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Predictive Modelling of PEMFC Degradation Against Hydrogen Crossover Using Machine Learning Models in Matlab

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Abstract **-** The complexity and nonlinearity of the connection between the hydrogen crossover and OCV decay led to extremely challenging establishment of their relationship and the possible prediction of their values using both experimental and conventional computational modeling approaches. Thus, machine learning becomes invaluable in providing low cost and efficient surrogate models that can capture and comprehend the effects of influential factors affecting the trends in the hydrogen crossover and OCV which makes the system highly complicated. To address the challenges in the characterization of PEMFC performance degradation as affected by the temporal hydrogen crossover, this work developed five ML-based models to carry out the OCV predictions using Matlab. The results of model performance evaluation, statistical analysis, and model fit performance suggest that Gaussian Process Regression-based model gave the best prediction accuracy among the other models using algorithms such as the Tri-Layered Neural Network, Ensemble, Decision Tree, and Kernel, with R and R^2 values of 1.0000 and 1.0000, respectively. During the deployment of GPR-based model, these values were observed to decrease to 0.9893 and 0.9787, respectively; still, an indicative of a well-performing model. Inversely, the model has showed more generalizability towards new experimental data and has minimized overfitting which makes it an excellent model for deployment. This finding is also aligned with the minimal RMSE, MSE, and MAE values of 0.0053, 0.00002765, and 0.0030, respectively. Given this, this work demonstrated the usability of machine learning to address the complexity of PEMFC degradation.

Keywords**:** PEMFC, machine learning, degradation, Matlab, hydrogen crossover, open circuit voltage, modeling

1. Introduction

The challenge on the development of proton exchange membrane fuel cell (PEMFC) technology particularly on issues about its durability and flexibility in operation not only undermines its performance but also threatens the promise of clean and more sustainable energy of the future. Improving the PEMFC particularly on reducing the costs, extending the operation at higher temperature to improve fuel cell efficiency, and achieving high reliability remain pivotal in its development [1]. Performance, durability, and cost are considered as the "iron triangle" of PEMFC [2] - [4]; thus, R&D efforts should focus on these developmental aspects to have an excellent PEMFC design. Hamrock and Yandrasits (2006) discussed that most of the research initiatives primarily focus on improving the durability of PEMFC membranes to achieve the desired level of structural reliability based on the requirements of commercialization for applications such as the automotive and stationary power stations [5]. Zhang et *al.* (2012) highlighted the importance of developing PEMFC capable of performing at high temperature and low relative humidity operations to improve water management, improve reaction kinetics [6]; hence, increasing fuel conversion efficiency, mitigate the potential carbon monoxide poisoning, and provide more flexibility to varying environmental and operating conditions. Other hurdles to PEMFC development are the purity and stability of hydrogen supply which greatly affect its technological and economic feasibility [7].

The relentless efforts on studying the PEMFC durability clearly emphasize the diminishing effect of gas crossover, particularly the H_2 fuel, to its performance and structural integrity. This phenomenon occurs due to some factors including the physico-chemical properties of the proton exchange membrane (PEM), membrane permeability, membrane hydration level, and operating conditions (i.e., temperature, pressure, and relative humidity) [8]. On the other hand, membrane defects and degradation due to mechanical, chemical, and thermal stresses greatly contribute to the intensification of hydrogen crossover rates more than just the permeability of hydrogen through the membrane [9]. Consequently, hydrogen crossover

results in the accelerated fuel cell component degradation including the membrane, catalyst layer, and carbon support, increased formation of free radicals, membrane thinning, formation of Pt band which decreases mechanical stability and proton conductivity of the membrane, and ultimately performance loss due to unconverted H_2 fuel to useful work and increased overpotentials [10]. Therefore, the quantification of hydrogen crossover is crucial in the pursuit of developing highly durable PEMFC, maximization of performance, and reduction of costs. However, existing experimental characterization methods face pressing issues related to accurate measurements as influenced by the complexity of experimental setups [11] and intricacy of the measuring instrument affecting the reproducibility of the results across different operating conditions [12] - [13], the difficulty in the measurement of local distribution of hydrogen crossover across the membrane electrode assembly (MEA) [14], insufficient understanding about the in-operando variations of membrane properties [15], and the lack of comprehensive baseline data for the extent of gas permeation across the membrane [16].

Prognostic methods of determining the effect of hydrogen crossover on the fuel cell performance are gaining much attention recently due to its low cost and high efficiency in modelling physical systems [17]. Equations-based models have been widely explored in the literature [18] - [20] but the real-world complexities and nonlinearity of fuel cell degradation are extremely difficult to be captured by these methodologies [21]. Furthermore, these models are subject to researcher's biases and assumptions which might inaccurately represent the actual mechanisms governing fuel cell degradation [22]. Thus, data-driven modelling approaches are seen to be effective in handling the nonlinear behavior of fuel cell performance decay as affected by various degradation parameters. Machine learning (ML) models offer promising breakthroughs in the research and development of PEMFC. These models accelerate the optimization of complex system parameters while accurately predicting their behaviors, provide deeper understanding about the factors affecting the changes in the fuel cell performance, and enable the development of more advanced and intelligent optimization approaches [23]. With better understanding of the interconnected degradation parameters using physics-informed ML models, fuel cell component design optimization is made possible at minimal cost, time, and computational resources [24]. Novel methodologies for predicting the PEM behavior and in-depth physical interpretation of the data-driven insights provided by the ML models about the internal mechanisms governing the PEMFC systems can be developed [25]. Therefore, the opportunities of ML application is huge with the number of experimental data in the literature from various explorations done within the PEMFC domain.

While experimental methods introduce valuable insights about the effect of hydrogen crossover on the fuel cell performance, some limitations of these approaches exist. For instance, the potential influence of contaminants present in the fuel on the membrane permeability could be extremely difficult to characterize experimentally [26]. Furthermore, the influence of other factors such as the gas pressure, the membrane ageing, varying current densities and operating temperatures, and the morphology and structural integrity of the membrane during hydrogen crossover measurements could present huge variation in their values [27] - [28]. The uniformity of clamping pressure and cell assembly could significantly affect the measured hydrogen crossover [29] but most of the reported studies do not consider the implication of this instance to their characterization. On the other hand, ML-based models are trained based on the hidden patterns revealed from the training data and the effect of these nuances in the experimental measurement of hydrogen crossover could be addressed in the machine learning process.

Therefore, this work focused on developing ML-based models based on the state-of-the-art machine learning algorithms to predict the open circuit voltage (OCV) decay as affected by the hydrogen crossover using Matlab. Specifically, five algorithms were used to carry out the predictions: Gaussian Process Regression (GPR), Tri-layered Neural Network (TLNN), Ensemble, Decision Tree, and Kernel ML algorithms. The models were subjected to performance evaluation based on their root mean square error (RMSE), mean square error (MSE), and mean absolute error (MAE) values. Furthermore, they were validated using the validation dataset and were subjected to statistical analysis against the experimental OCV values. Correlation, goodness-of-fit, and measure of spread were as well determined to further assess the performance of each model and their validity to be used for application. Finally, the models were deployed to predict the OCV values using new experimental data. On the hindsight, the results of this work add value to the current explorations done in the direct applications of machine learning to the PEMFC technology.

2. Methodology

2.1. Data

This work utilized the experimental data of hydrogen crossover currents and OCV decay from [30] as baseline data for for training, validating, and testing ML-based models for OCV predictions.

The PEMFC in their work is operated at 80°C and at 100% relative humidity to simulate the common operating conditions used in the literature. The hydrogen crossover current densities were measured at 0.35V using linear sweep voltammetry (LSV) from 0 - 0.7V at 2 mV/s scan rate. The MEA used was from Gore-SELECT® with membrane thickness of 12μm, active surface area of 25 cm^2, and Pt catalyst loadings of 0.1 mg/cm^2 for anode and 0.4 mg/cm^2 for cathode.

For the AST procedure, the MEA was subjected to both chemical and mechanical stress test at 90°C and OCV and hydrogen crossover measurements were recorded. Each completed AST cycle lasted for 24 hours.

The entire dataset consisted of time, hydrogen crossover current density, and OCV values. Random sampling was done to divide the dataset into training (60%), validation (20%), and test (20%) data. Training dataset was used to develop each model and the validation data was used to assess the performance of the models. Whereas, test data was used as experimental data representing actual PEMFC operation.

2.2. Machine Learning Models

Five ML-based models were developed in this work using algorithms namely the GPR, TLNN, Ensemble, Decision Tree, and Kernel methods. Each model was trained to predict the OCV, as the response variable, based on temporal variation of hydrogen crossover current density, as the predictor. Thus, both time and hydrogen crossover current densities were the selected features and their influence on the OCV values are taken into account by the ML-based models. Fig. 1 shows the modelling workflow employed in this work from data collection up to the model deployment. On the other hand, Table 1 briefly summarizes the difference between each model and their corresponding pros and cons.

Table 1: Comparison between various ML algorithms used to train the models in this work.

Algorithm	Overview	Advantages	Disadvantages	Source
Gaussian Process Regression	Technique based on non-parametric Bayesian regression wherein relationships between the input and output variables are modeled via Gaussian distribution.	Works well with small datasets, flexible in modeling complex relationships, and gives uncertainty estimates useful in decision-making process.	Challenging physical interpretation due to the complexity of Gaussian processes, inefficient for large datasets, and requires tedious selection of hyperparameter values and kernel functions for accurate modeling.	[31]
Tri-Layered Neural Network	Employs a feedforward architecture consisting of an input, a hidden, and an output layer which is useful when modeling complex, nonlinear relationships between the predictor and response variables.	Model nonlinear relationships with high accuracy, can handle large datasets efficiently, and flexible in different applications.	Requires careful engineering of hyperparameters, prone to overfitting, and challenging interpretability due to the model's complex nature.	[32]
Ensemble	Combination of various models which enhance the prediction accuracy and robustness.	Less sensitive to overfitting unlike the other models, provide more generalized prediction, and can handle outliers and noise effectively.	Less efficient due to the combination of multiple models, challenging interpretability, and ensemble parameters require careful tuning.	[33]
Decision Tree	A simple but powerful model that can handle both numerical and categorical data modeling.	Highly interpretable and can be visualized, flexible in handling numerical and categorical data, and sensitivity to outliers and missing data.	Highly sensitive to small changes in the data, less smooth compared to other models, and prone to overfitting.	[34]
Kernel	Uses high dimensionality in mapping the input variables using kernel functions then performing linear operations within that space.	Effective with multi-dimensional and nonlinear data, more generalizable when regularization is applied, and flexible in handling different types of data.	Inefficient in handling large datasets, highly sensitive to small changes in hyperparameters and kernel function, and less interpretable especially when dealing with nonlinear relationships.	[35]

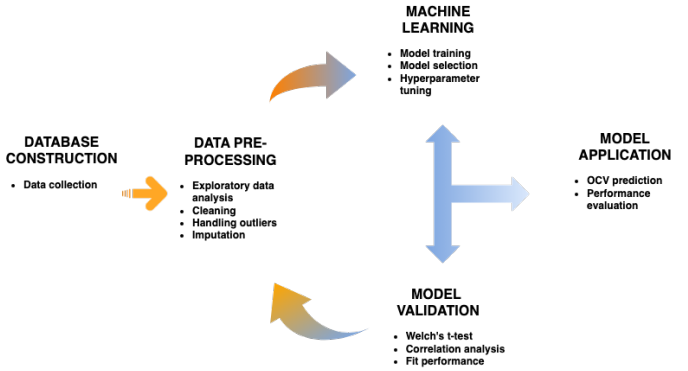

Fig. 1: Modeling workflow used in this work.

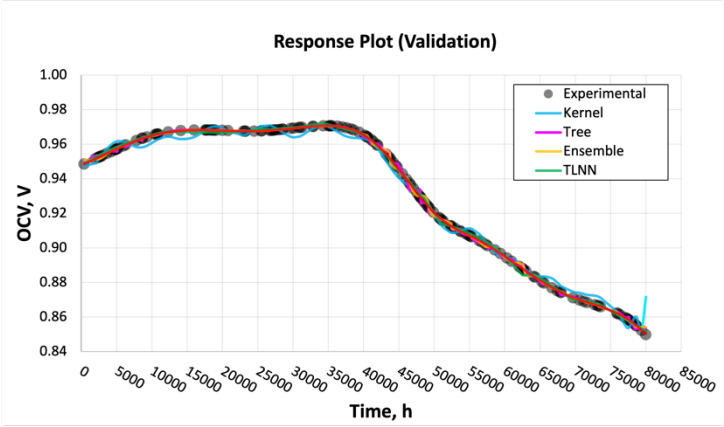

Fig. 2. OCV response of each model versus the experimental data.

2.3. Model Performance Evaluation and Statistical Analysis

The performance of each model was evaluated against the validation dataset based on their RMSE, MSE, and MAE values, as shown in Eqs. (1) - (3). The model that produces the least error values is considered as the best model for predicting the OCV as affected by temporal hydrogen crossover. Furthermore, the coefficient of determination (R^2) and Pearson's correlation coefficient (R) were determined to verify the correlation and goodness-of-fit of each model to the experimental data. Lastly, all models were subjected to Welch's t-test to confirm if there is a statistically significant difference between the model values and actual data of OCV.

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(y_i - \hat{y})^2} \tag{1}$$

$$MSE = \frac{1}{N}\sum_{i=1}^{N}(y_i - \hat{y})^2 \tag{2}$$

$$MAE = \frac{1}{N}\sum_{i=1}^{N}|y_i - \hat{y}| \tag{3}$$

2.4. Model Deployment

The actual performance of an ML model can be justified by the introduction of new data generated beyond the training and validation. In this work, the best performing model was tested using a different dataset and the performance was re-evaluated to check its accuracy when predicting new experimental values. The model performance during deployment was summarized and compared to the validation results during the training.

3. Results and Discussions

Each model was evaluated using the validation dataset randomly sampled from 0- to 80,000-cycle data of temporal hydrogen crossover current density and OCV. The result of model performance is summarized in Table 2. Based on the performance metric values, the GPR-based model gives the least prediction errors among the other models developed in this work. Whereas, ensemble, TLNN, and decision tree models also exhibited excellent approximations of the experimental OCV since their RMSE values range from 0.0011V to 0.0012V which are relatively small and deemed acceptable level of prediction errors. Although the Kernel-based model showed the greatest prediction error value, still, it can indicate a decent predictive model.

Table 2. Performance metric values during model validation.

Parameter	Values				
	GPR	TLNN	Ensemble	Tree	Kernel
RMSE	0.00006783	0.001100	0.001100	0.001200	0.003600
MSE	0.000000004601	0.000001262	0.000001108	0.000001521	0.00001281
MAE	0.00003949	0.0009280	0.0006496	0.0008482	0.002800

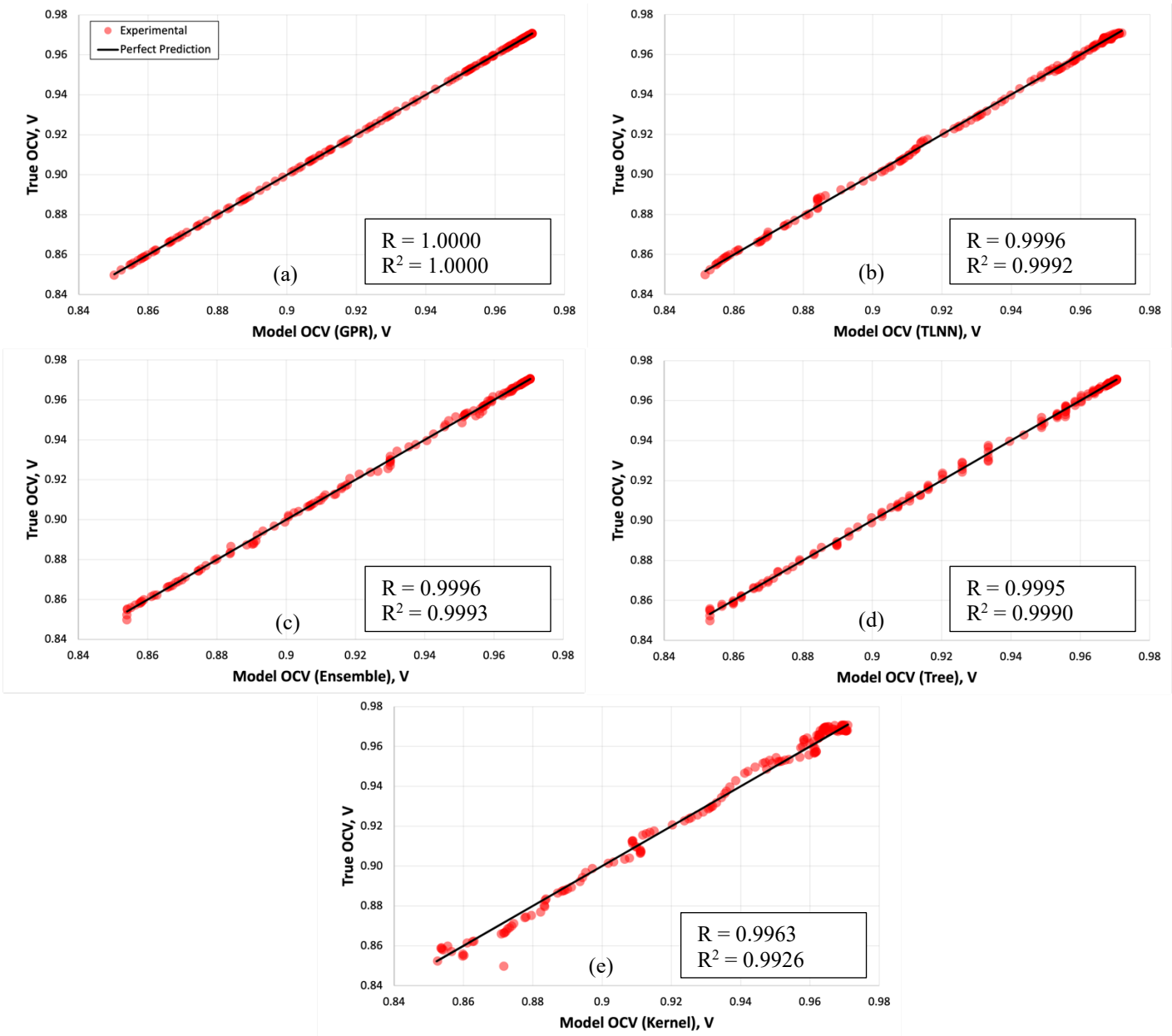

Fig. 3. Correlation between true and predicted OCV values: (a) GPR, (b) TLNN, (c) Ensemble, (d) Decision Tree, (e) Kernel.

The response of each model to temporal hydrogen crossover is illustrated in Fig. 2. The trend of the data suggests that GPR-based OCV value is seen to have the most excellent fit to the experimental data among the other models. Whereas, models such as the Kernel and Ensemble showed observable deviation from the true OCV values. On the other hand, TLNN and Decision Tree-based models are seen to decently fit the experimental data although minimal deviation from the true data can be noticed in their trends as well. One apparent observation from the figure is that the Kernel-based model exhibited the greatest fluctuation and huge prediction errors especially at the end of the AST program. Therefore, the need for further tuning of kernel hyperparameters and choosing a more appropriate kernel function that would increase its predictive performance should be considered. The goodness-of-fit of each model to the experimental data is illustrated in Fig. 3. The R and R^2 values ranging from 0.9926 - 1.0000 and 0.9963 - 1.000, respectively, justify the finding that GPR-based model provides the best prediction accuracy, aligned with the prediction error values.

Although [30] provided insights on the individual trend of the hydrogen crossover and the OCV, the relationship between them was not thoroughly investigated especially in the consideration of the nuances brought about by their individual measurement and their effects to the overall connection between the two variables. For instance, the authors have identified that the increase in the hydrogen crossover was correlated with the decrease in the OCV values. However, the individual effect of operating parameters such as the temperature on the hydrogen crossover and OCV and how it affects the overall relationship between them was not inspected. The increase in operating temperature result to increased solubility and diffusion coefficient of hydrogen, making it more permeable through the membrane [36]. Furthermore, the increased hydrogen crossover due to the influence of operating temperature intensifies the formation of free radicals brought about by the direct reaction between the H_2 and O_2 at the cathode side which compromises the structural integrity of both the membrane and the catalyst layer [37]. Operating PEMFC beyond the optimum temperature could ultimately lead to performance decay due to kinetic losses (via increased molecular mobility which promotes more hydrogen crossover) and ohmic losses (via membrane dehydration) [26]. Aside from this, the presence of contaminants within the PEMFC system could potentially alter the membrane properties leading to increased hydrogen permeation through it and could negatively impact the electrochemical reactions and their kinetics leading to loss in OCV [38] which could have been accounted by [30] but was not tackled in their work. On the other hand, the influence of the mechanical stability of the frame materials, the clamping pressure, and the overall assembly of fuel cell on the increased hydrogen crossover was recognized but the extent of their impact was not thoroughly delved. Given all these influential factors, the relationship between the temporal hydrogen crossover and the OCV decay is indeed complex and nonlinear. Thus, it would make sense that the functionality of GPR-based model has worked excellently with the dataset studied. Also, the GPR-based model was able to comprehend the potential effects of the external factors beyond just the initially set model predictors (i.e., time and hydrogen crossover current density).

The result of statistical analysis is presented in Table 3. Based on the Welch's t-test result, the p-values range from 0.9900 – 0.9990 which indicate that there is weak evidence to reject the null hypothesis and that there is no statistically significant difference between the model and experimental OCV values. Still, GPR-based model achieved the highest p-value among the models developed. In terms of the distribution of model data, the standard deviation and variance values range from 0.0.0384 to 0.0398 and 0.0015 to 0.0016, respectively. Meaning, there is a comparable level of distribution across all models.

Table 4 shows the results of performance evaluation and statistical analysis for GPR-based model against new experimental data during model deployment. The values show promising results of the performance of GPR-based model when tested on a different dataset. Although, there is a decrease in model performance seen during the deployment; however, the values are still within the level of acceptability and indicate a well-performing model for OCV prediction.

Nonetheless, the GPR-based model showed greater generalizability with the introduction of new experimental data subjected to OCV prediction and does not demonstrate overfitting as seen in Fig. 4. The probabilistic nature of GPR makes it an excellent model to flexibly handle uncertainties, noise, and nonlinearity between the relationships in the dataset [39]. Furthermore, GPR-based model often works remarkably even with small datasets which makes it low-cost and efficient surrogate models to the other intensive frameworks and experimental validation methods [40].

Table 3. Statistical analysis results per model.

Parameter	Values				
	GPR	TLNN	Ensemble	Tree	Kernel
Null hypothesis	Not rejected	Not rejected	Not rejected	Not rejected	Not rejected
p-value	0.9990	0.9902	0.9929	0.9972	0.9900
Standard deviation, σ	0.0397	0.0398	0.0396	0.0397	0.0384
Variance, σ^2	0.0016	0.0016	0.0016	0.0016	0.0015

Table 4. Summary of the performance evaluation during deployment.

Parameter	Value
RMSE	0.0053
MSE	0.00002765
MAE	0.0030
Null hypothesis	Not rejected
p-value	0.9995
Standard deviation, σ	0.0360
Variance, σ^2	0.0013

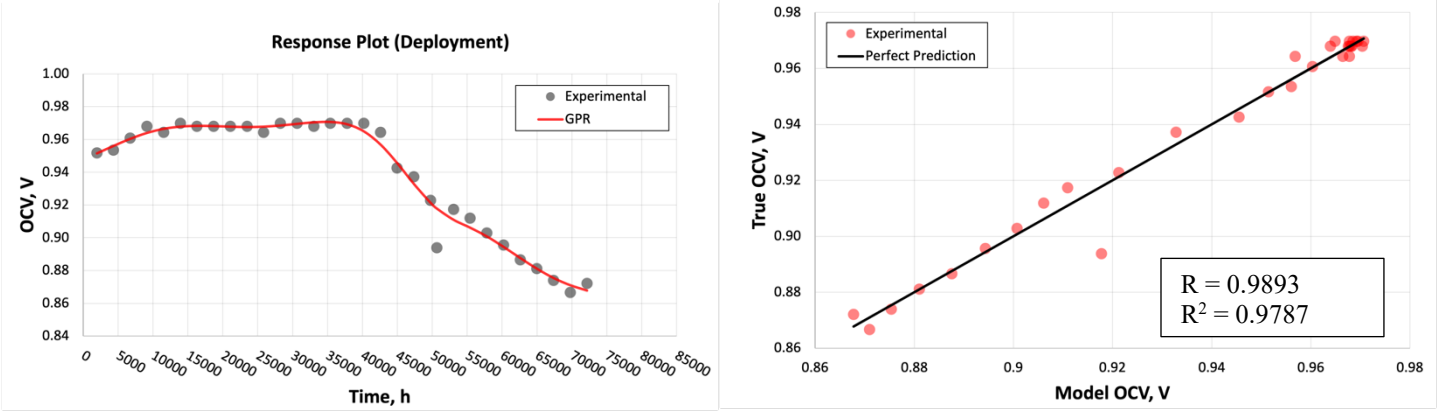

Fig. 4. Response (left) and correlation (right) plots for the OCV estimation of GPR-based model using new data.

4. Conclusion

This work focused on the development of ML-based models to predict the OCV against the temporal hydrogen crossover using Matlab. The dataset from [30] was used to train the ML models via employing five algorithms namely the Gaussian Process Regression (GPR), Tri-Layered Neural Network, Ensemble, Decision Tree, and Kernel methods. The results from the performance evaluation during model validation, statistical analysis, and fit performance consistently distinguished the GPR-based model as the best predictive model for OCV against the experimental data for temporal hydrogen crossover with R and R^2 values of 1.0000 and 1.0000, respectively. However, a decrease in these values to 0.9893 and 0.9787 was seen during the deployment performance evaluation. Although this is the case, the prediction error values given by the RMSE, MSE, and MAE values of 0.0053, 0.00002765, and 0.0030, respectively are still within the acceptance level and indicative of a well-performing model. Furthermore, the GPR-based model have shown ability to generalize on new data and minimize overfitting as seen in the trends of both the experimental and model OCV values which makes it an excellent model for deployment. The validity of the model is only limited within the range of the dataset used during training and prediction beyond 80,000 cycles must be subjected to further validation. Although the GPR-based model has effectively comprehend the hidden patterns in the dataset including the possibility of the external factors affecting the trend in hydrogen crossover and OCV that were not thoroughly delved in the work of [30], it is good to explicitly identify the direct effects other degradation parameters such as the operating conditions of PEMFC, degradation mechanisms (e.g., the release of fluorine ion at the cathode effluent), gas diffusivity and solubility in the membrane, cycling patterns, and etc. on the OCV to improve the prediction accuracy and to provide more context about the underlying mechanism governing the decay in fuel cell performance. Essentially, this work has demonstrated the effectiveness of using data-driven modeling methods to address the issues on the cost of analysis, time inefficiency, and intricacy of handling the complex, nonlinear PEMFC degradation.

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