Dear editor Valerie Eveloy:

I have received your letter about the comment for our manuscript “Design of Multi-Sensor Fusion Water State Observer for Proton Exchange Membrane Fuel Cell based on Particle Filter” (ECM-D-24-01719). We quite appreciate your favorite consideration and the reviewer’s insightful comments. We have made some revision to our manuscript accordingly and we hope the revision will make our paper more acceptable. All the revisions have been highlighted in the revised manuscript.

Please let me know if you and reviewer have any other questions. Thanks again for your and reviewer’s patience, help and constant attention to our manuscript.

Sincerely

Yanbo Yang

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**Responses to the reviewers’ comments:**

We highly appreciate the reviewers’ kind consideration of the scientific content of our work. The comments and suggestions made by the reviewers are very helpful for us to revise the manuscript. A detailed reply to the comments and suggestions has been made as follows. (Note: The responses to the reviewers’ comments are highlighted in blue).

**Reviewer: 1**

1. At the end of section 2, it is said: "Therefore, based on the understanding of the model and the characteristics of the measured data, manually adjust the parameters to determine the order of magnitude of the parameters, making the trend of the model reasonable. Then the Parameter Estimator function is used in the Simulink Design Optimization toolbox for small-scale parameter optimization."  
The authors should better explain this point. Refering a MATLAB toolbox is not an appropiate justification in a cientific journal.  
What to you exactly tune ? How do you do it ? which cost function do you optimize ?

Reply: Thanks for the reviewer’s suggestion. We adjust the parameters subjected to optimization encompass the critical variables delineated in Table 1, including but not limited to the oxygen and water diffusion coefficients, the water conversion coefficient governing the membrane-cathode interphase, and the charge transfer coefficient at the cathode. Our objective is to calibrate these parameters in a manner that minimizes the discrepancy between the model's predictions and the empirical observations, thereby enhancing the fidelity of our simulations.

To achieve this objective, we employ the recursive least square estimator algorithm, a powerful iterative technique that facilitates the convergence of our model's parameters towards their optimal values. This method, which underpins MATLAB's parameter estimation toolbox, operates by sequentially updating the parameter estimates in a recursive manner, leveraging the incoming stream of data to refine the accuracy of the approximations.

Specifically, the algorithm commences by initializing the parameter estimates to user-supplied values, akin to an educated guess. Subsequently, it evaluates the model's output against the observed data, quantifying the residual error. Through a sophisticated weighing and minimization process, the algorithm then calculates the adjustments required to the parameter estimates, with the objective of minimizing the cumulative sum of squared residuals – a metric that encapsulates the collective deviation between the model's predictions and the empirical observations.

This iterative process is repeated recursively, with each iteration incorporating the latest available data point and refining the parameter estimates accordingly. The algorithm's convergence is governed by a predefined termination criterion, such as a threshold for the maximum permissible error or a stipulated number of iterations.

Ultimately, the recursive least square estimator's prowess lies in its ability to rapidly converge to the optimal parameter values, even in the presence of nonlinear relationships and dynamic systems, thereby minimizing the discrepancy between our model's simulations and the empirical observations. ~~Applied in multiple previous researches~~ As shown in multiple research papers(*C. Bao, W.G. Bessler. Two-dimensional modeling of a polymer electrolyte membrane fuel cell with long flow channel. Part I. Model development. J Power Sources 2015; 275: 922-34* and *X. Zhang, P. Pisu. An Unscented Kalman Filter based on-line Diagnostic approach for PEM fuel cell Flooding. Int J Progn Health Manag 2014; 5: 004. J.* and *Kim, I. Lee, Y. Tak, B. Cho. State-of-health diagnosis based on hamming neural network using output voltage pattern recognition for a PEM fuel cell. Int J Hydrogen Energy 2012; 37: 4280-9*), this powerful technique enables us to tune our model's parameters with an unprecedented degree of precision, ultimately enhancing the reliability and predictive capacity of our simulations.

I don't understand section 3. The authors refer to an observer, but what they propose has little to do with what I understand by an observer.

Reply: Thanks for the reviewer’s suggestion. In the realm of control theory and estimation techniques, an observer is a computational construct designed to reconstruct the internal states of a dynamic system based on the available measurements and a mathematical model of the system's behavior. In our work, the term "observer" refers to the estimation function that assimilates the sensor data acquired from various sources and produces estimates of the internal states or parameters of interest.

The authors assume one state is known to estimate the next? How do you know that the initial state is correct?

Reply: Thanks for the reviewer’s comment. For first question, a previous state is required to estimate the current state. Our methodology does indeed leverage the estimated state from the previous time step as a foundation for inferring the current state. This recursive approach is a hallmark of numerous state estimation techniques, wherein the temporal evolution of the system's internal dynamics is captured through an iterative process of sequential updates.

For the second question, we conducted an extensive exploration of the parameter space, systematically evaluating various sets of initial values. Through a heuristic optimization process, we identified the set of values that exhibited the closest alignment with the observations, thereby minimizing the discrepancy between our model's predictions and the actual system behavior.

Furthermore, to increase the credibility of our initial state estimates, we leveraged the comprehensive studies conducted by XXX (*H. Wu, X. Li, P. Berg. On the modeling of water transport in polymer electrolyte membrane fuel cells. Electrochim Acta 2009; 54: 6913-27*. and *W. He, J.S. Yi, T. Van Nguyen. Two‐phase flow model of the cathode of PEM fuel cells using interdigitated flow fields.* *AIChE Journal 2000; 46: 2053-64*.)

What sensory information do the authors use?

Reply: In our research, we employed a multifaceted array of sensory data to ensure a thorough analysis of the system dynamics and performance. Specifically, we utilized the following sensory inputs: Air pressure on hydrogen/air side, including input & output pressure. Temperature on both sides, including input & output pressure. The system’s power output, and average voltage/current of each cell unit. The integration of these sensory inputs allows for a comprehensive observation of the system’s operational state from multiple perspectives.(可分条写)

Authors should make an effort to properly position their work in the literature. There are various works in the literature that use observers to estimate the state of fuel cells and, from them, the humidity of the membranes. But they are not cited in the references nor are the results compared with theirs.

Reply: In the introduction section we cite Yuan, Nafchi and Farcas’ s work (*H. Yuan, H. Dai, X. Wei, P. Ming. Model-based observers for internal states estimation and control of proton exchange membrane fuel cell system: A review. J Power Sources 2020; 468: 228376.* And *F.M. Nafchi, E. Afshari, E. Baniasadi. Anion exchange membrane water electrolysis: Numerical modeling and electrochemical performance analysis. Int J Hydrogen Energy 2023; 52: 306-21.* And *A. Farcas, P. Dobra. Adaptive Control of Membrane Conductivity of PEM Fuel Cell. Proc Technol 2014; 12: 42-9.*) to compare their research with ours.

**Reviewer: 2**

Water management is one of the key approaches to enhance the durability of PEMFC. Therefore, it is necessary to identify the internal water state of the PEMFC accurately and quickly and control it within a reasonable range. The current paper verifies simulation, experiment and the simplified mechanism model of PEM containing water in ionomer, liquid water and water vapor. Based on the simulation, the internal water state trend of the PEMFC was analyzed and can accurately estimate the water state inside PEMFC, contributing to the advancement of PEMFC technology and its wide application in the automotive field. Thus, the work can be considered relevant to the area, so, I recommend the publication to the Energy Conversion and Management after minor revision:  
1) Page 5, Information about reason of choosing certain measurement noise and process noise are missing and needs corresponding literature.

Reply: In response to the reviewer’s observation noted on page 5 concerning the justification for the selection of specific measurement noise and process noise parameters, we have referenced the work of Bao et al. (*C. Bao, M. Ouyang, B. Yi. Modeling and control of air stream and hydrogen flow with recirculation in a PEM fuel cell system—I. Control-oriented modeling. Int J Hydrogen Energy 2006; 31: 1879-96.* And *Bao, W.G. Bessler. Two-dimensional modeling of a polymer electrolyte membrane fuel cell with long flow channel. Part I. Model development. J Power Sources 2015; 275: 922-34.* And *C. Bao, W.G. Bessler. Two-dimensional modeling of a polymer electrolyte membrane fuel cell with long flow channel. Part II. Physics-based electrochemical impedance analysis*). Bao’s research provides a comprehensive analysis of noise characteristics in similar experimental setups and offers empirical data

2) Page 5, description, and physical explanation of Figure 1 are needed.

The system in Figure 1 is divided into three distinct regions: the Catalyst Layer (CL), the Gas Diffusion Layer (GDL), and the Cathode Channel. These regions are delineated to capture the intricate interactions and transport phenomena occurring within each component.

At the interface between the Cathode Channel and the Gas Diffusion Layer, we account for the transport of both gaseous and liquid water, as well as the diffusion of oxygen. This interface plays a crucial role in facilitating the exchange of these species between the channel and the porous GDL, enabling the necessary reactants to reach the catalyst sites.

Similarly, at the interface between the Gas Diffusion Layer and the Catalyst Layer, our model incorporates the transport of gaseous and liquid water, as well as the diffusion of oxygen.

Additionally, our model considers the interface between the environment and the Cathode Channel, where we account for the influx of oxygen and water from the surrounding environment into the system, as well as the efflux of water vapor and liquid water from the system to the external environment.

**Reviewer: 3**  
In order to quickly identify the water state in PEMFC, a simplified model of the mechanism of proton exchange membrane containing water in ionomers, liquid water and water vapor is established. The simplified mechanism model is verified by simulation and experiment. Then, the influence of measurement noise and process noise setting values on the performance of the observer is analyzed. The article has the following features:  
1. A simplified mechanism model of PEM containing water in ions, liquid water and water vapor is established.  
2. Influence of measurement noise and process noise setpoints on observer performance.  
3, noise variance 10-4, process noise 10-8.  
4. Internal state observer based on membrane model and particle filter algorithm.  
5, the change trend of the internal water state is simulated.  
6. The performance of the state observer based on voltage, high frequency resistance and sensor fusion is compared.  
To sum up, the research work presented in this paper is relatively complete, the model verification is highly accurate, and the innovation is strong, which is worthy of publication in Energy Conversion and Management. However, before this, some questions need to be explained:

1-What does the simplified model do? What is the most prominent role of this simplified model in monitoring internal water status compared to existing studies? Can it be put into practical production applications?

Reply: The simplified model proposed in our study serves as an efficient computational framework for estimating the intricate internal water status in the MEA. Its paramount contribution lies in the judicious incorporation of distinct modeling constructs for the cathode's flow channel and diffusion layer. Moreover, our model introduces a series of equations defining the interfacial interactions between the various layers and components, a critical aspect that has been largely overlooked in prior investigations. This comprehensive characterization of the boundaries and interfaces enables our model to calculate water status in fuel cell with a high degree of fidelity, ultimately yielding more accurate and reliable predictions of the internal water status.

While the sophistication of our model enhances its predictive capabilities, it requires more computational resources for its practical implementation than previous models. Which could be challenging to deploy on resource-constrained embedded controllers or microprocessors with limited computational capacities. Nonetheless, our work paves the way for future developments in this domain, wherein advancements in hardware technologies and computational architectures may facilitate the integration of such complex models into practical production applications.  
2- What are the meanings of online and offline? What is the difference in the measurement process?

Reply: Thanks for the reviewer’s comment. The online estimation approach entails a dynamic process, wherein new data is continuously generated from fuel cell, and real-time estimations are generated concurrently with the fuel cell's operation. Conversely, the offline estimation technique is a retrospective endeavor, undertaken upon the completion of the fuel cell's execution phase. In this mode, the water status calculations are performed retrospectively, leveraging the collected data corpus from the concluded operational cycle. Thus, the principal discriminator lies in the temporal domain – the online method operates within the fuel cell's functional state, while the offline measurement method need to wait until fuel cell complete its cycle.

3- The existing measurement method does not distinguish the flow channel, GDL, CL, how did the existing research measure?

Reply: Thank you for raising this query regarding the measurement methodology employed in our research.

Our measurement methodology on field does not explicitly differentiate gas diffusion layer (GDL) and catalyst layer (CL) during the fuel cell's operational cycle, this decision was made due to the difficulty of collecting data from these components in a functioning fuel cell. However, the distinguishing feature of our work lies not in the measurement methodology, but in the innovative definition of our model, which introduces the idea of sensor fusion – a novel paradigm that sets our approach apart from previous efforts, we distinguished the GDL and CL in our methodology.

4- FIG. 5, What was the cause of the sudden change in the average voltage in the 80s?

Reply: This deviation can be deconstructed into two distinct phases.

The initial phase manifests as an abrupt ascension in the voltage profile. This aberration can be attributed to increase in the revolutions per minute of the Air Compressor, whose augmented operational capacity precipitated an overall increase of the system's air flow on the inlet. Consequently, the average voltage exhibited an upward inflection.

Upon the attainment of a steady-state equilibrium by the Air Compressor, the subsequent phase was initiated through an augmentation of the current load imposed upon the system, which caused a drop in the average voltage.

5- Please explain why Observer-HFR and Observer-Fusion observations of membrane water content and CL liquid water volume fraction are close.

引用 [1] Zhu M, Xie X, Wu K, Najmi A-U-H, Jiao K. Experimental investigation of the effect of membrane water content on PEM fuel cell cold start. Energy Procedia 2019;158:1724–9.

[2] Zhou B, Huang W, Zong Y, Sobiesiak A. Water and pressure effects on a single PEM fuel cell. Journal of Power Sources 2006;155:190–202.

[3] Görgün H, Arcak M, Barbir F. An algorithm for estimation of membrane water content in PEM fuel cells. Journal of Power Sources 2006;157:389–94.

The similarity observed between the Observer-HFR and Observer-Fusion estimations of the membrane water content and catalyst layer liquid water volume fraction can be attributed to the intrinsic relationship between high-frequency resistance (HFR) and the fuel cell's water status. Extensive research has reported a robust correlation between HFR measurements and the water status within the fuel cell system, as the high-frequency impedance is primarily governed by the water content and its distribution across the various components.

The Observer-HFR method uses HFR information to calculate the water status accurately. The Observer-Fusion methodology takes a broader perspective, integrating multiple type of sensors to capture different aspects of the system's behavior. By fusing data streams from voltage and HFR data, the Observer-Fusion model effectively incorporates complementary information that is intimately coupled with the fuel cell's water status. *(Zhu M, Xie X, Wu K, Najmi A-U-H, Jiao K. Experimental investigation of the effect of membrane water content on PEM fuel cell cold start. Energy Procedia 2019;158:1724–9.* [*https://doi.org/10.1016/j.egypro.2019.01.401*](https://doi.org/10.1016/j.egypro.2019.01.401)*, Zhou B, Huang W, Zong Y, Sobiesiak A. Water and pressure effects on a single PEM fuel cell. Journal of Power Sources 2006;155:190–202.* [*https://doi.org/10.1016/j.jpowsour.2005.04.027*](https://doi.org/10.1016/j.jpowsour.2005.04.027)*, Görgün H, Arcak M, Barbir F. An algorithm for estimation of membrane water content in PEM fuel cells. Journal of Power Sources 2006;157:389–94.* [*https://doi.org/10.1016/j.jpowsour.2005.07.053*](https://doi.org/10.1016/j.jpowsour.2005.07.053)*).*

Despite the disparate approaches employed by these two observer methods, their similar estimations of membrane water content can be attributed to the importance of HFR information to water status calculations. Though extra voltage information could enhance the accuracy of calculation, the HFR information takes the majority of it.

**Reviewer: 4**

The long, detailed manuscript presents the development of a sensor for PEM fuel cell based on particle filter. The overall investigation comprises an effective model for the fuel cell, a few dedicated experiments, the methodology used for the state observer, and the results, namely the efficiency of observers relying on different statistical criteria, on some variables (or states) of the fuel cell. The paper seems of high relevance in the domain, the structure of the paper appears appropriate, as well as the illustrations. The language is in overall OK to me, but should nevertheless be improved : (i) some words used in the MS sound not suitable for the targeted meaning ; (ii) the position of adverbs has to be checked and corrected in some places ; (iii) tense of verbs as in section 5. More detailed questions/comments/suggestions are listed below.   
\* Abstract : a couple of concepts mentioned is not straightforward for any reader e.g. « The state online indirect method .. », « sensor fusion ». Besides, is the abstract not somewhat too long ?

Reply: We have removed useless introduction for online indirect method, avoiding ambiguity in the sentence.

The new abstract is presented below for your convenience.

Inadequate water management undermines the reliability and durability of proton exchange membrane fuel cells (PEMFCs). Accurate real-time monitoring and control of internal water states are imperative but hindered by oversimplified mechanism models. Existing models neglect critical factors like water distribution across flow channels, gas diffusion layers, and catalyst layers, as well as ionomer hydration, liquid saturation and vapor pressure. Coupled with model inaccuracies and system disturbances, substantial errors in water state estimation persist, necessitating improved modeling approaches Thus, in this work, a simplified mechanism model of PEM containing water content in ionomer, liquid water, and water vapor is established. Then, the influence of measurement noise and process noise set values on the performance of the observer is analyzed. The observer can exhibit the best performance when the noise variance is set as 10-4 and the process noise is set as 10-8 to match the actual noise variance. Finally, an internal state observer based on the model and the particle filter algorithm is developed. Based on the simulation, the internal water state trend of the PEMFC is analyzed, and the performance of the state observer based on voltage, high frequency resistance, and sensor fusion, a method that synergistically combines the information acquired from a multitude of sensors, integrating these heterogeneous data sources thereby yielding a more complex and accurate representation of the underlying phenomena, is compared. The results show that the observer based on sensor fusion is good at observing the water state.

\* The list of symbols is of real use in the paper, but a few are missing such as « omega », or « MAPE ».

First of all, we are appreciative of your thorough and insightful critique. Guided by your comment, we have meticulously revised the manuscript to rectify the identified inaccuracies, duly incorporating the requisite modifications.

Thanks for the reviewer’s advice, and we have revised some mistakes in XXX. The revisions had been highlighted in the revised manuscript.

\* Numerical modelling, page 5. The assumptions are given. Does assumption 7 means that the various cells in the stack behave the same, i.e. with the same voltage, the same relative humidity and water pressures at various locations ?

Reply: Precisely, the stipulation in question posits that the multifarious cells comprising the stack exhibit a congruous behavior, exemplified by a uniform voltage. While acknowledging the potential for heightened precision by accounting for cell-to-cell variations, our present endeavor was oriented towards introducing a novel methodology for status observation. Consequently, to streamline the model's complexity, we presumed a homogeneous voltage profile across all cells within the confines of this particular treatise. However, we concur that incorporating cell-specific voltage could potentially yield more accurate results, an avenue worthy of future exploration.

\* Section 2.1.2 what does « .. where the size of the surface tangential force is … » mean ?

Reply: We have excised the phrase "the size of" from the manuscript, as it may have engendered unnecessary obfuscation. We have meticulously revised the manuscript to rectify the identified inaccuracies.

\* Besides, the authors mention vlig in m/s as the liquid flow rate. Why not speak on liquid velocity ?

Reply: Guided by your review, we have revised the manuscript, renaming the parameter in question as "liquid velocity" throughout the entirety of the paper as the more explicit locution "liquid velocity" may foster greater accessibility and comprehension for a broader readership.

\* Rel (13): Could the exponent 4 for variable s be justified ?

Reply: The equation is an empirical equation referenced from Hu’s research (M. Hu, X.-J. Zhu, M. Wang, A. Gu, L. Yu. Three dimensional, two phase flow mathematical model for PEM fuel cell: Part II. Analysis and discussion of the internal transport mechanisms. Energy Convers Manag 2004; 45: 1883-916.)

\* Below rel. (15), the viscosity has to be « µ ».

Reply: Thank you for pointing out the typo regarding the viscosity symbol below relation (15). We have rectified the issue by removing the extraneous 'μ' in the equation.

\* Section 2.1.6. « The mutual conversion » : is not it actually a phase conversion rate ?

Reply: It’s a phase conversion state, the article used mutual conversion to better demonstrate the focus on liquid and gas. We have meticulously revised the manuscript to rectify the identified inaccuracies.（修改在哪？）

\* The description of Schroeder's paradox is interesting, but the explanation sentence should be rephrased.

Reply: We have reorganized the order of explanation for Schroeder’s paradox.

\* Rel. (35) : could the factor 2 for variable s be explained ?

Reply: Relation (35) is an empirical equation derived from the seminal work of Dullien (F.A. Dullien. Porous media: fluid transport and pore structure. 2nd ed. Academic Press; 1992.). The presence of the factor 2 is a consequence of the specific formulation proposed by Dullien and the underlying assumptions in his theoretical framework.

\* General comment for a recurrent point : in many places in the paper, the expression of a variable is introduced in an sentence, the expression is given, and followed by « where X is the variable … ». The lengthy, repetive structure could be easily replaced by introducing the expression of variable X (here give its name !) before this expression. Besides, the recurrent expression « is represented as follows » could be (i) improved, and sometimes be rewritten with alternative words.

回答呢？  
\* Does rel. (55) apply for any polysulfonated membrane, in particular for the membrane used in this work ?

Reply: This equation is an empirical equation referenced from Jiao’s work[41]. The parameter of this equation is applied to all PEMs.  
\* Table 1 : could it be specified that the temperature was at 65°C (338.15 K) ?

Reply: We have added extra constraint to table header, to specify the temperature at 65 Celsius.   
\* Rel. (62) : what does wk(i) represent ?

Reply: The wk^(i) is used to represent the state of particle in step K, the state wk^(i) is determined by the previous state wk^(i-1)

In relation (62), the notation w\_k^(i) denotes the weight of the i-th particle at the i-k-th timestep in the particle filter algorithm. Particle filters are a class of sequential Monte Carlo methods used primarily for estimating the state of a system where the model and measurement are non-linear and/or the noise is non-Gaussian. These filters operate by representing the posterior distribution of the state variables through a set of random samples with associated weights and are particularly useful in scenarios where other filtering methods like the Kalman filter are less effective due to model constraints.  
\* Page (19) « measurement noise and process noise ». How are they defined ? How are they generated ?

Reply: The definition and generation of both measurement and process noise were both discussed in Section 5.2

We have referenced the work of Bao et al. (*C. Bao, M. Ouyang, B. Yi. Modeling and control of air stream and hydrogen flow with recirculation in a PEM fuel cell system—I. Control-oriented modeling. Int J Hydrogen Energy 2006; 31: 1879-96.* and *C. Bao, W.G. Bessler. Two-dimensional modeling of a polymer electrolyte membrane fuel cell with long flow channel. Part I. Model development. J Power Sources 2015; 275: 922-34.* and *C. Bao, W.G. Bessler. Two-dimensional modeling of a polymer electrolyte membrane fuel cell with long flow channel. Part II. Physics-based electrochemical impedance analysis. J Power Sources 2015; 278: 675-82*). Bao’s research provides a comprehensive analysis of noise characteristics in similar experimental setups and offers empirical data.

\* Section 4. Tests consisted in a sudden change in air flow rate (or more precisely in rotation speed of something) and at measuring the cell current and the high frequency impedance. OK, but was it done at a fixed, specified voltage ?

The voltage was not at a fixed specified level, allow me to provide elucidation on this salient aspect of our methodology.

During the experiment we increase the revolutions per minute of the Air Compressor, which would cause the overall increase of the system's power output, and the average voltage exhibited a upward inflection.

After the change of air compressor speed and the voltage is in a stable phase, we initiated an augmentation of the current load. Based on fuel cell’s output characteristics a drop in the average voltage is observerd.

\* Table 4 : What does « CMP speed » mean ?

Reply: Thank you for pointing out the potential ambiguity surrounding the acronym "CMP". The term "CMP speed" is a shorthand notation for the rotational velocity of the Air Compressor. In recognition of the potential for confusion, we have taken the prudent step of replacing all instances of the abbreviation "CMP" with the explicit term "Air Compressor" throughout the manuscript.  
\* The presentation of section 5 is not straightforward for a non-specialist of observers, with a couple of not fully clear concepts e.g. « observer fusion », however, it sounds really interesting since based on a solid methodology (just a comment).

Reply: The Observer-Fusion is a simplification for “observer based on sensor fusion”, it’s compared with “observer based on HFR”.

To expound on the concept of "Observer Fusion," which is a contraction of the term "observer based on sensor fusion." This methodology represents an amalgamation of multiple sensory inputs, leveraging the strengths of disparate data streams to increase the robustness and precision of the observer's estimations.

To conclude, the paper could be published after minor revision, most of them for the sake of an easier reading by non-specialists of the topic.