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Expert diagnosis of polymer electrolyte fuel cells

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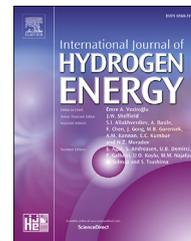
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Expert diagnosis of polymer electrolyte fuel cells



B. Davies^{*}, L. Jackson, S. Dunnett

Department of Aeronautical and Automotive Engineering, Loughborough University, Leicestershire, United Kingdom

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ABSTRACT

Diagnosing faulty conditions of engineering systems is a highly desirable process within control structures, such that control systems may operate effectively and degrading operational states may be mitigated. The goal herein is to enhance lifetime performance and extend system availability. Difficulty arises in developing a mathematical model which can describe all working and failure modes of complex systems. However the expert's knowledge of correct and faulty operation is powerful for detecting degradation, and such knowledge can be represented through fuzzy logic. This paper presents a diagnostic system based on fuzzy logic and expert knowledge, attained from experts and experimental findings. The diagnosis is applied specifically to degradation modes in a polymer electrolyte fuel cell. The defined rules produced for the fuzzy logic model connect observed operational modes and symptoms to component degradation. The diagnosis is then tested against common automotive stress conditions to assess functionality.

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Introduction

Polymer electrolyte fuel cells (PEFCs) exist as an emergent technology that could provide environmentally friendly electrical power for a range of applications. These scalable systems can be used to power small portable electronics [1,2], large stationary building power [3,4], or used as power plants for electric vehicles, which is potentially a global market [5–7]. However, PEFC systems are yet to achieve full commercialisation.

System reliability is one factor which is seen to limit successful applications, particularly in the automotive regime [6,8]. The US Department of Energy have set durability targets, wherein fuel cell vehicles are expected to survive comparably to internal combustion powertrains; nominally 5000 h total lifetime, or 150,000 miles equivalent usage, before a 10% drop-off in power output [9]. Demonstrator projects have reported

up to half of this expected lifetime performance, leaving significant room for improvement [10].

The dynamic and poorly constrained nature of automotive usage leads to unideal operating conditions, and ultimately component degradation. Cyclic loading profiles cause platinum-catalyst dissolution [11]; poor quality air can introduce contaminants [12]; and variations in temperature can lead to problems associated with water balance in the membrane [13]. Such operational conditions can vary as much within a single usage as they may in different geographic locations.

Thus the development of control and diagnosis systems has grown in recent years, in an attempt to improve PEFC performance through health management. Reviews of durability and degradation issues are available in Refs. [14–16], as well as an introduction to the health management topic in Ref. [8]. The diagnosis of fuel cell component degradation is however a complex problem, with different methodologies

^{*} Corresponding author.

E-mail address: B.Davies2@lboro.ac.uk (B. Davies).

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available. Common techniques can be broadly classified into model-based and signal-based methods.

Analytical models are useful for simulating system performance, though they require in-depth knowledge of the internal processes to generate equations [17]. The parameters of these equations can change significantly across the operating envelope. Accuracy is dependent on the complexity of the model, though this can add significant computational cost and time. As such, these models may have limited applicability to real-time diagnosis, however can be of most use in off-line simulation and design.

Some success in diagnosis has been noted when considering a limited number of degradation modes. For example, Hernandez et al. [18] design an electrical model of the PEFC, allowing detection of flooding events. Similar models are used by Legros et al. [19] and Asghari et al. [20], combined with lab-based impedance spectroscopy testing to detect performance.

Signal-based methods in contrast do not need full understanding of system interactions, instead utilising correlations found in previously observed performance. This approach does however require many hours of testing to be completed for accuracy, especially if the goal is to capture all variable and degradation combinations. Techniques in this region include neural networks, Fourier transforms, and Bayesian networks [21]. These non-model based approaches remain relatively new for PEFC diagnostics.

Whether using model- or signal-based approach, these diagnosis techniques also often require specific characterisation testing to achieve a diagnosis. In the case of PEFC, such tests include electrochemical impedance spectroscopy (EIS) [22], cyclic voltammetry [23], gas chromatography [24], and neutron imaging [25], among many others. The majority of these tests frequently interrupt operation, and require additional equipment to perform. As such, these are undesirable for practical and commercial applications.

A third diagnostic approach is in behavioural representation of the system, based on heuristic knowledge. This approach equates some input variables to output performance, and can be developed more rapidly than the full mathematical counterparts. This diagnostic system can call upon expert understanding and observations from fuel cell operators, without requiring the specific tests mentioned. The intention is to have a continuous health-monitoring system, without interrupting normal usage patterns for characterisation tests.

Fuzzy logic enables this heuristic diagnostic system. Fuzzy logic is able to evaluate a range of controlled and measured variables from the system, and infer the degradation processes without direct cell-internal measurements or tests [26]. This process is reliant on an effective base of knowledge [27], as is developed in this paper.

Fuzzy set theory was originally devised by L. Zadeh as a means of allowing computers to handle semantics in a similar way to human reasoning [29]. Early applications in alternative control have been successful for industrial plants [30], and vehicles such as ships and trains [31]. Fuzzy logic has seen usage for disease diagnosis in the medical realm [32], and some comparable applications in engineering systems [27].

Fuel cell diagnostics can benefit from the heuristic diagnostic approach because of the volume of knowledge

established during the recent years of development. Validation for new components and materials include many operational conditions and observed degradation behaviour, which can be used to populate the fuzzy logic knowledge base. For example, experimental tests have studied membranes under thermal stress [33], platinum dissolution due to high potentials [34], and mechanical forces which crush the fragile electrode materials [35], among many more. The conclusions drawn from these studies are to be utilised in the diagnostic rules base.

The diagnostic system presented in this paper covers the main degradation phenomena expected in practical PEFC usage. This includes rules for degradation of the polymer membrane, platinum catalyst, and gas diffusion materials, and water management performance issues. The diagnostic process accepts continuous sensor monitoring without altering the PEFC operation or performing the characterisation test previously identified.

Fuzzy logic has been utilised within PEFC operation for control applications, where the fuzzy logic is able to manage parameter variability well [36]; this includes applications of fuzzy logic in fuel cell power management [37–39]. As far as PEFC diagnosis is concerned, fuzzy logic has been used by Zheng et al. in Ref. [40] in a pattern matching approach. Therein, fuzzy clustering is used on EIS measurements to identify symptoms of membrane drying. This is performed in situ on a functioning fuel cell, though there is some academic debate over applicability of EIS in commercial systems [41].

The work presented in this paper utilises fuzzy logic to diagnose PEFC degradation states based on operational measurements, and the available knowledge about conditions causing degrading states. This is distinct from many existing diagnostic approaches in foregoing the requirement for EIS or other characterisation procedures to detect faulty operation. The diagnostic algorithm is compiled for multiple degradation modes, and validated experimentally for water management issues; flooding gas paths and drying out of the membrane. Water management is selected for a number of reasons. Firstly, it is seen as one of the most common faults expected to effect transportation PEFC performance [14]. Also, for validation testing, this degradation state is largely reversible, repeatable, and fast-acting, meaning experiments should provide high quality data.

This paper is organised as follows: Section [Expert diagnosis system development](#) details the diagnostic process, as well as the rules-system and the key motivators for PEFC degradation. The experimental set-up is described in Section [Experimental set-up](#), with the initial experimental validation given in Section [Results](#). Finally, conclusions are drawn from this work in Section [Conclusions](#).

Expert diagnosis system development

Fuzzy logic

Most logic and modelling practices require well quantified numerical inputs, crisp numbers, measurements, and equations. Fuzzy logic however is an approach using “degrees of truth” across a variable range. These ranges can also be

defined linguistically (for example, temperature may be *hot* or *cold*), increasing transparency for human operators and developers.

The fuzzy system is an inference calculation between the supplied inputs and a database of rules. This relates measured variables to unmeasurable internal mechanisms of the PEFC component degradation. The output is then a measure of which degradation states are activated in the current operational state, based on the knowledge base. The process flow is summarised in Fig. 1.

The fuzzy logic system accepts crisp measurement data of temperature, humidity, electrical power, and fuel supply from the monitored system. The measurements go through a *fuzzification* process, which maps the discrete numerical values to the linguistic ranges understood by the diagnostics. For example, a stack temperature may be classified into “low”, “normal”, and “hot” ranges. These ranges are termed fuzzy sets within the logic process.

The fuzzified measurement information is acted upon by the central inference process. The accompanying *rules base* holds the expert knowledge for which degradation modes are caused by particular operational modes, or revealed by other external measurements. Thus, this database defines how powerful and accurate the diagnosis may be.

Alongside the rules base is the *inference engine*, which compiles the degree to which each rule is satisfied by the fuzzified measurement. This outputs the diagnosis for which degradation modes are occurring. The final process in this system is the *defuzzification* of the diagnostic output. This reverses the fuzzification process, returning a crisp numerical certainty for any particular mode of degradation; such as a 25% confidence that the membrane is causing performance degradation. This information can be interpreted by control systems or the user to change usage.

Fuzzy sets

The fuzzy set is an expansion of traditional set theory, which allows for the boundaries of each set to overlap. Thus, a measurement value may lie partially within two or more sets, and proportionately trigger the logic of both. This is where the fuzzy concept originates.

Fig. 2 shows the fuzzy sets used against the input measurements of a PEFC system. A common design is to have a “normal” set for on-design values, and sufficient sets on either side to capture the off-design operation. The overlap between these sets signifies how behaviour changes continuously over the operating envelope. The centres of the sets are however defined by known (observed or specified) behaviour from the rules-literature. Simple triangular and trapezoidal set functions are used throughout as a general solution.

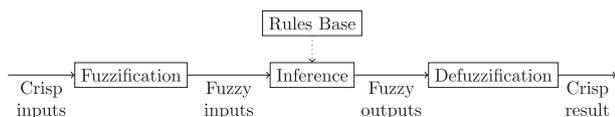


Fig. 1 – Fuzzy diagnostic system structure, adapted from Abonyi et al. [28].

An example is described for stack temperature measurements; normal operation is around 60 °C for a polymer electrolyte fuel cell, hot temperatures are above 100 °C, and cold temperatures are considered below 0 °C. The membrane requires liquid water content for effective operation, so the boundaries are defined by water phase transitions.

“Normal” temperature is represented by full truth at 60 ± 5 °C, and partial, fuzzy truth outside of this range (This 10 °C range represents the linguistic “around 60 °C” parameter, accounting for variability in true stack temperature measurement and control, as well as acceptable operational range defined by manufacturers). Fig. 2 shows the form of these ranges, under “stack temperature”, demonstrating the overlap between each fuzzy set. A measurement of 22 °C (possibly representing start up of a fuel cell vehicle under a summer average daily temperature) would represent 0.6 truth of cold and 0.4 truth of normal temperature condition. The fuzzy logic would thus use rules for cold and normal temperatures, combining the results proportionately by this ratio.

Of these fuzzy inputs, stack voltage, stack temperature, and feed humidity are taken as direct measurements from the fuel cell apparatus. Stack voltage cycles are counted simply as the operating voltage passes 0.9 V; this has been identified at the influencing factor, and duration of cycle is of secondary importance [42,43]. Stoichiometry is calculated using the commonly defined ratio between reactant feed and consumption, based upon current production [44]. Humidity change is the ratio of water molecules to membrane active sites, and defined empirically based on the relative humidity level [33,45]. These calculated parameters are processed prior to fuzzification.

Outputs of the diagnostic are also defined as fuzzy sets. Fig. 3 shows the output set used for all degradation modes considered in this system (shown only once for brevity). The six degradation modes are membrane chemical breakdown, membrane mechanical breakdown, platinum catalyst dissolution, catalyst carbon support corrosion, and water management issues gas-channel flooding, and membrane dehydration. These have been selected for commonly observed degradation modes, particularly in dynamic operation, and for the more fragile components in the PEFC construction [46].

The output sets follow the format of “none”, “evidenced”, “certain”, which is based on the diagnostic evidence considered in the rules base. These sets are defined by the author based upon percentage representation of certainty. That is, with less than 15% agreement between the measured operating conditions and those necessary for a certain degradation, there is effectively no evidence. “Evidenced” measurements represent up to 50% certainty that a given degradation mode is occurring, and fully certain conditions are for measurements upwards of 90% agreement with the degradation conditions. The output is therefore a [0,1] range stating how well the observed conditions represent any single degradation mode.

Diagnostic rules

The knowledge rules base takes the form of logical IF-THEN statements. The knowledge here is drawn from a variety of

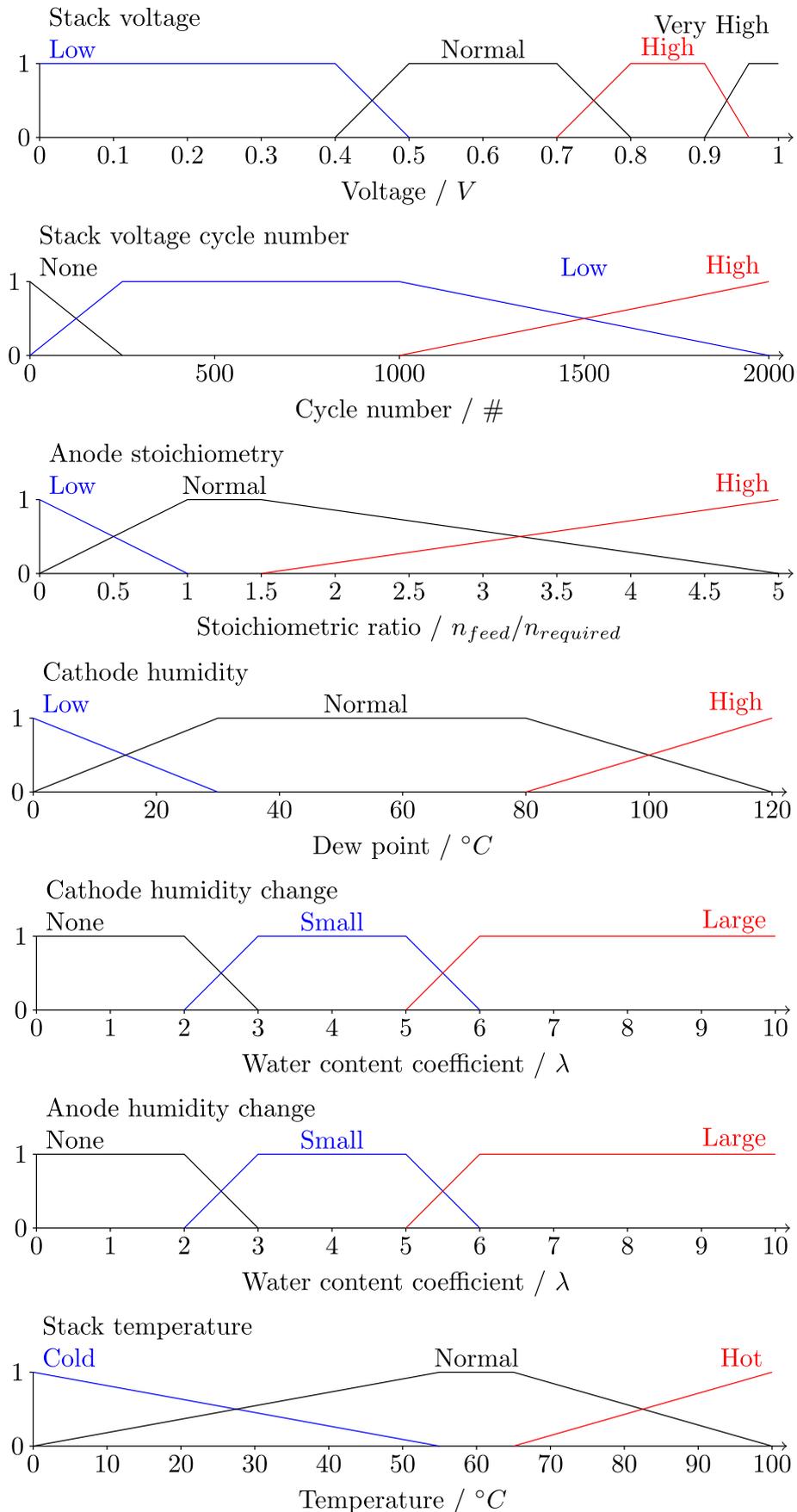


Fig. 2 – Fuzzy input sets.

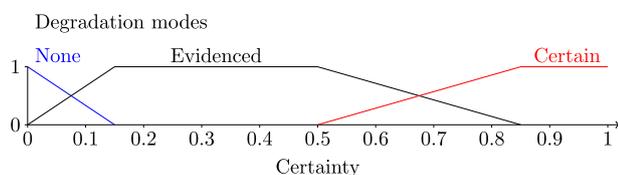


Fig. 3 – Fuzzy output set.

publications, as well as experimental experience. The rules base represents the significant contribution of this paper. The rules base is presented in Table 1. An explanation of the rules and the principle literature sources from which they are drawn is also described below.

The first three rules relate to open circuit operation, which is known to generate hydroxyl radicals ($\text{HO}\cdot$). This chemical agent is understood to be responsible for highly reactive attacks to the membrane polymer chain [47]. However, high voltage draw is required for the radical formation, so the potential for damage drops away quickly with decreasing voltage.

Rules two through six consider the platinum nano-particle catalyst in the PEFC; these are known to agglomerate together to reduce high surface energy. This can occur throughout the fuel cell operation (rules two and three), however the rate is significantly increased when the voltage is driven in a cyclical profile (rules four through six) [42,48,49]; an operational behaviour which will be of special importance to automotive applications.

The catalyst nano-particles are supported on carbon-structures which can suffer their own chemical attack. Rules seven and eight show that, under fuel starvation conditions, the fuel cell reaction can consume the carbon materials because of internal voltage conditions [50].

The membrane is mechanically constrained within the PEFC, and so dimensional changes can induce fatigue stress across its area. Rules nine through twelve reflect the fact that water content in the membrane can cause these dimensional changes and the stresses that ultimately lead to pinholes in the membrane [33]. Mechanical perforation is a total failure mode, as to allow direct mixing of the reactant gasses means not only eliminating electrical current, but also a severe thermal reaction within the stack.

Rules 13 through 17 of Table 1 detail problems that may arise from water management strategies. The polymer membrane requires a water content in order to achieve proton-conductivity; up to 300 times higher, compared to the dry membrane [51]. For this reason, reactant gasses are often humidified, to maintain hydration within the cell.

However, there is a balance to be achieved, between excess water condensing and blocking gas paths, and insufficient water in the membrane limiting conductivity [13]. Water management is thus an important control mechanism for efficient performance. As was eluded to earlier, condensation and evaporation of water is a quick degradation phenomena, relative to the chemical and fatigue failures in the other rules. Rules 13, 14, and 15 consider condensation and evaporation conditions through temperature and humidity changes,

whilst rules 16 and 17 add that high current loading generates water as a reaction product, which also contributes to water balance.

The concentrated output of these rules gives a possibility of any of the considered degradation modes being responsible for performance drop off. These rules are designed to use a reduced set of variable inputs, meaning a small number of sensors will be required on the PEFC system in application. Cell temperature and voltage are simple to measure to sufficient accuracy, though gas humidity is likely the most expensive sensor requirement. This consideration is in order to keep the cost of the diagnostic package low, hence not inflating the overall PEFC system cost.

In addition to identifying the degradation mode, the diagnosis provides a severity assessment. This is through comparison to a model of voltage performance, finding the difference between the measurement and prediction. The model is a simple one, which assumes perfect electrochemical performance, so any deviation should be assigned to the diagnosed degradation mode. The model is introduced in Section Predicting voltage.

The residual between the voltage measurement and prediction gives a value to the severity of the degradation; more severe degradation leads to worse voltage performance. Thus, severity is also bounded by fuzzy sets, as shown in Table 2. The upper limit is the US DoE target of less than 10% loss [9], with increments approaching this failure condition.

The severity level output is expected to be used to influence the user or control systems as to how quickly to act. The diagnostic response considers only the necessary conditions for degradation to occur, whilst the severity level characterises the observed damage.

Both measures should be used in conjunction to support control decisions. For example, a none-to-low certainty for all degradation mode outputs is expected for normal operation, meaning no change to control and usage of the PEFC is required. A high certainty diagnosis of a certain degradation mode means that damage is very likely to be happening within the fuel cell, and the operator must change usage to avoid further degradation. Such an output may be coupled with a low severity warning, meaning the damage has not yet effected performance; this combination may occur for long-duration degradation modes, such as platinum dissolution which can take many hours to show performance drop-off [52]. When a high-certainty diagnosis is coupled with a high-impact performance degradation, strong control actions should be made to mitigate or repair damage.

Experimental set-up

PEFC cell

A single cell 100 cm^2 PEFC is used to carry out experimental validation of the diagnostic system. This fuel cell is manufactured by Pragma Industries as a research and development stack. Although this PEFC is not one used commercially, the same materials and technologies are used in most state of the art systems, hence findings can be applicable to such equipment also. The stack uses established materials in its

Table 1 – Diagnostic rules base for PEFC degradation.

Rule	IF	THEN
1	Stack voltage is very high	Membrane chemical breakdown is certain AND Catalyst dissolution is evidenced
2	Stack voltage is high	Membrane chemical breakdown is evidenced AND Catalyst dissolution is evidenced
3	Stack voltage is normal	Membrane chemical breakdown is none AND Catalyst dissolution is evidenced
4	Stack voltage cycle number is high	Catalyst dissolution is certain
5	Stack voltage cycle number is low	Catalyst dissolution is evidenced
6	Stack voltage cycle number is none	Catalyst dissolution is none
7	Anode stoichiometry is normal OR Anode stoichiometry is high	Carbon support corrosion is none
8	Anode stoichiometry is low	Carbon support corrosion is evidenced
9	Anode humidity change is large AND Cathode humidity change is large	Membrane mechanical stress is certain
10	Anode humidity change is large OR Cathode humidity change is large	Membrane mechanical stress is evidenced
11	Anode humidity change is small AND Cathode humidity change is small	Membrane mechanical stress is none
12	Anode humidity change is none OR Cathode humidity change is none	Membrane mechanical stress is none
13	Stack temperature is cold OR Cathode humidity is high	Flooding is certain AND Dehydration is none
14	Stack temperature is normal OR Cathode humidity is normal	Flooding is none AND Dehydration is evidenced
15	Stack temperature is hot OR Cathode humidity is low	Flooding is none AND Dehydration is certain
16	Stack voltage is normal OR Stack voltage is high	Flooding is none
17	Stack voltage is low	Flooding is evidenced

Table 2 – Severity rules base.

IF	THEN
Voltage difference is 0–1%	Non-severe
Voltage difference is 1–3%	Low severity
Voltage difference is 3–5%	Medium severity
Voltage difference is 5–10%	High severity
Voltage difference is 10+ %	Severity warning

construction; Nafion polymer membrane, platinum nano-particle catalyst, carbon diffusion materials, silicone sealing gaskets, composite flow field plates, and metallic structure elsewhere. In addition, the flow field plates have channels for a water coolant circuit.

Table 3 presents the main technical information about the PEFC used in this study.

Ancillary systems

The PEFC is operated in an 800 W test bench, developed in-lab for the experimental requirements. The included subsystems are:

Electronic load

The load current can be varied through a resistive load, up to 150 A.

Air supply

The flow rate, pressure, temperature, and humidification supply to the cathode can be controlled and monitored.

Hydrogen supply

The flow rate, pressure, temperature, and humidification supply to the anode can be controlled and monitored.

Nitrogen supply

Nitrogen can be used as a purge gas at both electrodes of the PEFC, which is necessary during installation and start-up-shut-down procedures of the lab-based rig.

Temperature control circuit

The water circuit through the fuel cell can be cooled or heated to control temperature.

Control and monitoring environment

The control of the test bench is fulfilled with National Instruments LabView software, and a purpose built application.

Table 3 – PEFC test cell technical details.

Parameter	Value
Membrane thickness	25 μm
Active area	100 \times 100 mm
Platinum loading	0.2 mg/cm ² (an/ca)
Gas diffusion thickness	415 μm
Flow channels	7-fold serpentine
Compressive torque	4 Nm
Reactant stoichiometry	1/3 @ nominal (an/ca)
Typical voltage range	0.6–0.65 V
Stack temperature	45 $^{\circ}\text{C}$

Experimental methods

Experimental tests have been carried out for water management in the PEFC cell. Normal performance is first established through steady-state operation for a duration, at nominal conditions given in Table 3. This helps quantify variations in performance from this initial output, as well as checking recovery after testing; short term water management issues are considered largely reversible forms of degradation.

Flooding states are triggered by decreasing stack temperature so that water condensation is favoured. The divergence between stack temperature and gas dew point indicates that flooding is the most likely loss mechanism. Membrane dry-out is stimulated by the reverse; increasing stack temperature beyond the dew point temperature, thus encouraging evaporation.

Predicting voltage

Mann et al. have developed a general electrochemical model for fuel cell performance, useful for first-pass calculations in the design process [53], see Equation (1). This calculation accepts variables of fuel cell temperature and current loading, and is useful for calculating expected voltage performance of the PEFC, and hence quantifying any deviation as attributed to degradation phenomena.

$$E = E_{rev} - \frac{RT}{n\alpha F} \cdot \ln\left(\frac{i + i_{loss}}{i_0}\right) - i \cdot r_{cell} + B_C \cdot \ln\left(1 - \frac{i}{i_{max}}\right) \quad (1)$$

where E is the cell voltage, E_{rev} is the theoretical reversible voltage, R the universal gas constant, T the stack temperature, n the reaction charge number, α the charge transfer coefficient, F the Faraday constant, i the current density, i_{loss} the hydrogen crossover current, i_0 the reaction exchange coefficient, r_{cell} the internal cell resistance, B_C is an empirical parameter taking into account gas accumulation at the cathode, and i_{max} the limiting cathode current.

This expression is designed to predict the fuel cell performance for steady-state electronic loading under variable temperature conditions. The above mathematical prediction assumes perfect component condition, including no water management problems within the PEFC. Thus, any deviation in measured voltage is accounted as a degradation, and should be identified in the severity rules base, Table 2.

A note on temperature control

Due to limitations of the experimental set up, the desirable 60 °C stack temperature is not attainable. The small scale PEFC is heated during testing, using the integral water circuit. The result is that the fuel cell does not achieve optimal “normal” temperature, as was discussed in Section Fuzzy sets. The humidification system is similarly temperature limited to reflect best conditions at the given temperature.

For this reason, the fuzzy set for stack temperature is modified to represent the lower operating temperature. Instead of 60 °C, “normal” is defined for around 45 °C. The boundaries for “cold” and “hot” remain unchanged (as these are related to liquid water limits), however the fuzzy overlap between the sets are suitably adjusted. The new fuzzy set is as shown in Fig. 4.

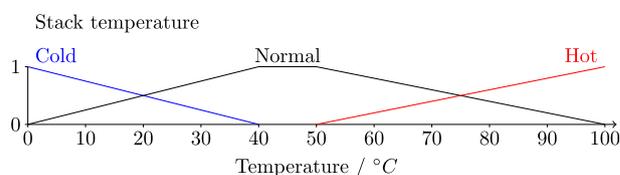


Fig. 4 – Modified fuzzy set for stack temperature.

The fuzzy rules utilising stack temperature remain unchanged; these rules consider the condensation of warm gas vapour on cooler fuel cell components. Therefore, by operating both the stack temperature and humidification system at lower temperatures, the integrity of the water management rules is maintained. This is also an example of the utility of the fuzzy logic system to use the same rules knowledge applied to different fuel cell systems, with flexibility in collaborating the fuzzy input sets.

Results

Flooding events

Fig. 5 shows the time progression through the flooding test. From the beginning of the experiment, stack temperature is decreased below the standard operating temperature, as seen in Fig. 5B. As the reactant feed dew point is held constant at 30 °C, the stack drops below this temperature after about 17 min into the test. This encourages condensation of hotter vapour on the cooler PEFC materials, leading to flooding events within the gas diffusion components.

Thus the stack transitions from a “normal” temperature at the beginning of the test, to increasingly “cold” temperatures. Up until 10 min, stack temperature is 45 °C, fully representing “normal” (fuzzy value of 1, Table 1) and no chance of flooding (fuzzy output of 1 from rule 14, and 0 from rules 13, and 15–17).

The diagnostic rule for a cold stack (Table 1, rule 13) is activated by the reducing temperature measurement, increasing the diagnostic output for flooding through the duration of the test. At 17 min stack temperature is 30 °C, representing 0.75 “normal” and 0.25 “cold” temperature; this increases the likelihood that flooding is occurring.

The output for flooding-diagnosis, in Fig. 5C, shows good response to the temperature change, predicting more flooding as stack temperature decreases. The initial cooling is detected as “evidenced” conditions for causing flooding, with approximately 0.5 output after 15 min. By the end of the test duration, flooding is diagnosed with almost certainty as the cause of voltage degradation.

At the same time as stack temperature decreases, Fig. 5A shows that the voltage output decreases also; this is expected behaviour predicted by the electrochemical model in Equation (1). However, the measured voltage decreases far more than the prediction. This behaviour is expected; as mentioned previously, the model-equation assumes perfect electrochemical performance within the PEFC. Thus the voltage degradation is attributed to the severity of flooding events.

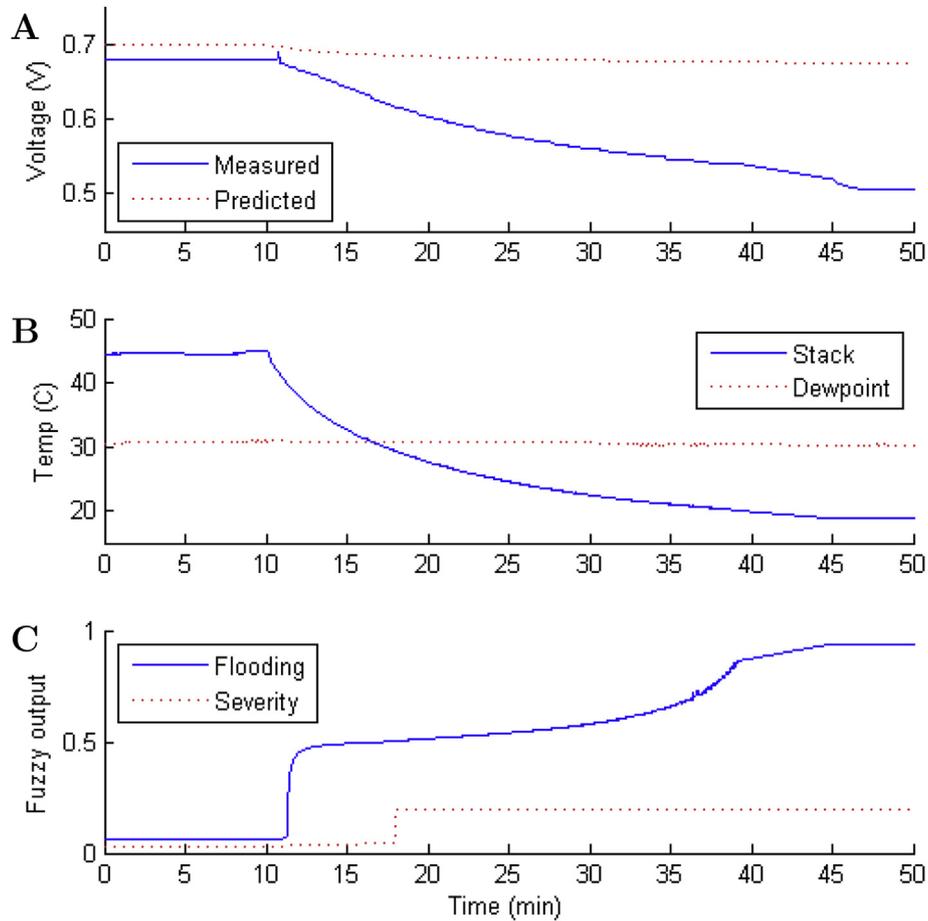


Fig. 5 – Diagnostic results for flooding test; A) Voltage predicted and measured result, B) Stack temperature and humidification dew point measurements, C) Output of expert diagnostic system.

Fig. 5C gives this severity fuzzy response, increasing from the “non-” to the “low severity” case after approximately 17 min.

The combined outputs of the expert diagnostic, of flooding possibility (rule 13) and severity rating (low-severity), inform the operator that flooding events are likely to be happening in the fuel cell, but with minimal performance degradation beginning from the 17 min mark. By the end of the time duration, flooding is certainly occurring in the “cold” PEFC stack, but remains low impact to the voltage performance. The user can use this information to make actions to reverse the flooding conditions before further voltage degradation is caused.

Membrane dry out

In the second test, results from which are shown in Fig. 6, the stack temperature is increased to cause membrane dehydration. Fig. 6B follows the temperature progression, from “cold” to “normal” operating conditions. Initially output voltage recovers, as in Fig. 6A; this control change is equivalent to the mitigation action for the flooding condition – warming the cell up again. As the stack temperature is above the feed humidification setting, this creates a dry-out condition within the cell.

Diagnosis rule 13 (Table 1) is activated at the start of the test; a stack temperature of 19 °C gives 0.525 truth of “cold” operation, meaning no evidence of dehydration. At the 10 min mark, stack temperature is up to 37 °C, “normal” with 0.925 truth. This is triggering the output of rule 14 in that membrane dehydration is “evidenced” at the higher temperature.

Under these conditions, the fuzzy diagnostic response for dehydration increases quickly, as shown in Fig. 6C. This acknowledges the higher temperature conditions, and capacity for membrane dehydration to occur, not its definite appearance. Thus the dehydration output is at the 0.5 “evidenced” level after 5 min. It is an important distinction that the diagnosis here is for the necessary conditions, as it takes several minutes for the dehydration effects to be apparent in the voltage performance.

It is for this reason the diagnosis and severity responses must be used together from Fig. 6C; the former infers the degradation mode that may be occurring, the latter qualifies its impact. The stack voltage measurement in Fig. 6A remains close to the prediction for the latter duration of the time period, meaning there are no dehydration effects seen, and the cell remains in good water-balance. Hence the severity rating for voltage degradation is firmly in the “non-severe”

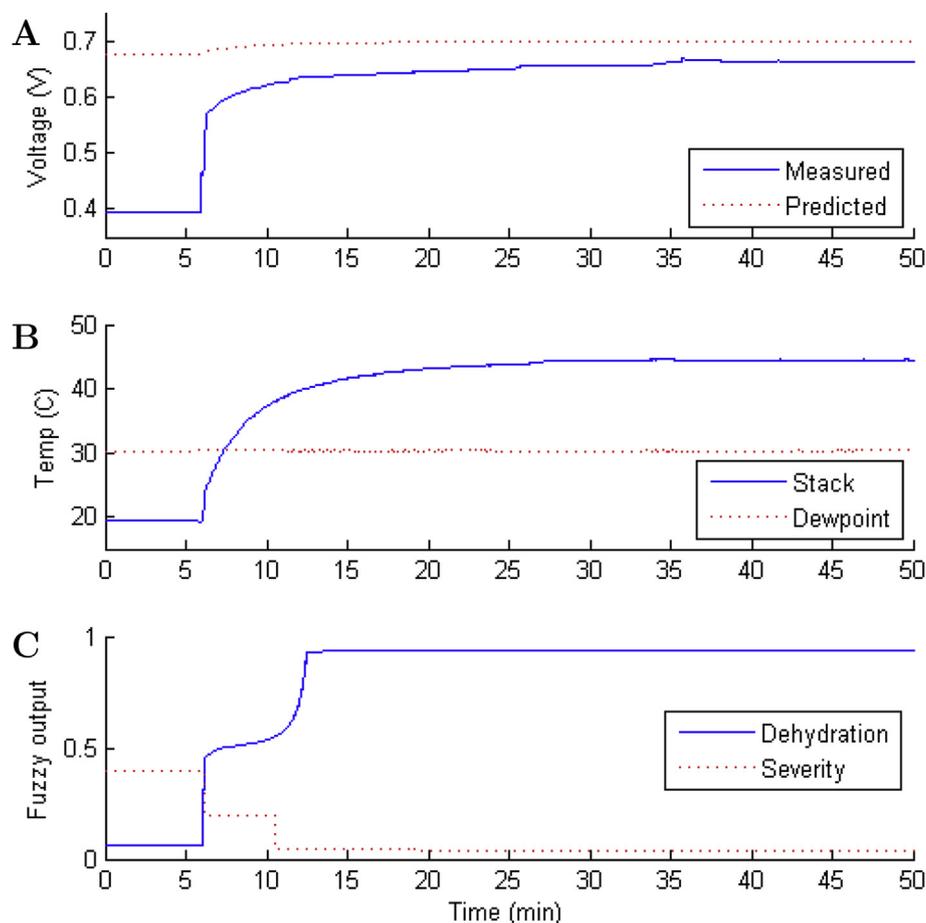


Fig. 6 – Diagnostic results for dehydration test; A) Voltage predicted and measured result, B) Stack temperature and humidification dew point measurements, C) Output of expert diagnostic system.

range. Only after a longer duration under these conditions will a significant dry-out degradation be felt by the PEFC.

Discussion

These experimental results have shown the usefulness of the expert diagnostic system; simple to comprehend and configure, whilst powerful for monitoring the PEFC dynamics and identify performance degradation. The flooding test response is the most positive; the diagnostic is quick to identify the conditions which cause a reduction to the electrical power output. The combined information of degradation mode and severity in Fig. 5 can allow the operator or control system to mitigate any further problems. These responses would be all the more pressing should the severity rating reach “high” or “warning” levels.

The diagnostic response for the membrane dry out condition shows the same quick response to degrading operating conditions. The difference with this validation test is where the fuel cell was not operated for sufficient time to reveal a voltage decrease. Further testing will be needed to tune this rule, to reflect the rates of dehydration as dependant on temperature level. Understanding this dynamic response will also benefit the development of the other diagnostic rules, yet to be experimentally validated.

Conclusions

An approach for the diagnosis of common PEFC degradation modes is proposed in this paper. This method utilises expert understanding and experience in the form of a fuzzy logic inference system. The goal of this diagnostic process is to provide real-time information about degradation phenomena and severity, allowing users and controlling systems to intelligently maintain the PEFC performance. This is achieved through continuous monitoring, forgoing traditional characterisation testing.

Diagnosis is achieved by comparing measured operating conditions to those known to cause specific degradation phenomena. The expert knowledge has been collated from various literature sources detailing component testing, degradation mechanisms, and accelerated system testing in PEFCs. This information is phrased as logical rules acted upon by fuzzy logic processing.

The expert diagnostic system is validated experimentally on a single-cell PEFC test rig. Although this is not the fuel cell expected to be used in practical applications, the nature of the technology allows findings to be applicable in all scales. Experimental tests are run to stimulate water management difficulties; namely flooding of gas diffusion pathways, and

drying out of the polymer electrolyte membrane. These degradation modes are known occurrences in dynamically operated PEFC systems, for example in automotive regimes.

The diagnostic programme responded quickly and positively to the experimental testing, identifying the possible conditions to cause given degradation modes, useful information when performance begins to degrade. In some conditions, the diagnostic responds excessively quickly, possibly leading to false-positives; the modelling for slowly-evolving degradation modes should be developed, to improve the diagnostic accuracy.

Future work will continue to validate the existing diagnostic rules, looking to test other degradation modes, and ultimately their combinations when this could happen in practice. Tuning the diagnostic modes will also be necessary for different scales of PEFC. Building on the diagnostic programme will be control responses of a broader health management system, which may automatically act to maintain PEFC power performance.

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REFERENCES

- [1] Wainright JS, Savinell RF, Liu CC, Litt M. Microfabricated fuel cells. *Electrochim Acta* 2003;48(20–22):2869–77. [http://dx.doi.org/10.1016/S0013-4686\(03\)00351-7](http://dx.doi.org/10.1016/S0013-4686(03)00351-7).
- [2] Lu GQ, Wang CY, Yen TJ, Zhang X. Development and characterization of a silicon-based micro direct methanol fuel cell. *Electrochim Acta* 2004;49(5):821–8. <http://dx.doi.org/10.1016/j.electacta.2003.09.036>.
- [3] Wang F-C, Kuo P-C, Chen H-J. Control design and power management of a stationary PEMFC hybrid power system. *Int J Hydrogen Energy* 2013;38(14):5845–56. <http://dx.doi.org/10.1016/j.ijhydene.2013.03.021>. <http://linkinghub.elsevier.com/retrieve/pii/S0360319913005958>.
- [4] Cottrell CA, Grasman SE, Thomas M, Martin KB, Sheffield JW. Strategies for stationary and portable fuel cell markets. *Int J Hydrogen Energy* 2011;36(13):7969–75. <http://dx.doi.org/10.1016/j.ijhydene.2011.01.056>.
- [5] Thomas CE. Fuel cell and battery electric vehicles compared. *Int J Hydrogen Energy* 2009;34:6005–20. <http://dx.doi.org/10.1016/j.ijhydene.2009.06.003>.
- [6] Garland NL, Papageorgopoulos DC, Stanford JM. Hydrogen and fuel cell technology: progress, challenges, and future directions. *Energy Procedia* 2012;28:2–11. <http://dx.doi.org/10.1016/j.egypro.2012.08.034>. <http://linkinghub.elsevier.com/retrieve/pii/S1876610212013902>.
- [7] Roy F, Garnit S, Crouvezier J. Fisypac project: the first vehicle integration of genepac fuel cell stack. *World Electr Veh J* 2009;3:1–7. <http://scholar.google.com/scholar?hl=en&btnG=Search&q=intitle:Fisypac+project+:+The+first+vehicle+integration+of+GENEPAC+fuel+cell+stack+{#}0>.
- [8] Jouin M, Gouriveau R, Hissel D, Pera M-C, Zerhouni N. Prognostics and health management of PEMFCState of the art and remaining challenges. *Int J Hydrogen Energy* 2013;38(35):15307–17. <http://dx.doi.org/10.1016/j.ijhydene.2013.09.051>. <http://linkinghub.elsevier.com/retrieve/pii/S036031991302274X>. <http://www.sciencedirect.com/science/article/pii/S036031991302274X>.
- [9] Garland N, Benjamin T, Kopasz J. DOE fuel cell program: durability technical targets and testing protocols. *ECS Trans* 2007;11(1):923–31. <http://dx.doi.org/10.1149/1.2781004>. <http://ecst.ecsdl.org/content/11/1/923.abstract>.
- [10] Wipke K, Sprik S, Kurtz J, Ramsden T, Ainscough C, Saur G. National fuel cell electric vehicle learning demonstration final report national fuel cell electric vehicle learning demonstration final report, NREL/DoE (July).
- [11] Ahluwalia RK, Arisetty S, Peng J-K, Subbaraman R, Wang X, Kariuki N, et al. Dynamics of particle growth and electrochemical surface area loss due to platinum dissolution. *J Electrochem Soc* 2014;161(3):F291–304. <http://dx.doi.org/10.1149/2.051403jes>. <http://jes.ecsdl.org/cgi/doi/10.1149/2.051403jes>.
- [12] Uddin MA, Park J, Ganesan S, Pasaogullari U, Bonville LJ, Molter T. A cationic contamination in PEFC cathode: a cause and effect study. *ECS Trans* 2015;66(24):29–36. <http://dx.doi.org/10.1149/06624.0029ecst>. <http://ecst.ecsdl.org/cgi/doi/10.1149/06624.0029ecst>.
- [13] Ji M, Wei Z. A review of water management in polymer electrolyte membrane fuel cells. *Energies* 2009;2(4):1057–106. <http://dx.doi.org/10.3390/en20401057>. <http://www.mdpi.com/1996-1073/2/4/1057/>.
- [14] Yousfi-Steiner N, Moçotéguy P, Candusso D, Hissel D, Hernandez A, Aslanides A. A review on PEM voltage degradation associated with water management: impacts, influent factors and characterization. *J Power Sources* 2008;183(1):260–74. <http://dx.doi.org/10.1016/j.jpowsour.2008.04.037>. <http://linkinghub.elsevier.com/retrieve/pii/S0378775308007337>.
- [15] Wilkinson DP, St-Pierre J. Durability. In: *Handb. fuel cells*. John Wiley & Sons; 2010.
- [16] Wu J, Yuan XZ, Martin JJ, Wang H, Zhang J, Shen J, et al. A review of PEM fuel cell durability: degradation mechanisms and mitigation strategies. *J Power Sources* 2008;184(1):104–19. <http://dx.doi.org/10.1016/j.jpowsour.2008.06.006>. <http://www.sciencedirect.com/science/article/pii/S0378775308011968>.
- [17] Petrone R, Zheng Z, Hissel D, Péra M, Pianese C, Sorrentino M, et al. A review on model-based diagnosis methodologies for PEMFCs. *Int J Hydrogen Energy* 2013;38(17):7077–91. <http://dx.doi.org/10.1016/j.ijhydene.2013.03.106>. <http://linkinghub.elsevier.com/retrieve/pii/S0360319913007465>.
- [18] Hernandez A, Hissel D, Outbib R. Modeling and fault diagnosis of a polymer electrolyte fuel cell using electrical equivalent analysis. *Energy Convers IEEE* 2010;25(1):148–60. http://ieeexplore.ieee.org/xpls/abs_all.jsp?arnumber=5353706.
- [19] Legros B, Thivel PX, Druart F, Bultel Y, Nogueira R. Diagnosis and modelling of proton-exchange-membrane fuel cell via and acoustic-emission measurements. *Cell* 2009;1–3 (July).
- [20] Asghari S, Mokmeli A, Samavati M. Study of PEM fuel cell performance by electrochemical impedance spectroscopy. *Int J Hydrogen Energy* 2010;35(17):9283–90. <http://dx.doi.org/10.1016/j.ijhydene.2010.03.069>. <http://linkinghub.elsevier.com/retrieve/pii/S0360319910005501>.
- [21] Zheng Z, Petrone R, Péra M, Hissel D, Becherif M, Pianese C, et al. A review on non-model based diagnosis methodologies for PEM fuel cell stacks and systems. *Int J Hydrogen Energy* 2013;38(21):8914–26. <http://www.sciencedirect.com/science/article/pii/S0360319913008550>.
- [22] Onanena R, Oukhellou L, Candusso D, Harel F, Hissel D, Aknin P. Fuel cells static and dynamic characterizations as tools for the estimation of their ageing time. *Int J Hydrogen Energy* 2011;36(2):1730–9. <http://dx.doi.org/10.1016/>

