC. The prognostics approaches, such as data driven, model driven and fusion approach, have been studied and reviewed, including their advantages and disadvantages. Successful applications of prognostics in other engineering systems, including batteries which are also an electrochemical device, have been highlighted. The key aspects, such as degradation mechanisms, prognostics modelling, accelerated testing, different health monitoring parameters and prognostics, are studied. In particular, both prognostics approaches and health monitoring techniques in PEMFC are reviewed. Finally, this study highlights the importance of incorporating different failure models into a global model so that prognostics can be applied to PEMFC more precisely to improve the lifespan.

2. Prognostics

Prognostics is a technology used to monitor degradation in engineering systems, predict when failure may occur, improve reliability, and provide a cost effective strategy for scheduled maintenance [40]. Prognostics of engineering

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systems or products has become very important as degradations in the individual parts may cause a severe (and irreversible) damage to the entire system,
120 environment and users. Ultimately, it may lead to failures and will result in significant costly repairs, that could otherwise have been avoided. Adopting prognostics techniques require continuous monitoring of performance, loading cycles and precursors of failures, and detecting any anomalies in these parameters.

125 Figure 3 illustrates the three main approaches to prognostics, which are (i) Data driven, (ii) Model driven and (iii) Fusion approach. Fusion approach is a combination of both (i) and (ii) methodologies. Figure 3 also shows the classifications of prognostics approaches. Data driven approach can be further classified into statistical and machine learning techniques. Statistical techniques
130 can be either parametric or non-parametric. Machine learning techniques can be either supervised learning, where test data is available or unsupervised learning, where test data is not available. Model driven approach can be based on physics of failure models or system models. Physics of Failure (PoF) models are based on the underlying physical phenomena of failures which requires detailed FMMEA
135 study. System model relates the system's output to its input, and it can be derived from first principles or test data. Fusion approach entails a combination of data driven and model driven approaches which incorporates the benefits and eliminates the drawbacks from both approaches [26].

2.1. Data Driven Approach

140 Data driven approach is considered as a black box approach to prognostics as it does not require system models or system specific knowledge to start the prognostics [41]. Monitored and historical data are used to learn the systems' behaviours and used to perform the prognostics. Hence the data driven approach is suitable for the systems which are complex and whose behaviours
145 cannot be assessed and derived from first principles. The implementation of data driven techniques for the purpose of health monitoring and prognostics is generally based on the assumption that the statistical characteristics of system's
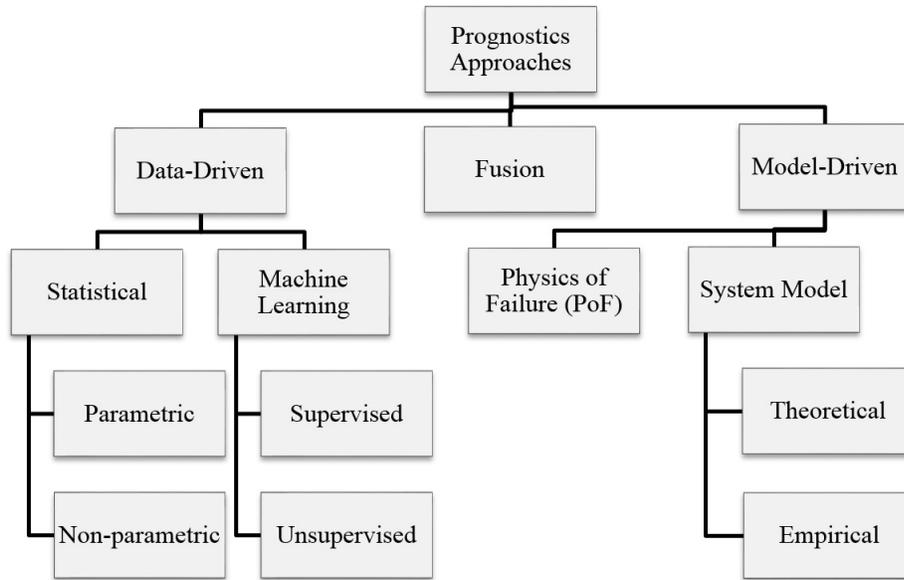
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Figure 3:   Classification of prognostics approaches

performance will not be changed until fault occurs [41]. Therefore, the main advantage of the data driven approach is that the underlying algorithms are quicker to implement and computationally more efficient to run compared with other techniques [42]. However, it is necessary to have historical data and knowledge of typical operational performance data, the associated critical threshold values and their margins. Data driven techniques completely rely on the analysis of data obtained from sensors and exploit operational or performance related signals that can indicate the health of the monitored system. Data driven strategies to prognostics have been applied in a number of engineering applications [43, 44, 45, 46, 47, 48, 49, 50].

The principal disadvantage of the data driven approach is that the confidence level in the predictions depends on the available historical and empirical data. Historical and empirical data are required in the data driven approach to define the respective threshold values. In some instances it is difficult to obtain or have historical data available, for example in the case of a new system or device that

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may require long time to test. When a test takes a long time to complete, the data collection becomes expensive and deployment of the system will take an ₁₆₅ unrealistic long period of time. However, there are techniques and procedures, that can be used to overcome this disadvantage [51, 52]. Three of the strategies that could be used to address this challenge are based on the use of:

1. Hardware-in-the-Loop simulations (HiL): Hardware-in-the-Loop is a computer simulation which is used to test a real product or system by connecting it to the hardware that applies simulated loads as in a real application. It is very fast and cheap to implement. In addition, several failure parameters (i.e., operational and environmental) can be controlled independently. HiL can also be used to develop algorithms, test and validate the algorithms, benchmarking and to develop metrics for prognostics [51].

2. Accelerated Life Test (ALT): Accelerated life test is designed to cause the product to fail more quickly than under normal operating conditions by applying an accelerated (elevated) stress condition which is responsible for a particular failure mechanism. ALT becomes an important method in the development of the prognostics. Several environmental and loading conditions can be applied independently to accelerate failures [31, 52, 53, 54, 55].

3. Online Learning (Semi supervised/Unsupervised learning): Online learning is based on the assumption that a new system performance data represents the healthy system and that they do not fail for a certain period of time. This type of approach can also be called semi supervised or unsupervised learning as only healthy data or no reference data is available. Reinforcement learning approach is also suitable for this strategy [56].

2.2. Model Driven Approach

The model driven approach uses mathematical equations that predict the ₁₉₀ physics governing failures and therefore is sometimes referred to as the Physics of Failure (PoF) approach. It requires knowledge of the failure mechanisms, geometry of the system, material properties and the external loads that are
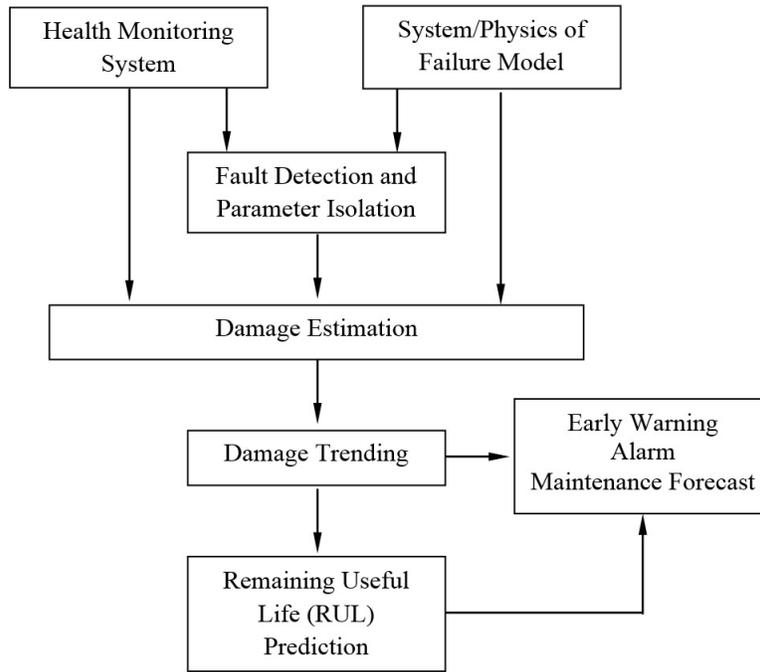
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Figure 4: Model driven approach to prognostics

applied to the system. An accurate mathematical model can benefit the prognostics process, where the difference between the output from a mathematical
195 model and the real output of the system can be used to find the anomalies, malfunctions, disturbances, etc. [26, 57]. Using the difference between the model and the data values for a performance parameter, the early warnings for failures and RUL can be predicted. Many prognostics works have been published based on the model-driven approach [47, 48, 58, 59, 60, 61, 62, 63]. A block diagram
200 of a typical model based approach is shown in Figure 4. Typical model driven approach is based on the system/physics of failure model for which the health monitoring system will provide required sensor data. Once a fault is detected by feeding the sensor data into the model, the damage parameter is isolated and a damage is estimated. The damage trend will then be used to estimate
205 the RUL [26].

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2.3. Fusion Approach

The fusion approach is based on the advanced features of both the data driven and model based methods. This approach requires an accurate mathematical model of the system for the physics based failure approach, and enough historical data and knowledge of typical operational performance data, for the data driven approach. The aim of the fusion approach is to overcome the limitations and disadvantages of both model and data driven approaches to estimate the remaining useful life [26]. Therefore, the accuracy of the fusion approach should be higher than both model and data driven approaches when used individually [26], although for a real-time analysis it may not be suitable due to the significant computational resources required. The fusion approach has been reported to be used in many applications before [26, 64, 65, 66, 67, 68, 69].

3. Applications of Prognostics to PEMFC

Although prognostics is used in many engineering systems and products including batteries [70, 71, 72, 73, 74, 75, 76, 77], to assess the remaining useful life and enhance the durability and reliability, prognostics is rarely discussed with respect to fuel cells. Jouin et al. (2013a, 2013b) discussed the benefits of applying the prognostics techniques to monitor the SOH and estimate the RUL of PEMFC, aiming at improving their durability and reliability, and hence extend their life spans [78, 79]. They also discussed layer approach (based on different tasks) to prognostics and health management (PHM) and degradation mechanisms in these review papers. Lack of experimental and failure data, and complete models which incorporates all wear mechanisms were also reported as main challenges [78, 79].

Most fuel cells need a health monitoring system in place to assess their performance. They are generally used to give early warnings or change the control strategy if an anomaly is detected in the performance variable or in any key monitoring parameter [23]. Sensors are used to monitor the parameters that need to be watched. Because a fuel cell is very sensitive to the supplied fuel, oxidant,

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load current and the amount of water and heat produced, health monitoring becomes a vital component to control the fuel cell system [2]. Furthermore, prognostics and health monitoring can provide information to a prognostics system to estimate RUL, schedule maintenance and improve the sustainability of the fuel cell.

240   This review is focused on PEMFC and organised under the subsections of degradation mechanisms, modeling for prognostics, accelerated testing, monitoring parameters and techniques, diagnosis and RUL estimation. Before starting the prognostics process, it is necessary to understand the most effective degradation mechanisms of PEMFC. Under this section degradation mechanisms responsible for failures in PEMFC are reported. For the model driven approach to prognostics, PEMFC failure models are necessary. Models could be derived from the first principle or test data. PEMFC models used for diagnosis and prognostics are discussed next. Accelerated test is discussed for the purpose of failure data collection, and understand the effect of different degradation mechanisms in the performance of PEMFC. Next, monitoring parameters and techniques for health monitoring and prognostics of PEMFC are investigated from the literature. Finally, from the prognostics models, test data, monitoring parameters and monitoring techniques, how PEMFC can be diagnosed regarding its failure conditions and how RUL is estimated are discussed, respectively.

3.1. Degradation Mechanisms

    A Membrane Electrode Assembly (MEA) is the key component of the fuel cell. The MEA consists of electrodes and membrane which is sandwiched between two electrodes. A fuel cell stack has many MEA stacked together. Failure in any components of the MEA i.e., membrane or electrodes, will cause complete failure of the stack even though all the other MEA are functional. Although there are no mechanical moving parts in the fuel cell, fuel cell components undergo degradation processes similar to other mechanical components. It is therefore necessary to understand the degradation mechanisms of the fuel cell components, particularly the membrane. Membrane degradation can be cate-

gorized into the followings: (1) Chemical degradation, (2) Mechanical degradation and (3) Membrane shorting [80, 81, 82]. Chemical degradation occurs when the membrane decomposes because of electrochemical reaction caused by poisonous substances and radicals which are produced in the cathode and anode during chemical reactions [83]. Some of the typical radical elements are peroxide (HO^-) [84], carbon monoxide (CO) [85] and hydroperoxide (HOO^-) [86, 87, 88]. Mechanical degradation occurs, when membrane undergoes mechanical degradation such as fracture because of thermal stress [80], humidity [89, 90, 91], pressure and mechanical stress. Membrane shorting occurs when membrane allows current to pass through. Mechanical degradation may cause early failures because of manufacturing defects and improper MEA fabrication [86]. Mechanical failure may also be caused by excessive or non-uniform pressure [86]. Fuel cell undergoes thermal and humidity cycling which may lead to additional mechanical stress on the MEA [91]. These degradations could lead to a decrease in the performance of the fuel cell and ultimately failures in membrane; hence a complete failure in the fuel cell stack.

Platinum (Pt) based catalysts are used in the electrodes to increase the rate of chemical reaction. These catalysts typically undergo variable potential cycling from 0.6 to 1.0 V. It may also undergo higher potential spikes and higher potential voltage during uncontrolled operations [92]. Sudden increases in the load current may result in a reduction of the catalyst area [80]. Pt dissolution is generally caused by chemical oxidation by the oxygen in the cathode electrode [93]. These conditions may expose the catalyst into electrochemical stress and may result in irreversible degradation in the catalyst. Some of the other operating conditions which may accelerate the failures are relative humidity [94, 91], reactant starvation [95, 96], carbon monoxide poisoning at the anode catalyst [96, 97, 98], cathode flooding and membrane drying [98, 99, 100, 101, 102]. Fuel cells run close to their open circuit voltage, accelerating different degradation mechanisms in the membrane and catalyst [103].

Many degradation mechanisms were investigated theoretically and experimentally. But the data related to which failure mechanisms cause more failures

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and which failure mechanisms cause less failures are not available. Failures and their corresponding failure mechanisms need to be investigated in relation to fuel cell applications such as stationary, automotive, backup, combined heat and power, etc. Hence it is necessary to carry out application specific failure modes, mechanisms and effects analysis.

3.2. Modeling for Prognostics

Once the failure modes, mechanisms and their effects are understood, physics of failure or underlying physical processes of failure need to be modeled. Prognostics-oriented fuel cell catalyst aging model has been reported by Zhang and Pisu (2014) [104]. Catalyst degradation model is based on on the platinum dissolution kinetic model proposed by Darling and Meyers [105, 106] and simplified for the purpose of prognostics. Aging parameters for this model are electrochemical surface area and membrane gas crossover. Burlatsk et al. (2012) have reported a mathematical model to predict the life of PEMFC under hydration dehydration cycling. Stresses associated with hydration and dehydration cycle have been modeled mathematically, particularly a model of relative humidity (RH) distribution in gas channels, a model of membrane stress and a model of damage accrual [107]. These model predict membrane lifetime as a function of RH cycle amplitude and membrane mechanical properties. A viscoplastic model of Nafion® has been reported in the literature by Solasi et al. (2008) [108]. Experimental results were used to develop a nonlinear time-dependent constitutive model to predict the hygro-thermomechanical behaviour of Nafion®. Rong et al. (2008a & 2008b) have developed a rate-dependent isotropic plasticity model with temperature and humidity dependent material properties to understand the viscoplasticity properties of catalyst layer components. Numerical simulations were used to investigate the crack initiation in the material [109, 110].

PEMFC models have been developed by a number of researchers mainly for the purpose of control. Generally, models for each part of the fuel cell such as catalyst layers, gas diffusion layers, etc. were developed and then integrated to simulate and understand the behaviours of fuel cells under different operat-

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ing conditions [111, 112, 113]. But underlying physical processes of failures or physics of failure models have not been fully developed i.e. integrated failure model for most of its failure mechanisms. Hence it is very important to develop failure models for each failure mechanisms and integrate them into a model which can predict most of the failures.

3.3. Accelerated Testing

Accelerated testing is a useful tool to collect failure data, understand the failure mode and mechanisms, and develop prognostics strategies. Accelerated testing on PEMFC can be carried out under load cycling [114, 115, 116], RH cycling [114, 117, 118, 119, 120], elevated temperature [121], thermal cycle [122], load ripples [123], anode flooding, membrane drying, carbon corrosion [88], Pt dissolution [55, 88] etc. Under the accelerated testing, effects of one parameter can be investigated effectively by keeping all the other parameters at normal conditions and changing the particular parameter between two possible extreme values. Accelerated testing can also be carried out for radical elements and external poisoning by introducing these elements into the fuel cell.

Accelerated testing under combined RH cycling and load cycling has been reported by Wu et al. (2014). Under these conditions, severe chemical degradation was observed in the membrane using transmission electron microscopy (TEM) and scanning electron microscopy (SEM) cross sectional images [114]. Petrone et al. (2015) proposed a new approach and protocol to accelerated testing for the purpose of prognostics and lifetime prediction based on adaptable load cycling [124]. It is important to have such general protocol to conduct accelerated testing on PEMFC for the purpose of prognostics and life time prediction.

3.4. Monitoring Parameters and Techniques

3.4.1. Voltage

Cell voltage is among the cheapest and easiest ways to implement monitoring techniques. It also is one of the quickest approaches to monitor a fuel cell, as

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voltage measurements do not require expensive and specialized sensors. Most of the fault modes of the fuel cell cause a voltage drop. Anode flooding at low current densities was investigated by O'Rourke et al. (2009) based on cell voltage measurements [125]. These cell voltages were then compared with median cell voltage. If the difference between any cell voltage and median cell voltage was higher than a predetermined value, then the cell could be under anode flooding. This method is a more efficient approach than the low frequency ($< 6Hz$) impedance measurement.The low frequency impedance measurement technique may take a longer period of time to identify flooding. Once flooding occurs and the cell voltage has already been decreased, it is likely that the irreversible phenomenon has already been started [125].

Membrane drying and cell flooding were investigated by Frappe et al. (2010) [126]. In this work, it was observed that membrane drying increased the membrane resistance because of insufficient water. Cell flooding blocks a part of the active area therefore the active area of the fuel cell reduces. Frappe et al. (2010) proposed that it was possible to monitor only a group of cells instead of monitoring every individual cell in the stack. Thus, sample groups of cells were selected to monitor at the inlet, outlet and center of the stack. State-of-health indicator of the fuel cell stack was proposed as the voltage difference between the center group and the inlet/outlet group. Based on this indicator, Frappe et al. (2010) proposed that if there were no voltage variations (the difference is zero), then this implied no fault condition; if all the voltages dropped together at the same time (again the difference is zero), then this implied a load variation; if the voltage of the center group dropped, there was possible drying; and if the voltage of the inlet/outlet group dropped, there was possible flooding. This approach was backed up with experimental data and results [126].

Xue et al. (2006) investigated model based condition monitoring of a PEMFC based on a lumped parameter dynamic fuel cell model and by employing the Hotelling T^2 statistical analysis [58]. Fault detection of the PEMFC was facilitated by comparing the real time fuel cell output voltage measurements with the baseline voltage by employing the Hotelling T^2 statistical analysis. The baseline

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voltages were used to evaluate the output T^2 statistics under normal operating conditions. Upper control limit for the fault was established and fault condition was declared if the T^2 statistics of real-time voltage measurements exceeded the upper control limit [58].

₃₉₀ *3.4.2. Impedance*

Alternating current (AC) impedance is generally used to estimate the state-of-health (SOH) of fuel cells and batteries [127, 128]. AC impedance can also be used as an in-situ health monitoring technique for PEMFC. An AC impedance measurement technique coupled with a model-based approach was suggested by ₃₉₅ Fouquet et al. (2006) [127]. In this study, a $150cm^2$ hydrogen PEMFC stack consisting of six cells was used. Test data was then fitted to parameters of a Randles-like equivalent circuit. In order to improve the quality of the fit, classical Randles was replaced with a constant phase element instead of a standard plane capacitor. It was reported that this modified model of Randles equiva-₄₀₀ lent circuit was an efficient and robust way to monitor the SOH of hydrogen PEMFC with respect to water content on MEA. Flooded and dry conditions were identified with respect to the variation of the parameters of the proposed modified Randles equivalent circuit model for PEMFC [127]. Unlike Fouquet et al. (2006), Kurz et al. (2008) reported a predictive control strategy based ₄₀₅ on impedance measurements [128]. From the impedance measurements at two different frequencies, the voltage drop caused by the flooding and drying phenomena was detected. Kurz et al. (2008) used this information to run the fuel cell at an optimal operating point [128].

Rubio et al. (2007, 2008) proposed a current interruption method to estimate ₄₁₀ the PEMFC model parameters such as double layer capacitance, diffusion resistance, charge transfer resistance, diffusion related time constant and membrane resistance (i.e. impedance) [129, 130]. It was found a correlation between the cathode flooding phenomenon and the diffusion resistance. It was also reported that impedance model parameters can be used to diagnose other degradation ₄₁₅ phenomena such as membrane drying, cathode drying, membrane degradation,

and anode poisoning. The current interruption method was shown to be easy to use as an in-situ health monitoring method [129, 130]. An implementation of continuous real-time impedance spectroscopy was carried out by Bethoux et al. (2009) [131]. This work showed that small sinusoidal current of known ampli-
tude and frequency could be sent into the fuel cell stack while it was operating. A low pass filter was proposed to retrieve the signal back from the fuel cell stack without any electrical disturbances to the load. Using the collected data, complex impedance can be estimated and equivalent Randles circuit parameters can be computed [131].

Cooper and Smith (2006) investigated four electrical measurement techniques for on-line measurement of ohmic resistance: current interrupt, AC resistance, high frequency resistance and electrochemical impedance spectroscopy (EIS) [132]. Internal resistance measurements of PEMFC based on all four techniques were compared. Current interrupt and high frequency resistance methods correlated each other well if the high frequency measurement technique was in the suitable range. For hydrogen PEMFC operating with moderately humidified reactants, the ohmic resistance measurements determined with the current interrupt technique, high frequency resistance and EIS were within $10-30\%$. It appears that there are considerable differences between these three techniques and there is no agreement about which method is the most suitable for on-line electrical measurement of ohmic resistance of hydrogen PEMFC [132]. Fast EIS approach can also be used to assess the performance of PEMFC in the real time [133]. For this purpose chirp signal which frequency changed with time, was used. Fourier transformation and Wavelet Coherence techniques were used to analyse the output signal [133].

Rubio et al. (2010) classified PEMFC degradation phenomena based on time scale, i.e., time taken to observe a variation in the performance of the fuel cell [98]. Cathode flooding, membrane drying, catalyst poisoning, and contamination of the hydrogen or oxidants were reported as short time scale phenomena. Slow rate chemical degradations such as corrosion and membrane degradation were mentioned as long time scale phenomena. PEMFC internal resistance was

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reported as a monitoring parameter and Rubio et al. (2010) showed that different phenomena could be observed from the relative increment of the PEMFCs internal resistance [98]. Generally short time scale phenomena of degradation are reversible if the control system takes an appropriate action (such as purging) on time so that the fuel cell recovers from the short time scale degradation phenomena. But the long time scale degradation phenomena is generally irreversible and the rate of degradation depends on the rate of chemical reaction such as corrosion and membrane degradation.

3.4.3. Temperature

Temperature measurement of PEMFC is important for the safety purposes and for the health monitoring purposes. Thermal stability of the PEMFC is important for better performance [134] and long life. Hence most of real world PEMFC systems will have temperature monitoring components and the collected data will be used to control the fuel cell operation for optimum power generation. Generally, temperature measurements are taken at multiple places which could be identified through numerical heat transfer simulations [135]. Location of these temperature measurements may vary with the structure of the stack, type of cooling i.e. water or air, size of the stack, material properties of fuel cell components, etc.

Correlation between local temperature and local current density was observed by G. Zhang et al. (2010) and local temperature rises with the local current density with decreasing operating voltage. Hence temperature and current data can provide detailed information about the stack condition and can be used for prognostics and health monitoring of the fuel cell.

3.4.4. Acoustic Emission Technique

Acoustic emission (AE) is the propagation of transient elastic waves in a solid medium, which are generated when the solid structure is subjected to irreversible changes such as crack or plastic deformation. Acoustic emission can be used to monitor deformation in the membrane due to the water contents. This

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was investigated and reported by B. Legros et al. (2010). AE has been used as diagnostic tool for water management i.e. hydration and dehydration. AE showed good sensitivity to different operating conditions such as gas humidification levels and MEA water uptake. It means AE could be used as online non-invasive monitoring strategy for PEMFC [136]. B. Legros et al. (2011) have also investigated electrochemical noise (EN) as a tool for the diagnosis of the PEMFC when it undergoes flooding and drying [137]. EN was increased during the drying of the fuel cell and also with current level.

3.5. Prognostics and RUL Estimation

A behavioural model suitable for PEMFC prognostics was developed by Lechartier et at. (2015) [138]. This model consists of two parts: (1) static model which represents polarization curve based on Butler-Volmer law and dynamic model which represents electrical equivalence of the fuel cell dynamic behaviour. Parameters were updated from the electrical measurements such as polarization curve and EIS. Static model was validated by comparing the experimental data and simulated results. Good fitting was observed in the dynamic model parameters. The combined behavioural model was also simulated. Performance of the model prediction was very good in new stack rather than in an old stack. A few problems needed to be addressed such as difference between the operating conditions for the polarization curve and aging process, decomposition of the current parameter and integration of some other input to improve the performance [138].

A complete analysis of prognostics to PEMFC was carried out by Jouin et al. (2016) [139]. A detail framework was proposed considering all the factors influencing two important outputs i.e. output power and lifetime of the stack. This detail analysis starts with vocabulary definition followed by literature review on degradation and incorporating most recent understanding of the degradation phenomena to establish a complete degradation and failure analysis of PEMFC. Electrodes and membrane were identified as most concerned and most affected components in the PEMFC. Certain degradations phenomena were selected and

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modeled to predict the power and lifetime accurately. Although validation results have shown higher correlation for four different datasets, it is required to test the model on actual systems such as automotive applications so that it could be validated for actual application [139, 140]. This work shows better improvement on prognostics work related to PEMFC. Framework should be incorporated with other degradation models and failure mechanisms to make complete and common framework for the prognostics of PEMFC. RUL estimation completely relies on the criteria for end-of-life and it may vary from system to system, from manufacturers to manufactures etc., as there are many parameters influence the lifetime of PEMFC. It makes difficult to define thresholds to each parameter that influences the lifetime [141]. Hence agreed definitions for healthy and failed PEMFC should be established first. For example 0 - 5% of power loss is good state of health (SOH), 5 - 10% of power loss is acceptable SOH and more than 10% power loss can be taken as degraded SOH [141]. This needs to be optimised and agreed by the manufacturers, system integrators, researchers etc. to maximise the return on the investment on the PEMFC systems.

Zhang and Pisu (2012) presented what possibly is the first systematic work on prognostics and RUL estimation of PEMFC [142]. They investigated a physics-based model for the purpose of prognostics based on an electro-chemical surface area (active area) under different operating conditions [142]. This work was based on the spatially lumped model and kinetic expression for platinum oxidation and dissolution presented by Darling and Meyers (2003, 2005) [105, 106] and on the 64-particle catalyst degradation model proposed by Zhang and Pisu (2012) [104]. The method was demonstrated by simplifying the model to a first order dynamic model where the dynamics of platinum oxide coverage during load cycling was neglected. Hence, this work is a model based approach. Low pass filter and Unscented Kalman Filter (UKF) were used to capture the slow degradation in the residual between the model of the catalyst and actual catalyst [142]. Later, Zhang and Pisu (2014) developed a diagnostic-oriented fuel cell model which incorporated the fault of water flooding inside the fuel cell.

UKF were then developed for channel flooding and gas diffusion layer flooding [143]. Extended Kalman filter (EKF) were used to estimate the RUL based on load current [144, 145]. An empirical degradation model was built from experimental data and parameter analysis for this purpose. An inverse first-order reliability model was used to extrapolate the SOH of PEMFC for the uncertainty quantification of the RUL estimation [145].

Jouin et al. (2014a) proposed a particle filtering framework for the prognostics of PEMFC [146]. A voltage drop representing irreversible degradations was used as the aging indicator of the PEMFC. Two sets of test data were used in this work: (1) aging under a constant 70A current load; and (2) aging under a high frequency small ripple current load of 70A. Tests under similar stable environmental conditions were carried out on a 5-cell PEMFC stack with an active area of $100cm^2$. The voltage evolution with time was modelled and used as the state model. Three different state models which could be used to represent voltage evolution were used: (1) linear model, (2) exponential model and (3) logarithmic model. Performance metrics of these three models were compared and the logarithmic model's predictions were found more accurate with greater stability [146]. Later, Jouin et al. (2014b) applied particle filtering framework based on power evolution with time and estimated the RUL using the same data [147]. Power evolution, acceleration of power degradation and recoveries of the power degradation were considered, modelled and incorporated with Particle Filtering (PF) framework by Jouin et al. (2014b) [147]. Predictions from this particle filtering framework produced less errors compared with other particle filtering frameworks [148]. Particle filter approach based on polarization equation, introduction of time dependency of the mission profile, and degradation of different components of the stack, was applied to micro Combined Heat and Power (μCHP) [149].

An adaptive particle filter algorithm based approach to prognostics and RUL estimation was proposed by Kimotho et al. (2014) for the IEEE PHM data challenge [150]. Kimotho et al. (2014) introduced a self-healing factor after each characterization and the adaptation of the degradation model parameter to fit

the change in the degradation behaviour at various stages of the PEMFC lifetimes [150]. SOH prediction based on physics driven and data driven model was reported by Kim et al. (2014). Voltage degradation model and four parameter equivalent circuit models were developed based on underlying physical phenomena. These model then were trained for the training data set, tested and validated on the test data set. The four parameters from the equivalent circuit model exhibited linear relationship with the voltage degradation which could be used to estimate the RUL of the PEMFC [151].

Further work on particle filters has been reported by Jouin et al. (2016) [152]. A global particle filter for power aging and three other particle filters were dedicated to specific parameters in the global filter. Unknown coefficients of the models had been estimated least square fitting to initialise all four particle filters. RUL predictions were within the 5% of error for a validation test of more than 500 hours. Quantity of data was needed for learning reported as a major drawback i.e. more than 1100 hours of test data was needed for learning which is very high compared with the lifetime of a PEMFC system [152].

An Adaptive Neuro-Fuzzy Inference System (ANFIS) was applied to the voltage drops caused by the degradation during normal operation of PEMFC stacks by Silva et al. (2014) [153]. A data driven approach based on an Echo State Network (ESN) which uses of a dynamical neurons reservoir, was studied as a prognostics system enabling an estimation of the remaining useful life of a PEMFC by Morando et al. (2014) [154]. This work was also based on cell voltage. In particular, the mean cell voltage was used to forecast the degradation of PEMFC [154]. A sensitivity analysis for this Echo State Network (ESN) based on data driven prognostics approach was studied by Morando et al. (2014), where the Analysis of Variance (ANOVA) statistical technique was used [155].

A novel PEMFC performance-forecasting model based on a modified relevance vector machine (RVM) was reported by Wu et al. (2016) [156]. Experimental aging voltage data was used to model RVM for a PEMFC stack. Model was then applied to voltage-degradation data from two experiments for a 1.2kW fuel cell stack. Results of the performance-forecast model were com-

pared with the counterpart model of classic support vector machine (SVM).
Although a good agreement between both models was observed, better predictions were made using modified RVM model compared with the SVM based model particularly when there were limited data available for prediction [156].

A data driven approach based on a constraint based Summation Wavelet-Extreme Learning Machine (SW-ELM) algorithm was developed by Javed et al. (2015). This algorithm was developed using stack voltage drop as an useful prognostics indicator and assuming aging process is irreversible [157, 158]. Algorithm was validated using Prognostics and Health Management (PHM) challenge data for 2014 which was published by PHM Society. Satisfactory performance was observed considering the model complexity, computational time and accuracy of the predictions [157]. This particular SW-ELM is computationally less expensive and has been recommended that it could be easily integrated into the system to carried out real-time RUL estimation to reduce the maintenance cost [158]. An ensemble structure of SW-ELM with a new incremental learning scheme was applied to μCHP to estimate the SOH of the stack under variable load conditions for a a year [159]. A constraint based ensemble strategy of connectionist SW-ELM was proposed [160]. Ensemble strategy was generalized on two rapid learning connectionist networks i.e., Extreme Learning Machine (ELM) and Leaky-Echo State Network (Leaky-ESN). This approach was applied on two PEMFC stacks which had life spans of 1150 and 1750 hours and prognostics predictions were initiated around their half-life period. Due to the limited learning data, all three connectionist algorithms did not perform well until the constraints were used. However predictions from SW-ELM were closer to the actual value than the predictions from ELM and Leaky-ESN [160]. Another Wavelet based approach namely Discrete Wavelet Transformation (DWT) was proposed for real time prognostics of PEMFC [161].

As fuel cells are complex electrochemical devices and multiphysics systems, to drive a model from the first principle might be a difficult task. However, some research work has been carried out in order to model the PEMFC for the purpose of controlling and capturing their dynamic behaviour [111, 112, 162, 163, 164].

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⁶³⁰ Other researches were also reported on fuel cell modeling for the purpose of prognostics and health monitoring of PEMFC to capture the fault based on the water flooding inside the fuel cell [143] and electrochemical surface area and membrane gas crossover [104].

4. Conclusions

⁶³⁵ An overview of the current state-of-the-art on the fields of health monitoring and prognostics on PEMFC systems was presented in this paper.

It was found that, although some important research works have already been reported on prognostics and estimation of the RUL of PEMFC stack systems, this field of knowledge is still at its early stage and needs a further significant development.

One of the main problems today is related to the lack of test and failure data available. Hence, FMMEA of PEMFC is a subject that is not completely understood yet. For example, in the case of the transportation industry, vibrations and air pollution are known to speed up the degradation of the fuel cell, often resulting in unexpected failures. However, how these parameters correlate with the damages are not exactly known yet.

It is also not clear which failure mechanisms cause most failures in the PEMFC in general and which failure mechanisms are frequent in particular application. These information can be useful because development of the prognostics strategies could be prioritized based upon frequent failure mechanism for a particular application or system.

FMMEA of PEMFC is essential for the design and optimisation of fuel cell systems, as it can improve performance from the early introduction of mitigation mechanisms from known failure modes. Accelerated life tests (ALT) can be used to better understand failure mechanisms by accelerating only the parameter that needs to be studied. These parameters include vibration, temperature, flooding, CO poisoning, but there may be others. However, ALTs can be costly since these are destructive tests that must be carried out in a technology that

is still considerably expensive.

It is clear that this is an area that will receive much growing attention within the following years, so there is an opportunity for researchers to develop this technology further and collaborate with industry partners to improve the overall life expectancy, efficiency and maintainability of PEMFC systems.

With the better understanding of FMMEA, failure models can then be further developed and integrated into a global model in order to achieve more reliable prognostics' forecasts. Control system strategies for PEMFC can then be based on this global model in order to improve the life expectancy, efficiency and maintainability of PEMFC.

Collaboration between researchers, designers and manufacturers is therefore paramount for information and data to be shared with respect to health monitoring and prognostics. Thus, the work presented herein are a contribution to the understanding that research and development on health monitoring and prognostics on PEMFC stack systems are vital so that emission free PEMFC technology can become a real tangible alternative for energy generation in the nearby future.

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