Degradation-Aware Optimal Design of Grid-Connected PEM Electrolyzer Hydrogen Plants

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***Abstract*—This paper presents a novel formulation for optimizing the design and operation of the grid-connected Proton Exchange Membrane Water Electrolyzer (PEMWE)-based Hydrogen Plants (PEMWE-HP), considering their lifetime degradation impacts. First, the PEMWE voltage and membrane degradation are modeled, and their effects on PEMWE efficiency are analyzed. Then, a degradation-aware optimization framework is developed, encompassing optimizing the ratings of the PEMWE-HP components (i.e., PEMWE, AC/DC converter, compressor, and hydrogen storage) and the PEMWE-HP hourly energy consumption concurrently with the internal parameters of the PEMWE (i.e., cell area, membrane thickness, and cathodic pressure). The proposed formulation is validated using the IEEE 30-bus benchmark system. Results demonstrate that neglecting the degradation models leads to suboptimal sizing of the PEMWE-HP and the PEMWE internal design. Moreover, this omission results in a misleading estimation of the Levelized Cost of Hydrogen (LCOH), with inaccuracy reaching up to 30%.**

***Index Terms*—hydrogen plants, PEM water electrolyzer, degradation, optimization**

I. Introduction

Hydrogen has emerged as a promising alternative to fossil fuels and as an effective energy carrier due to its high energy density.Yet, hydrogen production is still relatively far from competing with fossil fuels in terms of cost and large-scale deployment, prompting numerous research and development efforts to develop highly efficient and cost-effective electrolyzer technologies [1]. Among different electrolyzer technologies, proton exchange membrane water electrolyzer (PEMWE) is considered a promising one as it offers several advantages over other technologies, including a compact design, high current density (over $2\,\mathrm{A/cm^2}$), and a small footprint , making it highly attractive for many industrial applications [2]. However, PEMWE uses high-activity noble metals, which increases its cost compared to other technologies. As such, several studies in the literature focused on reducing the Levelized Cost of Hydrogen (LCOH) production from PEMWE via enhancing its design and operational performance [2].

The work in [3] optimizes the sizing of a centralized PEMWE-based hydrogen plant (PEMWE-HP) for the provision of grid ancillary services and to supply the transportation sector. Similarly, in [4], the authors optimize the deployment of PEMWE-HP to enhance the power grid's generation capacity at a minimal cost. The authors in [5] introduce a comprehensive planning model that integrates the electricity, hydrogen, and transportation systems to reduce the capital expenditures, energy consumption, travel time, and carbon emissions. Nevertheless, the works in [3]–[5] model the PEMWE as having a constant conversion rate in kg/kWh, regardless of its loading level or operating conditions. This simplification neglects the variations in PEMWE efficiency and may result in suboptimal or inaccurate sizing.

The study in [6] optimizes the scheduling of PEMWE-HP operation concurrently with the virtual power plants to minimize the LCOH. The authors in [7] optimize the operation of a grid-connected PEMWE-HP, integrated with an on-site PV system and Battery Storage, to minimize the LCOH. Although the studies [6], [7] account for variations in PEMWE efficiency with its loading, they utilize simple empirical models that overlook critical PEMWE safety constraints, as well as key design and operational factors, such as membrane thickness, cell area, and cathodic pressure.

The work in [8], conducted by some of the present authors, develops a model that simultaneously optimizes both PEMWE-HP sizing and operation, while accounting for efficiency changes with the loading level and incorporating PEMWE safety constraints. Additionally, it optimizes PEMWE's internal parameters (i.e., membrane thickness, cell area, and cathodic pressure) based on its power supply and hydrogen demand profiles. However, the study in [8] does not adequately account for the impact of PEMWE degradation on its optimal internal design, sizing, and operation. PEMWE degradation is a critical factor in determining its operational lifetime, as efficiency loss becomes a major limiting factor after approximately 2,000 hours of operation. This is primarily attributed to the increasing rate of cell voltage degradation and to the membrane deterioration, which results in elevated gas crossover with prolonged operation [9]. Several studies in the literature, such as [10]–[13], have investigated the modeling of PEMWE degradation from an electrochemical perspective. Yet, none of these studies consider how degradation may affect the optimal sizing and operation of large-scale grid-connected PEMWE-HPs. In particular, the techno-economic impacts of PEMWE degradation—such as its influence on the LCOH and the sizing of various components within the EHP—remain unaddressed.

To address these shortcomings, this paper presents a comprehensive formulation for optimizing the sizing and scheduling of a large-scale grid-connected PEMWE-HP shown in Fig. 1—comprising a PEMWE, AC/DC converter, compressor, and hydrogen storage—while accounting for both voltage and membrane degradation models of the PEMWE. Moreover, the

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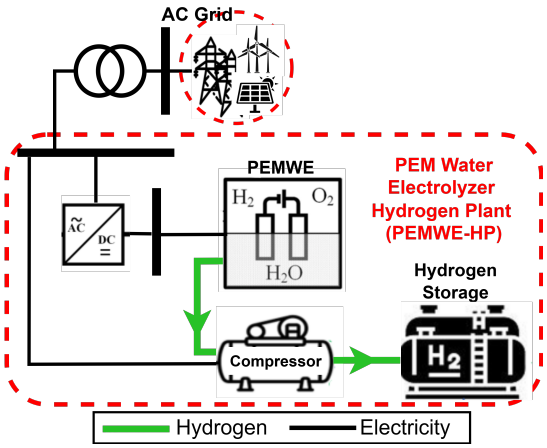

Fig. 1: Layout of the PEMWE-HP considered in the formulation.

proposed formulation is employed to evaluate the impact of incorporating these degradation models on the optimal design of PEMWE internal parameters, including membrane thickness, cell area, and cathodic pressure.

The rest of the paper is organized as follows: In section II, the PEMWE degradation models are presented and their impacts on the PEMWE's efficiency and safety constraints are analyzed. The formulation of the optimization model is explained in section III. In section IV, case studies and results are provided, and section V summarizes the paper's contributions and key findings.

II. Degradation Modeling of PEMWE and Its Influence on Efficiency and Safety Constraints

PEMWEs degrade over time due to prolonged or dynamic operation. This degradation reduces their efficiency and narrows their safe operating range. This section models this degradation and analyzes its impact on the PEMWE efficiency and the H_2/O_2 ratio.

PEMWE efficiency, η_{PEM}, consists of two components [1]: Voltage efficiency, η_V, and Faraday's efficiency, η_F. To incorporate the effects of time-dependent degradation, an operating hours index, h, is introduced into the efficiency calculations as follows:

$$\eta_{PEM}(h) = \eta_V(h) \cdot \eta_F(h) \tag{1a}$$

where,

$$\eta_V(h) = \frac{V_{LHV}}{V_{cell}(h)}, \quad \eta_F(h) = 1 - \frac{J_x(h)}{J} \tag{1b}$$

where V_{LHV} is the equivalent voltage of the lower heating value, V_{cell} is the PEMWE cell voltage, J_x is the equivalent current density of the gases (i.e., H_2, and O_2) crossover through the membrane, and J is the PEMWE operating current density.

The cell voltage consists of three components: the Nernst voltage (V_{ner}), activation overpotential (V_{act}), and resistive voltage (V_{res}) as below:

$$V_{cell} = V_{ner} + V_{act}(h) + V_{res}(h) \tag{2}$$

V_{ner} represents the minimum theoretical voltage required to split water; hence, its calculation is independent of the PEMWE degradation as follows [1]:

$$V_{ner} = 1.229 - 0.9 \times 10^{-3}\,(T_{cell} - 298) + \frac{RT_{cell}}{2F} \ln\left(\sqrt{\frac{\pi_{O_2}}{\pi_o}}\frac{\pi_{H_2}}{\pi_o}\right) \tag{3}$$

where, T_{cell}, R, F, π_{O_2}, π_{H_2}, and π_{O_2} are the operating temperature, gas constant, Faraday constant, O_2 anodic pressure, H_2 cathodic pressure, and reference pressure, respectively. The activation voltage is the overpotential needed to overcome the electrochemical reaction energy barrier.

Gas bubbles formation during PEMWE operation negatively affects its performance in two ways [10]: i) bubbles adhering to the electrode surface reduce the active catalytic area, thereby limiting electrochemical reaction rates and increasing the activation voltage; while ii) bubbles within the electrolyte impede proton transport, raising ionic resistance and contributing to higher resistive voltage. As such, both activation and resistive voltages depend on the operating hour index as provided in (4) and (5).

$$\begin{aligned} V_{\text{act}}(h) &= V_{\text{anode}} + V_{\text{cathode}} \\ &= \frac{RT_{\text{cell}}}{2\alpha F} \log\left(\frac{J}{(1-\theta)J_0 + \theta J_b}\right) + \frac{RT_{\text{cell}}}{F} \log\left(\frac{J}{J_0}\right) \end{aligned} \tag{4a}$$

$$\text{where, } \theta = 0.54 \cdot \frac{\log(3600\,h) + 3.556}{24.33 + \log(3600\,h)} + 0.36 \tag{4b}$$

J_0 and J_b are the exchange current densities for the electrode and bubble-covered surfaces, respectively. θ represents the fraction of the electrode surface covered by bubbles and is a function of the operating hours h. It is worth noting that bubble formation at the anode is more significant than the small bubbles formed at the cathode; therefore, θ affects only the anodic component of V_{act} [10]. The resistive voltage, provided in (5), comprises a non-degraded component specific to the membrane and a degradation component, which is derived from experimental high-frequency resistance (HFR) measurements [11]. This degradation component is modeled as a logarithmic function of operating hours.

$$V_{\text{res}}(h) = \left(\left(R_o + \frac{d_m(h)\delta_m}{\sigma_m}\right) + 0.0749 \cdot \log(3600\,h)\right) J \tag{5}$$

where R_o is the electrodes resistance, and d_m, δ_m, and σ_m represent the membrane thickness, swelling, and conductivity, respectively.

As the membrane degrades, it becomes thinner, which may slightly reduce the resistive voltage. However, the primary impact of membrane degradation is on Faraday's efficiency, as faradaic losses due to gas crossover are strongly influenced by membrane thickness. Thinner membranes lead to increased gas crossover. The time evolution of membrane thickness is modeled by the following differential equation [12]:

$$\frac{d_m}{dh} = \Delta d_m \cdot R_{\text{fluor}}(h) \tag{6}$$

where Δd_m is the membrane thinning per mole of fluoride released, and R_{fluor} is the fluoride release rate. R_{fluor} is a function of the operating hours as follows [13]:

$$\text{R}_{\text{fluor}}(h) = \max\left(0,\ \sum_{i=0}^{4} a_i(d_m) \cdot J^i - 0.0006 \cdot h\right) \tag{7}$$

The coefficients $a_i(d_m)$ are polynomial terms that vary with d_m. As such, the degraded membrane thickness after h operation hours is obtained as below:

$$d_m(h) = d_m(0) - \frac{d_m}{dh} \cdot h \tag{8}$$

As shown earlier in (1b), Faraday's efficiency is a function of gases crossover equivalent current density, $J_x(h)$, that is highly dependent on $d_m(h)$ as follows [1]:

$$J_x(h) = J_{H_2,x}(h) + J_{O_2,x}(h) = 2\left(F\,\mathrm{Pr}_{H_2,T}\,\frac{\pi_{H_2}+\pi_{O_2}}{d_m(h)\delta_m} + \frac{a_x}{d_m(h)\delta_m}J\right) \quad (9)$$

The H_2/O_2 ratio, Ψ_{H_2}, which represents the percentage of hydrogen relative to oxygen within the anodic compartment of the PEMWE, is a critical safety parameter, as it can form an explosive mixture [8]. This ratio, defined in (10), is highly dependent on the hydrogen crossover equivalent current density, $J_{H_2,x}$, which is a component of J_x. Consequently, it is significantly affected by membrane degradation.

$$\Psi_{H_2}(h) = \frac{J_{H_2,x}(h)}{J_{H_2,x}(h) + 0.5J} \quad (10)$$

To better understand the impact of both voltage degradation on the PEMWE performance, Fig. 2 (a) and (b), compare the cell voltage of PEMWE and the voltage efficiency at the beginning of life and after 10,000 hours of operation. As shown in the figure, after 10,000 hours operation, as current density increases, the cell voltage rises significantly while efficiency highly decreases compared to the beginning of life. Fig. 2 (c) illustrate the how hydrogen crossover increases for all current densities after 10,000 operational hours compared to at the beginning of life due to the degraded membrane. This clearly proves the great importance on the incorporation of time-dependent degradation mechanisms into long-term PEMWE modeling.

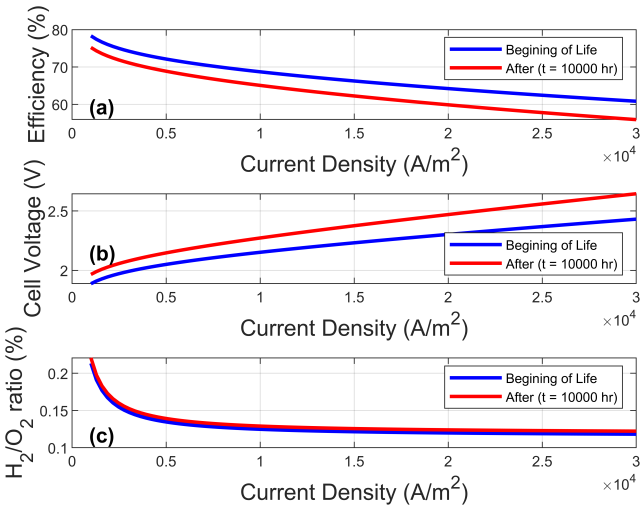

Fig. 2: (a) Efficiency, (b) Cell voltage and (c) hydrogen crossover at the beginning of life and after t=10,000 hrs of operation, for N117 membrane.

III. Formulation of the Optimization Problem

The proposed formulation aims to minimize the LCOH by optimizing the sizing of the four components in the PEMWE-HP (PEMWE, AC/DC converter, compressor, and hydrogen storage), as well as the hourly scheduling of the PEMWE power and hydrogen storage flow. This section first outlines the inputs and decision variables of the optimization problem, followed by the detailed formulation of the objective function and its associated constraints.

Inputs: The inputs of the proposed formulation are the i) hourly profile of the maximum power ($P^{max,t}$) that the PEMWE-HP is allowed to import; ii) hourly profile of the hydrogen demand (F_d^t); and iii) N_{MCS} Monte Carlo scenarios for the hourly profile of the electricity price ($Price_{elec}^t$). It is worth noting that the first input (the hourly profile of the maximum power) is the maximum power that can be drawn at the PEMWE-HP bus. This profile is obtained by solving the sub-optimization problem explained in [1], which considers the grid configuration, power flow, voltage, and thermal line capacity constraints, as well as the probability density functions of solar irradiation and wind speed. The Monte Carlo scenarios of the electricity price are generated based on historical electricity prices data, with each pricing scenario mc assigned a probability of PR^{mc}. Details on the generation of these scenarios and their probabilities are provided in [1].

Decision Variables: The decision variables include: i) the sizes of the four main components of the PEMWE-HP —namely, the PEMWE rating (P_{PEM}^{rate}), AC/DC converter rating (P_{Conv}^{rate}), compressor rating (P_{Cmp}^{rate}), and hydrogen storage capacity (M_{HS}); ii) the hourly power consumption of the PEMWE (P_{PEM}^t); and iii) the PEMWE internal design parameters, including the cell area (A_{cell}), membrane thickness (d_m), and cathodic pressure (π_{H_2}).

Objective Function: The optimization model aims to minimize the LCOH expressed as:

$$Min\left(\frac{CAPEX + \Sigma_y^Y\left(\frac{\sum_{mc=1}^{N_{MCS}} PR^{mc}\cdot OPEX(mc,y)}{\sum_{mc=1}^{N_{MCS}} PR^{mc}\cdot(1+r)^y}\right)}{\Sigma_y^Y\left(\frac{\Sigma_{t=1}^{\tau} F_{PEM}^t}{(1+r)^y}\right)}\right) \quad (11)$$

where Y is the PEMWE-HP life time in years, r is the yearly discount rate, $CAPEX$ is the capital expenditures obtained as:

$$CAPEX = P_{Conv}^{rate}C_{Conv} + P_{PEM}^{rate}C_{PEM} + P_{Cmp}^{rate}C_{Cmp} + M_{HS}C_{HS} \quad (12)$$

$OPEX(mc, y)$ represents the operating expenditures in year y of the PEMWE-HP lifetime, based on the electricity price under scenario mc, and is calculated as follows:

$$OPEX(mc,y) = \Sigma_{t=1}^{\tau}(P_{PEM}^t + P_{Cmp}^t)Price_{elec}^t(mc) + F_{HS}^t O_{HS} + P_{PEM}^t O_{PEM} + P_{Cmp}^t O_{Cmp} \quad (13)$$

C and O are the capital and operational costs of the corresponding components. For simplicity, while the PEMWE power consumption varies hourly within a year, the same hourly profile is assumed to repeat across all years of the PEMWE-HP lifetime. Accordingly, the PEMWE efficiency at each hour t is calculated as the average efficiency at that hour over the entire Y years of the plant's lifetime. Hence, F_{PEM}^t, the hydrogen flow produced by the PEMWE, is calculated as follows:

$$F_{PEM}^t = \frac{P_{PEM}^t\ \Sigma_{y=1}^{Y}\eta_{PEM}^t(y\times 8760)}{\Delta H_{LHV}\cdot Y} \quad (14)$$

P_{Cmp}^t is the compressor power consumption given as:

$$P_{Cmp}^t = \frac{2RT_{in}k\ F_{PEM}^t}{(k-1)\eta_{Cmp}}\left(\left(\pi_{HS}/\pi_{H_2}\right)^{(k-1)/k} - 1\right) \quad (15)$$

where R, T_{in}, π_{HS}, k, and η_{Cmp} are the gas constant, inlet gas temperature, hydrogen storage pressure, polytropic coefficient and compressor efficiency, respectively.

Constraints: The objective function provided in (11) is subject to power, PEMWE operation, and hydrogen production constraints. The power consumption of the PEMWE and the compressor must be limited to the maximum power as follows:

$$P^t_{PEM} + P^t_{Cmp} \leq P^{max,t} \tag{16}$$

The PEMWE operation is limited to the minimum and maximum current densities, and cathodic pressures as in (17) and (18).

$$0 \leq J^t \leq J^{max}_{PEM} \tag{17}$$

$$\pi^{min}_{H_2} \leq \pi^t_{H_2} \leq \pi^{max}_{H_2} \tag{18}$$

For safety purposes, the H_2/O_2 ratio must remain within the safe operating range. Accordingly, similar to the PEMWE efficiency, the H_2/O_2 ratio at each hour t is calculated as the average of the H_2/O_2 ratio at that hour across the entire Y years of the plant's lifetime, as given below:

$$\Sigma^Y_{y=1}\Psi^t_{H_2}(y\times 8760)/Y \leq \Psi^{safe}_{H_2} \tag{19}$$

The state of hydrogen in the storage is constrained by the minimum and maximum permissible limits as follows:

$$SoH^{min} \leq SoH^t \leq SoH^{max} \tag{20a}$$

where,

$$SoH^t = \frac{\sum^{q=t-1}_{q=1}(1-\gamma_{HS})(F^q_{PEM} - F^q_d) + F^t_{PEM} - F^t_d}{M_{HS}} \tag{20b}$$

γ_{HS}, F^t_d is the storage dissipation rate and hydrogen demand.

IV. Case Studies and Results

The proposed formulation is validated using the IEEE-30 bus benchmark transmission system [1], where a PEMWE-HP is connected to bus 18. The maximum power profile, P^{max}, is obtained from [1]. Historical data of the electricity prices given by the Independent Electricity System Operator (IESO) in Ontario, Canada, has been used to generate 1000 scenarios for four typical days representing the seasons of the year [14]. A transportation system hydrogen demand of four typical days is obtained as the mean of the hydrogen demand of each season using the data given in [1]. Three case studies are considered for the design of the PEMWE-HP, defined as follows: **i) Case I:** the baseline case without consideration of degradation models; **ii) Case II:** includes only voltage degradation; **iii) Case III:** incorporates both voltage and membrane degradation models. For all the cases, the membrane is chosen from the four Nafion commercially available thicknesses (N211-25.4μm, N212-51μm, N115-127μm, N117-183μm).

A. Case I: Optimal Design of EHP (Base Case)

For the four commercially available Nafion membrane thicknesses, Table I presents the corresponding optimal design of the PEMWE-HP, including the sizes of the four main components (i.e., PEMWE, AC/DC converter, compressor, and hydrogen storage), as well as the PEMWE internal parameters (i.e., cell area, membrane thickness, and cathodic pressure). The table also reports the optimal LCOH for each design.

As the membrane thickness increases from N211 to N117, the required operating pressure also rises. This is because thicker membranes can withstand higher pressure within the safety constraints. This leads to decreased compressor size, whereas thinner membranes require larger compressors due to higher hydrogen output rates at lower pressures. In contrast, thicker membranes operate more efficiently under high pressure, requiring smaller compressors. However, these gains are counterbalanced by the need for larger converter and electrolyzer units, as thicker membranes reduce proton transport, necessitating larger surface areas to sustain output. Despite these trade-offs, N212 shows the least LCOH, followed by N115. Fig. 3 (a) illustrates the efficiency curve of the optimal PEMWE design, while Fig. 3 (b) shows that the H_2/O_2 ratio of that optimal PEMWE remains within the safety limit of 10%.

B. Case II: Optimal Design of EHP Considering Voltage Degradation

Considering the time-dependent parameters for voltage degradation, which affect cell voltage and voltage efficiency, the optimized PEMWE-HP designs obtained for different membrane thicknesses are as stated in Table I. As shown in the table, the optimal PEMWE-HP design requires a thicker membrane of N117 compared to Case I, which uses a N212 membrane. However, in Case II, with consideration of voltage degradation, this optimal design shows that the LCOH estimation is higher than in Case I by 4%.

This change in the optimal membrane thickness compared to Case I is attributed to the fact that, in the voltage efficiency model, membrane degradation has a more pronounced effect on thinner membranes, leading to higher operational costs and a decline in system efficiency over time. As provided earlier in (5), the total resistance of the membrane electrode assembly consists of fixed and time-dependent degradation-induced resistances. For thinner membranes, the first term (fixed resistance) is inherently small while the time-dependent one constitutes a larger fraction of total resistance as degradation progresses. This disproportionately increases the voltage drop V_{res}, thereby reducing the total PEMWE efficiency η_{PEM}. A lower η_{PEM} necessitates greater electrolyzer power input to maintain the same hydrogen output, resulting in increased cost from elevated energy consumption.

Fig. 3 (a) presents the efficiency curve of the optimal PEMWE in Case II, which is consistently lower than that of Case I across all current densities. This reduction is attributed to the higher cell voltage resulting from the use of a thicker membrane and the effects of degradation. As shown in Fig. 3 (b), the H_2/O_2 ratio in Case II is also lower than in Case I, due to the reduced gas crossover associated with the thicker membrane.

C. Case III: Optimal Design of EHP Considering Voltage and Membrane Degradations

Considering the membrane degradation, which mainly affects Faraday's efficiency in addition to the voltage degradation, the optimized PEMWE-HP design is obtained as stated in Table I. Similar to Case II, the optimal design in Case III also requires a thicker N117 membrane. This is driven by both voltage degradation and membrane thinning over time, necessitating the use of a thicker membrane during the design stage. Furthermore, the additional reduction in PEMWE efficiency—caused by membrane degradation and increased gas crossover—leads

TABLE I: Optimized Results for Base Case, Voltage Degradation, and Combined Degradation Across All Membranes

Parameter	Base Case (Case I)				Voltage Degradation (Case II)				Combined Degradation (Case III)			
	N211	N212	N115	N117	N211	N212	N115	N117	N211	N212	N115	N117
Cathodic Pressure (bar)	6.56	13.14	30.09	37.21	6.26	13.43	31.22	41.51	5.82	13.65	36.06	51.88
Area (m^2)	15.76	17.41	18.24	20.30	16.52	17.05	17.77	17.72	18.13	17.00	17.30	17.73
Hydrogen Storage (100 kg)	226.45	218.52	229.91	242.69	207.65	207.80	223.27	238.74	202.998	193.536	206.789	224.987
Compressor Size (MW)	30.80	25.53	18.10	18.02	32.94	24.78	17.29	14.80	33.41	22.61	14.80	12.48
Converter Size (MW)	88.09	101.33	117.72	140.05	107.09	111.16	116.52	116.42	117.48	110.86	113.56	116.71
Electrolyzer Size (MW)	85.44	98.29	114.19	135.85	103.88	107.83	113.03	112.93	113.95	107.53	110.16	113.22
Price ($/kg H_2)	6.83	6.54	6.59	6.76	7.81	7.35	6.94	6.80	8.92	8.04	7.31	7.09

to higher PEMWE-HP power consumption, resulting in an increase in the LCOH by 4.5% compared to Case II and 8.5% compared to Case I. Fig. 3 (a) shows that the peak efficiency of optimal PEMWE is decreased from 70% for Case I to 60% in Case II. Fig. 3(b) shows that, unlike voltage degradation, accounting for membrane degradation (thinning) in Case III leads to increased gas crossover for the optimal PEMWE design compared to Cases I and II.

To further highlight the importance of accounting for degradation, the PEMWE design using the thinnest membrane (N211), in Case III, results in a 30% higher LCOH compared to Case I, which neglects degradation effects. In other words, ignoring degradation can lead to a 30% error in LCOH estimation.

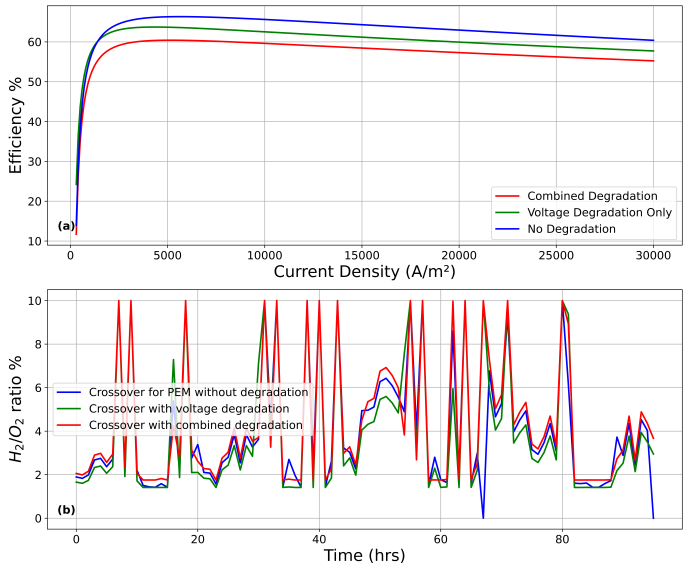

Fig. 3: (a) Efficiency and (b) Hydrogen crossover of PEMWE Electrolyzer for different degradation cases.

V. Conclusion

This paper proposes a novel mathematical formulation that optimizes the design and operation of the HP components (i.e., PEMWE, AC/DC converter, compressor, and hydrogen storage) and the internal parameters of the PEMWE (i.e., cell area, membrane thickness, and cathodic pressure) while accounting for the PEMWE degradation. The results indicate that ignoring degradation makes a huge difference in system design and cost estimation for hydrogen, such that the deviations can hit 30% at most. For instance, while membrane N212 is the optimal choice when degradation is not considered, the optimal PEMWE design shifts to N117 membrane when degradation effects are taken into account. Moreover, including degradation in the optimization increases the Levelized Cost of Hydrogen (LCOH) of the optimal PEMWE design by 8.5%, highlighting the utmost importance of incorporating long-term performance when optimizing PEMWE-based hydrogen plants. Considering the integration of the designed EHP with renewable intermittent resources is a possible future work.

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