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Development of an Oxygen Pressure Estimator Using the Immersion and Invariance Method for a Particular **PEMFC System**

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Abstract: The fault detection method has been used usually to give a diagnosis of the performance and efficiency in the proton exchange membrane fuel cell (PEMFC) systems. To be able to use this method a lot of sensors are implemented in the PEMFC to measure different parameters like pressure, temperature, voltage, and electrical current. However, despite the high reliability of the sensors, they can fail or give erroneous measurements. To address this problem, an efficient solution to replace the sensors must be found. For this reason, in this work, the immersion and invariance method is proposed to develop an oxygen pressure estimator based on the voltage, electrical current density, and temperature measurements. The estimator stability region is calculated by applying Lyapunov's Theorem and constraints to achieve stability are established for the oxygen pressure, electrical current density, and temperature. Under these estimator requirements, oxygen pressure measurements of high reliability are obtained to fault diagnosis without the need to use an oxygen sensor.

Keywords: estimator development; Lyapunov's Theorem application; non-linear system; PEMFC system; sensor replacement

1. Introduction

Fuel cell (FC) system is an advanced power system necessary for a clean, sustainable, and environmentally friendly future, because FCs are promising candidates as an alternative to conventional fossil fuels, due to their higher energy density, energy efficiency, and very low emissions [1–3]. The main operation of the FCs is to transform gaseous fuel chemical energy into electricity. Besides, the FCs can be used as alternative stationary and mobile power source [4,5]. The main types of FCs are proton exchange membrane, direct methanol, solid oxide, molten carbonate, phosphoric acid, alkaline, and microbial [6].

In particular, the proton exchange membrane fuel cell (PEMFC) has attracted the attention of researchers in the last few decades due to its characteristics as low operating temperature, low noise, quick start-up capability, light mass, and high-power density [2,4,6,7]. The PEMFCs have recently passed the test phase and have slightly reached the commercialization stage due to the impressive research effort [8]. However, the two biggest limitations preventing the PEMFC system from further commercialization are its reliability and durability [7].

A lot of studies on PEMFC performance have been carried out, since three-dimensional simulation models to more detailed measurement techniques, such as electrochemical impedance spectroscopy [9,10]. To have a PEMFC diagnosis, the fault detection method has been used commonly



to guarantee correct and safe operation in the PEMFC system [7,11,12]. However, to achieve such a diagnosis, several sensors have been used to measure different parameters like the mass flow, oxygen pressure, hydrogen pressure, compressor velocity, electrical current, water pressure, voltage, and temperature of the stack [11,13,14].

A lot of researchers have worked on the development of sensors with high reliability [15–18]. These devices must present characteristics, such as high sensitivity and selectivity, robustness, fast response time, operation at high temperature and low power consumption [19–21]. However, in real applications, the reliability of sensors during the system operation is variable. Thus, inaccurate sensor measurements can provide misleading results in PEMFC fault diagnosis, which can end in failures and damages of the PEMFC system [7,11]. To solve this problem, novel methods have been proposed to reduce errors in PEMFC fault diagnosis [9,10]. For this reason, an efficient method to replace the oxygen sensor is proposed in this work, since the oxygen management system is an important subsystem, which is used for supplying proper oxygen pressure in the PEMFC stack cathode. Besides, the complexity and nonlinearity of the oxygen pressure are difficult to model [22]. So, using the voltage, electrical current density, and temperature measurements and applying the immersion and invariance (gradient estimator) method it is possible to develop an oxygen pressure estimator for getting high-reliability oxygen measurements avoiding the use of oxygen sensor for PEMFC system fault diagnosis.

The paper is organized as follows, the formulation of a gradient estimator to develop the oxygen pressure estimator is described in Section 2. The PEMFC potential-current behavior is discussed in Section 3. The oxygen pressure estimator applied to a PEMFC system is presented in Section 4. The simulation and results are introduced in Section 5. Finally, some concluding remarks are presented in Section 6.

2. Formulation of Gradient Estimator

The immersion and invariance (gradient estimator) method has been proposed to solve problems of stabilization and adaptive control of nonlinear systems, which are present in any real practical problem [23–26]. The key step for the estimator development using this method is the construction of a monotone mapping, which explicitly depends on some of the estimator tuning parameters [27,28]. For these reasons, in this work, this method has been used to develop the oxygen pressure estimator.

The estimator design is formulated by proposing a function where the system behavior representation distinguishes between measurable and not measurable signals. As shown in [28,29], there is a general kind of function dependent on two variables θ and ξ expressed by

$$F(\theta,\xi) = G(\theta) + H(\xi) + K(\theta,\xi)$$
(1)

with $\theta > 0$ and $\xi > 0$, where ξ and θ are known and time-dependent variables, such that measurable signals $F(\theta, \xi)$ and $H(\xi)$ are represented by

$$y(t) = F(\theta, \xi) - H(\xi).$$
⁽²⁾

Indeed, the representation in the non-linear regression form will be

$$y(t) = \phi(\theta, \xi), \tag{3}$$

where

$$\phi(\theta,\xi) := G(\theta) + K(\theta,\xi). \tag{4}$$

Given this formulation, the following proposition can be stated.

Proposition 1. Consider the function $\phi(\theta, \xi)$, where $F(\theta, \xi)$ and $H(\xi)$ are known and the variable corresponding to the non-linear regression model satisfies that the partial derivative of $\phi(\theta, \xi)$ with respect to θ is greater than zero. Then, the gradient estimator is given by

$$\hat{\theta} = \gamma(y(t) - \phi(\hat{\theta}, \xi)) \tag{5}$$

with $\gamma > 0$ ensuring that

$$\lim_{t \to \infty} \hat{\theta} = \theta, \tag{6}$$

for all initial condition $\hat{\theta}_0$ such as $\frac{\partial \phi(\hat{\theta}_0, \xi)}{\partial \theta} > 0.$

Proof. To show that the immersion and invariance estimator converges to the desired value, it is necessary to use the monotonicity property of the function $\phi(\theta, \xi)$ concerning θ . Then, as:

$$\frac{\partial \phi(\theta,\xi)}{\partial \theta} > 0, \tag{7}$$

the function is strictly monotonically increasing and also fulfills

$$(\hat{\theta} - \theta) \left[\phi(\hat{\theta}, \xi) - \phi(\theta, \xi) \right] > 0 \quad \forall \hat{\theta} \neq \theta,$$
(8)

taking the Lyapunov's function candidate

$$V(\hat{\theta}) = \frac{1}{2\gamma} (\hat{\theta} - \theta)^2, \tag{9}$$

its time-derivative along the trajectories of (2)-(5) is given by

$$\dot{V} = -(\hat{\theta} - \theta)[\phi(\hat{\theta}, \xi) - \phi(\theta, \xi)] < 0 \quad \forall \hat{\theta} \neq \theta.$$
(10)

Note that the negative definiteness of \dot{V} immediately follows from (8). Then, the proof is completed by using Lyapunov's Second Stability Theorem. \Box

3. PEMFC Potential-Current Behavior

An accurate mathematical model to represent the PEMFC potential V_c has been reported in [30], where V_c is a depending function of stack current, cathode pressure, reactant partial pressures, PEMFC temperature, and membrane humidity using a combination of physical and empirical relationships, and can be expressed in terms of the Nernst's potential E_{th} and the three main types of potential drops; activation V_{act} , ohmic V_{ohm} , and concentration V_{con} .

$$V_c(\theta,\xi) = E_{th}(\theta) - V_{ohm}(\xi) - V_{act}(\theta,\xi) - V_{con}(\theta,\xi),$$
(11)

where θ denotes the oxygen pressure (*atm*), and ξ the electrical current density in the cell ($A \cdot \text{cm}^{-2}$). Nernst's potential E_{th} . The Nernst's potential or open-circuit potential is the maximum power obtained by one cell corresponding to exchange Gibbs free energy as a result of the difference between reactant products and Gibbs's free energy. It can be described by the following equation [30–32].

$$E_{th}(\theta) = E_0 + B_1(T_0 - T) + B_2 T \ln\left[\frac{P_{H_2}\theta^{1/2}}{P_{H_2O}}\right],$$
(12)

where T_0 and T are the initial temperature and the cell temperature, respectively (*K*), P_{H_2} is a positive constant that represents the hydrogen pressure (*atm*), and E_0 is the reference potential (*V*). B_1 and B_2

are positive constants that depend on stack temperature and potential (V/K) [30]. Water pressure is represented by P_{H_2O} (*atm*).

Ohmic potential drop V_{ohm} . The ohmic potential drop arises from the resistance of the polymer membrane to the transfer of protons and from the resistance of the electrode and the collector plate to the transfer of electrons [30–32].

$$V_{ohm}(\xi) = \frac{\xi}{A_{fc}} R_{ohm},\tag{13}$$

where $R_{ohm} > 0$ is the internal electrical resistance (Ω) and A_{fc} is the cell active area. Besides, the ohmic resistance can be expressed as a function of the membrane conductivity (cm⁻¹ · Ω^{-1}), σ_m .

$$R_{ohm} = \frac{t_m}{\sigma_m},\tag{14}$$

where t_m is the thickness of the membrane (*cm*), and σ_m is a function of membrane water content λ_m and the cell temperature *T*.

$$\sigma_m = b_1 \exp\left[b_2\left(\frac{1}{303} - \frac{1}{T}\right)\right],\tag{15}$$

where b_1 is a function of membrane water content and b_2 is a constant [30].

$$b_1 = b_{11}\lambda_m - b_{12},\tag{16}$$

where b_2 , b_{11} , and b_{12} are usually determined empirically. In this work, the values for b_2 , b_{11} , and b_{12} are taken from [33].

Activation potential drop V_{act} . The activation potential drop comes when the movement of electrons needs to break and form chemical bonds in the anode and cathode (i.e., part of the available energy is lost in driving the chemical reaction that transfers the electrons to and from the electrodes). Although the activation overvoltage occurs at both PEMFC electrodes, the reaction of hydrogen oxidation at the anode is faster than the reaction of oxygen [30–32].

$$V_{act}(\theta) = V_o + V_a(\theta)(1 - \exp[-c_1\xi]), \tag{17}$$

where c_1 is a constant. The functions V_o and V_a are both dependent on oxygen pressure and temperature. They have been calculated empirically by

$$V_o = V_o^0 + B_1(T_0 - T) - 1.07551B_2T + \frac{3B_2T}{2}\ln\left(\frac{P_{ca} - P_{sat}}{P_{atm}}\right),$$
(18)

where V_o^0 is the initial potential drop (*V*) at zero current density. P_{ca} and P_{atm} are the pressures of the cathode and atmospheric, respectively (*atm*). The water saturation pressure P_{sat} (*mPa*) is expressed as

$$\log_{10} P_{sat} = \frac{-1.69}{10^{10}} T^4 + \frac{3.85}{10^7} T^3 - \frac{3.39}{10^4} T^2 + 0.143T - 20.92.$$
(19)

The function V_a is given as:

$$V_a(\theta) = B_3 \left(\frac{\theta}{0.1173} + P_{sat}\right)^2 + B_4 \left(\frac{\theta}{0.1173} + P_{sat}\right) + B_5,$$
 (20)

where the constants B_3 , B_4 , and B_5 are dependent on the stack temperature and the voltage (V/K) and usually are determined empirically [30].

Concentration of potential drop V_{con} . The concentration of potential drop corresponds to the concentration gradients formed due to mass diffusions from the flow channels to the reaction sites (catalyst area). The factors underlying this potential drop are high current densities, slow transportation of reactants and products, and water film covering the catalyst surfaces to the anode and cathode [30–32].

$$V_{con}(\theta,\xi) = \xi \left(\frac{c_2\xi}{I_{max}}\right)^{c_3},\tag{21}$$

where $c_3 \in \mathbb{R}_+$ is a constant, I_{max} is the maximum electrical current density in the cell and c_2 is an oxygen pressure function [30].

$$c_{2} = \begin{cases} B_{6} \left(\frac{\theta}{0.1173} + P_{sat} \right) + B_{7} & if \ h(\theta) < 0, \\ B_{8} \left(\frac{\theta}{0.1173} + P_{sat} \right) + B_{9} & if \ h(\theta) \ge 0, \end{cases}$$

$$(22)$$

where

$$h(\theta) = \theta + 0.1173 P_{sat} - 0.2346 atm,$$

and B_6 , B_7 , B_8 , and B_9 are constant values that depend on the stack temperature and are usually determined empirically.

Lemma 1. *The discontinuous function* c_2 *defined in* (22) *can be approximated by the continuous function* C_2 *given below.*

$$C_2 = D_1 \left(\frac{\theta}{0.1173} + P_{sat} \right) + D_2,$$
 (23)

where

$$D_1 = \frac{1}{2}B_6 + \frac{1}{2}B_8 + \frac{1}{2}(B_6 - B_8)\tanh[h(\theta)]$$

and

$$D_2 = \frac{1}{2}B_7 + \frac{1}{2}B_9 + \frac{1}{2}(B_7 - B_9)\tanh[h(\theta)]$$

The parameters values of the PEMFC voltage model are taken from [30] (see Table 1).

Table 1. Parameters for the PEMFC voltage model [30].

Symbol	Parameter	Value	
A_{fc}	Cell Active Area	100 cm ²	
$\vec{E_0}$	Reference Potential	$1229 \times 10^{-3} \text{ V}$	
Imax	Maximum Current Density	$2.2 \text{A} \cdot \text{cm}^{-2}$	
P_{atm}	Atmospheric Pressure	1 atm	
P_{ca}	Cathode Pressure	2 atm	
P_{H_2}	Hydrogen Pressure	1 atm	
P_{H_2O}	Water Pressure	$396 imes 10^{-3}$ atm	
t_m	Membrane Thickness	$125 imes10^{-4}~{ m cm}$	
T_0	Initial Temperature	298.15 K	
V_o^0	Initial Potential Drop	$279 imes 10^{-3} \mathrm{V}$	
b_1	Membrane Humidity Function	$686 imes 10^{-4} \ { m cm}^{-1} \cdot \Omega^{-1}$	
b_2	Constant	350 K	
b_{11}	Constant	$513 \times 10^{-5} \text{SO}_3^- \cdot \text{H}_2 O^{-1} \cdot \text{cm}^{-1} \cdot \Omega^{-1}$	

Symbol

 b_{12}

 c_1

Сз

 B_1

 B_2

 B_3

 B_4

 B_5

 B_6

 B_7

 B_8

B9

 λ_m

ont.		
	Value	
	$326 imes10^{-5}\mathrm{cm}^{-1}\cdot\Omega^{-1}$	

 $10 \ A^{-1}$

2 (-)

 $85\times 10^{-5}~V{\cdot}K^{-1}$

 $43085 \times 10^{-6} \text{ V} \cdot \text{K}^{-1}$

 $-1618 \times 10^{-8} \mbox{ TV} \cdot \mbox{ K}^{-1} \mbox{+} 1618 \times 10^{-5} \mbox{ V}$

 $18\times 10^{-4} T \: V{\cdot}K^{-1} - 166\times 10^{-3} \: V$

 $-58 \times 10^{-5} \mathrm{T~V} \cdot \mathrm{K}^{-1} + 5736 \times 10^{-4} \mathrm{V}$

 $358\times 10^{-5}~T~K^{-1}-622\times 10^{-3}$

 $-725 \times 10^{-6} \text{ T K}^{-1} + 1.68$

 $433\times 10^{-6}~T~K^{-1}-68\times 10^{-3}$

 $-8\times 10^{-4}~{\rm T}~{\rm K}^{-1}+54\times 10^{-2}$

 $14 \ H_2 O \cdot (SO_3^-)^{-1}$

Table 1. Cont.

4. Application of Oxygen Pressure Estimator to a PEMFC System

Parameter

Constant

Constant

Constant

Temperature and Potential Function

Temperature and Potential Function

Temperature and Potential Function

Temperature and Potential Function

Temperature and Potential Function

Temperature Function

Temperature Function

Temperature Function

Temperature Function

Membrane Humidity

The oxygen pressure estimator presented in this section is derived from the results presented in Sections 2 and 3. The measurable signal is defined by applying the Equations (2)-(11).

$$y(t) = V_c(\theta, \xi) + V_{ohm}(\xi), \tag{24}$$

where

$$\phi(\theta,\xi) := E_{th}(\theta) - V_{act}(\theta,\xi) - V_{con}(\theta,\xi) .$$
⁽²⁵⁾

Now a proposition related to the PEMFC system is presented.

Proposition 2. Consider the function $\phi(\theta, \xi)$, with C_2 and θ are greater than zero, such that, inequality (26) is satisfied

$$B_2 T(P_{ca} - P_{sat} - 3\theta) - 2\left(\frac{\partial V_a}{\partial \theta}\right)\theta(P_{ca} - P_{sat}) \ge 0.$$
⁽²⁶⁾

Then, ξ *can be expressed in terms of* θ *and T as follows:*

$$\xi = \left(D \left[\frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} \right] \right)^{\frac{1}{c_3 + 1}},$$
(27)

where

$$D = \frac{I_{max}^{c_3}}{c_3 C_2^{c_3 - 1}} \left(\frac{\partial C_2}{\partial \theta}\right)^{-1}$$

and $\frac{\partial \phi(\theta,\xi)}{\partial \theta} > 0.$

Proof. The proof starts with the partial derivative of ϕ with respect to θ , which is given by

$$\frac{\partial \phi(\theta,\xi)}{\partial \theta} = \frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} \left(1 - \exp[-c_1 \xi] \right) - \frac{\partial C_2}{\partial \theta} \left(\frac{c_3 C_2^{c_3 - 1} \xi^{c_3 + 1}}{I_{max}^{c_3}} \right).$$

Now, taking the set of values (θ, ξ) , that satisfy inequality (28),

$$0 \leq \frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} \left(1 - \exp[-c_1 \xi] \right) - \frac{\partial C_2}{\partial \theta} \left(\frac{c_3 C_2^{c_3 - 1} \xi^{c_3 + 1}}{I_{max}^{c_3}} \right).$$
(28)

Since $0 < 1 - \exp[-c_1\xi] < 1$ for ξ and $c_1 > 0$, then

$$0 \leq \frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} - \frac{\partial C_2}{\partial \theta} \left(\frac{c_3 C_2^{c_3 - 1} \xi^{c_3 + 1}}{I_{max}^{c_3}} \right) < \frac{\partial \phi(\theta, \xi)}{\partial \theta}.$$

So, the admissible limit values (θ, ξ) that satisfy inequality (28) can be found when this is equal to zero.

$$0 = \frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} - \frac{\partial C_2}{\partial \theta} \left(\frac{c_3 C_2^{c_3 - 1} \xi^{c_3 + 1}}{I_{max}^{c_3}} \right),$$

setting

$$D = \frac{I_{max}^{c_3}}{c_3 C_2^{c_3 - 1}} \left(\frac{\partial C_2}{\partial \theta}\right)^{-1},$$

thus,

$$\xi = \left(D \left[\frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} \right] \right)^{\frac{1}{c_3 + 1}}$$

As $\frac{\partial C_2}{\partial \theta} > 0$, then,

$$\frac{B_2T}{2}\left(\frac{P_{ca}-P_{sat}-3\theta}{\theta(P_{ca}-P_{sat})}\right)-\frac{\partial V_a}{\partial \theta}\geq 0,$$

since $\theta > 0$ and $P_{ca} - P_{sat} > 0$, then,

$$B_2T(P_{ca}-P_{sat}-3\theta)-2\left(\frac{\partial V_a}{\partial \theta}\right)\theta(P_{ca}-P_{sat})\geq 0.$$

Now the following proposition is introduced as a result of the combination of Proposition 1 and Proposition 2. This result shows the estimator and its stability using Lyapunov's functions.

Proposition 3. Consider the function $\phi(\theta, \xi)$, C_2 and θ are greater than zero, such that, inequality (26) is satisfied and with ξ expressed as:

$$\xi = \left(D \left[\frac{B_2 T}{2} \left(\frac{P_{ca} - P_{sat} - 3\theta}{\theta(P_{ca} - P_{sat})} \right) - \frac{\partial V_a}{\partial \theta} \right] \right)^{\frac{1}{c_3 + 1}}.$$

Then, the gradient estimator of oxygen pressure is given by

$$\hat{\theta} = \gamma(y(t) - \phi(\hat{\theta}, \xi)), \tag{29}$$

with $\gamma > 0$, ensuring that

$$\lim_{t \to \infty} \hat{\theta} = \theta. \tag{30}$$

Proof. For the values of θ and ξ stated in the hypothesis of Proposition 2, it is obtained that the partial derivative of $\phi(\theta, \xi)$ with respect to θ is greater than zero. Then, by Proposition 1, the gradient estimator of oxygen pressure is given by

$$\hat{\theta} = \gamma(y(t) - \phi(\hat{\theta}, \xi)),$$

 $\lim_{t\to\infty}\hat{\theta}=\theta.$

with $\gamma > 0$, ensuring that

5. Simulations and Results

The Runge–Kutta fourth-order algorithm, described in [34], and the values of the parameters given in the Table 1 were used to perform the simulations. The first step was to determine the stability region for the estimator under the established constrains of the Propositions 2 and 3. The estimator stability region is given within the interval (0 atm, 0.45 atm) and the simulation results of such constraints are shown in Figures 1 and 2. The behavior of the partial derivative of ϕ with respect to θ as a function of θ and ξ for different temperatures is shown in Figure 1.



Figure 1. Behavior of the partial derivative of ϕ concerning θ .

The behavior of θ and ξ considering the established constraints for different temperatures is shown in Figure 2.

Within stability region, the oxygen pressure estimator and the PEMFC potential-current simulations were performed using oxygen pressure equal to 0.3 *atm* and different values for θ_0 and γ . The oxygen pressure estimator shows an asymptotic convergence to the proposed value for oxygen pressure. The estimator behavior can be appreciated for different values θ_0 in Figure 3, and different values of γ in Figure 4.

The electrical current density calculated based on the estimator proved an asymptotic convergence to the electrical current density calculated for oxygen pressure equal to 0.3 *atm*, the simulation is shown for different values of θ_0 in Figure 5, and for different values of γ in Figure 6.

The cell potential calculated based on the estimator evidenced an asymptotic convergence to the potential calculated for oxygen pressure equal to 0.3 *atm*, the simulation results for different values of θ_0 are shown in Figure 7 and for different values of γ in Figure 8.



Figure 2. Behavior of the partial derivative of ϕ concerning θ .



Figure 3. Estimator behavior with different values θ_0 .



Figure 4. Estimator behavior with different values γ .



Figure 5. Simulation of electrical current density stability with different values θ_0 .



Figure 6. Simulation of electrical current density stability with different values γ .



Figure 7. Simulation of potential stability with different values θ_0 .



Figure 8. Simulation of potential stability with different values γ .

Finally, the power or potential-current performance curve based on the estimator demonstrated an asymptotic convergence to the power for oxygen pressure equal to 0.3 atm, the simulation is shown for different values of θ_0 in Figure 9, and for different values of γ in Figure 10. This curve has proved to be of vital importance for the PEMFC system fault diagnosis [35].



Figure 9. Simulation of power stability with different values θ_0 .



Figure 10. Simulation of power stability with different values γ .

6. Conclusions

To avoid oxygen sensors for PEMFCs, an oxygen pressure estimator has been developed based on the immersion and invariance (gradient estimator) method, and its stability conditions are established using Lyapunov's Theorem. Additionally, in this work, the PEMFC electrical current density has been characterized in terms of oxygen pressure and temperature under certain constraints.

The oxygen pressure estimator presents an absolute convergence within the stability region to the measurable value of oxygen pressure. However, the corresponding working condition can be different because it is directly related to laboratory environmental conditions. So, the next step is to evaluate the performance of the proposed estimator under different PEMFC conditions to improve the oxygen pressure estimator.

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