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Modelling of polymer electrolyte membrane electrolyzer degradation and reliability

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Hydrogen production using electrolyzers can contribute to reduction in global emissions. A Polymer Electrolyte Membrane electrolyzer (PEME) splits water into hydrogen and oxygen, offering advantages in dynamic operation that enables rapid responses to fluctuations in power input and operating conditions. This reduces start-up time and allows immediate hydrogen generation. Ensuring the reliability and safety of PEMEs is critical for efficient hydrogen production. Degradation and failures of electrolysis cells can lead to hydrogen crossover, posing safety concerns and corrosion which reduce gas diffusion and conductivity, affecting performance. Although PEMEs have a lifetime of 40,000-60,000 hours, availability remains low due to frequent operational downtime and maintenance. This paper proposes a Petri net (PN) model which, in addition to reliability assessment, considers the degradation and maintenance processes of the stack. PNs are suitable for modelling complex, concurrent systems, making them ideal for capturing the dynamic interactions within the electrolyzer. By capturing such interactions, the PN approach is used for modelling both normal operations and potential failure scenarios. Such an approach can aid the hydrogen industry in making better asset management decisions, improving electrolyzer availability and safety. It can also inform the risk assessment process, enabling strategic investments in reliability and operational efficiency.

Keywords: Hydrogen, PEM electrolyzer, degradation, reliability.

1. Introduction

Polymer Electrolyte Membrane electrolyzers (PEMEs) play a vital role in sustainable hydrogen production by offering rapid responses to fluctuating energy inputs, particularly from renewable sources. Ensuring their reliability and efficiency is crucial for maintaining consistent hydrogen production and minimizing operational downtime. Despite the importance of reliability analysis in PEMEs, so far there is noticeable inadequate research in this area. However, studies on PEM fuel cells (PEMFCs), which share some structural and operational similarities with electrolyzers, provide valuable insights into the potential application of advanced reliability modelling techniques, such as Petri nets.

Petri nets (PN) have been widely used to model complex systems due to their ability to represent dynamic, concurrent, and stochastic processes effectively. In the context of PEMFCs, researchers have demonstrated the utility of Petri nets for reliability analysis. Wieland et al. (Wieland et al. 2009) modelled PEMFC stack reliability using a PN framework, classifying system behaviours into reversible, spontaneous, and degrading events. However, their approach oversimplified stack operations, overlooking realistic time-dependent degradation dynamics. Similarly, Whiteley et al. (Whiteley et al. 2015) developed a degradation model integrating Petri nets with operational data, enabling more precise predictions of voltage degradation.

A more comprehensive application was demonstrated by Fecarotti et al. (Fecarotti, Andrews, and Chen 2016), who used Petri nets to model not just the fuel cell stack but also supporting subsystems, such as hydrogen and cooling components. Their model incorporated failure and maintenance procedures to evaluate reliability and performance metrics. However, limitations in physical system dynamics hindered a detailed understanding of subtle degradation mechanisms. Vasilyev et al. (Vasilyev et al. 2021) extended this work by combining deterministic and stochastic models, offering insights into how operational cycles impact system lifetime.

While these studies establish the potential of PNs in reliability modelling, their focus on PEMFCs underscores the research gap in electrolyzer applications. This study aims to address this gap by developing a novel Petri net model tailored to PEME degradation and maintenance processes. It considers the electrochemical performance and monitoring of the cell voltage to determine the cell-stack degradation. The model's ability to simulate dynamic interactions of the cells offers a promising approach for enhancing the reliability and operational efficiency of hydrogen production systems.

The manuscript is organised as follows. Sec. 2 introduces PEME stack and degradation, Sec. 3 presents basic PN concepts. Sec. 4 gives the PEM model which includes the condition monitoring, degradation and maintenance processes. Finally, the results and discussion are provided in Sec. 5.

2. PEM stack and degradation

The main components that make up the electrolysis cell include: the membrane, electrocatalyst layer, porous transport layer and the bipolar plate. Cell and stack failure can result from degradation mechanisms of the components of the cell.

The membrane plays a crucial role in separating gaseous reaction products, facilitating proton transport, and supporting catalyst layers. Therefore, the membrane must possess desirable properties, such as chemical stability, mechanical strength, thermal stability, proton conductivity, and resistance to gas crossover (Feng et al. 2017; Papakonstantinou et al. 2020). Considering safety and reliability, the durability of the membrane is crucial, as its failure can result in hydrogen and oxygen recombination, as well as the potential buildup of explosive mixtures (Feng et al. 2017).

The catalyst layer facilitates the electrochemical reactions that occur during the electrolysis process. It splits water molecules into hydrogen and oxygen by catalyzing the transfer of electrons between the anode and cathode (Khan et al. 2018). The porous transport layer (PTL) simultaneously transports charges and heat between the electrode and the bipolar plate in the solid structure and gas/water in the pore space (Babic et al. 2017).

The bipolar plate (BP) is a metallic flow field based on a channel structure. It serves multiple functions within an electrolyzer stack, including the separation of individual cells, facilitation of heat and current transfer between cells, and distribution of reacting agents throughout the electrolyzer (Marcelo Carmo et al. 2013; Babic et al. 2017) (Lædre et al. 2017).

In a study conducted by Millet et al., (Millet et al. 2012) two failure mechanisms were identified: hydrogen-oxygen combustion and perforation of the membrane electrode assembly (MEA). If combustion occurs in a stack, the temperature inside is high enough to cause the melting of many cell components. The degradation of the membrane can result in its rupture, subsequently causing the accumulation of greater quantities of H_2/O_2 atmospheres within the cells, stacks, and liquid-gas separation units. Gas cross-permeation across the membrane causes the hydrogen generated at the cathode to become contaminated with oxygen, while the oxygen produced at the anode becomes contaminated with hydrogen. These can potentially cause complete destruction of the entire stack. The use of thicker membranes can be employed to monitor and mitigate this risk.

Hydrogen crossover reduces the purity of the produced hydrogen gas and can result in the mixing of hydrogen with oxygen on the cathode side which can lead to safety hazards or degradation of the electrolyzer components (Omrani and Shabani 2021; Martin et al. 2022). Therefore, crossover can lead to a reduction in the efficiency and performance of the electrolyzer, in addition to the mixing of gases, it can also result in the decreased selectivity of the electrochemical reactions. Various strategies are employed to reduce gas crossover, which are determined by the interaction between hydrogen and the membrane matrix. These methods can include passive approaches like enhancing membrane thickness, reactive approaches like facilitating hydrogen consumption by introducing Pt into either the CL or the current collector, or a combination of both (Bessarabov and Millet 2018).

Performance monitoring of a PEME electrolyzer involves tracking various operational parameters, and degradation rate is one of the most critical indicators of long-term efficiency and reliability. The degradation rate is strongly correlated with the operating conditions and is often accelerated by higher current density and temperature as reported in studies by (Suermann, Bensmann, and Hanke-Rauschenbach 2019) and (Papakonstantinou et al. 2020).

Intermittent operation from renewable energy has shown to impact the lifetime and performance of a PEME (Weiß et al. 2019). In their study, the authors showed that cycling into open circuit voltage (OCV) periods leads to a significant performance loss, particularly at high current densities of 3 A/cm^2, compared to periods where the cell voltage is not varied and potentiostatic at 1.3 V. The degradation rate was found to increase with the number of OCV periods over the 1000 h of test and attributed to the degradation of the anode catalyst.

Various key performance indicators (KPIs) for monitoring the condition of PEM electrolyzers are provided by polarization curve and cell voltage monitoring. Deep learning models, such as Graph Neural Networks (GNN) and distributed federated learning (DFL), can be used to analyze these KPIs to estimate the remaining useful life (RUL) and predict degradation trends (Zhang et al. 2024). A structured approach to maintenance is also made possible by using Petri net models to analyze deterioration states and maintenance procedures, based on the polarization curve and cell voltage variations.

These studies highlight the various factors contributing to performance loss in a PEM electrolyzer. The next chapters introduce the concepts of Petri Nets and how the condition monitoring process were modelled.

3. Petri Net concepts

A Petri Net (PN) is a flexible tool used for modeling and analyzing systems, combining graphical representation with mathematical precision. Structurally, it consists of two distinct types of nodes: **places** (P) and **transitions** (T), connected by arcs (A). Places are represented by circles and symbolize conditions or states, transitions are rectangles that represent events or actions, and arcs denote relationships between them. Tokens, often depicted as dots within places, illustrate the state or dynamic behavior of the system, such as resource availability or process execution. PNs are particularly effective for studying systems characterized by concurrency, synchronization, and resource sharing, making them invaluable for analyzing both static and dynamic properties of distributed, parallel, or stochastic systems.

An example of a simple PN is given in Fig. 1.

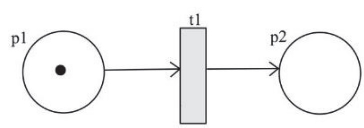

Figure 1. Petri net example

A transition is enabled when all its input places contain the required number of tokens as determined by the weights of their connecting arcs. The PEME model is described in the next section.

4. PEME model

The PEME model comprises of the condition monitoring module, degradation module and maintenance module.

4.1. Condition monitoring module

4.1.1. Polarization curve

The total voltage of a PEME cell, *Vcell*, is a sum of the open-circuit voltage (*E*), and overpotentials: activation (*Vact*), concentration (*Vcon*), and ohmic (*Vohm*) as shown in Eq. (1). It depicts the overall cell/stack performance at different load conditions.

$$V_{cell} = E + V_{ohm} + V_{act} + V_{trans} \quad (1)$$

These overpotentials represent losses due to different mechanisms, making this a critical equation for understanding overall efficiency of the cell and energy losses.

The Open-Circuit Voltage, *E*, is determined using:

$$E = E_0 + \frac{RT}{2F}\left[ln\left(\frac{pH_2\sqrt{pO_2}}{a_{H_2O}}\right)\right] \quad (2)$$

This equation calculates the open-circuit voltage using the standard electrode potential (E_0), operating temperature (*T*), gas partial pressures (pH_2, pO_2), and water activity (a_{H_2O}). It describes how thermodynamics influences voltage based on gas conditions and system temperature.

$$E_0 = 1.229 - 0.9 \times 10^{-3}(T - 298) \quad (3)$$

Eq. (3) adjusts (E_0) for deviations in temperature (*T*) from a reference value of 298 K. It reflects how chemical potential and system operating temperature influence the baseline voltage.

$$V_{act} = \frac{RT}{\alpha_{an}F}\text{arcsinh}\left(\frac{i}{2i_{an,0}}\right) + \frac{RT}{\alpha_{ca}F}\text{arcsinh}\left(\frac{i}{2i_{ca,0}}\right) \quad (4)$$

The activation overpotential is described using the Butler-Volmer equation shown in Eq. (4). It simplifies the activation overpotential in terms of current density (*i*), exchange current density (i_0), and charge transfer coefficients (αan, αca). This is crucial for quantifying losses due to electrode reaction kinetics.

The concentration overpotential equation is shown in Eq. (5)

$$V_{con} = \frac{RT}{4F}ln\left(\frac{CO_2}{C_{O_2,0}}\right) + \frac{RT}{2F}ln\left(\frac{CH_2}{C_{H_2,0}}\right) \quad (5)$$

It models the overpotential resulting from concentration gradients of reactants and products. It quantifies losses due to mass transport limitations using gas concentrations at the electrode-membrane interface.

The ohmic losses, *Vohm*, is shown in Eq. (6)

$$V_{ohm} = RiA \quad (6)$$

The ionic resistance is *R* and the activated membrane area is represented by *A*.

The polarisation curve of a single cell PEME is shown in Fig. 2. The cell voltage increases due to the overpotentials which become more significant at higher current densities.

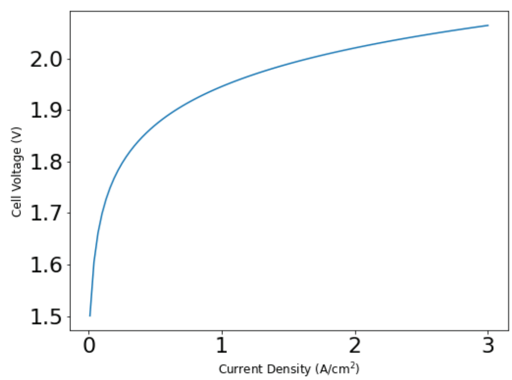

Figure 2. PEME polarization curve

This curve typically shows a steep increase at low current densities (activation region), a more linear increase in the middle (ohmic region), and another steep increase at high current densities (concentration region).

4.1.2. Cell voltage monitoring

The cell voltage of the PEME is monitored to identify individual cell performance, faults and to optimise the operation of the stack. A significant advantage of cell voltage monitoring (CVM) is its non-intrusive nature, allowing monitoring without disrupting normal operation.

Voltage deviations from expected levels can serve as indicators of degradation or failure modes, such as pinholes in membranes or carbon corrosion. The discrepancies between cells can result in imbalances that affect overall stack performance. This makes CVM an invaluable technique for improving stack availability and reliability. However, to pinpoint the root causes of degradation, CVM is often complemented by advanced characterization methods like electrochemical impedance spectroscopy (EIS) and high-frequency resistance (HFR) measurements. These techniques provide deeper insights into resistance contributions and dynamic phenomena within the stack.

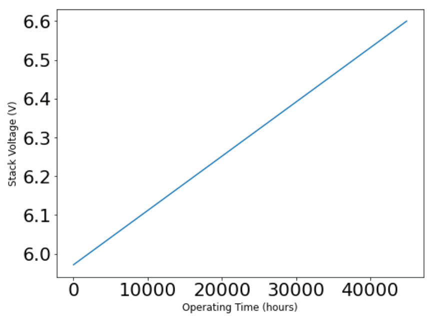

Figure 3. Stack voltage of a 3-cell PEM electrolyzer over time

A plot of the 3-cell stack voltage against operating time is shown in in Fig. 3. A linear degradation rate of 14μV/h (M Carmo et al. 2013) is used in this paper. The linear trend is consistent with the assumption that degradation occurs uniformly over time.

The increasing stack voltage is attributed to the increased resistance in the electrolysis process, primarily due to membrane thinning, catalyst degradation, and other stack component failures. These phenomena force the electrolyzer to require a higher voltage to maintain the same hydrogen production rate.

In the proposed approach, the polarization curve and individual cell voltage information feed into the degradation and maintenance PN modules which evaluates the condition of the stack over time. This is used to reduce system downtime, for efficient operation and maintenance.

4.2. Degradation and maintenance module

The degradation of the stack is modelled to ensure safety, and to optimize maintenance decisions. The places represent the degradation states of the stack, and the transitions represent the transition times between different states, as depicted in Fig. 4.

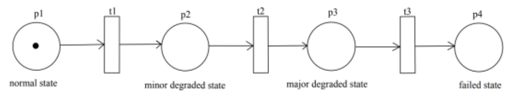

Figure 4. PN of the degradation process of PEME stack

Due to lack of lifetime data, the transitions are assumed to follow an exponential distribution with parameter, λ, and the Mean Time to Failure, μ, derived from the condition monitoring module.

$$\lambda = \frac{1}{\mu} \quad (7)$$

The sample time of a transition in the degradation PN is expressed as:

$$t = -\mu[\ln(X)] \quad (8)$$

The lifespan of Proton Exchange Membrane Electrolyzer (PEME) systems typically ranges between 10 and 20 years, with a degradation rate of less than 14 μV/h, translating to an annual average degradation of 2–4% (Bareiß et al. 2019). The degradation of the stack is measured by voltage decay, and the system is considered to have failed when the voltage of any cell reaches an end-of-life (EoL) threshold of 2.2 V. Since electrolyzer cells are connected in series, the total stack voltage is the sum of the individual cell voltages. Consequently, the degradation rate is calculated based on the time required for a single cell to reach the EoL voltage of 2.2 V at mid-point current density.

Table 1. Parameters of distributions for degradation transitions

	t1	t2	t3
μ	1.11E-04	4.46E-05	2.23E-05

To further define times to move between states, μ (shown in Table 1) for a cell to move to the minor degraded state is determined using the time when the stack voltage reaches 20% of its EoL value, similarly the major degraded state is described as reaching 50% of the EoL voltage.

Overall, in Fig. 5, the stack and individual cells go through four states: normal, minor degraded, major degraded, and failed. Degradation transitions (*t*1, *t*2, *t*3) follow the exponential distribution, while replacement transitions (*t*4, *t*5, *t*6) use normal distributions modeling replacement times with assumed values of 2 reflecting a 2-hour time period. The system starts in the normal state and may degrade progressively to failed state using transitions from *t*1 to *t*3, or if replacement is carried out the stack moves back to the normal state using the transitions from *t*4 to *t*6.

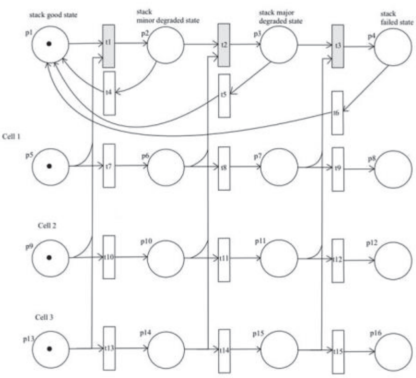

Figure 5. PN of the degradation and replacement process of 3-cell stack PEME

In terms of individual cell modelling, the normal operational states of cells 1, 2, and 3 are represented by places *p*5, *p*9, and *p*13, respectively. Using cell 1 as an example, it transitions from a normal state (*p*5) to a minor degraded state via transition *t*7. Further degradation occurs through transition *t*8, leading the cell to a major degraded state (*p*6), and finally, the cell transitions to a failed state (*p*7) through *t*9. Similarly, cells 2 and 3 experience degradation through transitions *t*10, *t*11, *t*12 (for cell 2) and *t*13, *t*14, *t*15 (for cell 3).

If any individual cell within the stack undergoes degradation, the performance deterioration is uniformly reflected across the entire stack, indicating a cascading impact on overall functionality. In this model, the stack is then replaced. In practice, when a failure occurs in a PEM electrolyzer cell within a stack, the affected cells are typically replaced rather than repaired. The intricate design and complexity of cell components make on-site repairs challenging, costly, and often less reliable compared to replacement. Additionally, electrolyzer manufacturers (OEMs) commonly recommend replacing the entire stack if the voltage or power demand increases by 20% or more compared to initial operating conditions (Mayyas et al. 2019). In addition, cell failure can significantly compromise the integrity of the electrolyzer because they can propagate rapidly, leading to extensive damage throughout the stack and posing safety risks, such as gas crossover (Millet et al. 2012).

This study focuses on corrective maintenance, where it is assumed that the functionality of cells are fully restored after a failure has happened. The condition monitoring module is directly linked to the maintenance process. After stack replacement, the places with degraded states are reset. Different failure states are associated with their maintenance actions.

5. Results and Analysis

A Monte Carlo simulation estimates the time spent in each state by iteratively triggering transitions, accumulates the time spent in each state, and normalizes them by the total simulation time to compute system availability.

The Monte Carlo simulation was configured with a maximum of 10,000 iterations and a simulation end time of 50,000. To ensure accuracy and computational efficiency, convergence was monitored using the checkStability function in Python. This function was employed to determine when the simulation results stabilized, allowing for early termination if convergence was achieved before reaching the maximum number of iterations. The simulation continued until either the maximum iteration limit (maxIter = 10,000) was reached or convergence was detected through the checkStability function.

The results of the PN model in Fig. 6 and Fig. 7 demonstrate the distribution of time spent in different states of the PEME stack. The horizontal axis represents the average availability, while the vertical axis represents the probability density, indicating the frequency of specific availability values for each state in the Monte Carlo simulations.

For the normal state, the distribution is highly concentrated around a value of 1, indicating that the system spends most of its operational time in this state. The minor degraded state has a much narrower distribution around a lower value, suggesting that cells transition briefly through this state before either progressing to a more severe state or being repaired. This transient nature is due to the rapid replacement actions triggered by early degradation detection, ensuring stack does not remain in this state for long.

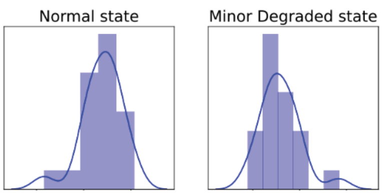

Figure 6. Distribution of availability values for normal state and minor degraded state

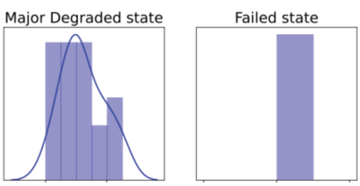

Figure 7. Distribution of availability values for major degraded state and failed state

In contrast, the major degraded state exhibits a broader distribution compared to the minor degraded state, indicating that cells remain in this state longer. This prolonged duration may result from the severity of degradation or delays in replacement. However, the overall proportion of time spent in this state remains low compared to the normal state, showcasing the effectiveness of the replacement process. Finally, the failed state distribution is narrowly concentrated near zero, indicating that the system rarely enters this state. This outcome highlights degradation is detected, and stack is replaced before leading to complete failure.

Overall, the dominance of the normal state, combined with the low occupancy of degraded and failed states, illustrates a modelling approach for analyzing a well-maintained PEME stack. The model emphasizes early detection and efficient replacement processes to prevent catastrophic failures. This behavior aligns with real-world scenarios where effective maintenance strategies ensure system reliability and operational safety.

6. Conclusions

This paper presented a comprehensive approach to modeling the reliability, degradation, and maintenance processes of PEMEs. By integrating a condition monitoring module to analyze polarization curves and individual cell voltage over time, the study enables detection of degradation and failure states. The proposed Petri net model captures the dynamic interactions within the electrolyzer stack, incorporating degradation of the individual cells and replacement processes. This framework not only enhances understanding of PEME operation and failure scenarios but also provides valuable insights for improving maintenance strategies, increasing system availability, and ensuring operational safety.

Future research will extend this work by including detailed inspection and maintenance strategies, and further expand the model to optimize electrolyzer performance and support strategic decision-making in the hydrogen industry.

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