Hybrid Modeling of PEM Fuel Cell: Machine Learning of Equilibrium Water Content within a 1D Cell Model

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Abstract

Low-temperature proton-exchange membrane fuel cells (PEMFCs) require careful water management in order to keep the membrane hydrated for high proton conductivity and to keep the gas channels from flooding with liquid water. Accurate dynamical models are needed for model-based approaches for controller and soft sensor design. In the case that some properties of the fuel cell components are not well known or physical models for them are not available, a hybrid model, combining a first-principles-based model with data-based submodels, may be applied. This work explores how a specific submodel of a PEMFC, namely the equilibrium water content of the membrane, can be determined from measurement data that would be readily available from a fully assembled fuel cell. Synthetic data generated from a reference PEMFC model are used to train the hybrid model. Results for data with different sampling rates and levels of added noise are compared to study the required data quality for successful training.

**Keywords**: PEMFC, hybrid modeling, machine learning

* 1. Introduction

The pivotal component of a PEMFC is the membrane. It provides the separation of gases between the anode and cathode gas channels, as well as the transport of protons. Ensuring sufficient membrane hydration is essential to maintain high proton conductivity and, consequently, achieve efficient power generation (Singh et al., 2022). Model-based water management requires the establishment of reliable dynamical models of PEMFC. Special attention should be directed to the membrane submodel, as it often includes empirical dependencies derived from specific experiments conducted in the earlier phases of PEMFC development (Liso et al., 2016). One challenge here is the accurate description of the water entering the membrane. The equilibrium water content of the membrane depends on the water vapor activity and the temperature in the gas phase. The reliability of empirical relations determined for specific membrane materials or operating ranges is a concern if they are applied outside of these conditions. Determining this relation directly from measurement data of a fully assembled fuel cell instead might increase model fidelity with limited additional effort.

In this paper, a hybrid PEMFC model is proposed (Martensen et al., 2023), offering the advantage of preserving the physical states within the system and the possibility to learn the equilibrium water content from data. Here, it is assumed that the equilibrium water content function can be approximated using an artificial neural network (ANN). To determine the parameters of the ANN, synthetic data sampled from the output signals of a reference model are used. Output signals that can typically be measured on a fully assembled PEMFC are selected for this purpose. The influence of data sampling rates and measurement noise on the training process and resulting function is studied.

* 1. Materials and Methods
		1. Reference model of a PEMFC

In this case study, a reference model is used to generate synthetic data in place of actual measurements. The PEMFC reference model is based on (Mangold et al., 2010), while the details of the membrane model can be found in (Neubrand, 1999). This is a 1D along-the-channel, first-principle, single-phase, non-isothermal model that is divided into the following parts: anode and cathode gas channels, gas diffusion layers, catalyst layers, membrane and electrode. The resulting model consists of a set of partial differential equations (PDEs) and algebraic equations.

For numerical simulations, the spatial coordinate was discretized using the finite volume method with $N\_{cv}$ control volumes. The resulting system of differential and algebraic equations (DAEs) has the following structure:

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| --- | --- |
| $$\left[\begin{matrix}I&0\\0&0\end{matrix}\right]\left[\begin{matrix}\dot{x}\_{d}\\\dot{x}\_{a}\end{matrix}\right]=\left[\begin{matrix}f\_{d}(t,x\_{d},x\_{a},u)\\f\_{a}(t,x\_{d},x\_{a},u)\end{matrix}\right]$$ | (1) |

with initial conditions for $t\_{0}=0$

|  |  |
| --- | --- |
| $$\begin{matrix}x\_{d}\left(t\_{0}\right)=x\_{d,0} ,\\f\_{a}\left(t,x\_{d,0},x\_{a,0},u\right)=0 ,\end{matrix}$$ | (2) |

where $x\_{d}\in R^{11N\_{cv}}$ is the vector of dynamic states that consists of the concentrations of species in the anode (H2, H2O) and cathode (O2, N2, H2O) gas channels, water content in the membrane, electrical potential difference, internal energies in gas channels, and total energy of the solid, $x\_{a}\in R^{12N\_{cv}+1}$ is the vector of algebraic states that consists of the mole fractions of the species in the catalyst layers, electrical potential and current density of the membrane, temperatures of gases in the channels and solid, pressures in the gas channels, and voltage of the fuel cell, $u\in R^{11}$ is the vector of inputs that consists of input molar flows of reactants and water, temperature of gases at the inlets, coolant temperature, output pressures of the channels, and electrical current.

Combining the dynamic and algebraic states together and considering the output equation, the reference model of PEMFC can be rewritten in compact form as

|  |  |
| --- | --- |
| $$\begin{matrix}M\dot{x}=f\left(t,x,u\right) ,\\y=h\left(t,x,u\right) ,\end{matrix}$$ | (3) |

where $y\in R^{3}$ is the vector of outputs that consists of the voltage and output flows of water vapor from the gas channels.

The water content at the membrane surface is assumed to be in equilibrium with the gas phase of the neighboring catalyst layer. There is a number of different equilibrium water content functions or so-called sorption isotherms that can be found in the literature (Dickinson et al., 2020). For the reference model, two polynomial relations are used that have been fitted to experiments for the temperature points 30 °C (Springer et al., 1991) and 80 °C (Hinatsu et al., 1994):

|  |  |
| --- | --- |
| $$\begin{matrix}Λ\_{30}=36 a\_{H\_{2}O}^{3}- 39.85 a\_{H\_{2}O}^{2}+17.81 a\_{H\_{2}O}+0.043 , \\Λ\_{80}=14.1 a\_{H\_{2}O}^{3}- 16 a\_{H\_{2}O}^{2}+10.8 a\_{H\_{2}O}+0.3 ,\end{matrix}$$ | (4) |

where $a\_{H\_{2}O}$ is the relative humidity in the gas phase.

For temperatures between these values, linear interpolation is applied, resulting in the following function of two variables, see Fig. 1,

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| --- | --- |
| $$Λ=f\_{Λ}\left(a\_{H\_{2}O},T\right) $$ | (5) |

that is used in the anode and cathode catalyst layer for each control volume.

* + 1. Hybrid model of a PEMFC

The hybrid model is trained on data in order to reproduce the physical behavior of a PEMFC, represented in this case study by the reference model.

Each instance of $f\_{Λ}$ in the reference model is replaced in the hybrid model by

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| --- | --- |
| $$\hat{f}\_{Λ}=a\_{H\_{2}O} f\_{ANN}(v,θ) ,$$ | (7) |

where $f\_{ANN}$ is a feedforward ANN with a single hidden layer with hyperbolic tangent activation function and a linear output layer, $v= \left[\begin{matrix}a\_{H\_{2}O}&T\end{matrix}\right]^{T}$ is the vector of the ANN inputs and $θ\in R^{N\_{θ}}$ is a parameter vector that consists of the weights and biases of the ANN layers.

In the resulting hybrid model structure

|  |  |
| --- | --- |
| $$\begin{matrix}\begin{array}{c}M\dot{x}=\tilde{f}\left(t,x,\tilde{x},u\right) ,\\\tilde{x}=f\_{\tilde{x}}\left(x\right) ,\end{array}\\y=h\left(t,x,u\right) ,\end{matrix}$$ | (6) |

$\tilde{x}$ are auxiliary variables in the right-hand-side function that are calculated by the submodel $f\_{\tilde{x}}\left(x\right)$. The function $f\_{\tilde{x}}$ comprises all appearances of the equilibrium water content function $Λ=f\_{Λ}\left(a\_{H\_{2}O},T\right)$ in the discretized model.

* + 1. Parametrization problem

In this paper, reconstruction of the function from the PEMFC output vector is considered. The objective function based on the mean squared error (MSE) is used as follows:

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| --- | --- |
| $$J\left(θ\right)=\frac{1}{N\_{exp}} \sum\_{i=1}^{N\_{exp}}(y\_{i}^{s}-\hat{y}\_{i}^{s}(θ))^{2},$$ | (8) |

where, $y^{s}\in R^{3×N\_{exp}}$ is the min-max scaled output vector from the collected data, $N\_{exp}$ is number of sample points, and $\hat{y}^{s}\in R^{3×N\_{exp}}$ is the min-max scaled output vector of the hybrid model calculated for the parameter set of ANN $θ$.

* 1. Numerical setup

The reference and hybrid models are implemented in MATLAB 2022b (The MathWorks, Inc., Natick, MA, USA). For the discretization $N\_{cv}=20$ is used, which results in 220 dynamic and 241 algebraic states. To solve these models efficiently, the ode15s solver with sparse mass matrix *M* and additional Jacobian pattern options are used. In addition, for the right-hand-side functions of the state equations in (3) and (6), MEX-functions are generated and used for the simulations.



Figure 1 – Locations of data points for internal variables (left, every 20th point is plotted) and shape of reference equilibrium water content $f\_{Λ}$ (right).

The reference model is used to generate output data of the PEMFC for the parametrization of the hybrid model. The function $\hat{f}\_{Λ}$ is supposed to approximate the $f\_{Λ}$ well over its entire domain, which is restricted to values $0.05\leq a\_{H\_{2}O}\leq 0.95$ and $313\leq T\leq 373$ K. For this purpose, an input trajectory is manually designed such that, according to the reference model, the domain of $f\_{Λ}$ is well covered. The resulting input trajectory contains 14 set points with 2 min of transition time and 8 min of steady-state time. To smoothen the trajectories during the transition, shape-preserving piecewise cubic interpolation was used. In Fig. 1, coverage of the domain for $f\_{Λ}$ is depicted. The sample data $d=\left(y,t\_{k}\right)$ for input trajectory$u$ contains the output matrix $y\in R^{3×N\_{exp}}$ and time vector $t\_{k}\in R^{N\_{exp}}$. The sampling time $t\_{s}=1$ s is chosen for the nominal case 1. To study how the quality of data influences training, three additional data sets with a different sampling time and added normally distributed noise are generated:

* case 2: $t\_{s}=5$ s, no added noise;
* case 3: $t\_{s}=1$ s, added normally distributed noise ($σ=0.5\%$ for voltage, $σ=2\%$ for output flows);
* case 4: $t\_{s}=5$ s, added normally distributed noise ($σ=0.5\%$ for voltage, $σ=2\%$ for output flows).

The ANN $f\_{ANN}$ includes 10 neurons in the hidden layer resulting in $N\_{θ}=41$ parameters that need to be estimated from training. For the ANN inputs and outputs min-max scaling is used. The initialization of the ANN parameters needs to be approached with caution to ensure that a numerical solution for the overall model can be calculated. Here, the parameters of the hidden layer are initialized randomly and the parameters of the output layer are initialized such that $\hat{f}\_{Λ}$ is approximately linear in $a\_{H\_{2}O}$ and constant in $T$, yielding the initial set of ANN parameters $θ\_{init}$.

The training of the ANN is based on minimizing the objective in (8). Here, MATLAB’s fminunc is used with default options except step tolerance set to$ 10^{-16}$ and specified objective gradient. The objective gradient is calculated using the complex-step method.

* 1. Results

The model was trained separately for cases 1-4, starting from the same initial parameters $θ\_{init}$. The results are visualized in Fig. 2 and Fig. 3. Fig. 2 demonstrates the convincing agreement between the output of the trained hybrid model and the sample data with added noise, i.e. case 2. Fig. 3 shows the errors between equilibrium water content in the trained hybrid model and the reference model. The mean values of the errors $\left|\hat{f}\_{Λ}-f\_{Λ}\right|$ are summarized in Table 1. Despite the decrease in data quality in the considered cases, only insignificant errors are observed in the domain of interest, except near its boundaries, where the density of data points is lower.



Figure 2 – Outputs of PEMFC for sample data with added noise and trained hybrid model. Cell voltage (left), water vapor flow at anode (middle) and cathode (right).



Figure 3 – Error $\hat{f}\_{Λ}-f\_{Λ} $between equilibrium water content in the trained hybrid model and the reference model. Training on datasets for the nominal case 1 (top left), case 2 (top right), case 3 (bottom left), and case 4 (bottom right).

Table 1 – Mean values of the absolute error $\left|\hat{f}\_{Λ}-f\_{Λ}\right|$

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Cases | 1 | 2 | 3 | 4 |
| Mean error | 2.31e-2 | 2.26e-2 | 3.89e-2 | 4.14e-2 |

* 1. Conclusions

In this work, a hybrid model for a PEMFC containing a data-based submodel for the equilibrium water content of the membrane has been presented. It has been shown that the equilibrium water content can be accurately approximated from the output signals that can be measured on a fully assembled PEMFC. In addition, reduced sampling rates and added measurement noise did not lead to significantly reduced accuracy or overfitting issues.

Future work will be dedicated to the training of the hybrid model on real measurements of a PEMFC and corresponding design of experiments, as well as the extension to a larger number of data-based submodels.

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