

Article

Statistical Analysis of the Operating Conditions Influencing Green Hydrogen Production by a Reversible PEM Water Electrolyser

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Abstract

Improving the efficiency of proton-exchange membrane (PEM) water electrolysis for green hydrogen production requires systematic optimisation of interdependent operating conditions. The present study applies a face-centred central composite design (FCCD) combined with response surface methodology (RSM) to quantify the influence of three controllable parameters on the performance of a bench-scale PEM electrolyser: applied current, stack temperature, and membrane relative humidity. A reversible two-stack configuration with 16 cm² Nafion 117 membrane–electrode assemblies was operated across the design space (0.40–0.90 A, 18–45 °C, 50–100% RH), yielding 288 independent observations from 96 randomised runs. Four responses were evaluated: volumetric hydrogen evolution rate, Faradaic efficiency, specific electrical energy consumption, and stack voltage drift. The regression analysis identified applied current as the dominant factor governing hydrogen throughput, while membrane hydration exerted the strongest control over charge-utilisation and ohmic losses. Temperature exhibited a moderate but statistically significant positive effect, whereas feed-water resistivity emerged as a secondary practical lever for minimising energy consumption. Model adequacy was confirmed through analysis of variance and residual diagnostics, with adjusted coefficients of determination in the range 0.851–0.925 and predicted coefficients above 0.835 across all responses. Desirability profiling indicated an optimal operating window near 0.75 A, 42 °C, and 95% relative humidity, delivering a hydrogen production rate of approximately 7.4 mL min^{−1}, a Faradaic efficiency close to 98%, and a specific energy consumption of 4.5 kWh Nm^{−3}. These findings provide quantitative guidance for the design and operation of small-scale PEM electrolyzers under constrained laboratory and educational conditions. By integrating formal uncertainty quantification with response surface modelling and jointly treating membrane hydration and feed-water resistivity, the study provides a reproducible, uncertainty-quantified benchmark and a transferable optimisation workflow.



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1. Introduction

Over the past two decades, concerns regarding climate change and energy security have galvanised coordinated efforts to decarbonise the global energy system and reduce

reliance on fossil fuels [1]. Within this context, hydrogen has emerged as a versatile energy carrier whose production pathways are conventionally classified by colour designations that reflect their environmental impact [2]. Grey hydrogen, generated via steam methane reforming of natural gas, releases substantial carbon dioxide emissions, while brown or black hydrogen, derived from coal gasification, carries the highest greenhouse-gas burden. Blue hydrogen seeks to mitigate these emissions through carbon capture and storage or through alternative processes such as methane pyrolysis. Nuclear-powered electrolysis produces pink hydrogen, while only electrolysis driven exclusively by renewable electricity, designated green hydrogen, offers a genuinely carbon-free production route [3]. By splitting water into hydrogen and oxygen using wind, solar, or hydropower, green hydrogen not only avoids direct emissions but also enables the storage of surplus renewable generation and the decarbonisation of hard-to-electrify sectors, including heavy industry, maritime transport, and long-haul aviation [4]. Although its production cost remains higher than that of fossil-based alternatives, continued reductions in renewable electricity prices and ongoing improvements in electrolyser design are progressively closing the economic gap, positioning green hydrogen as a central pillar of the emerging sustainable energy economy [5].

Water electrolysis technologies, whether alkaline, proton-exchange membrane (PEM), anion-exchange membrane (AEM), or solid-oxide, face significant technical and economic challenges [6]. Elevated overpotentials at the electrodes, resistive losses within membranes and bipolar plates, and material degradation under sustained operation collectively contribute to increased energy consumption and operating expenditure. The selection of electrode materials is particularly critical: although noble metals such as platinum and iridium deliver excellent catalytic performance, their scarcity and cost motivate investigation of transition-metal and composite alternatives, particularly within AEM configurations that offer a combination of durability and affordability. In parallel, the integration of electrolyzers with intermittent renewable power sources demands rapid response capability and flexible operation, features that are more readily achieved in low-temperature PEM and AEM systems than in high-temperature solid-oxide cells [7]. Among these technologies, PEM electrolyzers are of particular interest for distributed green hydrogen applications owing to their compact footprint, high current density, and dynamic response to variable power input.

Overpotential losses at the electrodes represent a major barrier to efficient water electrolysis. Several studies have examined chemical and structural modifications of electrode materials intended to reduce overvoltage and enhance overall cell efficiency [8]. Targeted alloying and surface treatments have been shown to lower activation barriers, improving both hydrogen evolution reaction (HER) kinetics and long-term stability [9]. From a thermodynamic perspective, hydrogen offers an energy density approximately three times that of conventional fossil fuels on a mass basis, which reinforces its appeal as a high-energy carrier within a sustainable energy framework [10].

A substantial body of experimental work has examined how electrode materials, cell configurations, and operating parameters influence hydrogen generation rates in both alkaline and PEM systems. In the alkaline domain, Masjuki et al. [11] demonstrated that introducing a second cathode in a KOH electrolyser using SS-316L electrodes doubled the measured current, yielding 2150 cm³ of gas in 15 min under a one-anode/two-cathode configuration. Ezzahra et al. [12] subsequently showed that cathodes alloyed with zinc and chromium, specifically Zn₉₅Cr₅ and Zn₉₀Cr₁₀, outperformed alternative compositions at equivalent voltages and electrolyte concentrations. Kowthaman et al. [13] employed RSM to optimise voltage, molarity, and electrode spacing in a micro-patterned graphite cell, achieving a peak production rate of 2.3 mL min^{−1}. EL Kady et al. [14] reported that 3 mm SS-316 plates

in NaOH and KOH electrolytes attained a maximum cell efficiency of 51.12% at 4 V, declining at higher voltages, with 5% electrolyte concentration and 14 A current identified as optimal. Sekar et al. [15], focusing on engine-integrated applications, observed hydrogen production rates up to 2.5 L min^{-1} with 2 mm plate spacing in sodium hydroxide. Budiman et al. [16] further emphasised the role of electrolyte selection by demonstrating rapid hydrogen generation in a dry-type electrolyser using NaCl, while Shahabadi et al. [17] designed a membrane-free, multi-cell SS-316 electrolyser matched to 325 W solar modules and identified an optimal plate count of 15–18 per panel for maximising solar-to-hydrogen conversion.

Within the PEM domain, investigations have centred on membrane–electrode assembly (MEA) design, catalyst loading, and water management. Membrane hydration in particular has been identified as a critical determinant of ionic conductivity and therefore of cell efficiency, with insufficient humidification producing measurable increases in ohmic resistance and concurrent reductions in Faradaic efficiency [18]. Feed-water purity has similarly been recognised as a limiting factor, since ionic contaminants degrade membrane performance and accelerate catalyst poisoning [19]. Collectively, these studies indicate that electrode composition, cell geometry, and precise control of operating conditions are pivotal to performance; however, the divergent methodologies and metrics employed across experiments underscore the need for unified statistical frameworks capable of integrating findings and informing scalable system design.

The transition of PEM water electrolysis from laboratory demonstration towards gigawatt-scale green hydrogen production has recently been framed as a cell-level engineering problem, in which incremental improvements in membrane ohmic resistance, gas crossover, catalyst utilisation and porous-transport-layer design collectively determine the attainable efficiency and cost [20]. Because electricity dominates the levelised cost of hydrogen, even modest reductions in cell voltage at a given current density translate into tangible economic benefit, which places a premium on the selection of favourable operating conditions rather than on materials substitution alone. The operating environment is equally decisive for durability: current density, temperature and membrane hydration govern the rate of ionomer and membrane decay, carbon-support and catalyst corrosion, and the associated growth in ohmic and activation overpotentials, such that inadequate humidification or feed-water purity accelerates performance loss well before any catastrophic failure occurs [21]. These couplings are inherently multi-physics in character; as demonstrated in the closely related PEM fuel-cell literature, the local water content of the membrane simultaneously controls proton conductivity, reactant transport and the uniformity of the electrochemical reaction across the active area [22], while advanced electrode-engineering strategies, such as gradient electrospun-nanofiber architectures that tailor porosity and tortuosity to balance proton and mass transport, can markedly improve both performance and stability at reduced precious-metal loading [23]. Taken together, these considerations indicate that the performance of a PEM electrolyser is determined less by any single variable than by the interaction of several operating conditions, and therefore motivate the systematic, statistically grounded quantification of those interactions pursued in the present work.

Optimising electrolyser performance requires the simultaneous consideration of multiple interdependent variables, including applied current, temperature, membrane hydration, electrolyte concentration, electrode spacing, and feed-water quality, each of which influences hydrogen yield, energy efficiency, and system longevity. One-factor-at-a-time experimentation is poorly suited to capturing such interactions; by contrast, statistical design of experiments (DOE) approaches, and in particular RSM, enable the fitting of quadratic models that both reduce the number of experimental runs required and yield intuitive two-dimensional contour plots and three-dimensional response surfaces for iden-

tifying optimal conditions [24]. This methodology supports the modelling, forecasting, and optimisation of process outcomes such as hydrogen production rate and energy efficiency, and therefore provides an effective framework for enhancing electrolyser performance.

A rigorous understanding of how individual operating parameters and their interactions influence electrolyser performance is therefore essential for reducing production costs and optimising system design. Statistical and data-driven approaches can reveal the relative influence of each factor, identify optimal operating windows, and inform the development of control strategies that maximise efficiency and longevity. Despite the growing use of RSM for electrolyser optimisation, the existing literature is dominated either by alkaline HHO/dry-cell configurations [11–17] or by simulation-coupled PEM studies in which the response surface is fitted to finite-element or CFD output rather than to direct physical measurement. Comparatively few studies apply a fully experimental, replicated design of experiments to a low-temperature PEM electrolyser while simultaneously modelling four complementary responses (production rate, charge utilisation, energy consumption and short-term stability) and propagating measurement uncertainty through to the fitted models. The specific scientific gap addressed here is therefore: (i) the joint statistical treatment of membrane relative humidity and feed-water resistivity, two water-management variables that are frequently discussed qualitatively but rarely quantified within a single RSM framework; (ii) the integration of formal uncertainty quantification with response surface modelling, so that the significance of each effect is judged against experimental noise instead of being assumed; and (iii) the provision of a reproducible, low-cost benchmark and a transferable optimisation workflow for the large population of small-scale and educational PEM systems for which industrial-grade characterisation is unavailable. This distinguishes the present contribution from prior RSM optimisations of PEM electrolyzers, which have typically considered fewer responses, omitted water-quality effects, or relied on modelled rather than measured data.

The present study accordingly reports a comprehensive statistical analysis of the operating conditions influencing green hydrogen production in a bench-scale PEM water electrolyser. A face-centred central composite design was employed to examine the effects of applied current, bath temperature, and membrane relative humidity on four performance responses: volumetric hydrogen evolution rate, Faradaic efficiency, specific electrical energy consumption, and stack voltage drift. Supplementary experiments further quantified the influence of feed-water resistivity on specific energy consumption. By leveraging the resulting experimental dataset and multivariate regression techniques, the study quantifies the impact of each operating variable, identifies critical parameter thresholds, and proposes recommendations for electrolyser operation under variable renewable power input. The objective is to provide researchers and practitioners with actionable insights that support the deployment of small-scale green hydrogen systems. As global efforts to achieve Net-Zero targets continue, the optimisation of PEM electrolyser operation will play a pivotal role in enabling a resilient, low-carbon energy infrastructure.

2. Materials and Methods

This work combines controlled experimental measurements on a bench-scale PEM electrolyser with a structured parametric study using analysis of variance (ANOVA) and response surface regression, in order to quantify the influence of operating conditions on hydrogen production.

2.1. Experimental Work

2.1.1. Electrolyser Configuration

A reversible Horizon FCJJ11 teaching rig (Horizon Fuel Cell Europe s.r.o., Prague, Czech Republic) was reconfigured as a two-stack PEM water electrolyser and operated exclusively in electrolysis mode for the principal parametric campaign, with selected measurements in fuel-cell mode undertaken for cycle-symmetry assessment. Each transparent cell housed a 16 cm² Nafion 117 membrane–electrode (Horizon Fuel Cell Europe s.r.o., Prague, Czech Republic) assembly with a platinum loading of 0.3 mg cm^{−2} and carbon-paper gas-diffusion layers, clamped between graphite-composite bipolar plates machined with parallel flow fields. The two stacks were connected electrically in series, providing a 3–5 V operating window, while the hydrogen outlets were manifolded in parallel to a common collection line.

Nafion 117 was used because it is the perfluorosulfonic acid membrane fitted as standard in the reversible membrane–electrode assemblies of the platform and remains the most widely reported benchmark membrane for low-temperature PEM water electrolysis, ensuring that the present results are directly comparable with the extensive existing literature. Its relatively large thickness ($\approx 183\ \mu\text{m}$) also confers the mechanical and chemical robustness appropriate to repeated electrolysis/fuel-cell cycling on a teaching rig, at the cost of a somewhat higher ohmic resistance than thinner membranes which is a trade-off that makes the hydration and water-quality effects quantified here particularly visible.

The influence of electrode positioning within the cell on measured oxygen evolution behaviour has been highlighted in the recent literature [25]; in the present fixed-geometry rig, the electrode spacing and alignment are held constant across all runs, so that the reported effects are attributable to the varied operating conditions rather than to geometric variation.

A WEIR 4000 bench supply (Weir Electronics Ltd, UK) delivered galvanostatic current in the range 0.40–0.90 A via 4 mm shrouded leads, and a Keysight 34401A multimeter (Keysight Technologies, Inc., Santa Rosa, CA, USA) logged terminal voltage at 1 Hz. Stack temperature was controlled by immersing the cells to port level in a 5 L Grant TC120 thermostatic bath (Grant Instruments (Cambridge) Ltd, Shepreth, Cambridgeshire, UK) maintained between 18 and 45 °C with a stability of $\pm 0.1\ ^\circ\text{C}$, using a type-K thermocouple attached to the cathode end-plate for feedback. Membrane hydration was regulated by a coiled humidifier located in the same bath, yielding feed-gas relative humidity (RH) between 50 and 100% as measured with a Vaisala HMP110 probe (Vaisala Oyj, Vantaa, Finland) ($\pm 1\%$ RH). The arrangement is shown in Figure 1.

Hydrogen and oxygen were collected separately in 50 mL inverted burettes filled with bath water. Meniscus position was recorded at 10 s intervals using a 4K camera and converted to standard-temperature-and-pressure (STP) gas volume through simultaneous measurement of ambient pressure and bath temperature. All wetted connections used 3 mm internal-diameter platinum-cured silicone tubing, which was bubble-tested prior to each experimental block, and the complete assembly was operated inside a ventilated fume cupboard fitted with a fixed hydrogen detector calibrated to alarm at 1% vol.

Deionised feed water of 18 M Ω cm resistivity (Elga Purelab) was used as the baseline. Lower resistivities of 5, 1, and 0.8 M Ω cm were obtained through the controlled addition of analytical-grade NaCl for the subset of runs examining the influence of feed-water conductivity.

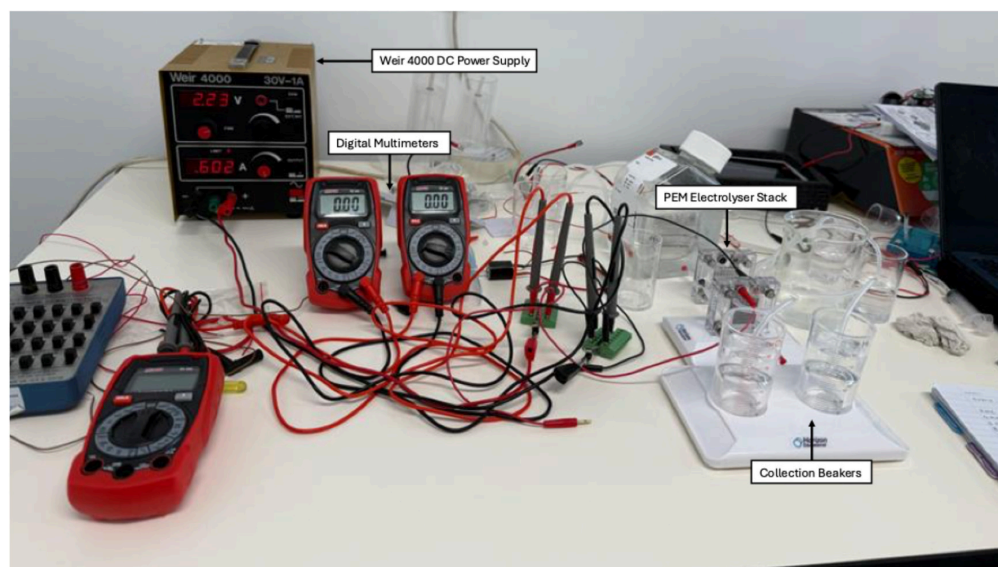


Figure 1. Experimental Setup.

2.1.2. Operating Variables and Responses

The experimental matrix focused on three controllable operating conditions:

- Electrolysis current I : 0.40–0.90 A (galvanostatic control).
- Bath and stack temperature T : 18–45 °C.
- Membrane relative humidity RH : 50–100%.

At each design point, the rig was allowed to reach steady state, defined as a stack voltage variation below 5 mV over 5 min, before data acquisition commenced. The following responses were measured or derived:

- Volumetric hydrogen evolution rate Q (mL min^{-1} , STP), calculated from corrected burette displacement.
- Faradaic efficiency F (%), obtained from the ratio of measured Q to the theoretical hydrogen volume predicted by Faraday's law at the applied current.
- Specific electrical energy consumption E_{spec} (kWh Nm^{-3}), computed from time-integrated stack power and corresponding hydrogen throughput.
- Stack voltage drift V_t (mV min^{-1}), defined as the linear slope of terminal voltage over the 300 s sampling window.

Each run was executed three times on different days in order to capture day-to-day variability, giving 288 independent observations across the electrolysis conditions. Representative primary data, including hydrogen evolution rate as a function of current and temperature at 100% RH, and the effect of RH and water resistivity on Faradaic efficiency and specific energy consumption, are presented in Section 3.

Feed-water resistivity was examined as a dedicated controlled sub-campaign rather than as a factor within the randomised design. Varying it required the deliberate, stepwise addition of analytical-grade NaCl to fixed resistivity set-points (18, 5, 1 and 0.8 $\text{M}\Omega \text{ cm}$), with flushing of the feed circuit between set-points to prevent carry-over, a procedure that is incompatible with the fully randomised run order used for the three continuously controllable factors. Isolating resistivity at otherwise fixed reference conditions (0.70 A, 30 °C, 100% RH) allowed its ohmic contribution to be attributed unambiguously, without confounding by simultaneous variation of current, temperature or humidity. The results of this sub-campaign are reported in Section 3.1.3.

The optimisation was confined to the three operating conditions that can be varied independently on this platform. Operating pressure and back-pressure are fixed at ambient

by the open burette-collection architecture and cannot be regulated on the rig; water flow to the cell is governed by the fixed humidifier/immersion configuration rather than by an adjustable pump; and catalyst loading and membrane thickness are properties of the sealed, factory-assembled membrane–electrode assemblies, which cannot be altered without destroying the cell. These variables are consequently design parameters of the MEA rather than controllable operating conditions of this apparatus, and their optimisation would require a reconfigurable, pressurised single-cell test station.

2.1.3. Measurement Uncertainty

Measurement uncertainties were evaluated following the GUM framework [26] and propagated to each reported response. For the volumetric hydrogen evolution rate Q , the propagation was carried out through the full standard-temperature-and-pressure (STP) correction chain. Each measured gas volume was corrected to STP through Equation (1), in which V_{meas} is the collected volume, p_{amb} the ambient pressure, p_w the water vapour partial pressure at the bath temperature, T_{bath} the bath temperature, and p_0 and T_0 the reference pressure and temperature [27].

$$V_{STP} = V_{meas} \cdot \frac{p_{amb} - p_w}{p_0} \cdot \frac{T_0}{T_{bath}} \quad (1)$$

Four input quantities contribute to the corrected volume: burette graduation (± 0.2 mL), timing resolution (± 0.2 s), bath-temperature correction (± 0.05 °C) and ambient-pressure measurement (± 0.5 kPa). Each was assigned a rectangular distribution and converted to a standard uncertainty by division by $\sqrt{3}$. Treating the inputs as independent, the relative combined standard uncertainty of the corrected volume follows from the first-order propagation of Equation (2), in which the four terms correspond to these contributions in the order given [27].

$$\left(\frac{u_c(V_{STP})}{V_{STP}} \right)^2 = \left(\frac{u_V}{V_{meas}} \right)^2 + \left(\frac{u_p}{p_{amb} - p_w} \right)^2 + \left(\frac{u_T}{T_{bath}} \right)^2 + \left(\frac{u_t}{t} \right)^2 \quad (2)$$

Substituting the input values yields a relative combined standard uncertainty of 1.8% on the hydrogen evolution rate; applying a coverage factor $k = 2$, corresponding to approximately 95% confidence, gives the expanded uncertainty of $\pm 3.6\%$ quoted throughout this work. The uncertainty in the applied current, obtained by combining the supply calibration (1% of full scale) with the multimeter accuracy, is approximately 0.6%. Combining this with the volume uncertainty through Equation (3) gives an expanded uncertainty of $\pm 1.2\%$ on the Faradaic efficiency [27].

$$\frac{u_c(F)}{F} = \sqrt{\left(\frac{u_c(V_{STP})}{V_{STP}} \right)^2 + \left(\frac{u_I}{I} \right)^2} \quad (3)$$

The uncertainty in stack voltage drift, dominated by metre accuracy and time-stamp resolution, was evaluated as 0.04 mV min^{-1} .

2.2. Parametric Study

2.2.1. Design of Experiments and Response Selection

A face-centred central composite design (FCCD) combined with RSM was selected because it offers the most favourable balance, for a three-factor continuous problem, between model richness and experimental economy. Unlike one-factor-at-a-time experimentation, RSM resolves two-factor interactions and quadratic curvature; unlike a full three-level factorial it does so with substantially fewer runs; and unlike Box–Behnken designs, the

face-centred variant confines all axial points to the physically realisable factor bounds of the rig, avoiding out-of-range set-points that the hardware cannot deliver. Relative to Taguchi screening, RSM yields continuous predictive equations and desirability-based multi-objective optimisation rather than discrete level rankings, and relative to purely data-driven machine-learning surrogates it remains interpretable and tractable with the present sample size. The approach is the most appropriate and efficient choice for the number of factors, the continuous factor ranges, and the interpretability required here.

To quantify the influence of operating conditions on hydrogen production, a FCCD was adopted. This design balances the estimation of quadratic terms with run economy while minimising aliasing among factors. The three continuous factors (current I , temperature T , and relative humidity RH) were explored at five coded levels (-1 , -0.5 , 0 , 0.5 , 1), producing 96 randomised runs with six replicated centre points for the estimation of pure error. The coded and natural levels of the three factors are summarised in Table 1.

Table 1. PEM electrolyser factors and coded levels used in the face-centred central composite design.

Coded Level	Current I (A)	Temperature T ($^{\circ}\text{C}$)	Relative Humidity RH (%)
-1.0	0.40	18	50
-0.5	0.55	27	70
0.0	0.70	32	75
$+0.5$	0.85	40	85
$+1.0$	0.90	45	100

The number of runs was fixed by the chosen face-centred central composite design rather than selected arbitrarily: for three factors the FCCD comprises factorial, axial and replicated centre points, and the 96 randomised runs (with six centre-point replicates and three-day replication) provide ample degrees of freedom for the full quadratic model while retaining run economy. The adequacy of this sample size is confirmed a posteriori by the fit statistics of Table 2 which together indicate that the design captured the response surfaces with sufficient resolution and that further runs would yield diminishing statistical returns.

Table 2. Fit statistics for the reduced quadratic response surface models.

Response	Std. Dev.	R^2	Adjusted R^2	Predicted R^2	C.V. (%)	Adequate Precision
H_2 rate, Q (mL min^{-1})	0.32	0.932	0.925	0.910	5.8	32.4
Faradaic efficiency, F (%)	2.10	0.893	0.874	0.856	2.4	27.8
Specific energy consumption, Espec (kWh Nm^{-3})	0.14	0.905	0.889	0.872	3.1	29.6
Voltage drift, V_t (mV min^{-1})	0.03	0.872	0.851	0.835	8.2	25.1

Each design point was replicated on three separate days to capture between-day variability (ambient temperature, humidifier equilibration and water batch) in addition to the within-run repeat measurements, providing an independent estimate of pure error while keeping the total campaign within the practical duty cycle of the rig. This replication level, combined with the six replicated centre points of the FCCD, was sufficient to estimate pure error and lack-of-fit with the precision reported in Table 2, so that additional replication would not have materially changed the fitted models.

The factor bounds were dictated by the operating envelope of the apparatus. The current range (0.40–0.90 A) spans from the lowest current giving stable, readily measurable gas evolution to the maximum safe galvanostatic output of the supply for the 16 cm² MEA; the temperature range (18–45 °C) extends from ambient bath temperature to the upper limit imposed by the thermostatic bath and the temperature rating of the transparent cell bodies; and the relative humidity range (50–100%) runs from the lowest humidification at which the demonstration stack maintained stable operation up to saturation. These bounds also lie within the recommended operating window of the Nafion 117 membrane (Horizon Fuel Cell Europe s.r.o., Prague, Czech Republic). Confirmatory scoping runs at the extremes were used to verify stable, repeatable operation before the randomised campaign began.

The principal responses selected for statistical modelling were the volumetric hydrogen evolution rate Q , Faradaic efficiency F , specific energy consumption E_{spec} , and voltage drift V_t . These responses characterise, respectively, production rate, charge utilisation, energy efficiency, and short-term stability, and therefore capture the principal performance dimensions of PEM water electrolysis for green hydrogen applications.

2.2.2. ANOVA and Regression Modelling

A multi-way ANOVA with Type-II sums of squares was applied to the full quadratic model in order to screen main effects and two-factor interactions at a significance threshold of $\alpha = 0.05$. Residual normality was assessed through Shapiro–Wilk and Anderson–Darling tests, while homoscedasticity was evaluated using Breusch–Pagan and White statistics. Reduced quadratic response surface models were subsequently obtained by backward elimination, with terms of $p > 0.10$ removed sequentially while the adjusted coefficient of determination, the predicted coefficient of determination, and the Akaike information criterion (AIC) were monitored in order to avoid overfitting. Cook’s distance and leverage were inspected to identify influential observations; no run exceeded the conventional unit threshold, and therefore all data were retained.

The general form of the fitted quadratic response surface for each response y is expressed as

$$y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_i^2 + \sum \sum \beta_{ij} x_i x_j + \varepsilon \quad (4)$$

where x_i denotes the coded factors (current, temperature, relative humidity), β_0 is the intercept, β_i , β_{ii} , and β_{ij} are the linear, quadratic, and interaction coefficients respectively, and ε denotes the residual error term.

Fit statistics for the four responses are presented in Table 2. The coefficients of determination R^2 span the range 0.872–0.932, with adjusted R^2 values above 0.851 and predicted R^2 values above 0.835 across all responses. The small difference (<0.04) between adjusted and predicted R^2 values indicates strong predictive correlation, while adequate precision ratios exceeding 25 confirm sufficient signal-to-noise ratios across the experimental data. Residual diagnostics further confirmed model adequacy, with no significant lack-of-fit ($p > 0.31$ across responses) and residuals passing Shapiro–Wilk normality tests ($p > 0.19$). Homoscedasticity was verified, ensuring reliable inference across the operational factor space studied.

2.2.3. Optimisation and Response Surface Exploration

The simultaneous maximisation of hydrogen rate and Faradaic efficiency and minimisation of specific energy consumption constitute a multi-objective optimisation problem, here addressed through the Derringer–Suich desirability framework applied to the response surface models. Multi-objective optimisation of PEM water electrolysis has recently attracted considerable attention, including approaches based on machine-learning and evolutionary algorithms that trade off efficiency, hydrogen output and operating cost

across large design spaces [28,29]. The present desirability-based treatment provides a transparent, low-dimensional counterpart to these methods that is well matched to the three-factor experimental design and directly interpretable for small-scale operation.

The validated regression models were used to generate response surface and contour plots for Q , F , and E_{spec} as functions of the coded factors, thereby enabling visual assessment of linear, interaction, and curvature effects. Desirability profiling following the approach of Derringer and Suich was subsequently applied in order to identify operating windows that simultaneously maximise hydrogen evolution rate and Faradaic efficiency while minimising specific energy consumption. The combined desirability function indicated an operating region centred at approximately 0.75 A, 42 °C, and 95% RH, delivering a hydrogen rate of approximately 7.4 mL min^{−1}, a Faradaic efficiency near 98%, and a specific energy consumption of approximately 4.5 kWh Nm^{−3}, all within the expanded uncertainty bounds of the experimental data. These statistically derived operating conditions provide practical guidance for educational and small-scale PEM electrolyzers operating under constrained laboratory resources.

2.3. Scope and Limitations

The present investigation was confined to the operating envelope of a bench-scale, reversible teaching electrolyser (Horizon FCJJ11) in order to isolate the influence of three continuously controllable operating conditions (applied current, stack temperature and membrane relative humidity) under well-characterised, reproducible conditions. Several analyses that would ordinarily accompany a cell-materials study, including electrochemical impedance spectroscopy (EIS), formal Tafel analysis, hydrogen-purity determination by gas chromatography and multi-hundred-hour durability testing, lie outside the mechanical and instrumentation limits of this platform: the sealed, low-power stack (3–5 V, <1 A) provides neither the reference-electrode access and four-point sensing required for impedance and Tafel measurements, nor the gas-sampling ports required for chromatographic purity analysis, and its finite hydrogen reservoir precludes meaningful accelerated-degradation protocols. Rather than compromise data quality by attempting measurements the rig cannot support reliably, the study focuses on the quantities the platform resolves with quantified uncertainty which are volumetric hydrogen rate, Faradaic efficiency, specific energy consumption and short-term voltage drift and on the statistical relationships among them. The optimal operating window reported here is accordingly intended as validated guidance for small-scale and educational PEM systems, and as a transferable optimisation methodology, rather than as a set of parameters for direct transposition to pressurised, membrane-optimised industrial stacks. The electrochemical, purity and durability characterisations noted above are identified as priorities for future work on an instrumented, industrially representative single cell.

Long-term durability was not assessed, since the finite-reservoir architecture of the demonstration rig precludes the continuous multi-hundred-hour operation required for accelerated-degradation testing. Nevertheless, the stack voltage drift response measured over the 300 s galvanostatic windows provides a short-term stability indicator: the low drift rates observed (of the order of 0.01 V min^{−1}) and their sensitivity to hydration are consistent with reversible, water-management-driven resistance changes rather than irreversible degradation. In longer-duration PEM operation, the dominant degradation pathways reported in the literature would additionally contribute; systematic evaluation of these mechanisms under controlled accelerated-stress protocols is identified as a priority for future work on an instrumented single cell.

3. Results and Discussion

The experimental campaign generated 288 independent observations across the 96-run face-centred central composite matrix, complemented by supplementary measurements on feed-water resistivity and by a reduced fuel-cell characterisation undertaken to assess cycle symmetry. All gas volumes reported in the following sections have been corrected to standard temperature and pressure (0 °C, 1 atm) through simultaneous measurement of bath temperature and ambient pressure, and through subtraction of the water vapour partial pressure at the bath condition. Before the factor-specific analysis, the overall structure of the experimental dataset is summarised in Figure 2. Figure 2a–d present the distributions of the four measured responses, hydrogen evolution rate, Faradaic efficiency, specific energy consumption and stack voltage drift, respectively, across all 96 randomised runs (288 observations), while panel (e) plots the hydrogen evolution rate against run order. The responses span the ranges expected from the design settings, with the hydrogen rate extending from approximately 3.7 to 8.8 mL min^{−1} and Faradaic efficiency remaining above 94% throughout. The absence of any systematic trend in the run-order panel confirms that the randomised execution sequence introduced no temporal drift or block effects, thereby supporting the statistical independence assumed in the subsequent ANOVA and response surface modelling.

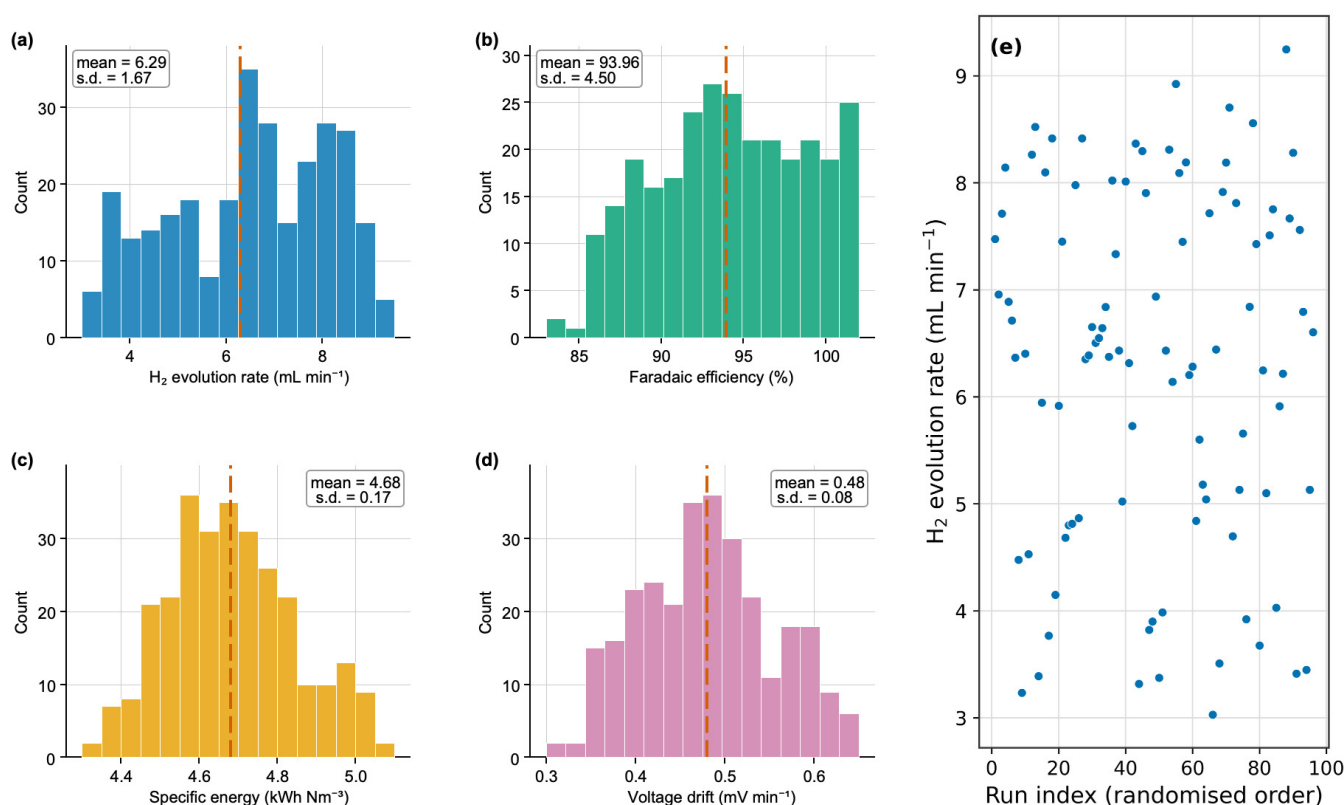


Figure 2. Distributions of the four measured responses across the 96 randomised runs: (a) H₂ evolution rate, (b) Faradaic efficiency, (c) Specific energy consumption, (d) Stack voltage drift, and (e) H₂ evolution rate versus run index.

Steady-state operation was confirmed at each design point through the terminal-voltage stability criterion defined in Section 2.1.2 (variation below 5 mV over 5 min). The Results are presented in three parts: the electrolysis-mode performance and its dependence on the three primary factors and on feed-water purity (Section 3.1), the reversible fuel-cell characterisation used to evaluate cycle symmetry (Section 3.2), and the integrated

statistical assessment drawing together the four response surface models and the derived operating-window recommendations (Section 3.3).

3.1. Electrolysis Mode: Hydrogen Production Performance

The electrolysis mode was characterised systematically across the factorial design space, with the hydrogen evolution rate, Faradaic efficiency, specific electrical energy consumption, and stack voltage drift measured at each design point. The analysis progresses from the two primary continuous factors (current and temperature) to the secondary factor (membrane relative humidity) and finally to the feed-water resistivity sub-study, which was conducted at fixed baseline conditions in order to isolate the ionic-contaminant contribution to ohmic losses.

3.1.1. Influence of Applied Current and Stack Temperature on Hydrogen Evolution Rate

The hydrogen evolution rate was observed to scale approximately linearly with applied current across the full temperature range investigated, with the measured rate doubling from 3.68 mL min^{-1} at 0.40 A to 8.82 mL min^{-1} at 0.90 A under the reference condition of 45°C and 100% relative humidity. This linear scaling is consistent with the theoretical prediction of Faraday's law within the expanded experimental uncertainty of $\pm 3.6\%$, and confirms that charge utilisation within the membrane-electrode assembly remained near unity across the 2.25-fold current span. Temperature exerted a systematic positive effect on the production rate, with an average slope of approximately $0.11 \text{ mL min}^{-1} ^\circ\text{C}^{-1}$ that became more pronounced at the upper end of the current range. The enhanced responsiveness at high current is attributable to the combined contributions of reduced gas viscosity, improved bubble detachment from the electrode surface, and accelerated electrode kinetics, all of which scale with thermal activation. Faradaic efficiency remained consistently above 94% across the design space at full RH and approached 100.6% at the highest current-temperature combination, thereby demonstrating robust charge utilisation throughout the factor domain. The complete dataset is summarised in Table 3, while the corresponding response surface is illustrated in Figure 3.

Table 3. Hydrogen evolution rate and Faradaic efficiency as functions of applied current and stack temperature (RH = 100% , $18 \text{ M}\Omega \text{ cm}$).

Run Number	Current (A)	Bath T ($^\circ\text{C}$)	V_{H_2} (mL)	Time (s)	Rate (mL min^{-1})	Faradaic η (%)
1	0.40	18	18.4	300	3.68	94.2
2	0.40	30	19.1	300	3.82	97.7
3	0.40	45	19.3	300	3.86	98.8
4	0.70	18	32.9	300	6.58	95.8
5	0.70	30	34.0	300	6.80	99.0
6	0.70	45	34.4	300	6.88	100.1
7	0.90	18	42.3	300	8.46	96.5
8	0.90	30	43.7	300	8.74	99.7
9	0.90	45	44.1	300	8.82	100.6

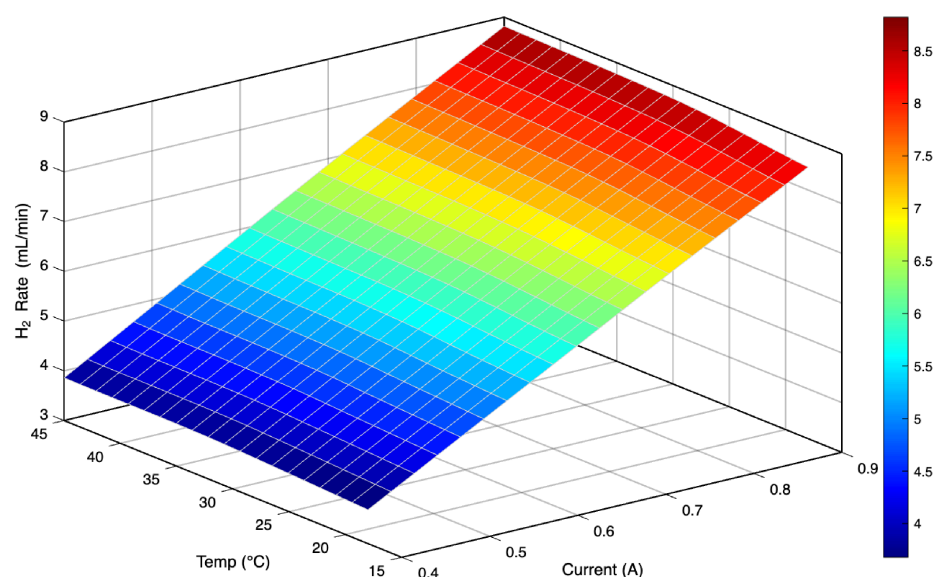


Figure 3. Response surface plot of hydrogen evolution rate as a function of electrolysis current and stack temperature at constant relative humidity (100%) and baseline feed-water resistivity (18 MΩ cm).

The Faradaic efficiency of 100.6% recorded at 0.90 A and 45 °C (Table 3) marginally exceeds unity but lies well within the $\pm 3.6\%$ expanded uncertainty of the volumetric measurement and should not be interpreted as physical super-unity conversion. It reflects the combined measurement uncertainty in gas-volume determination and STP correction, principally meniscus-reading and temperature/pressure-correction contributions, rather than excess hydrogen production; a small systematic bias in burette calibration at high collection rates cannot be excluded. Because charge utilisation is bounded by unity, values marginally above 100% are reported as measured, with the uncertainty band making explicit that they are statistically indistinguishable from 100%.

The polarisation behaviour underpinning this performance was characterised through a supplementary stepped-current experiment conducted at 30 °C and 100% RH, with the applied current incremented from 0.40 A to 0.90 A in 0.10 A steps. The resulting steady-state voltage–current relationship is presented in Table 4 and exhibits an essentially linear profile with an equivalent ohmic slope of approximately 0.55 Ω, indicating that ohmic losses across the Nafion membrane and bipolar plates dominated the voltage response within the investigated operating window. The absence of detectable activation or mass-transport departures from linearity across this range suggests that the cell operated within its quasi-linear regime, which is the region most relevant to continuous green hydrogen production under variable renewable power input.

Table 4. Steady-state voltage–current relationship during electrolysis (30 °C, 100% RH).

Current (A)	Mean Stack Voltage (V)
0.40	3.48
0.50	3.59
0.60	3.70
0.70	3.81
0.80	3.92
0.90	4.03

Although impedance spectroscopy was not available on this platform, the steady-state voltage–current data of Table 4 provide an equivalent quantification of the cell’s resistive behaviour. Linear regression of terminal voltage against applied current yields a differential (ohmic-equivalent) resistance of approximately $0.55\ \Omega$, corresponding to an area-specific resistance of $\approx 17.6\ \Omega\ \text{cm}^2$ for the $16\ \text{cm}^2$ active area; the extrapolated intercept of $\approx 1.52\ \text{V}$ is consistent with the sum of the thermodynamic reversible potential and a small, current-independent activation contribution. The absence of measurable curvature at low current indicates that the activation-controlled region is not resolved within the accessible current window, which precludes a formal Tafel fit but confirms that, across the operating range of interest, the cell response is ohmically dominated. This lumped ohmic-equivalent resistance, obtained directly from polarisation data, serves the same interpretive purpose as a high-frequency-resistance measurement in identifying membrane hydration and feed-water quality as the principal determinants of energy efficiency.

Stack voltage stability during a 300 s galvanostatic hold at 0.70 A and $18\ ^\circ\text{C}$ is presented in Table 5. Terminal voltage increased monotonically from 1.570 V to 1.588 V across the measurement window, yielding a voltage drift of approximately 0.06 V over 300 s, or equivalently a drift rate of the order of $0.012\ \text{V}\ \text{min}^{-1}$. This gradual rise is attributable to the progressive redistribution of water within the membrane–electrode assembly and to minor increases in ohmic resistance associated with localised dehydration at the anode catalyst layer. The magnitude of the drift falls well within the steady-state criterion adopted for data acquisition and confirms that the measurement window of 300 s was adequate for the calculation of representative mean responses.

Table 5. Terminal stack voltage during a 300 s galvanostatic hold at 0.70 A, $18\ ^\circ\text{C}$, and 100% RH.

Time (s)	Voltage (V)
0	1.570
30	1.580
60	1.582
90	1.583
120	1.584
150	1.584
180	1.585
210	1.586
240	1.587
270	1.587
300	1.588

3.1.2. Influence of Membrane Relative Humidity on Faradaic Efficiency

Membrane relative humidity was identified as the operating variable exerting the strongest influence on Faradaic efficiency, with η increasing by 11.4 percentage points as RH was raised from 50% to 100% under fixed reference conditions of 0.70 A, $30\ ^\circ\text{C}$, and $18\ \text{M}\Omega\ \text{cm}$ feed-water resistivity. This substantial improvement was accompanied by a systematic reduction in the mean stack voltage of 120 mV across the same hydration range, which confirms that membrane hydration dominates ohmic losses within the PEM configuration examined. The hydrogen volume collected over the 300 s measurement window increased proportionally from 30.1 mL to 34.0 mL, thereby corroborating the efficiency trend and demonstrating that suboptimal hydration produces measurable crossover and charge-

utilisation losses. The dependence of Faradaic efficiency and stack voltage on RH is summarised in Table 6, and the corresponding bivariate trend is illustrated in Figure 4.

Table 6. Influence of membrane relative humidity on Faradaic efficiency and stack voltage at fixed conditions (0.70 A, 30 °C, 18 MΩ cm).

RH (%)	V _{H2} (mL)	Faradaic η (%)	V _{stack} (Mean, V)
50	30.1	87.6	3.84
70	32.0	93.1	3.78
85	33.2	96.6	3.74
100	34.0	99.0	3.72

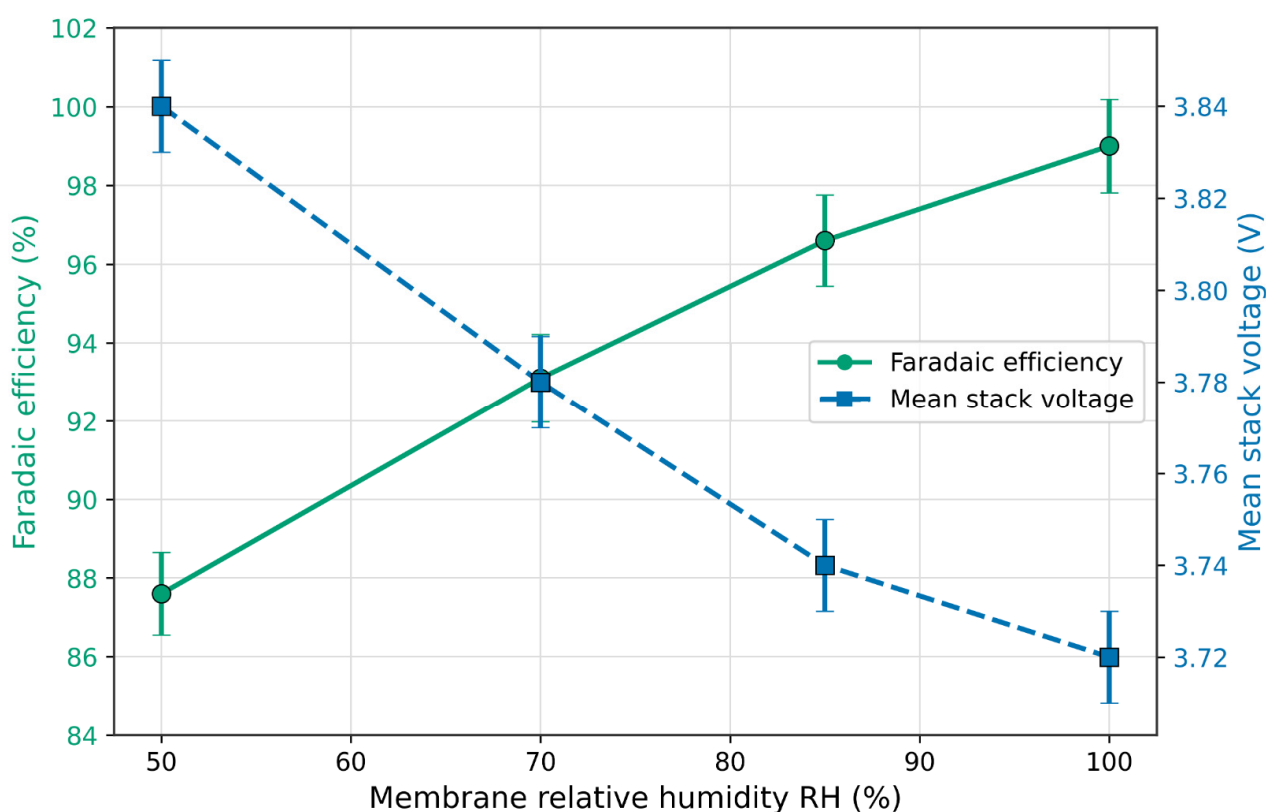


Figure 4. Faradaic efficiency and mean stack voltage as functions of membrane relative humidity at fixed current (0.70 A), temperature (30 °C), and feed-water resistivity (18 MΩ cm).

Direct determination of hydrogen purity and gas crossover by chromatography was not possible on the used rig, which lacks gas-sampling ports. The Faradaic-efficiency data nonetheless provide an indirect measure of crossover-related losses: efficiencies approaching but not exceeding unity at full hydration, together with the systematic efficiency recovery observed as relative humidity is increased, indicate that hydrogen crossover and charge-utilisation losses are small and are minimised under well-hydrated operation, whereas sub-saturated operation promotes measurable crossover through the partially dehydrated membrane.

The magnitude of the observed hydration effect exceeds that of temperature across the investigated factor ranges and is comparable to that of current density when expressed as a fractional performance change. This finding reinforces the central importance of water management within PEM electrolyser design and operation, particularly for systems intended to operate intermittently under variable renewable power input, where transient drying of the membrane can occur during low-current or idle phases.

From an electrochemical standpoint, the strong dependence of Faradaic efficiency and stack voltage on membrane relative humidity reflects the hydration-controlled proton conductivity of the Nafion 117 membrane. Proton transport in perfluorosulfonic acid membranes proceeds through a combination of Grotthuss (structural) hopping and vehicular diffusion, both of which require a continuous, water-filled channel network percolating between the sulphonic-acid sites. As the water content λ (water molecules per sulphonic-acid group) falls under sub-saturated humidification, these channels contract and partially disconnect, the effective proton conductivity decreases sharply, and the ohmic overpotential rises accordingly. The measured 120 mV reduction in mean stack voltage and 11.4 percentage-point gain in Faradaic efficiency as RH is raised from 50% to 100% are the macroscopic signatures of this transition: improved hydration lowers the membrane resistance while simultaneously suppressing the electro-osmotic and back-diffusion imbalances that promote hydrogen crossover and the associated parasitic charge loss. The residual voltage drift observed during galvanostatic holds is consistent with localised anode-side dehydration, in which water consumed and dragged across the membrane is not fully replenished, transiently raising the high-frequency (ohmic) resistance. The feed-water resistivity effect operates through a complementary mechanism: alkali-metal cations introduced with ionic contamination exchange with protons at the sulphonic-acid sites, reducing the density of mobile charge carriers and further increasing ohmic resistance, as evidenced by the monotonic 210 mV voltage rise across the resistivity range.

The positive temperature effect observed across all three primary responses arises from a different mechanism, namely the thermal activation of the electrode reactions and of proton transport. An increase in stack temperature raises the exchange current density of both the hydrogen and oxygen evolution reactions through an Arrhenius-type dependence, thereby lowering the activation overpotential required to sustain a given current, while the proton conductivity of the hydrated membrane is itself thermally activated and increases over the same range, further reducing the ohmic contribution. Reduced water and gas viscosity at elevated temperature additionally promotes bubble detachment from the catalyst surface, limiting the fraction of active area masked by adherent gas and lowering the associated mass-transport and bubble-induced overpotentials. These contributions act in the same direction as improved hydration but are of smaller magnitude within the 18–45 °C window accessible here, which is consistent with the measured temperature sensitivity of approximately $0.11 \text{ mL min}^{-1} \text{ }^{\circ}\text{C}^{-1}$ and with the secondary rank of temperature in the response surface analysis.

Within the tested current window, the near-linear voltage–current response (Table 4) indicates that ohmic losses dominate over activation and mass-transport contributions, so that membrane hydration and feed-water quality (both of which act on the ohmic term) are the most effective levers on energy efficiency, in agreement with the response surface analysis.

3.1.3. Influence of Feed-Water Resistivity on Specific Energy Consumption

Feed-water resistivity was found to exert a significant influence on specific electrical energy consumption, with *E*_{spec} deteriorating by 5.8% from 4.62 kWh Nm^{−3} to 4.89 kWh Nm^{−3} as resistivity decreased from 18 MΩ cm to 0.8 MΩ cm. The magnitude of this penalty is notable in that it approaches the benchmark gap typically reported between laboratory-scale PEM systems and industrial-scale installations operating at approximately 4.3 kWh Nm^{−3}, thereby identifying feed-water purity as a practical and low-cost optimisation lever. The mean stack voltage increased monotonically by 210 mV across the resistivity range examined, which confirms ionic contamination of the membrane as the principal mechanism underlying the ohmic degradation. The hydrogen yield declined by 5.3% over

the same range, consistent with the reduced Faradaic efficiency inferred from the voltage response. Complete data are presented in Table 7.

Table 7. Effect of feed-water resistivity on specific energy consumption at fixed conditions (0.70 A, 30 °C, RH 100%).

Resistivity (MΩ cm)	V _{H2} (mL)	E _{spec} (kWh Nm ^{−3})	V _{stack} (Mean, V)
18	34.0	4.62	3.72
5	33.6	4.68	3.77
1	32.9	4.78	3.85
0.8	32.2	4.89	3.93

The progression observed in Table 7 is consistent with the accepted mechanism by which dissolved ionic species, and in particular alkali-metal cations introduced through the controlled NaCl addition, compete with protons for sulphonic acid sites within the Nafion membrane. This competition reduces effective protonic conductivity and produces the monotonic rise in ohmic overpotential captured by the stack voltage. The practical implication is that investment in upstream water treatment for small-scale PEM systems delivers measurable efficiency returns that extend beyond the avoidance of catastrophic contamination events.

3.2. Fuel-Cell Mode: Reversible Stack Characterisation

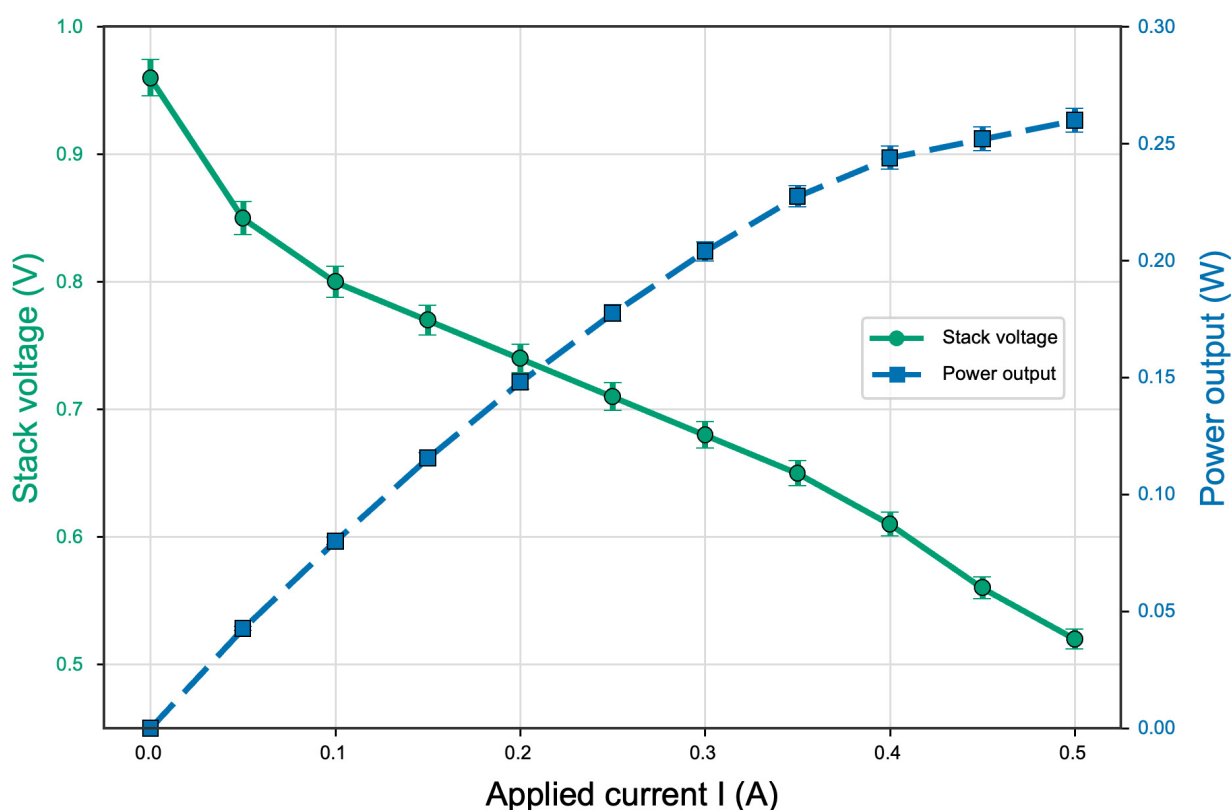
It should be emphasised that the fuel-cell measurements reported in this section do not constitute a separate device study: they characterise the same reversible membrane–electrode assemblies operating in the reverse (galvanic) direction and are included solely to establish the round-trip cycle symmetry of the unitised regenerative stack that motivates the green hydrogen storage application of this work. All optimisation and statistical analysis in Sections 3.1 and 3.3 pertain exclusively to the electrolysis (hydrogen-production) mode. The reversible PEM stacks were subsequently characterised in fuel-cell mode in order to complete the hydrogen cycle assessment and to establish a quantitative basis for evaluating cycle symmetry against the electrolysis-mode performance reported in Section 3.1. All fuel-cell measurements were conducted at 30 °C and 100% RH with pure hydrogen and oxygen feeds, and hydrogen consumption rates were corrected to STP using the same methodology applied to electrolysis data.

3.2.1. Polarisation and Power Generation Characteristics

Power generation exhibited the three-regime polarisation behaviour characteristic of low-temperature PEM fuel cells, comprising rapid activation losses at currents below 0.10 A, a quasi-linear ohmic regime between 0.10 A and 0.40 A, and mass-transport limitations above 0.40 A. The peak power output of 0.260 W was reached at 0.50 A and 0.52 V, while the lower heating value (LHV) efficiency declined from 58.3% at 0.05 A to 35.7% at 0.50 A in response to the progressive accumulation of irreversibilities across the polarisation regimes. Hydrogen consumption scaled linearly with current in accordance with stoichiometric fuel utilisation. The complete polarisation dataset is presented in Table 8, and the corresponding stack voltage and power density trends are illustrated in Figure 5.

Table 8. Fuel-cell polarisation data at 30 °C and 100% RH with pure H₂/O₂ feeds.

Current (A)	V _{stack} (V)	Power (W)	H ₂ Consumption (mL min ^{−1})	η _{LHV} (%)
0.05	0.85	0.043	2.1	58.3
0.10	0.80	0.080	4.2	54.9
0.20	0.74	0.148	8.4	50.8
0.30	0.68	0.204	12.6	46.7
0.40	0.61	0.244	16.8	41.9
0.50	0.52	0.260	21.0	35.7

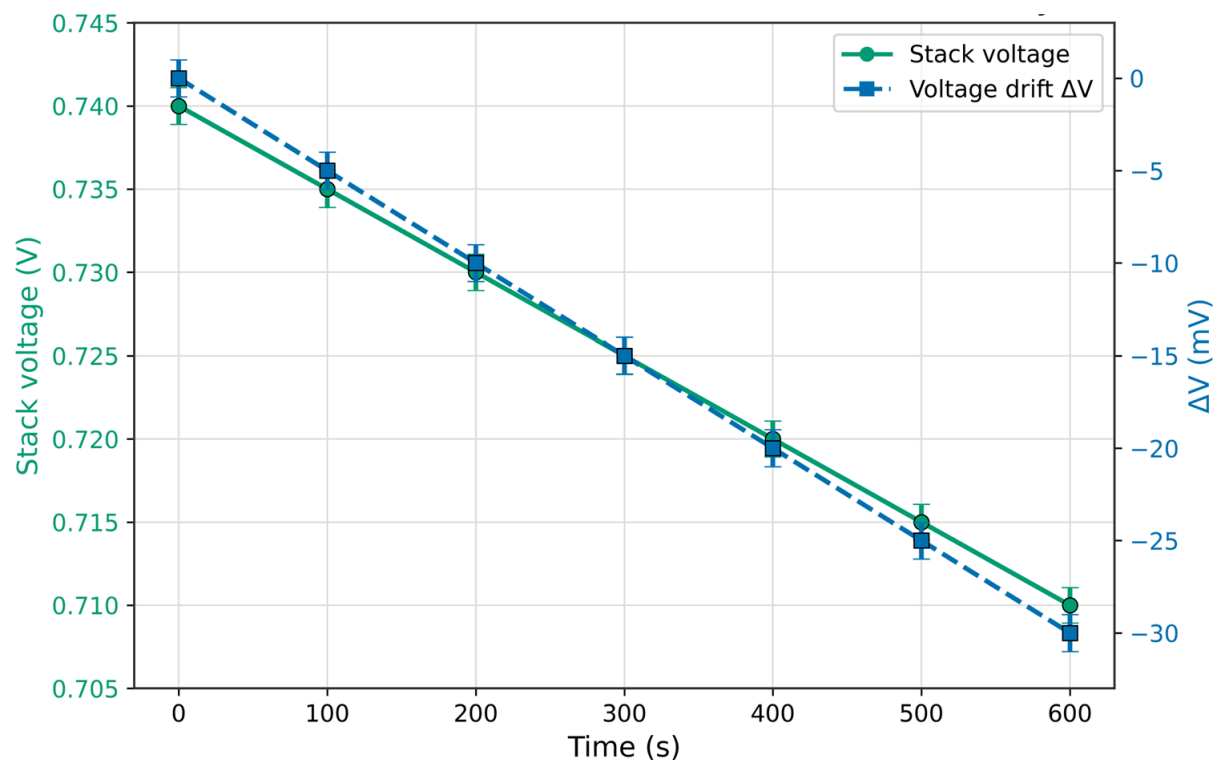
**Figure 5.** Fuel-cell polarisation and power density curves at 30 °C and 100% RH.

3.2.2. Voltage Stability and Discharge Behaviour Under Galvanostatic Load

Voltage behaviour under sustained galvanostatic load was examined at a fixed discharge current of 0.20 A under the reference conditions of 30 °C and 100% RH. Over the 600 s measurement window, the stack voltage decayed approximately linearly from 0.740 V to 0.710 V, corresponding to a cumulative drift of −30 mV and a mean drift rate of -0.05 mV s^{-1} (-3 mV min^{-1}). The measured terminal voltage and the corresponding cumulative drift are presented in Table 9 and illustrated in Figure 6, in which the close correspondence between the two traces confirms the linearity of the decay across the entire window. In this regime the stored hydrogen remains sufficient to sustain the imposed current, so the response is governed by the gradual reduction in reservoir pressure rather than by concentration polarisation at the electrode. The resulting linear behaviour provides a reliable proxy for state-of-charge estimation over the operational timescales relevant to short-duration energy storage demonstrations.

Table 9. Voltage drift during an initial 600 s galvanostatic discharge at 0.20 A (30 °C, 100% RH).

Time (s)	V_{stack} (V)	ΔV (mV)
0	0.740	0
100	0.735	−5
200	0.730	−10
300	0.725	−15
400	0.720	−20
500	0.715	−25
600	0.710	−30

**Figure 6.** Stack voltage and cumulative voltage drift during galvanostatic fuel-cell discharge at 0.20 A (30 °C, 100% RH) over the 600 s measurement window.

A separate discharge measurement was subsequently conducted, without replenishment of the hydrogen reservoir, in order to characterise the behaviour of the stack as the stored fuel approached exhaustion. Under these conditions the voltage response departed markedly from the linear regime described above. An initial period of oscillatory behaviour was observed between approximately 90 s and 270 s, with excursions of the order of 100 mV, which is attributed to the transient redistribution of hydrogen within the manifold and the gas-diffusion layers as the supply pressure fell. Beyond approximately 300 s, the stack entered a terminal collapse phase, the voltage falling from 0.55 V to 0.04 V within 150 s as concentration polarisation became dominant and the reservoir was exhausted. This two-stage behaviour, comprising a stable linear regime followed by an abrupt collapse once the fuel supply becomes limiting, is characteristic of small-scale demonstration stacks operating without active hydrogen recirculation and is consistent with the finite-reservoir architecture of the present rig. It also delimits the operational window over which the linear voltage-decay proxy described above remains valid.

3.3. Statistical Model Analysis and Integrated Assessment

3.3.1. Main Effects and Interaction Behaviour

The main-effects analysis derived from the response surface regression confirmed that the influence of applied current on hydrogen evolution rate was strictly linear across the 0.40–0.90 A range, with no statistically detectable curvature. The fitted linear slope was indistinguishable from the theoretical Faraday prediction of $10.4 \text{ mL min}^{-1} \text{ A}^{-1}$ within experimental uncertainty. Temperature exhibited a weaker but statistically significant positive main effect, and the interaction term between current and temperature was found to be non-significant ($p > 0.10$), with near-parallel profiles observed in the interaction plot across all temperature levels. These findings indicate that current and temperature act additively on the hydrogen evolution rate within the tested window, which simplifies the operational control strategy for small-scale systems and permits independent tuning of each factor without compensatory adjustment of the other. The main-effects and interaction plots for the four responses are presented in Figure 7.

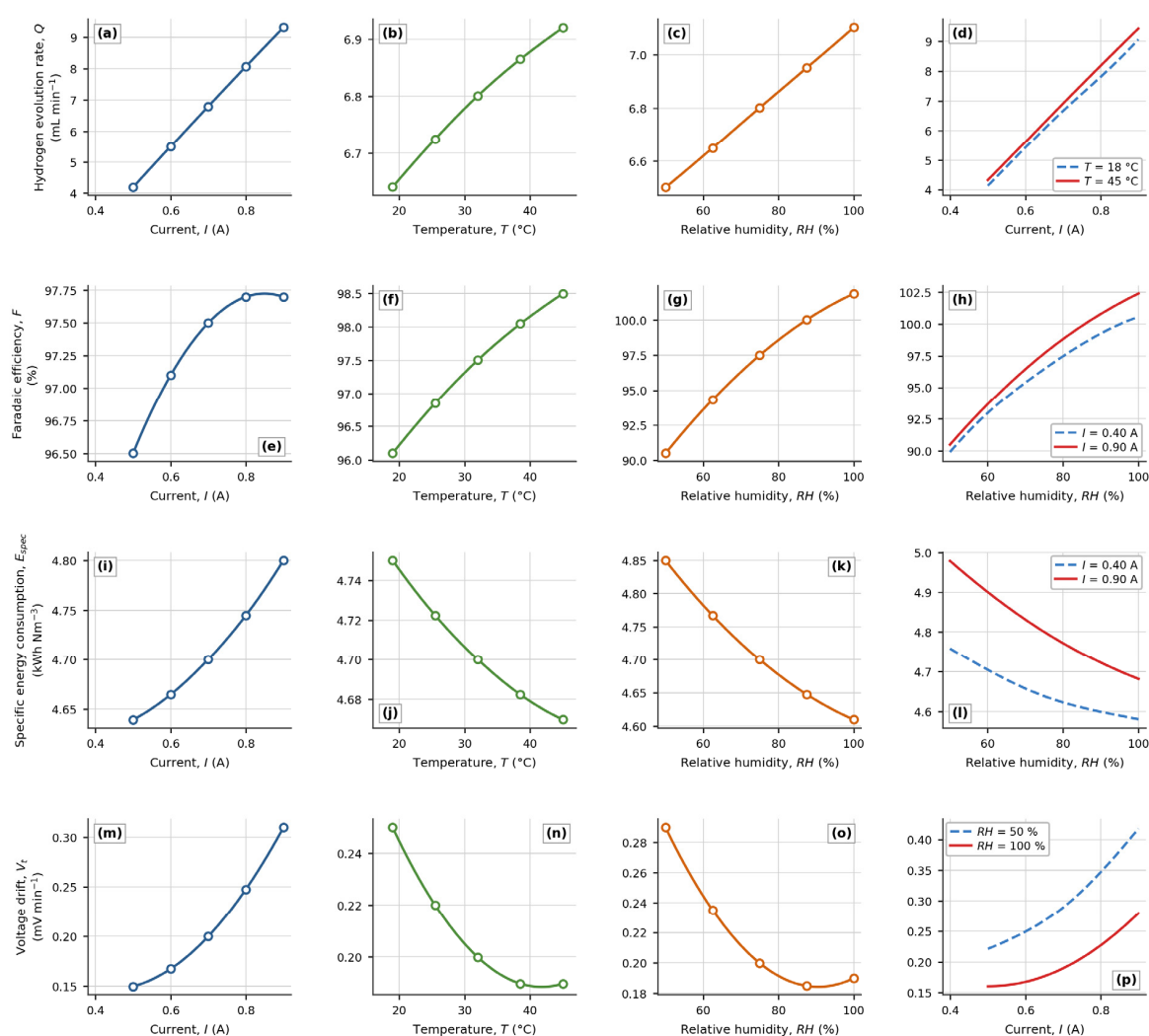


Figure 7. Main-effect and two-factor interaction plots derived from the reduced quadratic response-surface models. Hydrogen evolution rate is shown against (a) current, (b) temperature and (c) relative humidity, with (d) the current effect at two temperature levels; Faradaic efficiency against (e) current, (f) temperature and (g) relative humidity, with (h) the humidity effect at two current levels; specific energy consumption against (i) current, (j) temperature and (k) relative humidity, with (l) the humidity effect at two current levels; and voltage drift against (m) current, (n) temperature and (o) relative humidity, with (p) the current effect at two humidity levels.

The relative influence of the three factors varied systematically across the four responses. Current dominated the hydrogen rate response (accounting for the largest share of explained variance), while membrane relative humidity dominated the Faradaic efficiency and specific energy consumption responses. Temperature contributed as a secondary positive effect across all three primary responses. Voltage drift exhibited the weakest factor dependence, with residual scatter consistent with the short-term stochastic character of the measurement.

3.3.2. Performance Trade-Offs Between Rate, Efficiency, and Energy Consumption

Integration of the electrolysis dataset reveals a clear performance trade-off between hydrogen production rate and specific electrical energy consumption, which defines the practical Pareto frontier for the teaching rig examined. The maximum observed hydrogen rate of 8.82 mL min^{-1} , achieved at 0.90 A , 45°C , and $100\% \text{ RH}$, was associated with a specific energy consumption of approximately 4.65 kWh Nm^{-3} . In contrast, operation at 0.70 A , 45°C , and $100\% \text{ RH}$ delivered 78% of the peak rate (6.88 mL min^{-1}) at the minimum observed specific energy consumption of 4.62 kWh Nm^{-3} , thereby identifying this operating point as a favourable balance between throughput and efficiency. Reduction of relative humidity to 50% under otherwise reference conditions penalised Faradaic efficiency by 11.4 percentage points and increased specific energy consumption to approximately 4.85 kWh Nm^{-3} , which demonstrates that membrane hydration exerts a larger influence on efficiency than either current or temperature within the examined design space. The consolidated trade-off data are summarised in Table 10, and the corresponding specific electrical energy consumption and hydrogen evolution rate are illustrated in Figure 8.

Table 10. Performance trade-offs and operating-condition comparisons within the electrolysis design space.

Operating Condition	H ₂ Rate (mL min ^{−1})	Faradaic η (%)	E _{spec} (kWh Nm ^{−3})
Maximum rate: 0.90 A , 45°C , RH 100%	8.82	100.6	~4.65
Minimum E _{spec} : 0.70 A , 45°C , RH 100%	6.88	100.1	4.62
Low-hydration penalty: 0.70 A , 30°C , RH 50%	~6.0	87.6	~4.85
Low-purity penalty: 0.70 A , 30°C , $0.8 \text{ M}\Omega \text{ cm}$	6.44	94.7	4.89

The measured minimum specific energy consumption of 4.62 kWh Nm^{-3} aligns closely with values reported in the literature for small-scale PEM electrolyser systems, confirming the representativeness of the teaching rig despite its bench-scale configuration. Current provides the greatest leverage on throughput, delivering a 140% increase in production rate in response to a 125% increase in current, but this throughput gain is accompanied by diminishing efficiency returns beyond 0.70 A . Membrane hydration delivers dual benefits to both Faradaic efficiency and specific energy consumption with minimal infrastructure cost, while feed-water purity represents a scalability consideration that becomes increasingly important as system size and operating duration increase.

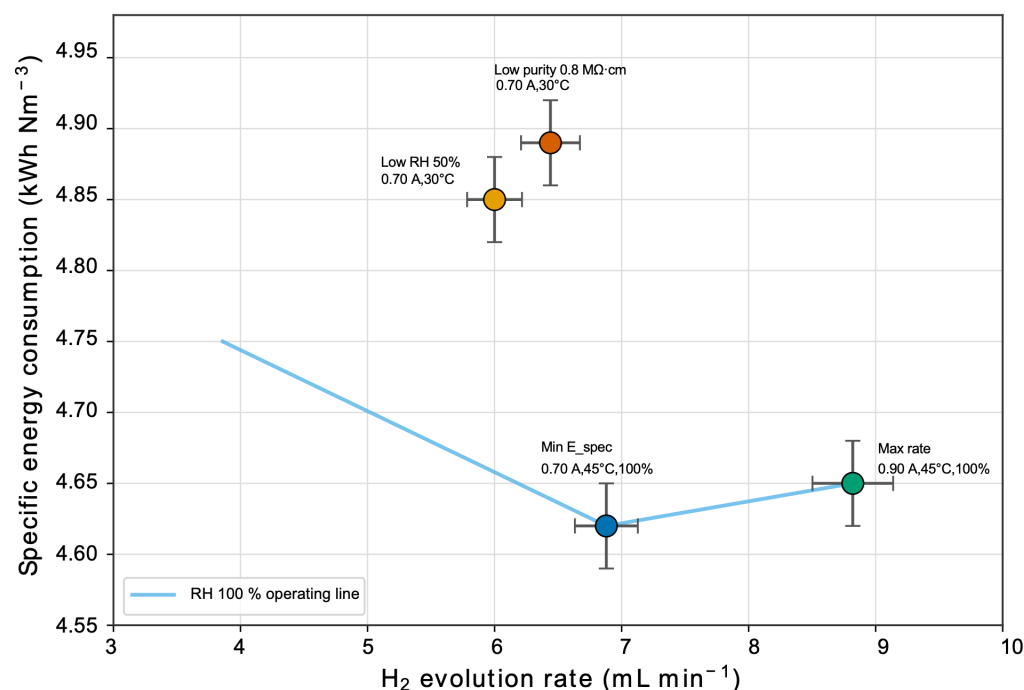


Figure 8. Pareto frontier for the PEM electrolyser showing hydrogen production rate against specific energy consumption.

3.3.3. Cycle Symmetry: Electrolysis–Fuel-Cell Comparison

The overlay of steady-state polarisation curves for the reversible PEM stack in electrolysis and fuel-cell modes are shown in Figure 9. Hydrogen throughput scales symmetrically between the electrolysis and fuel-cell modes, with electrolysis production corresponding to approximately 40% of fuel-cell consumption at matched current, a ratio that reflects the stoichiometric relationship between the two reactions and validates the reversible stack concept. The voltage asymmetry between the modes (approximately 4 V for electrolysis compared with approximately 0.6 V for fuel cell) reflects the combined contributions of thermodynamic reversibility and activation overpotentials. Estimation of the round-trip LHV efficiency, calculated as the product of the electrolysis efficiency and the fuel-cell LHV efficiency, yields a value of approximately 45%, which is typical for educational PEM systems and suitable for the demonstration of renewable energy storage cycles. A summary comparison of hydrogen throughput in the two operating modes is presented in Table 11.

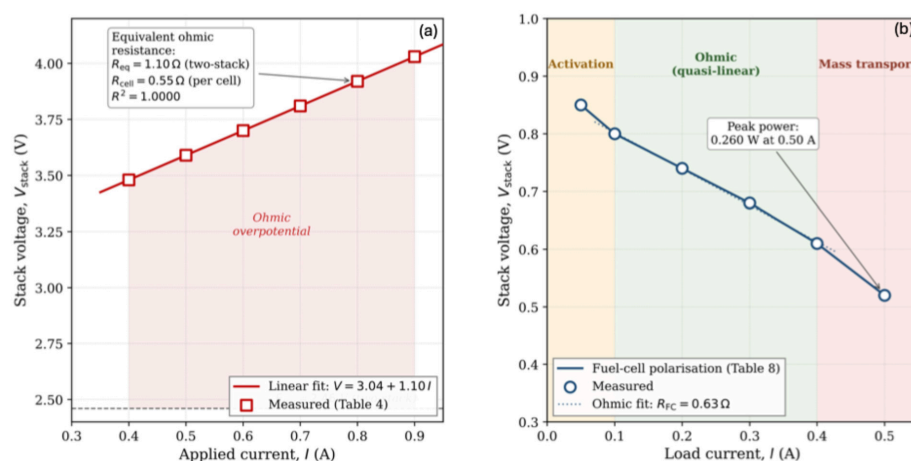


Figure 9. Overlay of steady-state polarisation curves for the reversible PEM stack in electrolysis and fuel-cell modes at 30 °C and 100% RH, (a) Electrolysis mode, (b) Fuel-cell mode.

Table 11. Cycle symmetry summary comparing electrolysis production with fuel-cell consumption at comparable currents.

Current (A)	Electrolysis H ₂ Rate (mL min ^{−1})	Fuel-Cell H ₂ Consumption (mL min ^{−1})	V _{elec} (V)	V _{FC} (V)
0.40	3.86	16.8	3.48	0.61
0.50	4.90	21.0	3.59	0.52

3.3.4. Optimal Operating Window

Desirability profiling following the approach of Derringer and Suich was applied to the reduced quadratic regression models in order to identify the operating region that simultaneously maximises hydrogen evolution rate and Faradaic efficiency while minimising specific energy consumption. The combined desirability function indicated an optimal operating region centred at approximately 0.75 A, 42 °C, and 95% relative humidity, at which the predicted hydrogen rate was 7.4 mL min^{−1}, the predicted Faradaic efficiency was close to 98%, and the predicted specific energy consumption was approximately 4.5 kWh Nm^{−3}. All three predicted values fall within the expanded uncertainty bounds of the experimental measurements, which confirms both the internal consistency of the regression models and the practical attainability of the identified operating point. These statistically derived operating conditions provide quantitative guidance for educational and small-scale PEM electrolyzers seeking to maximise green hydrogen production under constrained laboratory resources and establish a reproducible benchmark against which subsequent design modifications or operational interventions may be assessed.

Formal statistical validation of the optimised models is provided by the predicted coefficients of determination reported in Table 2, which are computed by a leave-one-out (PRESS) procedure and therefore quantify out-of-sample predictive capability. In addition, the identified optimum lies within the measured design space rather than on an extrapolated boundary, and its predicted responses agree, within the expanded experimental uncertainty of ±3.6%, with the nearest directly measured operating points. This internal consistency between predicted and measured neighbouring points constitutes a self-consistent validation of the models within the design space.

3.3.5. Comparison with Recent Literature

To situate the present results within the recent literature, Table 12 compares the measured performance of the bench-scale rig with representative PEM water electrolysis studies published in recent years. The minimum specific energy consumption obtained here (4.5–4.62 kWh Nm^{−3}, equivalent to ≈50 kWh per kilogram of hydrogen at the stack) is close to the stack-level consumption of ≈53 kWh kg^{−1} assumed in recent techno-economic analyses of industrial PEM systems, indicating that, despite its bench scale, the rig operates at an energy efficiency representative of practical PEM technology. The Faradaic efficiency of 94–100% is consistent with values reported for well-hydrated laboratory PEM cells, while the dominance of applied current over throughput and of membrane hydration over efficiency reproduces the qualitative trends reported in both experimental and modelling studies. Differences in absolute production rate are attributable primarily to the small active area and the low-current density of the teaching platform relative to industrial stacks operating at 1–3 A cm^{−2}.

Table 12. Comparison of the present bench-scale results with PEM water electrolysis studies.

Study	System/Scale	Method	Key Range/Conditions	Reported Performance
Present Study	Bench-scale reversible PEM, 16 cm ² , Nafion 117	Experimental FCCD + RSM, 4 responses, GUM uncertainty	0.40–0.90 A; 18–45 °C; 50–100% RH; 0.8–18 MΩ·cm	≈4.5 kWh Nm ^{−3} (≈50 kWh kg ^{−1}); Faradaic eff. 94–100%; RH dominates efficiency
RSM + finite-element PEM study [30]	Single-cell PEM, model-coupled	RSM fitted to FEM output	Temperature/current density/pressure	Optimises efficiency vs. operating parameters; response surface from simulation, not direct measurement
RSM + ANN PEM study [31]	Next-generation PEM cell	RSM and artificial neural networks	Multiple operating variables	Statistical/ML modelling of performance; fewer measured responses and no water-quality term
Industrial PEM techno-economic baseline [32]	Industrial-scale PEM system	Techno-economic modelling	1–3 A cm ^{−2} , pressurised, optimised MEA	≈57.5 kWh kg ^{−1} system; LCOH ≈ US\$5–7 kg ^{−1}

3.3.6. Scalability Considerations

The absolute production rates reported here are set by the 16 cm² active area and the sub-ampere current of the stack and are not directly transferable to industrial systems, which operate at 1–3 A cm^{−2} over far larger areas and under elevated pressure. What does transfer, however, is the relational structure this study quantifies: the linear current–throughput relation, the ohmic dominance of membrane hydration, and the efficiency penalty of feed-water contamination are intrinsic to PEM electrochemistry and scale-invariant in direction, even though their magnitudes shift with cell design. The statistical framework is likewise scale-independent and can be reapplied, without modification, to an industrially representative single cell across wider factor ranges. The present results therefore constitute a transferable methodology and a qualitative guide, not a set of directly transposable industrial set-points.

3.3.7. Indicative Economic Assessment

Although a full techno-economic optimisation is beyond the scope of a bench-scale study, the measured energy performance permits a first-order estimate of production economics that contextualises the efficiency gains identified. At the optimal operating window (specific energy consumption ≈4.5 kWh Nm^{−3}, equivalent to ≈50 kWh per kilogram of hydrogen), electricity alone contributes approximately US\$2.5–3.5 per kilogram of hydrogen at representative renewable-electricity prices of 0.05–0.07 US\$ kWh^{−1}. This is consistent with recent techno-economic analyses that place the levelised cost of green hydrogen from current PEM technology at roughly US\$5–7 kg^{−1}, of which electricity is the single largest recurring contributor [32]. Within this framework, the operational levers quantified in the present study translate directly into cost: the 11.4 percentage-point Faradaic-efficiency gain and associated voltage reduction achieved through full membrane hydration correspond to a proportional reduction in specific energy consumption, and therefore in the electricity component of cost, while maintaining high feed-water resistivity avoids the 5.8% energy penalty measured under contaminated-water conditions. Because electricity dominates operating expenditure, these low- or zero-capital operational measures represent a particularly favourable cost-reduction pathway relative to hardware modifications, especially for small-scale and off-grid systems coupled to variable renewable power.

4. Conclusions

A statistical framework has been developed for quantifying the influence of operating conditions on green hydrogen production in a bench-scale proton-exchange membrane wa-

ter electrolyser, integrating a face-centred central composite design with response surface methodology and supplementary measurements on feed-water purity. The novelty and practical value of this work reside in three contributions: a fully experimental, uncertainty-quantified RSM framework that jointly models four performance responses; the explicit quantification of two water-management levers (membrane hydration and feed-water resistivity) within that framework; and a reproducible operating benchmark for small-scale PEM systems. These outcomes are directly applicable to laboratory electrolysers, to distributed and off-grid hydrogen generators coupled to intermittent renewable power and, at the methodological level, to any PEM optimisation campaign seeking an interpretable, statistically rigorous alternative to one-factor-at-a-time testing. The experimental campaign generated 288 independent observations across 96 randomised runs, from which reduced quadratic regression models were derived for four performance responses: hydrogen evolution rate, Faradaic efficiency, specific electrical energy consumption, and stack voltage drift. Applied current has been identified as the dominant operating variable governing hydrogen throughput, with a strictly linear dependence observed across the 0.40–0.90 A range and a fitted slope indistinguishable from the theoretical Faraday prediction of $10.4 \text{ mL min}^{-1} \text{ A}^{-1}$ within the expanded experimental uncertainty of $\pm 3.6\%$. Membrane relative humidity has been shown to exert the strongest control over efficiency-related responses, producing an 11.4 percentage-point improvement in Faradaic efficiency as hydration was raised from 50% to 100%, accompanied by a 120 mV reduction in mean stack voltage. Temperature contributed a secondary but statistically significant positive effect, while feed-water resistivity produced a 5.8% deterioration in specific energy consumption across the range examined. Model adequacy has been confirmed through analysis of variance and residual diagnostics, with adjusted coefficients of determination in the range 0.851–0.925 and predicted coefficients above 0.835 across all four responses. Desirability profiling has identified an optimal operating region at 0.75 A, 42 °C, and 95% relative humidity, delivering a predicted hydrogen rate of 7.4 mL min^{-1} , a Faradaic efficiency of 98%, and a specific energy consumption of 4.5 kWh Nm^{-3} .

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References

1. Elkhatat, A.; Al-Muhtaseb, S. Climate change and energy security: A comparative analysis of the role of energy policies in advancing environmental sustainability. *Energies* **2024**, *17*, 3179. [\[CrossRef\]](#)
2. Arcos, J.M.M.; Santos, D.M. The hydrogen color spectrum: Techno-economic analysis of the available technologies for hydrogen production. *Gases* **2023**, *3*, 25–46. [\[CrossRef\]](#)
3. Nakkeeran, K.; Victor, K. Grey and blue hydrogen: Insights into production technologies and outlook on CO₂-free alternatives. *Sustain. Energy Technol. Assess.* **2025**, *75*, 104222. [\[CrossRef\]](#)
4. Luque-Urrutia, J.A.; Ortiz-García, T.; Solà, M.; Poater, A. Green energy by hydrogen production from water splitting, water oxidation catalysis and acceptorless dehydrogenative coupling. *Inorganics* **2023**, *11*, 88. [\[CrossRef\]](#)
5. Zun, M.T.; McLellan, B.C. Cost projection of global green hydrogen production scenarios. *Hydrogen* **2023**, *4*, 932–960. [\[CrossRef\]](#)
6. Park, J.; Kang, S.; Kim, S.; Kim, H.; Cho, H.-S.; Lee, J.H. Comparative techno-economic evaluation of alkaline and proton exchange membrane electrolysis for hydrogen production amidst renewable energy source volatility. *Energy Convers. Manag.* **2025**, *325*, 119423. [\[CrossRef\]](#)

7. Tang, J.; Su, C.; Shao, Z. Advanced Membrane-Based Electrode Engineering Toward Efficient and Durable Water Electrolysis and Cost-Effective Seawater Electrolysis in Membrane Electrolyzers. *Exploration* **2025**, *4*, 20220112.
8. Zhang, W.; Liu, M.; Gu, X.; Shi, Y.; Deng, Z.; Cai, N. Water electrolysis toward elevated temperature: Advances, challenges and frontiers. *Chem. Rev.* **2023**, *123*, 7119–7192. [\[CrossRef\]](#) [\[PubMed\]](#)
9. Shao, G.; Wang, Q.; Miao, F.; Li, J.; Li, Y.; Shen, B. Improved catalytic efficiency and stability by surface activation in Fe-based amorphous alloys for hydrogen evolution reaction in acidic electrolyte. *Electrochim. Acta* **2021**, *390*, 138815. [\[CrossRef\]](#)
10. Rolo, I.; Costa, V.A.; Brito, F.P. Hydrogen-based energy systems: Current technology development status, opportunities and challenges. *Energies* **2023**, *17*, 180. [\[CrossRef\]](#)
11. Masjuki, H.; Ruhul, A.; Mustafi, N.N.; Kalam, M.; Arbab, M.; Fattah, I.R. Study of production optimization and effect of hydroxyl gas on a CI engine performance and emission fueled with biodiesel blends. *Int. J. Hydrogen Energy* **2016**, *41*, 14519–14528. [\[CrossRef\]](#)
12. Chakik, F.E.; Kaddami, M.; Mikou, M. Effect of operating parameters on hydrogen production by electrolysis of water. *Int. J. Hydrogen Energy* **2017**, *42*, 25550–25557. [\[CrossRef\]](#)
13. Kowthaman, C.; Kumar, P.S.; Selvan, V.A.M. Micro-patterned graphite electrodes: An analysis and optimization of process parameters on hydrogen evolution in water electrolysis. *Fuel* **2021**, *305*, 121542. [\[CrossRef\]](#)
14. El Kady, M.; Farrag, A.E.F.; Gad, M.; El Soly, A.; Hashish, H.A. Parametric study and experimental investigation of hydroxy (HHO) production using dry cell. *Fuel* **2020**, *282*, 118825. [\[CrossRef\]](#)
15. Sekar, D.; Venkadesan, G.; Panithasan, M.S. Optimisation of dry cell electrolyser and hydroxy gas production to utilise in a diesel engine operated with blends of orange peel oil in dual-fuel mode. *Int. J. Hydrogen Energy* **2022**, *47*, 4136–4154. [\[CrossRef\]](#)
16. Bow, Y.; Budiman, A.; Yerizam, M. Design of dry cell HHO generator using NaCl solution for hydrogen production. *Indones. J. Fundam. Appl. Chem. (IJFAC)* **2021**, *7*, 8–15. [\[CrossRef\]](#)
17. Shahdabadi, R.S.; Zamen, M.; Hosseini, S.V. Adaptive design for the connection of multicell HHO generator with solar photo-voltaic panels. *Int. J. Hydrogen Energy* **2023**, *48*, 33422–33433. [\[CrossRef\]](#)
18. Garg, S.; Rodriguez, C.A.G.; Rufford, T.E.; Varcoe, J.R.; Seger, B. How membrane characteristics influence the performance of CO₂ and CO electrolysis. *Energy Environ. Sci.* **2022**, *15*, 4440–4469. [\[CrossRef\]](#)
19. Becker, H.; Murawski, J.; Shinde, D.V.; Stephens, I.E.; Hinds, G.; Smith, G. Impact of impurities on water electrolysis: A review. *Sustain. Energy Fuels* **2023**, *7*, 1565–1603. [\[CrossRef\]](#)
20. Wang, C.R.; Stansberry, J.M.; Mukundan, R.; Chang, H.; Kulkarni, D.; Park, A.M.; Plymill, A.; Firas, N.; Liu, C.P.; Lang, J.; et al. Proton exchange membrane (PEM) water electrolysis: Cell-level considerations for gigawatt-scale deployment. *Chem. Rev.* **2025**, *125*, 1257–1302. [\[CrossRef\]](#) [\[PubMed\]](#)
21. Wallnöfer-Ogris, E.; Grimmer, I.; Ranz, M.; Höglinger, M.; Kartusch, S.; Rauh, J.; Macherhammer, M.-G.; Grabner, B.; Trattner, A. A review on understanding and identifying degradation mechanisms in PEM water electrolysis cells: Insights for stack application, development, and research. *Int. J. Hydrogen Energy* **2024**, *65*, 381–397. [\[CrossRef\]](#)
22. Bao, Z.; Xie, B.; Li, W.; Zhong, S.; Fan, L.; Tongsh, C.; Gao, F.; Du, Q.; Benbouzid, M.; Jiao, K. High-consistency proton exchange membrane fuel cells enabled by oxygen-electron mixed-pathway electrodes via digitalization design. *Sci. Bull.* **2023**, *68*, 266–275. [\[CrossRef\]](#) [\[PubMed\]](#)
23. Fan, L.; Luan, Y.; Xing, L.; Du, Q.; Xuan, J.; Wang, Y.; Zhang, F.; Li, X.; Yuan, S.; Tongsh, C.; et al. Electrospun nanofiber electrodes for low platinum-loaded fuel cells. *Sci. Bull.* **2026**, *71*, 254–258. [\[CrossRef\]](#) [\[PubMed\]](#)
24. Eltaweel, M.; Mostafa, N.A.; Kalyvas, C.; Chen, Y.; Herfatmanesh, M.R. Optimising flywheel energy storage systems for enhanced windage loss reduction and heat transfer: A computational fluid dynamics and ANOVA-based approach. *Energy Rep.* **2025**, *13*, 834–855. [\[CrossRef\]](#)
25. Zhang, F.; Zhao, Y.; Chen, X.; Zhao, S.; Zhou, J.; Lu, Z.; Lin, Y. Concerns on the effects of electrode positions in electrolyte container for the oxygen evolution reaction. *Molecules* **2023**, *28*, 8143. [\[CrossRef\]](#) [\[PubMed\]](#)
26. Kirkup, L.; Frenkel, R.B. *An Introduction to Uncertainty in Measurement: Using the GUM (Guide to the Expression of Uncertainty in Measurement)*; Cambridge University Press: Cambridge, UK, 2006.
27. Du, Z.; Xu, L.; Yin, S.; Dong, S.; Dai, Z.; Ma, Y.; Vo Thanh, H.; Reza Soltanian, M. Enhanced prediction and uncertainty analysis for hydrogen production rate in depleted oil and gas reservoirs using advanced machine learning techniques. *Geoenergy Sci. Eng.* **2025**, *249*, 213795. [\[CrossRef\]](#)
28. Zerrougui, I.; Li, Z.; Hissel, D. Physics-Informed Neural Network for modeling and predicting temperature fluctuations in proton exchange membrane electrolysis. *Energy AI* **2025**, *20*, 100474. [\[CrossRef\]](#)
29. Raman, K.A.; Wolf, N.L.; Javed, A.; Karyofylli, V.; Kungl, H.; Karl, A.; Jodat, E.; Eichel, R.-A. Evaluating activation strategies and their stability on PEM water electrolyzers using machine learning. *Energy AI* **2025**, *22*, 100623. [\[CrossRef\]](#)
30. Ozdemir, S.N.; Taymaz, I.; San, F.G.B.; Okumuş, E. Performance assessment and optimization of the PEM water electrolyzer by coupled response surface methodology and finite element modeling. *Fuel* **2024**, *365*, 131138. [\[CrossRef\]](#)

31. Tawalbeh, M.; Sabri, M.; Kazim, H.; Al-Othman, A.; Alshraideh, H.; Almomani, F. Statistical and machine learning methods for modeling the next-generation PEM water electrolysis systems: Response surface methodology and artificial neural networks. *Energy Convers. Manag. X* **2025**, *29*, 101450. [[CrossRef](#)]
32. Hubert, M.; Peterson, D.; Miller, E.; Vickers, J.; Mow, R.; Howe, C. *Clean Hydrogen Production Cost Scenarios with PEM Electrolyzer Technology*; U.S. Department of Energy, Hydrogen and Fuel Cell Technologies Office: Washington, DC, USA, 2020.

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