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PEMFC for aeronautic applications: A review on the durability aspects

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Abstract: Proton exchange membrane fuel cells (PEMFC) not only offer more efficient electrical energy conversion, relative to on-ground/backup turbines but generate by-products useful in aircraft such as heat for ice prevention, deoxygenated air for fire retardation and drinkable water for use on-board. Consequently, several projects (e.g. DLR-H2 Antares and RAPID2000) have successfully tested PEMFC-powered auxiliary unit (APU) for manned/unmanned aircraft. Despite the progress from flying PEMFC-powered small aircraft with 20 kW power output as high as 1 000 m at 100 km/h to 33 kW at 2 558 m, 176 km/h [1–3], durability and reliability remain key challenges. This review reports on the inadequate understanding of behaviour of PEMFC under aeronautic conditions and the lack of predictive methods conducive for aircraft that provide real-time information on the State of Health of PEMFCs.

Highlights: The main research findings are

– To minimize performance loss due to high altitude and inclination by adjusting cathode stoichiometric ratio.
– To improve quality of oxygen-depleted air by controlling operating temperature and stoichiometric ratio.
– Need to devise real time prediction methods conducive for determining PEMFC SoH in aircraft.

Keywords: Proton exchange membrane fuel cells, Auxiliary power units, Aircraft, Multifunctional fuel cells, Durability, State of Health

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1 Introduction

Depleting fossil fuels, global warming and new regulatory frameworks (e.g. health and environmental restriction laws) are drawbacks of the conventional energy supply system that necessitate implementation of new innovative solutions. Technology advances towards "more electric" devices and systems increase demand for on-board electrical power. For instance, smartphones, automated healthcare robotics, electrically assisted steering wheel, electric air conditioning and electric hand break in automobiles require predominantly more electrical energy as the power source to operate when compared to traditional technologies. "More electric" in the larger context of complex systems mean replacing pneumatic or hydraulic systems with lighter and more efficient electric systems. All these advancements increase the requirements for on-board electrical power generation. Aeronautics industry is no exception to the move towards "more electric". For example, Boeing 787 Dreamliner generates 1.5 MW which is almost an order of magnitude higher compared to the traditional airliner [4]. Hence the need to find alternative energy sources which not only generate electrical power but also address health and environmental requirements.

Proton exchange membrane fuel cells (PEMFC) exhibit properties suitable for aeronautic applications such as silent operation, emission of potentially useful by-products, solid electrolyte and no moving parts [5, 6]. The silent operation of fuel cells reduce noise in airports and alleviate hearing problems of airport personnel who are inadvertently exposed to high noise levels. Water and deoxygenated air can be used to generate water on-board and retard fire retardation on jet fuel tank respectively [7, 8]. Heat generated can be used for hot water and ice prevention in freezing conditions [7]. Consequently, PEMFC is studied and marketed as multifunctional fuel cell (MFC) in order to appeal to a wider market [9–11].

Given the advantages of PEMFC, NASA proposed fuel cell-powered propulsion in attempts to discover "21st Century" aircraft that are affordable, safe, environmentally compatible and silent [5]. In 2003, one of the major aircraft manufacturers Boeing bought into the idea of "more-electric" commercial aircraft [12]. Boeing proposed change of the 7E7/787 pneumatically-powered environmental control system (ECS), and hydraulically-powered flight control and cargo doors system into electrically-powered [13]. Airbus also envisaged switching hydraulically-powered secondary flight control of A380 ATRA (Advanced Testing and Research Aircraft) to electro-mechanical actuators [13]. Airbus further collaborated with DLR (Deutsches Zentrum für Luft- und Raumfahrt, German Aerospace Center) to establish a fuel cell-powered motorized glider (DLR Antares-H2) as a flying test laboratory [6]. Nowadays, research has shifted to testing the behaviour of PEMFC under field and simulated laboratory conditions [1, 2, 11, 14, 15].

This review presents an overview of progress made in establishing the application of PEMFC in commercial aircraft. The review focuses on highlighting successful field testing, challenges and required stack-level testing to validate feasibility of PEMFC-powered APU for commercial aircraft. Therefore, the aim of this review is to highlight work done and identify key factors that will assist in fast tracking industrialization and commercialization of PEMFC in the aeronautics industry. The paper is structured in a manner that allows easy flow of discussion from the milestones achieved in the 21st century, Multifunctional fuel cell (MFC) with operating conditions, stack-level testing, factors influencing fuel cell with respect to degradation, predictive methods to PEMFC State of Health.

1.1 Research milestones in the 21st century

Table 1 gives the timeline of research milestones for PEMFC-powered aircraft. The timeline is limited to manned aircraft, mostly focussing on PEMFC system testing, stack testing and marketing. The table seeks to acknowledge progress made in the field and assist in identifying future prospects that contribute towards commercialization of fuel cells.

1.2 Multifunctional PEMFC for aeronautic applications

One of the primary objective of "more-electric" aircraft is to replace auxiliary power units (APUs) powered by gas turbine generators attached to main engine propulsion with PEMFC-powered APU [13]. APU are used to generate electricity during ground operations in order to power electrical loads such as lighting, cabin environmental conditioning and main engine start-up. PEMFC-powered APU generates electrical energy through electrochemical processes which subsequently reduce the load on main engine and consumption of jet fuel [12]. It is well recognized that PEMFC cannot meet power demands for the whole aircraft, hence it is considered as a secondary power sources or sometimes hybrid with batteries [1, 27, 28]. In addition to the APU application, potential PEMFC capabilities on board are recharging batteries, generating electrical power and heat, producing drinkable water, replacing Ram Air Turbine and supplying deoxygenated air for inerting [7, 8, 20, 29]. Generating water, heat and inert gas could reduce take-off weight and thus save jet fuel. Use of water produced on-board rather than collected from different airport locations can enhance confidence on its quality. The electrical power generated on board by PEMFC can be used to power moving ailerons, nose wheel electric motor, brakes, flight control, cabin environmental control and pressurization, and emergency power sources [24]. Figure 1 and Table 2 contains additional electrical system of a more-electric commercial aircraft and their respective energy demand, respectively [13, 24]. At least 700 kW is required to power the additional electrical functions of a more electric aircraft compared to the conventional.

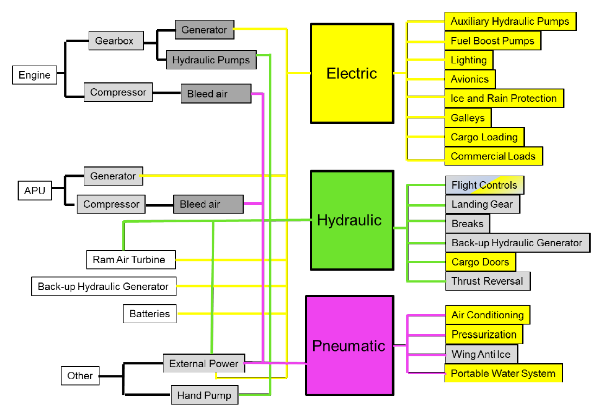

Figure 1: Demonstration of energy supply (left) and future electric systems for a more-electric aircraft

1.3 Operating conditions for aeronautic applications

PEMFC must withstand harsh operating and environmental conditions experienced in aeronautics in order to be fully considered as alternative energy supply devices. Key operating conditions are temperature, stoichiometric ratio, pressure and relative humidity. Environmental conditions are subfreezing or elevated temperatures, low

Table 1: Highlights of PEMFC applications in aircraft

Researchers/Articles (Year)	Milestones
[5] 2001	NASA proposed shift from hydraulics and pneumatics to environmentally-inspired electric power supplied by fuel cells, hydrogen or hybrids.
[12, 13, 16] 2003 - 2005	Boeing evaluated feasibility of SOFC and PEMFC for use in aircrafts' APU. PEMFC was selected as a better candidate for further evaluation due to its level of maturity.
[1] 2008	Boeing flew first 2-seater manned PEMFC/Li-ion batteries hybrid-powered aircraft. PEMFC maximum power output was 24 kW and aircraft reached 1 000 m altitude.
[17, 18] 2008	Developed a vibrating platform as a testing facility to study mechanical behaviour of fuel cells in a vibrating environment linked to aircraft applications. The data acquired was further used to demonstrate that experimental data can be used to simulate three-dimensional model reflective of the behaviour.
[19] 2008	DLR, Airbus and Michelin successfully tested 20 kW PEMFC emergency power system.
[2, 3] 2009, 2013	DLR flew world's first piloted aircraft exclusively powered by 33 kW PEMFC. The Antares DLR-H2 reached 2 558 m altitude.
[6, 7] 2009/10	DLR and Airbus identified potential uses of PEMFC as a multifunctional system for both on ground and cruise. For example, water and heat generation on board, and inerting jet fuel with cathode discharge deoxygenated gases.
[7, 20] 2010	Tested effects of aeronautic environmental effect on performance of PEMFC stack. Tests conducted were temperature, inclination, low pressure/high altitude, vibrations, oxygen content of the cathode exhaust gas and water generation.
[15, 21] 2010 -2013	An EU-funded project ENFICA-FC flew hydrogen Li-ion batteries/fuel cell-powered RAPID200 for the first time. Maximum power output for the PEMFC was 20 kW. The flight reached endurance of 45 minutes with a speed of up to 150 km/h.
[22] 2010	FC LAB and other institutes in France and Denmark designed climatic chamber for subzero testing of PEMFC used in aircraft.
[23] 2011	DLR and Airbus successfully tested A320 ATRA with electric nose wheel powered by 25 kW PEMFC.
[8] 2011	Revealed that water recovered from PEMFC nearly meets all USEPA and WHO drinking water requirements.
[14] 2012	Investigated and modelled feasibility of water generation on-board taking into account hydrogen pressure tanks and reformate systems. The study showed that fuel cells are capable of the functions and hydrogen pressure tanks are more suitable in long-range commercial aircraft.
[24] 2013	Examined feasibility of PEMFC use with existing Boeing-like aircraft technology and its effect on aircraft performance. Research findings were that PEMFC is compatible with existing technology but power-weight ratio negates performance. The heaviest hardware being fuel cell and hydrogen storage.
[25] 2014	Analysed challenges and policy issues delaying commercialization of fuel cells and hydrogen energy.
[26] 2014	German Aerospace Center, Institute of Engineering Thermodynamics designed and assembled low pressure test facility. In this study, low pressure of 700 mbar is equivalent to 2 400 m altitude. The test facility was used to estimate optimal operating conditions of fuel cell under ambient- and low-pressure conditions in terms of cell humidification. In addition, the study showed that low pressure decreased cell performance and efficiency.

Table 1: (Continuation) Highlights of PEMFC applications in aircraft

[27] 2015	Studied hybrid of PEMFC and batteries looking at layout and connections. The formulated dynamic model showed that direct connection between batteries and PEMFC without inverters is feasible.
[11] 2015	Studied effect of operating parameters on relative humidity of cathode exhaust gas, oxygen depleted gas used for inerting jet fuel. Comparison between single and twin system architecture with particular attention on pressure, stoichiometric ratio and stack temperature of 40 – 60^{o}C led to future outlook on water management and self-humidification of the stack for operating temperature outside the manufacturer's recommendation of 40 to 60°C (possibly 20 to 90°C).
[10] 2015	Suggested that fuel cells should be marketed as a solution to many problems rather than energy providers. Marketing fuel cells as multifunctional focusing on its advantages will appeal to a wider market and subsequently fasten commercialization.

Table 2: Additional electrical systems with their respective energy demand for a more-electric aircraft

Main electrical consumer	Previous form of energy supply	System electrical power demand (kW)	Phase of flight
Flight control	Hydraulic	80	All
Landing gear	Hydraulic	25	Descending and landing
Engine starter	Partly pneumatic	350	Initial taxi, take-off/climb
Wing anti ice	Pneumatic	200	All
Environmental control and system	Pneumatic	400	All
In-flight entertainment	Pneumatic	20	All
Total additional in-flight demand		**700 kW**	

pressure, vibrations and inclinations. Aeronautic studies mostly focus on environmental effects at system and stack level rather than operating conditions [20, 22, 30]. One of the reasons could be that it would be easier to study and define operating conditions for aeronautic applications after broadening the understanding of environmental effects on PEMFC performance and lifetime. Research on operating conditions is predominantly limited to stationary and terrestrial conditions [31–39]. Information obtained on effect of aeronautic conditions on PEMFC will assist in defining the optimum operating conditions. Experimental testing and modelling work currently conducted by Airbus and DLR researchers seeks to optimize management of discharge water, heat and cathode exhaust gas of a MFC [9, 11, 14].

1.4 PEMFC system testing under aeronautic conditions

Studies carried out since 2007 are mostly directed towards understanding behaviour of PEMFC at system level. Significant use of fuel cells in propulsion for manned aircraft has been mostly demonstrated by three projects: Boeing, Airbus with German Aerospace Center (DLR) and Environmentally Friendly Inter City Aircraft powered by Fuel cell (ENFICA-FC) [1, 2, 15]. The first published work on PEMFC-powered manned aircraft flown by Boeing was in 2007 [1]. A 20 kW PEMFC in hybrid with Li-ion battery was used to power a 2-seater manned aircraft. PEMFC provided half the 36 kW power demand for take-off and the full 15 kW required for cruise [1]. Airbus, DLR and Michelin in 2008 successfully used 25 kW PEMFC on a testbed A320 ATRA (Advanced Technology Research Aircraft) as emergency power system [19]. DLR in 2009 tested a piloted Antares DLR-H2 aircraft exclusively powered by approximately 33 kW PEMFC. The flight test was a success reaching altitudes up to 2 558 m without any significant

PEMFC performance loss [2]. ENFICA-FC flew first European Commission-funded fuel cell-powered aircraft. The aircraft was powered by PEMFC/Li-ion batteries hybrid with fuel cell maximum power output of 20 kW [21]. In 2011, Airbus and DLR successfully demonstrated electric nose wheel of A320 ATRA powered by 25 kW PEMFC [23]. The success of the tests proved capability of PEMFC to perform in aeronautic conditions. Hence the shift towards better understanding of PEMFC performance and durability at stack level under aeronautic conditions.

1.5 PEMFC stack testing under aeronautic conditions

Laboratory tests identified to explore environmental effects on PEMFC are orientation/inclination, high altitude/low pressure, vibrations, subfreezing and elevated temperatures. Feasibility studies of MFC focus on produced water, heat and cathode exhaust gas management. For instance, tests carried out by [17] to study mechanical behaviour of fuel cell on a vibrating platform simulating aircraft conditions showed no significant damage on the fuel cell. The damage was estimated based on leak tests which showed no notable pressure changes [17]. The tests were conducted on a short stack which exhibited "elastic" properties. The "elastic" stack resembles non-linear multi-body behaviour that could decrease with increase in size of the stack [18]. Therefore rigidity and balance-of-plant fittings could become an issue for larger stack [17]. On the other hand, orientation and inclination tests showed voltage decrease at an angle of 30° when operated at a low air stoichiometry of 1.6 [7]. Contrarily the low stoichiometry of 1.6 yielded better cathode exhaust gas quality with only 10% oxygen content [7].

High altitude/low pressure effects on fuel cells were studied under simulated conditions or within a system of small manned aircraft [26, 30, 40]. Relationship between pressure and altitude is defined as approximately 200, 2 000, 3 000 and 12 000 m above sea level and are equivalent to 1 to 0.9, 0.75, 0.7 and 0.2 bar respectively [7, 11]. Altitude tests showed notable performance loss at 0.7 bar/2200 m altitude [20, 26, 30]. Further testing showed that the loss can be minimized by increasing air stoichiometry to 2.5 for altitudes higher than 2 000 m [30]. Taking into account the effect of higher air stoichiometry on fuel cell durability, Hordé *et al.* recommended the use of fuel cells in pressurized conditions but bearing in mind the power required for compressors [30].

Effect of subfreezing temperature on cell performance and its ability to self-start under different simulated operating scenarios representative of aircraft applications was studied using a climatic test chamber designed by [22]. The scenarios were short reference tests (ambient temperature), low temperature tests (−34 to 15°C), ground survival tests (−40 to 20°C) and operating low temperature tests (−9 to −6°C). PEMFC generated enough heat (stack operating temperature of 15°C) for self-heating at a surrounding temperature down to −34°C in order to maintain required performance. Nevertheless, the stack failed at operating and surrounding temperatures of −9°C and −1°C respectively due membrane perforation caused by insufficient drying prior to freezing [22]. Low temperature PEMFC subjected to temperatures up to 120°C was customized and heat-transfer oils were used as coolants for the fuel cell to withstand the conditions [41].

1.6 Factors influencing PEMFC life under aeronautic conditions

Fuel cell life is influenced by operating conditions, material and configuration of components, impurities in the reactants, external environment and surrounding conditions within the system. "First fuel cell law" states that one cannot change one parameter in a fuel cell; change of one parameter causes a change in at least two other parameters, and at least one of them has an opposite effect of the one expected to be seen [42]. As a result, it is often difficult to single out the effect of one parameter as they occur either concurrently or simultaneously. Nevertheless studies are conducted to better understand factors that influence fuel cell life, predominantly for stationary and automotive applications rather than for aeronautics [11, 31, 36, 43–46]. Hence, factors discussed in this paper are not necessarily representative of the aeronautic environment but taken as a reference in accordance with the purpose of this review. Common effects of the factors are poor water management, thermal management, fuel and oxidant starvation, catalyst corrosion, contamination and undesired chemical reactions of cell components [36, 43, 47, 48].

Water management is the most studied phenomenon (i.e. flooding and dehydration) since ion conductivity of the proton-conducting membrane for a low temperature PEMFC (LT-PEMFC) strongly depends on water content [49]. Flooding occurs when the rate of water generation exceeds the rate of water removal and the opposite for dehydration. Flooding often occurs at cathode side where water is generated by oxygen reduction reaction and electro-osmotic drag (see Figure 2). Desired water content is defined as enough water to facilitate transport of protons from anode to cathode without flooding the MEA, approximately 90 to 110% relative humidity at the outlet of cathode [37, 50, 51]. The water movement within LT-PEMFC shown in Figure 2 advocated effect of water content and transport on other fuel cell functions (such as reactant gases supplies) [49].

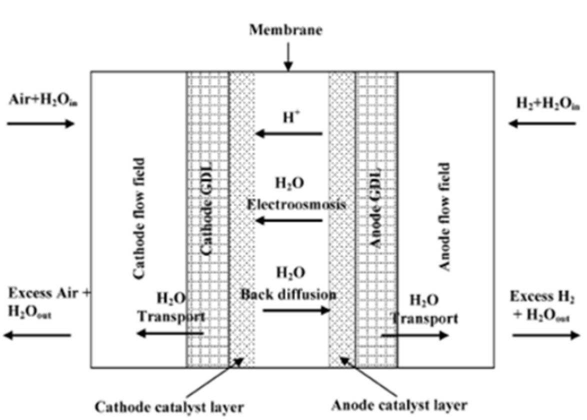

Figure 2: Water movement within a PEMFC.

Fuel/air starvation occurs when the fuel cell is operated at sub-stoichiometric reaction conditions. Starvation results from a sudden change in demand of reactant gases during load cycling or water accumulation [36]. Starvation causes high potential which in turn facilitates cell reversal. Cell reversal occurs when cathode potential decreases and overall potential becomes negative due to formation of oxygen and hydrogen at the anode and cathode respectively. For instance, Figure 3 shows platinum activity reduced by cell reversal as a result of fuel starvation after only 1 second of operation. 28% loss of ECSA was observed after 3 minutes of operation [52]. Furthermore, platinum particle size distribution increased from an average of 2.64 to 4.95 nm after 10 minutes of fuel starvation due to platinum sintering caused by cell reversal [52].

In the case of air starvation, ECSA decreased by 40% while platinum particle size distribution increased from 2 to more than 4 nm after 120 minutes [53]. There were no notable changes observed on the anode side, suggesting that hydrogen oxidation is not affected by air starvation at low electrode potential [53]. In addition, oxygen formed from water electrolysis reaction at the anode during hydrogen starvation can react with carbon to form carbon dioxide, a serious poison to the platinum catalyst [54]. The heat generated from the electrolysis reaction can create hotspots on the membrane that lead to breakthrough and cracks. Possible links between the phenomena are shown in Figure 4 demonstrating complexity that can rise from a change of one factor.

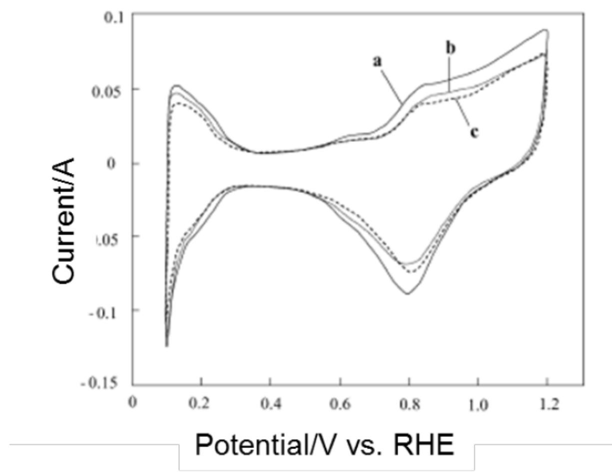

Figure 3: Cyclic voltammograms for cathode during cell reversal caused by fuel starvation: a) before, b) after 1 s and c) after 3 mins.

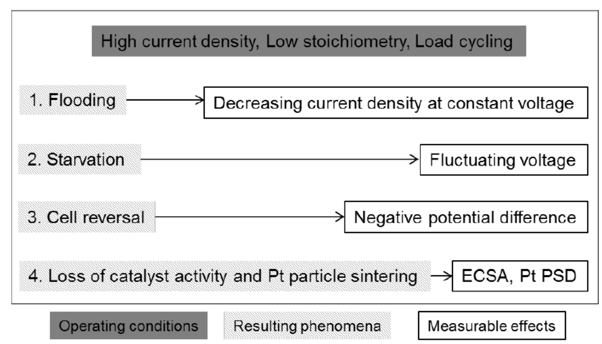

Figure 4: Demonstration of the link between various phenomena that contributes to performance loss and respective indicators

1.6.1 Temperature and relative humidity

Nominal operating range for low temperature PEMFC is within the range of 60 to 80°C [50]. PEMFC used for aeronautic applications is likely to be exposed to environmental/surrounding temperatures that range from subzero to elevated temperatures up to 90°C [11, 22]. Theoretically, high temperature improves fuel cell performance by increasing reaction kinetics while lowering activation losses and mass transport limitations [50]. Amirinejad *et al.* used a humidified single cell (150 to 200% relative humidity, RH) to study the effect of operating temperature (50 to 80°C) on fuel cell performance [31]. Combination of high temperature and high current density yielded better performance while the opposite occurred at low current density regions. The reason for the improved performance was that high temperature increases diffusivity and conductivity while decreasing mass transport resistance. At high temperature and high current density, the fuel cell produced enough water at the cathode side to humidify anode side by back-diffusion mechanism. Amirinejad *et al.* also showed that fuel cell performance is mostly affected by anode humidification compared to cathode humidification. The study conducted by Wasterlain *et al.* to explore combinations of optimal operating parameters (including temperatures of 40 and 60°C) showed that high temperature can either improve fuel cell performance or induce material degradation. Similarly to Amirinejad *et al.*, Wasterlain *et al.* noted that fuel cell performance depends on the ability of the membrane to keep enough water and provide adequate conductivity (see Figure 5). Consequently, performance improvement at higher temperatures of 60°C observed by Wasterlain *et al.* was minimal due to low fuel cell relative humidity of 11.7% and 37% for the anode and cathode respectively [44]. Moreover, fuel cell with 8-segmented cells operated at a constant temperature of 50°C and changing humidity (40, 60, 70 %RH) showed higher performance for 60 and 70 %RH. The fuel cells operated at 40 %RH had higher performance downstream due to humidification provided by water movement within the cells [55].

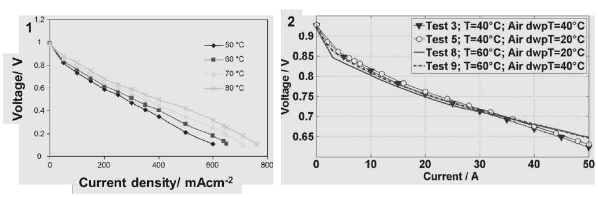

Figure 5: Effect of temperature on FC performance 1. 100 %RH, 1.5:2 Sa:Sc [31] and 2. 2:3 Sa:Sc for T = 40°C and 2:5 Sa:Sc for T = 60°C [44].

On the other hand, performance loss due to low humidity can be recovered when detected quickly. Voltage of fuel cells operated at low humidity of 37 and 77 %RH for 30 minutes dropped from 0.65 V to less than 0.5 V and 0.6 V respectively. Both cells fully recovered the voltage of 0.65 V after being re-humidified for 15 minutes [54]. Similarly, current drop on fuel cells operated at 60 and 70 %RH was recovered to even higher current output after 20 and 5 minutes respectively [55]. The drop in current was associated with possible flooding due to water moving downstream during load cycling [55]. A stack operated at a temperature range of 80 to 120°C with varying humidity of 60 and 15 %RH showed performance loss at low current density due to insufficient water generation. However, the performance loss was recovered after the stack was re-hydrated through cooling down [41].

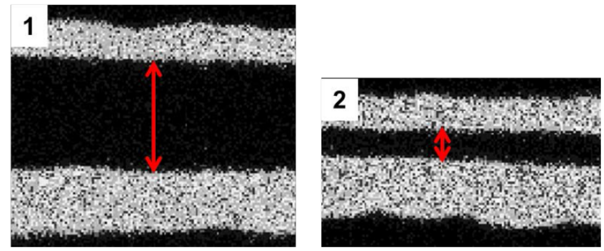

Figure 6: Electron probe images of Pt X-Ray: a) new MEA and b) MEA operated for 1 000 hours with significantly thin membrane layer (showed by the arrow) [59]. Both micrographs are to the same scale.

In addition to the effect of temperature on overall fuel cell performance, some studies examined the effect on individual components such as membrane, electrode and gas diffusion layer (GDL) [35, 46, 56, 56–60]. For instance, high temperature at low relative humidity causes membrane drying whereas water residues formed during subfreezing temperature block GDL surface. Healy *et al.* observed membrane degradation after operating a cell at 95°C with 75/50% anode/cathode relative humidity. Membrane thickness in Figure 6 was more than 4 orders of magnitude thinner after 1 000 hours of operation. Rate of fluoride release was 2 times more compared to a cell operated under mild conditions of 60°C and 100/100% anode/cathode relative humidity [59]. Le Canut *et al.* observed that flooding can have two effects on fuel cell components. The effects occur either inside GDL and flow channels or within pores of the catalyst layer thereby blocking pathways for gases and catalytic reactive site respectively [61]. High temperature coupled with high relative humidity can cause electrolyte degradation. Wasterlain *et al.* observed increased gas crossover due to operating the fuel cell at high temperature (60°C) and low relative humidity (36%RH) [44].

In the context of MFC for aircraft, Keim *et al.* and Werner *et al.* studied the effect of temperature on electrical power supply, oxygen content of cathode discharge gas (oxygen depleted gas, ODA) and produced water [9, 11]. Keim *et al.* explored the effect of temperature on dryness of ODA when using a honeycomb silica structure that facilitates drying [9]. ODA must be dry with at most 2 g(H_2O)kg^{-1}(ODA) specific humidity in order to avoid contaminating jet fuel [9]. The drying process within the honeycomb silica structure involves: 1. drying (ODA is dried by the silica structure to the required specific humidity, 2. regeneration (silica structure is regenerated by cooling to a set temperature) and 3. resting (the silica structure is allowed to rest while other silica structure units are active. Temperatures investigated were condensation, T_{cond}

(6.6 to 19.8°C) and regeneration, T_{reg} (94 to 113°C). T_{cond} is the temperature required to cool/condense ODA in order to separate air from water since water can damage silica gel. T_{reg} is the temperature required to cool down/regenerate the silica drying unit since hot silica structure does not create the necessary vapour pressure gradient for adsorption to take place and subsequent drying of ODA. Specific humidity of ODA increased with an increase in T_{cond}, resulting in failure to achieve 2 gkg^{-1} at T_{cond} equals to 19.8°C. The contrary was observed for T_{reg}, in which specific humidity decreased with an increase in T_{reg} and all tests achieved less than 2 gkg^{-1}specific humidity. Provisionally accepted operating temperatures to achieve less than 2 gkg^{-1} specific humidity of ODA are 14.9°C T_{cond} and 94°C T_{reg}. The temperatures were chosen based on the fact that highest T_{cond} and lowest T_{reg} require less energy. However, authors recommended further research on lower T_{reg} [9].

Werner *et al.* experimentally evaluated and modelled the impact of stack temperature on water management and oxygen content of cathode exhaust gas from single and twin fuel cell systems [11]. A single system consisted of one fuel cell stack whereas the twin system has two stacks with both cathodes connected in series, a separator and a condenser. The second fuel cell was supplied by conditioned exhaust air of the first fuel cell. The separator and condenser conditioned the exhaust air from the first cell for the second fuel cell. Manufacturer's recommended operating temperature range of 40 to 60°C was taken into account and stack was operated between 45 and 65°C. Relative humidity of cathode exhaust gas, taken as reflection of membrane humidity, within the range of 90 to 110% reflects high-performing and steady operating fuel cell stack [50, 51]. Werner *et al.* revealed that cooling temperatures of 45°C at 700 mbar and 55°C at 950 mbar can yield 108 and 98% relative humidity respectively for the single fuel cell system. It was not feasible to operate the single fuel cell system at less than 60°C and 2 000 mbar due to flooding. The twin fuel cell system was then used to examine feasibility of 2 000 mbar. Dried inlet air was used in order to maintain temperature at less than 60°C. The desired oxygen content of less than 12% could be achieved at higher stoichiometry. Hence Werner and others recommended exploring the effect of operating temperature outside the manufacturer's range, namely 20 to 90°C. Ruiu *et al.* examined performance of operating a stack up to 120°C but it was customized for high temperatures and heat-transfer oils were used for cooling at temperatures above 90°C instead of the conventional deionized water [41].

1.6.2 Stoichiometric ratio

Stoichiometric ratio is key in ensuring that sufficient reactant gases are supplied to fuel cell reactive sites in order to avoid detrimental effects caused by starvation. Commonly used stoichiometric ratio is 1.2:2 anode to cathode [62]. The choice of the ratio is probably based on stoichiometric factor of the overall PEMFC chemical reaction (see Eq. 1) and ratio of hydrogen to oxygen when air is used.

$$2H_2 + O_2 \rightarrow 2H_2O + \textit{electrical energy} \quad (1)$$

However demand for reactant gases varies with load and water content of the cell [63]. As a result, several studies are conducted to better understand the effect of stoichiometric ratio on fuel cell performance [30, 44, 45, 62, 64, 65]. Both studies by Kim *et al.*, and Candusso *et al.* reported that anode and cathode stoichiometric ratio had no significant effect on fuel cell performance [62, 66]. One of the reasons could be the limitation of analytical techniques since both studies only monitored current and voltage. Wasterlain *et al.* observed that increasing anode stoichiometry has no significant effect at 60°C as it yielded better water drainage. On the other hand, minor performance losses revealed by the polarization curve with 20 mV loss in the high current region at 40°C were due to excessive drying. The drying induced by high anode stoichiometry reduced ECSA but had no notable effect on membrane permeability and its mechanical structure. Contrarily, increased cathode stoichiometry resulted in better performance as long as there was sufficient humidification [44]. Figure 7 shows that cathode stoichiometry has no significant effect on resistance, except for mass transport [67]. The observation was attributed to the fact that only oxygen transport was affected since the membrane was hydrated at 80 %RH for 1 hour. Furthermore, cathode stoichiometry has a positive effect on fuel cell performance unless it is below 1.6 [45]. Similarly to Harms *et al., Figure 8* shows that a significant voltage drop was observed at cathode stoichiometry of 1.5 [65].

Considering the effect of pressure/altitude in aeronautic applications, Hordé *et al.* evaluated the effect of cathode stoichiometry from 1.5 to 2.5. The desired performance was achieved at cathode stoichiometry of 1.75 and 2.5 for 1200 and 2200 m altitude respectively [30]. However implications such as membrane drying and power consumption by compressors are concerns when increasing stoichiometry to more than 2.5 as required for higher altitude [30]. For MFC, cathode stoichiometric ratio of 2.5 yielded 13.8% oxygen content of ODA [11]. Cathode stoichiometry of 1.9 yielded the desired ODA with less than 12% oxygen content from single fuel cell system, although

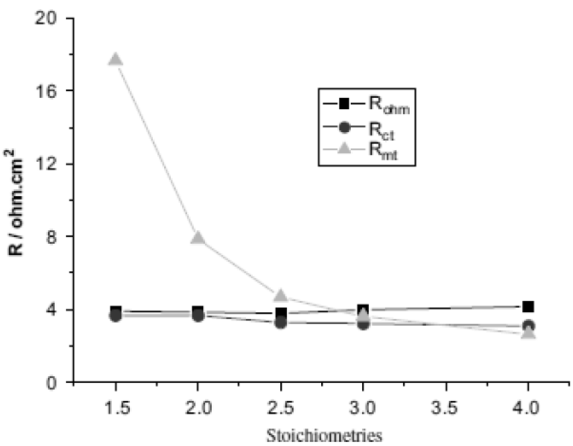

Figure 7: Effect of cathode stoichiometry on ohmic (RΩ), charge transfer (Rct) and mass transport (Rmt) resistances at anode stoichiometry of 1.5 and current density of 500 mAcm^{-2}

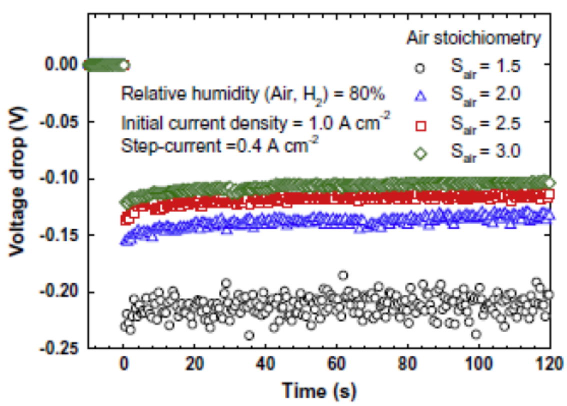

Figure 8: Transient response of cell voltage under load change at different cathode stoichiometry

it was below the manufacturer's recommended operating conditions. Contrarily, higher cathode stoichiometry of 3.6 and 2.8 for stack 1 and stack 2 from twin fuel cell system yielded oxygen content less than 12% [11]. Hence authors recommended further research on cathode stoichiometry that can yield the desired oxygen content (less than 12%) within the recommendations in order to minimize damage to the stack.

1.6.3 Load cycling

Load cycling (mostly between 0.6 and 1.0 V) results from change in power demand caused by driving cycles such as start-up, shutdown, idling and high voltage operation. Load cycling affects the water content of fuel cells and the demand for reactant gases, which ultimately affect the performance and lifetime [63, 68, 69]. More than 50% and 30% of degradation in vehicular applications is caused by load cycling and start-stop respectively [63]. Effect of load cycling is evident on individual cell components. For example, catalyst corrosion observed at voltages between 0.6 V and 0.9 V caused formation of platinum oxide which resulted in blocked catalyst surface [70, 71]. Fuel starvation attributed to load cycling caused permanent damage to the electrocatalyst, namely morphology transformation, carbon corrosion, reduced ECSA and Pt particles agglomeration as revealed by respective analytical techniques [69, 72]. Furthermore, Bose *et al.* observed change in chemical composition of MEA after 480 h of accelerated load cycling [69]. The significant increase in low frequency resistance and decrease in ECSA after 200 cycles confirms the degrading effect of load cycling as shown in Figure 9 [72]. It is worth noting that degradation caused by load cycling occurs faster at low voltage with 1.7 mAcm^{-2}cycle^{-1} and 0.21 mAcm^{-2}cycle^{-1} at 0.6 V and 0.8 V respectively [72]. The observations are attributed to high current density at low voltage which results in higher reactant gases demand and water generation. Consequently, higher water content blocks the gas diffusion layer and catalyst layer and thereby limiting transport of gases to reactive sites [73]. Relevance of the operating conditions is subject to the load profile of commercial aircraft. For instance, series of operation of aircraft are ground operation and loading, engine start, take-off and climb, cruise, and landing. All stages take approximately 30 minutes except cruise, which depends on the route. It is therefore necessary to first determine the load profile for a PEMFC-powered APU for commercial aircraft.

1.7 PEMFC life prediction methods under aeronautic conditions

Complex reactions taking place either concurrently within a fuel cell (i.e. chemical, electrical, mechanical and thermal) making it difficult to single out causes of failure, performance loss or shortened lifetime. Despite the complexities, methods are developed to predict Fuel Cell State of Health (SoH) in terms of performance loss, degradation and, fault detection and isolation (FDI) [45, 63, 74–79]. Diagnostic methods available are model or non-model based [71, 80–84]. Model-based methods are further classified as white, grey or black box depending on the nature of input and output. The Black box model is most suitable for PEMFC since it is directly derived from experiments, requires little computational effort and, capable of on-line

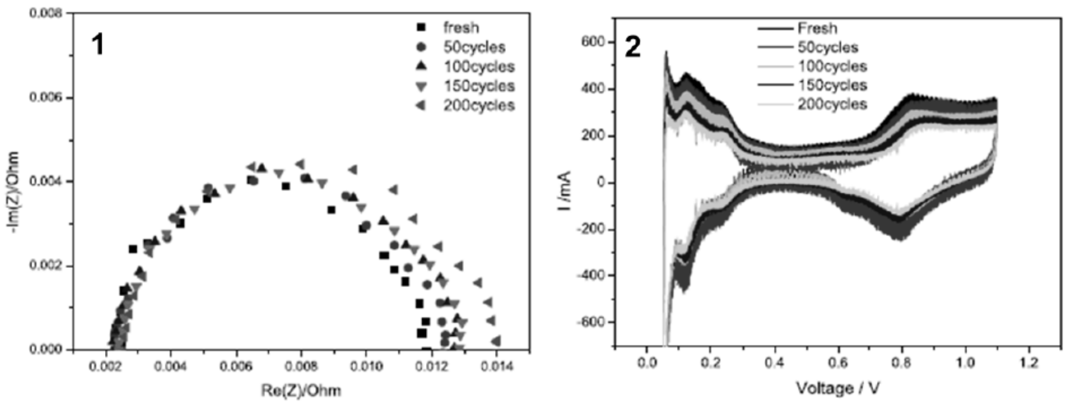

Figure 9: Electrochemical Impedance Spectrometry (1) and cyclic voltammograms (2) of fresh and MEA subjected to load cycling

monitoring, detection and diagnostic applications [81]. In cases of very short transient periods, model-based methods such as statistics and knowledge of physical or electrical phenomena become useful. Model-based methods can be used as a control strategy, although relevance and reliability of the methods depends on developments of dynamic ageing models [85].

On the other hand, non-model based methods are simple, flexible, capable of dealing with nonlinear problems and do not require system structure knowledge. Non-model based methods are further grouped as artificial intelligence, statistical method or signal processing [82]. An emerging area of science called Prognostics and Health Management (PHM) focuses on methods that assess State of Health (SoH), predict Remaining Useful Lifetime (RUL) and decide mission achievement from mitigation actions [80, 85]. PHM can be classified as a signal processing method since it involves active data acquisition to identify and isolate faults (fault diagnosis) that accelerate ageing of a fuel cell. The fault diagnosis methods involve four steps: data pre-processing, feature extraction, feature reduction and fault classification. One major drawback of the non-model based method is generating datasets for targeted fault conditions, which can be time-consuming in cases of multiple faults [82]. Dataset generation for data pre-processing includes acquiring data from different sensors and electrochemical characterization techniques such as Polarization Curve (IV curve), Electrochemical Impedance Spectroscopy (EIS), Cyclic Voltammetry (CV) and Linear Square Voltammetry (LSV). A thorough review on the electrochemical techniques is published by [86]. CV and LSV are auxiliary techniques that provide useful information on catalyst activity and membrane health (crossover) respectively. Main drawbacks of CV and LSV are that current cannot be drawn during measurements and the quality of data is affected by non-uniform cell voltage distribution within large stacks [77, 87, 88].

1.7.1 Polarization curve

Polarization curves are the most commonly used electrochemical method to characterise fuel cells. Polarization curves provide information on overall performance loss without differentiating various sources of the loss. For instance, the polarization curves on Figure 10 shows no significant difference between dry and flooded cells and it is almost impossible to identify causes of voltage drop observed on the dry and the flooded cells [33]. Despite the limitation, the polarization curve is useful in estimating overall degradation rate [32, 54, 89]. Degradation rate is a rate at which voltage decay over time and common limits for fuel cells operated under nominal conditions are between 2 and 10 μVh^{-1}, although 60 μVh^{-1} has also been reported [54]. Bezmalinovic *et al.* proposed the use of a polarization change curve as a degradation diagnostic tool. A polarization change curve is a plot of the difference between actual cell potential and potential at the beginning of life for the entire spectrum of current densities. Polarization change curve gives the linear relationship between current density and cell voltage, in which activation and resistive losses can be easily identified. The information obtained from the linear relationship allows the estimation of the electrochemically active surface area and instant prediction of electrocatalyst state of health [79]. The method however needs to be tested for reproducibility under different operating conditions and to be validated.

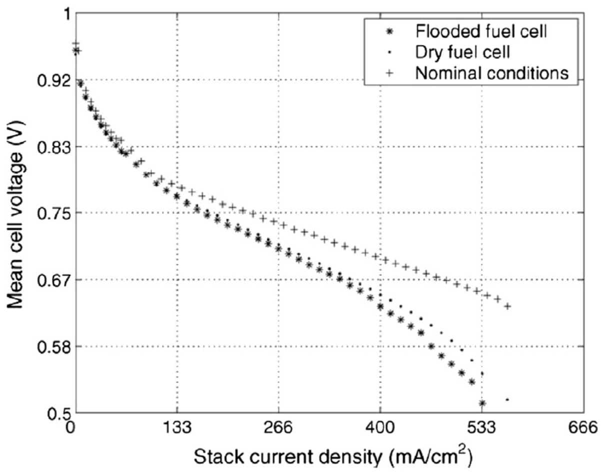

Figure 10: Measured polarization curve for a stack operating at normal, drying and flooding conditions

1.7.2 Electrochemical Impedance Spectroscopy

Electrochemical Impedance Spectroscopy (EIS) coupled with models compliments the polarization curve by differentiating the various losses [33, 44, 84, 90]. EIS data is presented as a Nyquist plot with equivalent circuit models (ECM) to ease interpretation and estimate electrochemical parameters such as resistances, time constants and capacitance [90–94]. The electrochemical parameters are reflective of state of health of fuel cell and its components such as drying/flooding of ionomer membrane and catalyst layer. Ohmic resistance (R_{Ω}) increases with dehydration which causes local hot spots on the catalyst surface and corrosion of carbon support [95]. Flooding at the cathode reduces the active surface area (ECSA) available for the reaction and limits oxygen diffusion, which in turn increases charge transfer resistance (R_{ct}) especially at high current densities [33, 96]. Furthermore, double layer capacitance (C_{dl}) is directly proportional to ECSA when the supported catalyst is evenly distributed over MEA surface. Hence the catalyst layer can be represented by C_{dl} and R_{ct} [90, 92].

Water content has a huge impact on low temperature PEMFC performance, hence most studies use drying/flooding as a measure of state of health. For instance, Fouquet *et al.* studied fuel cell state of health based on drying and flooding conditions using EIS and ECMs [33]. The study defined three subspaces related to nominal, drying and flooding conditions by measuring membrane and polarization resistances. Figure 11 shows that the resistances of the normal cell from Fouquet *et al.* and Shan *et al.* are qualitatively similar [33, 72]. Rubio *et al.* further proposed simplified ECM for the nominal, drying and flooding conditions within 1 Hz and 5 kHz frequency domain using an inexpensive and portable device [75, 97]. However, the frequency domain is questionable as to whether it reflects all phenomena occurring within fuel cells. Yuan *et al.* and Rezaei Niya *et al.* presented a thorough review of applications of EIS and equivalent circuit models in characterizing PEMFC [98, 99]. Although EIS can effectively diagnose fuel cell state of health such as cathode flooding, membrane drying and catalyst ECSA, the spectrometer is not ideal for on-board integration. Major drawbacks of EIS in the context of aeronautics are compactness, sensitivity to electrical network stability and operating conditions, and complex data that does not yet provide instant real-time diagnosis. Models can be used to develop a database for instant fault detection and isolation but their diagnostics are limited to tested operating conditions and the available library [37].

1.8 PEMFC State of Health under aeronautic conditions

PEMFC State of Health (SoH) is essential in determining durability, remaining useful life of a fuel cell and reliability. SoH can be monitored by better understanding the behaviour of PEMFC under aeronautic conditions, in terms of identifying factors that cause degradation, performance loss and shortened lifetime. In so doing, measurable parameters directly linked to the factors can be identified. Identified measurable parameters can be used to establish prediction methods and techniques capable of providing real time information on PEMFC state of health without interrupting energy supply. Several reviews have been published on fuel cell reliability, but predominantly limited to terrestrial applications [36, 43, 76, 78, 100]. The reviews on diagnostic tools by Wu *et al.*, Zheng *et al.* and Petrone *et al.* provide a general review of available techniques [81, 82, 86]. The reviews showed that there is no single technique capable of providing real-time diagnostic data, cost-effective, non-intrusive and insensitive to electrical network.

Table 3 presents a summary of the link between various factors as reported in the respective publications. Furthermore, Figure 12 schematically shows the effect of changing a parameter in PEMFC and possible measurable effects. The next logical step is to examine whether the links are applicable to aeronautic conditions and employ the relationship between measurable effects and operating conditions to develop predictive methods suitable for aircraft.

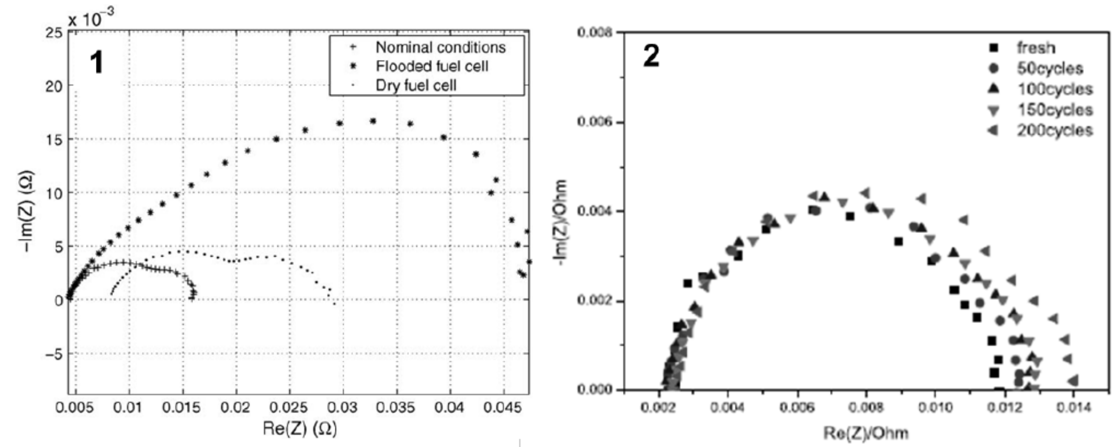

Figure 11: Nyquist plots of normal, flooded and dry cells. Note the similar plot sizes of normal cells

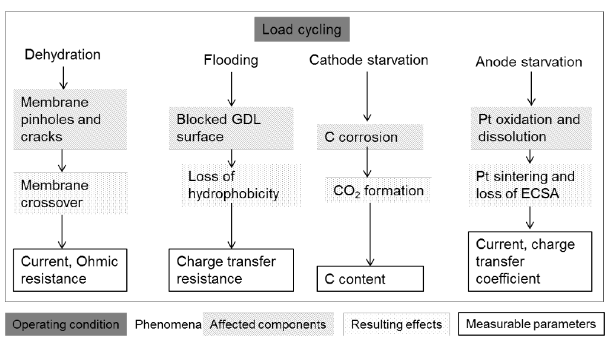

Figure 12: Schematic illustration of possible links between operating conditions and measurable parameters.

1.9 Summary and outlook

Proton exchange membrane fuel cells (PEMFC) directly convert chemical energy into electrical energy through the reaction of hydrogen and air in the presence of a catalyst. The by-products (water, deoxygenated air and heat) have essential uses in aircraft such as fire retardation, ice prevention and supply of drinkable water. Hence PEMFC are tested in the field in small manned aircraft, in auxiliary power units for commercial aircraft and as alternative power source in case of emergency. In addition to field testing, PEMFC are also tested in laboratories under simulated aeronautic conditions such as vibrations, high altitude, subfreezing temperatures and inclination. Hence this review seeks to report on the progress of PEMFC-powered APU for commercial aircraft highlighting success stories and shortfalls that are delaying industrialization and commercialization of the technology. Current research focusses on optimising operating conditions for MFC and better understanding of the behaviour of PEMFC under aeronautic conditions. Information obtained from the studies can be used to identify factors that influence fuel cell life and ultimately develop techniques that provide real-time data on PEMFC State of Health under aeronautic conditions. From the literature survey conducted, it can be concluded that:

1. PEMFC are not only considered for powering APU, but as multifunctional fuel cells (MFC) capable of producing drinkable water, generating useful heat, discharging oxygen-depleted air, recharging batteries, replacing Ram Air Turbine and power small electrical systems on-board (i.e. flight control and landing gear).
2. Field testing has successfully shown that PEMFC are capable of providing energy in case of emergency as well as power electric nose wheel in commercial aircraft. Simulated laboratory testing under aeronautic conditions (i.e. vibrations, inclination/orientation, high altitude/low pressure and surrounding temperature) showed that performance loss can be minimized by tuning operating conditions (stack temperature, stoichiometric ratio, relative humidity, etc.).
3. Behaviour of PEMFC under aeronautic conditions is not well documented. The few studies conducted confirm that fuel cell performance is affected by the aeronautic conditions. The extent of the effect is not yet quantified. Hence it is crucial to conduct more research in order to establish differences/similarities between terrestrial and aerial effects on fuel cells. MFC operating conditions that are required for optimum quality of the by-products also affects performance and life of fuel cell. At this stage, research focusses on the quality of the products rather than the overall life of fuel cell.
4. Establishing the effect of aeronautic conditions and MFC operating requirements is necessary to identify

Table 3: Summary of the links between various operating conditions and their repective effects on PEMFC components under terrestrial conditions

Factor	Phenomenon	Linked factor	Measurable effect	Affected component	Reference
Anode stoichiometry	Starvation	Load cycling/cell reversal	ECSA and particle size distribution	Catalyst layer	[52]
Load cycling	Flooding and starvation	Current density and reactant gases' demand	Degradation rate	Performance, GDL and catalyst layer	[55, 69, 72, 73]
Stack temperature	Drying	Relative humidity	Hydrogen crossover, membrane resistance and membrane thickness	Membrane	[59]
Relative humidity	Flooding and starvation	Load cycling	Membrane thickness, ECSA and mass transport resistance	Membrane, catalyst and GDL	[44, 101]

factors that influence fuel cell life and performance when used in aircraft. The information will be used to develop prediction techniques suitable for aircraft applications. Available techniques either do not provide comprehensive information on SoH, are invasive, not sensitive to non-homogeneous current distribution observed in larger stacks or difficult to fit in aircraft (i.e. too large with complex balance of plant).

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References

[1] Lapeña-Rey N *et al.*, Environmentally friendly power sources for aerospace applications, *J. Power Sources,* 2008, 181, 353-362
[2] DLR International Press Release, Fuel cells power first takeoff for DLR's Antares aircraft, *Fuel Cells Bulletin,* 2009, 3
[3] Rathke P, Thalau O, Kallo J, *et al.*, Long Distance Flight Testing with Fuel Cell Powered Aircraft Antares DLR-H2, unpublished.
[4] Boeing International Press Released, *787 Electrical System*, 2013, http://787updates.newairplane.com/787-Electrical-Systems/787-Electrical-System
[5] Seidel AJ, Sehra AK and Colantonio OR, NASA aeropropulsion research: Looking forward, 2001
[6] Friedrich KA, Kallo J, Schirmer J, *et al.*, Fuel Cell Systems for Aircraft Application, *ECS Transitions,* 2009, 25, 193-202
[7] Renouard-Vallet G, Saballus M, Schimitthals G, *et al.*, Improving the environmental impact of civil aircraft by fuel cell technology: concepts and technological progress, *Energy Environ. Sci.,* 2010, 1458-1468
[8] Tibaquirá JE, Hristovski KD, Westerhoff P, *et al.*, Recovery and quality of water produced by commercial fuel cells, *Int J Hydrogen Energy,* 2011, 36, 4022-4028
[9] Keim M, Kallo J, Friedrich KA, *et al.*, Multifunctional fuel cell system in an aircraft environment: An investigation focusing on fuel tank inerting and water generation, *Aerospace Science and Technology,* 2013, 29, 330-338
[10] Hardman S, Chandan A and Steinberger-Wilckens R, Fuel cell added value for early market applications, *J. Power Sources,* 2015, 287, 297-306
[11] Werner C, Busemeyer L and Kallo J, The impact of operating parameters and system architecture on the water management of a multifunctional PEMFC system, *Int J Hydrogen Energy,* 2015, 40, 11595-11603
[12] Daggett DL, Eelman S and Kristiansson G, Fuel cell APU for commercial aircraft, Proceedings of AIAA/ICAS International Air and Space Symposium and Exposition: The Next 100 Years *(14 - 17 July* 2003), Dayton, Ohio, 1-9
[13] Eelman S, del Pozo y de Poza I and Krieg T, Fuel cell APU's in commercial aircraft - an assessment of SOFC and PEMFC concepts, Proceedings of *24th International Congress of the Aeronautical Sciences (29 Aug - 3 Sept 2004),* Yokohama, Japan, 1-10

[14] Renouard-Vallet G, Saballus M, Schumann P, *et al.*, Fuel cells for civil aircraft application: On-board production of power, water and inert gas, *Chem. Eng. Res. Design,* 2012, 90, 3-10

[15] Romeo G, Borello F, Correa G, *et al.*, ENFICA-FC: Design of transport aircraft powered by fuel cell & flight test of zero emission 2-seater aircraft powered by fuel cells fueled by hydrogen, *Int J Hydrogen Energy,* 2013, 38, 469-479

[16] Nehter P and Winkler W, System analysis of fuel cell APUs for aircraft applications, Proceedings in *Workshop 1 - Aerospace Applications (31 Aug - 1 Sept* 2005*),* Hamburg, 1-10

[17] Rouss V, Lesage P, Bégot S, *et al.*, Mechanical behaviour of a fuel cell stack under vibrating conditions linked to aircraft applications part I: Experimental, *Int J Hydrogen Energy,* 2008, 33, 6755-6765

[18] Rouss V, Candusso D and Charon W, Mechanical behaviour of a fuel cell stack under vibrating conditions linked to aircraft applications part II: Three-dimensional modelling, *Int J Hydrogen Energy,* 2008, 33, 6281-6288

[19] Airbus International Press Release, Emission Free Power for Civil Aircraft successfully Demonstrates Fuel Cell in Flight, 2008, http://www.airbus.com/presscentre/press-release-detail/detail/emission-free-power-for-civil-aircraft-airbus-successfully-demonstrates-fuel-cells-in-flight/

[20] Kallo J, Renouard-Vallet G, Saballus M, *et al.*, Fuel cell system development and testing for aircraft applications, Proceedings of *18th World Hydrogen Energy Conference (16 - 21 May* 2010*),* Essen, Germany, 435-444

[21] Romeo G, The First European Commission Funded Aircraft Powered by a Hydrogen Fuel Cell took its First Flight, unpublished, 2010

[22] Bégot S, Harel F, Candusso D, *et al.*, Fuel cell climatic tests designed for new configured aircraft application, *Energy Conversion and Management,* 2010, 51, 1522-1535

[23] DLR International Press Release, *DLR Airbus A320 ATRA taxis uses fuel cell-powered nose wheel for the first time*, 2011

[24] Pratt JW, Klebanoff LE, Munoz-Ramos K, *et al.*, Proton exchange membrane fuel cells for electrical power generation on-board commercial airplanes, *Appl. Energy,* 2013, 101, 776-796

[25] Ehteshami SMM and Chan SH, The role of hydrogen and fuel cells to store renewable energy in the future energy network – potentials and challenges, *Energy Policy,* 2014, 73, 103-109

[26] Werner C, Gores F, Busemeyer L, *et al.*, Characteristics of PEMFC operation in ambient- and low-pressure environment considering the fuel cell humidification, *CEAS Aeronautical Journal,* 2014, 1-15

[27] Ebrahimi H, Gatabi JR and El-Kishky H, An auxiliary power unit for advanced aircraft electric power systems, *Electr. Power Syst. Res.,* 2015, 119, 393-406

[28] Verstraete D, Lehmkuehler K, Gong A, *et al.*, Characterisation of a hybrid, fuel-cell-based propulsion system for small unmanned aircraft, *J. Power Sources,* 2014, 250, 204-211

[29] Nishizawa A, kallo J, Garrot O, *et al.*, Fuel cell and Li-ion battery direct hybridization system for aircraft applications, *J. Power Sources,* 2013, 222, 294-300

[30] Hordé T, Achard P and Metkemeijer R, PEMFC application for aviation: Experimental and numerical study of sensitivity to altitude, *Int J Hydrogen Energy,* 2012, 37, 10818-10829

[31] Amirinejad M, Rowshanzamir S and Eikani MH, Effects of operating parameters on performance of a proton exchange membrane fuel cell, *J. Power Sources,* 2006, 161, 872-875

[32] S. J. C. Cleghorn SJC, Mayfield DK, Moore DA, *et al.*, A polymer electrolyte fuel cell life test: 3 years of continuous operation, *J. Power Sources,* 2006, 158, 446-454

[33] Fouquet N, Doulet C, Nouillant C, *et al.*, Model based PEM fuel cell state-of-health monitoring via ac impedance measurements, *J. Power Sources,* 2006, 159, 905-913

[34] Alink R, Gerteisen D and Oszcipok M, Degradation effects in polymer electrolyte membrane fuel cell stacks by sub-zero operation—An in situ and ex situ analysis, *J. Power Sources,* 2008, 182, 175-187

[35] Wahdame b, Candusso D, Harel F, *et al.*, Analysis of a PEMFC durability test under low humidity conditions and stack behaviour modelling using experimental design techniques, *J. Power Sources,* 2008, 182, 429-440

[36] Pei P and Chen H, Main factors affecting the lifetime of Proton Exchange Membrane fuel cells in vehicle applications: A review, *Appl. Energy,* 2014, 125, 60-75

[37] Chevalier S, Josset C, Olivier J, *et al.*, Experimental Validation of an Identification Procedure of PEMFC Stack State of Health Using EIS Combined with a Physical Impedance Modelling, *Chemical Engineering Transactions,* 2013, 33, 1015-1020

[38] Enz S, Dao TA, Messerchimidt M, *et al.*, Investigation of degradation effects in polymer electrolyte fuel cells under automotive-related operating conditions, *J. Power Sources,* 2015, 274, 521-535

[39] Lebreton C, Benne M, Damour C, *et al.*, Fault Tolerant Control Strategy applied to PEMFC water management, *Int J Hydrogen Energy,* 2015, 40, 10636-10646

[40] Uno M, Shimida T, Ariyama Y, *et al.*, Development and demonstration flight of a fuel cell system for high-altitude balloons, *J. Power Sources,* 2009, 193, 788-796

[41] Ruiu T, Dreizler AM, Mitzel J, *et al.*, Evaluation of a 2.5 kWel automotive low temperature PEM fuel cell stack with extended operating temperature range up to 120 °C, *J. Power Sources,* 2016, 303, 257-266

[42] Barbir F, PEM Fuel Cells: Theory and Practice. (2nd ed.) Elsevier, 2012

[43] Schmittinger W and Vahidi A, A review of the main parameters influencing long-term performance and durability of PEM fuel cells, *J. Power Sources,* 2008, 180, 1-14

[44] Wasterlain S, Candusso D, Hissel D, *et al.*, Study of temperature, air dew point temperature and reactant flow effects on proton exchange membrane fuel cell performances using electrochemical spectroscopy and voltammetry techniques, *J. Power Sources,* 2010, 195, 984-993

[45] Harms C, Köhrmann F and Dyck A, Study of the influence of key test parameters on the performance of a PEMFC stack, *Solid State Ionics,* 2015, 275, 75-79

[46] Qiu Y, Zhong H, Wang M, *et al.*, Effect of relative humidity cycles accompanied by intermittent start/stop switches on performance degradation of membrane electrode assembly components in proton exchange membrane fuel cells, *J. Power Sources,* 2015, 283, 171-180

[47] D. Liu and S. Case, Durability study of proton exchange membrane fuel cells under dynamic testing conditions with cyclic current profile, *J. Power Sources,* 2006, 162, 521-531

[48] Borup R, Meyers J, Pivovar B, *et al.*, Scientific Aspects of Polymer Electrolyte Fuel Cell Durability and Degradation, *Chemical Reviews,* 2007, 107, 3904-3951

[49] Li H, Tang Y, Wang Z, *et al.*, A review of water flooding issues in the proton exchange membrane fuel cell, *J. Power Sources,* 2008, 178, 103-117
[50] Larminie J and Dicks A, Fuel Cell Systems Explained. (2nd ed.) England: John Wiley & Sons Ltd, 2003
[51] Tolj I, Bezmalinovic D and Barbir F, Maintaining desired level of relative humidity throughout a fuel cell with spatially variable heat removal rates, *Int J Hydrogen Energy,* 2011, 36, 13105-13113
[52] Taniguchi A, Akita T, Yasuda K, *et al.*, Analysis of electrocatalyst degradation in PEMFC caused by cell reversal during fuel starvation, *J. Power Sources,* 2004, 130, 42-49
[53] Taniguchi A, Akita T Yasuda K, *et al.*, Analysis of degradation in PEMFC caused by cell reversal during air starvation, *Int J Hydrogen Energy,* 2008, 33, 2323-2329
[54] Knights SD, Colbow KM, St-Pierre J, *et al.*, Aging mechanisms and lifetime of PEFC and DMFC, *J. Power Sources,* 2004, 127, 127-134
[55] Weng F, Hsu C and C. Li, Experimental investigation of PEM fuel cell aging under current cycling using segmented fuel cell, *Int J Hydrogen Energy,* 2010, 35, 3664-3675
[56] Jeon Y, Na H, Hwang H, *et al.*, Accelerated life-time test protocols for polymer electrolyte membrane fuel cells operated at high temperature, *Int J Hydrogen Energy,* 2015, 40, 3057-3067
[57] Wang Z, Zuo P, Chu Y, *et al.*, Durability studies on performance degradation of Pt/C catalysts of proton exchange membrane fuel cell, *Int J Hydrogen Energy,* 2009, 34, 4387-4394
[58] Franck-Lacaze L, Bonnet C, Choi E, *et al.*, Ageing of PEMFC's due to operation at low current density: Investigation of oxidative degradation, *Int J Hydrogen Energy,* 2010, 35, 10472-10481
[59] Healy J, Hayden C, Xie T, *et al.*, Aspects of the Chemical Degradation of PFSA Ionomers used in PEM Fuel Cells, *Fuel Cells,* 2005, 5, 302-308
[60] Collier A, Wang H, Yuan X, *et al.*, Degradation of polymer electrolyte membranes, *Int J Hydrogen Energy,* 2006, 31, 1838-1854
[61] Le Canut J, Abouatallah RM and Harrington DA, Detection of Membrane Drying, Fuel Cell Flooding, and Anode Catalyst Poisoning on PEMFC Stacks by Electrochemical Impedance Spectroscopy, *Journal of the Electrochemical Society,* 2006, 153, A857-A864
[62] D. Candusso D, Harel F, De Bernardinis A, *et al.*, Characterisation and modelling of a 5 kW PEMFC for transportation applications, *Int J Hydrogen Energy,* 2006, 31, 1019-1030
[63] Pei P, Chang Q and Tang T, A quick evaluating method for automotive fuel cell lifetime, *Int J Hydrogen Energy,* 2008, 33, 3829-3836
[64] Urbani F, Barbera O, Giacoppo G, *et al.*, Effect of operative conditions on a PEFC stack performance, *Int J Hydrogen Energy,* 2008, 33, 3137-3141
[65] Kim B, Cha D and Kim Y, The effects of air stoichiometry and air excess ratio on the transient response of a PEMFC under load change conditions, *Appl. Energy,* 2015, 138, 143-149
[66] Kim S, Shimpalee S and van Zee JW, The effect of stoichiometry on dynamic behavior of a proton exchange membrane fuel cell (PEMFC) during load change, *J. Power Sources,* 2004, 135, 110-121
[67] Yan X, Hou M, Sun L, *et al.*, AC impedance characteristics of a 2 kW PEM fuel cell stack under different operating conditions and load changes, *Int J Hydrogen Energy,* 2007, 32, 4358-4364
[68] Yan Q, Toghiani H and Causey H, Steady state and dynamic performance of proton exchange membrane fuel cells (PEMFCs) under various operating conditions and load changes, *J. Power Sources,* 2006, 161, 492-502
[69] Bose A, Babburi P, Kumar R, *et al.*, "Performance of individual cells in polymer electrolyte membrane fuel cell stack underload cycling conditions," *J. Power Sources,* vol. 243, pp. 964-972, 12/1, 2013.
[70] Shao Y, Yin G and Gao Y, Understanding and approaches for the durability issues of Pt-based catalysts for PEM fuel cell, *J. Power Sources,* 2007, 171, 558-566
[71] Yousfi-Steiner N, Moçotéguy P, Candusso D, *et al.*, A review on polymer electrolyte membrane fuel cell catalyst degradation and starvation issues: Causes, consequences and diagnostic for mitigation, *J. Power Sources,* 2009, 194, 130-145
[72] Shan J, Lin R, Xia S, *et al.*, Local resolved investigation of PEMFC performance degradation mechanism during dynamic driving cycle, *Int J Hydrogen Energy,* 2016, 41, 4239-4250
[73] Liu M, Wang C, Zhang J, *et al.*, Diagnosis of membrane electrode assembly degradation with drive cycle test technique, *Int J Hydrogen Energy,* 2014, 39, 14370-14375
[74] Tian G, Wasterlain S, Candusso D, *et al.*, Identification of failed cells inside PEMFC stacks in two cases: Anode/cathode crossover and anode/cooling compartment leak, *Int J Hydrogen Energy,* 2010, 35, 2772-2776
[75] Rubio MA, Urquia A and Dormido S, Diagnosis of performance degradation phenomena in PEM fuel cells, *Int J Hydrogen Energy,* 2010, 35, 2586-2590
[76] Miller M and Bazylak A, A review of polymer electrolyte membrane fuel cell stack testing, *J. Power Sources,2011,* 196, 601-613
[77] Wasterlain S, Candusso D, Harel F, *et al.*, Development of new test instruments and protocols for the diagnostic of fuel cell stacks, *J. Power Sources,* 2011, 196, 5325-5333
[78] Yuan X, Li H, Zhang S, *et al.*, A review of polymer electrolyte membrane fuel cell durability test protocols, *J. Power Sources,* 2011, 196, 9107-9116
[79] Bezmalinovic D, Simic B and Barbir F, Characterization of PEM fuel cell degradation by polarization change curves, *J. Power Sources,* 2011, 294, 82-87
[80] Jouin M, Gouriveau R, Hissel D, *et al.*, Prognostics and Health Management of PEMFC – State of the art and remaining challenges, *Int J Hydrogen Energy,* 2013, 38, 15307-15317
[81] Petrone R, Zheng Z, Hissel D, *et al.*, A review on model-based diagnosis methodologies for PEMFCs, *Int J Hydrogen Energy,* 2013, 38, 7077-7091
[82] Zheng Z, Petrone R, Péra MC, *et al.*, A review on non-model based diagnosis methodologies for PEM fuel cell stacks and systems, *Int J Hydrogen Energy,* 2013, 38, 8914-8926
[83] Chevalier S, Trichet D, Auvity B, *et al.*, Multiphysics DC and AC models of a PEMFC for the detection of degraded cell parameters, *Int J Hydrogen Energy,* 2013, 38, 11609-11618
[84] Chevalier S, Auvity B, Olivier JC, *et al.*, Detection of Cells State-of-Health in PEM Fuel Cell Stack Using EIS Measurements Coupled with Physics Modelling, *Fuel Cells,* 2014, 14, 416-429
[85] Cadet C, Jemeï S, Druart F, *et al.*, Diagnostic tools for PEMFCs: from conception to implementation, *Int J Hydrogen Energy,* 2014, 39, 10613-10626
[86] Wu J, Yuan XZ, Wang H, *et al.*, Diagnostic tools in PEM fuel cell research: Part I Electrochemical techniques, *Int J Hydrogen En-*

ergy, 2008, 33, 1735-1746

[87] Lee K, Lee B, Yoo S, *et al.*, Development of a galvanostatic analysis technique as an in-situ diagnostic tool for PEMFC single cells and stacks, *Int J Hydrogen Energy,* 2012, 37, 5891-5900

[88] Brightman E, Hinds G and O'Malley R, In situ measurement of active catalyst surface area in fuel cell stacks, *J. Power Sources,* 2013, 242, 244-254

[89] Scholta J, Berg N, Wilde P, *et al.*, Development and performance of a 10 kW PEMFC stack, *J. Power Sources,* 2004, 127, 206-212

[90] Migliardini F and Corbo P, CV and EIS Study of Hydrogen Fuel Cell Durability in Automotive Applications, *International Journal of Electrochemical Science,* 2013, 8, 11033-11047

[91] Paganin VA, Oliviera CLF, Ticianelli EA, *et al.*, Modelistic interpretation of the impedance response of a polymer electrolyte fuel cell1," *Electrochim. Acta,* 1998, 43, 3761-3766

[92] Gomodam GM and Weidner JW, Analysis of Electrochemical Impedance Spectroscopy in Proton Exchange Membrane Fuel Cells, *International Journal of Energy Research,* 2005, 29, 1131-1151

[93] Brunetto C, Moschetto A and Tina G, PEM fuel cell testing by electrochemical impedance spectroscopy, *Electr. Power Syst. Res.,* 2009, 79, 17-26

[94] Mousa G, Golnaraghi F, DeVaal J, *et al.*, Detecting proton exchange membrane fuel cell hydrogen leak using electrochemical impedance spectroscopy method, *J. Power Sources,* 2014, 246, 110-116

[95] T. Kurz T, Hakenjos A, Krämer J, *et al.*, An impedance-based predictive control strategy for the state-of-health of PEM fuel cell stacks, *J. Power Sources,* 2008, 180, 742-747

[96] Rodat S, Sailler S, Druart F, *et al.*, EIS measurements in the diagnosis of the environment within a PEMFC stack, *J. Appl. Electrochem.,* 2010, 40, 911-920

[97] Rubio MA, Urquia A, Kuhn R, *et al.*, Electrochemical parameter estimation in operating proton exchange membrane fuel cells, *J. Power Sources,* 2008, 183, 118-125

[98] Yuan X, Wang H, Colin Sun J, *et al.*, AC impedance technique in PEM fuel cell diagnosis—A review, *Int J Hydrogen Energy,* 2007, 32, 4365-4380

[99] Rezaei Niya SM and Hoorfar M, Study of proton exchange membrane fuel cells using electrochemical impedance spectroscopy technique – A review, *J. Power Sources,* 2013, 240, 281-293

[100] de Bruijn FA, Dam VAT and Janssen GJM, Review: Durability and Degradation Issues of PEM Fuel Cell Components, *Fuel Cells,* 2008, 8, 3-22

[101] Wang F, Yang D, Li B, *et al.*, Investigation of the recoverable degradation of PEM fuel cell operated under drive cycle and different humidities, *Int J Hydrogen Energy,* 2014, 39, 14441-14447