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Multiple Synergistic Degradation Parameter Identification of PEM Fuel Cells Utilizing Threat Response Adaptive Differential Evolution Algorithm

Weiqing Ni, Zhenjie Liu, Jisen Li, Liyan Zhang, Qihong Chen and Dongqi Zhao * 

School of Automation, Wuhan University of Technology, Wuhan 430070, China; 363528@whut.edu.cn (W.N.); 312994@whut.edu.cn (Z.L.); 265414@whut.edu.cn (J.L.); zlywhut@whut.edu.cn (L.Z.); chenqh@whut.edu.cn (Q.C.)
* Correspondence: dongqi_zhao@whut.edu.cn

Abstract

Proton exchange membrane fuel cells (PEMFCs) experience significant performance degradation over long-term operation, hindering their commercial viability. Accurately identifying polarization curve parameters during aging is crucial for elucidating degradation mechanisms and enabling health monitoring, yet this task faces challenges such as parametric coupling and pronounced nonlinearity. This study tackles these identification challenges through the integrated application of a dynamic aging model, which captures the synergy between degradation mechanisms like platinum oxidation and membrane resistance increase, and the introduction of the novel Threat Response Adaptive Differential Evolution (TRADE) algorithm. The algorithm employs multidimensional threat assessment, a three-tier response strategy, and adaptive decision-making to achieve accurate and robust parameter identification. Validated with experimental data from commercial PEMFC stacks over a full ageing cycle, the TRADE algorithm achieves a root mean square error as low as 0.00675 V within the 100–1000 h range, demonstrating superior fitting performance and stability. Sensitivity analysis further reveals that activation overpotential is the dominant degradation mechanism throughout the entire cycle (contributing $\geq 70\%$), whereas the contribution of concentration overpotential rises substantially to 33% under high-current-density conditions. This study provides a robust modelling framework and an effective methodology for quantifying PEMFC ageing mechanisms, predicting remaining useful life, and optimizing system performance.

Keywords: proton exchange membrane fuel cell; ageing modeling; parameter identification; multi-mechanism degradation; sensitivity analysis



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1. Introduction

Proton exchange membrane fuel cells (PEMFCs) are promising energy conversion devices owing to their high efficiency and environmental benefits [1,2]. However, performance degradation over extended operation impedes their widespread commercialization. This degradation manifests as a decline in output voltage and alterations in polarization characteristics [3]. Recent studies highlight the complexity of degradation mechanisms, which involve catalyst oxidation, increased membrane resistance, and mass transfer limitations. These mechanisms collectively influence the activation, ohmic, and concentration overpotentials [4,5]. Consequently, the accurate modeling and parameter identification of these aging processes are essential for degradation diagnosis, lifetime prediction, and system optimization.

The polarization curve (V-I curve) is the most intuitive and comprehensive metric for characterizing PEMFC performance. Its distinct regions—activation, ohmic, and concentration—are governed by specific physical processes. Each region is associated with a set of parameters that evolve with cell aging [6]. The activation region at low current densities is governed by electrochemical kinetics, characterized by the exchange current densities ($i_{0,a}$, $i_{0,c}$) and the charge transfer coefficients (α_a , α_c). Aging mechanisms such as platinum oxidation and active site loss directly increase the activation overpotential by reducing the exchange current density (i_0). The linear ohmic region reflects the combined protonic and electronic resistances, predominantly the membrane resistance (R). This resistance increases owing to membrane thinning or chemical degradation. The sharp voltage drop at high current densities is governed by mass transport limitations, captured by the parameters m and n . These parameters are linked to diffusion layer properties and degrade with pore flooding or catalyst layer compaction [7]. Temperature critically influences the cell's electrochemical behavior. It exponentially governs the exchange current density via the Arrhenius relationship and linearly affects both the Nernst potential and ohmic losses. Furthermore, temperature alters gas diffusivity and water vapor pressure, which in turn governs the concentration overpotential.

Therefore, precise characterization of the aging polarization curve necessitates the identification and quantification of the key parameters governing these overpotential losses. This work focuses on mechanisms quantitatively linked to the model parameters: active site loss (governed by Pt oxidation, $V_{PtO}(t)$, and active area decay, $A(t)$), increased proton transport resistance (modeled by time-varying membrane resistance, $R(t)$), and mass transfer channel deterioration (represented by evolving parameters $m(t)$ and $n(t)$). Accurate identification of these parameters is therefore foundational for elucidating the coupled degradation processes [8].

To this end, researchers have developed various electrochemical models to describe PEMFC operational characteristics. These models typically incorporate a set of physical and empirical parameters, such as exchange current density, charge transfer coefficient, proton conductivity, and water management coefficients [9]. However, the values of many such parameters are difficult to obtain through direct experimental measurement. Furthermore, due to the complex multi-physics coupling effects within PEMFCs, the identification of these parameters often constitutes a nonlinear, multi-variable optimization problem with multimodal characteristics [10,11]. Traditional methods, including the least squares approach, are prone to becoming trapped in local optima and exhibit high sensitivity to the initial conditions.

In recent years, accurate parameter estimation for Proton Exchange Membrane Fuel Cells (PEMFCs) has garnered significant attention in modeling and optimization research. Complementing traditional mathematical programming and measurement methods (e.g., Sequential Quadratic Programming [12], the Current Switching Technique [13], and Electrochemical Impedance Spectroscopy [14]), meta-heuristic (MH) algorithms have become a prominent approach. Their prominence stems from their robustness and superior global search capabilities in addressing nonlinear, high-dimensional parameter optimization problems. Consequently, numerous studies have focused on developing and applying advanced MH algorithms to improve estimation accuracy and computational efficiency. For example, applied to stacks such as the Ballard Mark V, proposed algorithms include the Enhanced Hunger Games and Marine Predator Optimization algorithm (HGS-MPA) [15], the Dynamic Fick's Law Algorithm (DFLA) [16], and the Archimedes Optimization Algorithm (AOA) [17]. Similarly, for the BCS 500W stack, effective algorithms include the Chaotic Teaching Learning-Based Optimization algorithm (CTLBO) [18], the Hybrid Chaotic Parti-

cle Swarm Optimization and Slime Mold Algorithm (HCPSOSMA) [19], and the Enhanced Salp Swarm Algorithm (ESSA) [20].

While these advanced MH algorithms have demonstrated effectiveness in static or semi-static parameter estimation, accurately identifying parameters under continuous and synergistic ageing degradation—a dynamic, nonlinear, and strongly coupled process—remains a formidable challenge. Most existing studies focus on optimizing performance models at specific health states, with limited emphasis on tracking the temporal evolution of degradation parameters and delineating their intertwined effects throughout the fuel cell's lifetime.

To address these challenges, this study focuses on parameter identification for the evolving ageing degradation of PEMFCs, with three key contributions. First, a dynamic mathematical model incorporating multidimensional degradation terms is constructed by integrating key ageing mechanisms: platinum catalyst oxidation (PtO_2), active area decay, membrane resistance increase, and mass transfer channel blockage. This model enables precise characterization of the synergistic degradation among the three overpotential categories. Second, inspired by the “perception–decision–response” intelligent paradigm, the Threat Response Adaptive Differential Evolution (TRADE) algorithm is proposed. The algorithm quantifies population status via multidimensional threat perception, adapts three-tier response strategies to distinct optimization phases, and dynamically adjusts search behavior through an adaptive decision-making mechanism. This approach enhances the accuracy and robustness of complex parameter identification. Finally, the model and algorithm are validated using full-cycle ageing experimental data from a 3 kW commercial PEMFC stack. A sensitivity analysis is subsequently conducted to identify the dominant degradation sources.

2. PEMFC Aging Modeling

The performance degradation of PEMFCs due to ageing ultimately manifests as a synergistic increase in the activation (η_{act}), ohmic (η_{ohm}), and concentration (η_{conc}) overpotentials. This section establishes the linkage between the introduced ageing mechanisms and the three overpotential categories, thereby constructing a physically interpretable model. The model's rationale is grounded in the integration of fundamental physicochemical ageing mechanisms.

The aging-induced performance degradation of proton exchange membrane fuel cells (PEMFCs) is a complex, multifaceted phenomenon that arises from irreversible physicochemical changes in their core components. Accurate modeling of this degradation process is therefore not merely an exercise in curve fitting but a prerequisite for elucidating underlying mechanisms, enabling prognostics, and informing mitigation strategies [6,9]. A core challenge lies in translating micro-scale material changes—such as catalyst oxidation and membrane thinning—into macro-scale performance losses. These losses manifest synergistically as increases in the activation (η_{act}), ohmic (η_{ohm}), and concentration (η_{conc}) overpotentials [3]. This section presents a semi-empirical, dynamic aging model developed by integrating established degradation mechanisms into the fundamental cell voltage equation. For each mechanism, we first discuss its physicochemical origin based on experimental evidence, then justify its mathematical formulation, and finally link it to a corresponding overpotential category. This approach ensures the model's physical interpretability and the identifiability of its parameters from standard polarization curve data.

The steady-state polarization (V-I) curve provides the most comprehensive performance signature of a PEMFC. Under steady-state, isothermal conditions, the single-cell

output voltage is commonly described as the Nernst potential (E_{Nernst}) plus the sum of the three major overpotentials [21]:

$$V = E_{\text{Nernst}} + \eta_{\text{act,a}} + \eta_{\text{act,c}} + \eta_{\text{ohm}} + \eta_{\text{conc}} \quad (1)$$

widely adopted explicit form that decomposes these losses is given by [22]:

$$V = E_{\text{rev}} - V_{\text{PtO}} - \frac{RT}{2\alpha_a F} \ln\left(\frac{i + i_{\text{loss}}}{i_{0,a}}\right) - \frac{RT}{4\alpha_c F} \ln\left(\frac{i + i'_{\text{loss}}}{i_{0,c}}\right) - iR - m(e^{\text{ni}} - 1) \quad (2)$$

In Equation (2), the second and third terms correspond to the anode and cathode activation overpotentials ($\eta_{\text{act,a}}$ and $\eta_{\text{act,c}}$), respectively; the fourth term represents the ohmic overpotential (η_{ohm}); and the fifth term captures the concentration overpotential (η_{conc}). The central premise of the proposed aging model is that the key parameters in these terms—the exchange current densities ($i_{0,a}$, $i_{0,c}$), the limiting current density (i_{loss}), the membrane resistance (R), and the mass transfer coefficients (m , n)—are not constants. Instead, they evolve over time due to specific degradation mechanisms. These time-evolving parameters and their corresponding aging mechanisms are summarized in Table 1.

Table 1. Key Aging Parameters Extractable from the Polarization Curve.

Parameter	Physical Meaning	Governing Loss	Primary Aging Mechanisms
$i_{0,a}, i_{0,c}$	Exchange current density (anode/cathode); intrinsic kinetics.	Activation overpotential (η_{act})	Catalyst active site loss (Pt dissolution, oxidation, agglomeration).
α_a, α_c	Charge transfer coefficient (anode/cathode).	Activation overpotential (η_{act})	Change in catalyst surface state.
R	Total ohmic resistance (dominantly membrane).	Ohmic overpotential (η_{ohm})	Membrane chemical/mechanical degradation.
m, n	Mass-transfer limiting parameters.	Concentration overpotential (η_{conc})	Pore blockage (flooding), catalyst layer collapse.
E_{rev}	Reversible (open-circuit) voltage.	Nernst voltage	Fuel crossover, catalyst poisoning.
i_{loss}	Parasitic/crossover current loss.	Activation loss (implicit)	Membrane thinning/degradation, short circuits.

The open-circuit voltage is governed by the Nernst equation, which defines the theoretical maximum electromotive force of the cell. It is influenced by operational factors, primarily temperature and reactant partial pressures [23]:

$$E_{\text{Nernst}} = 1.229 - 0.85 \times 10^{-3}(T - 298.15) + 4.3085 \times 10^{-5}T \left[\ln(P_{\text{H}_2}) + \frac{1}{2} \ln(P_{\text{O}_2}) \right] \quad (3)$$

The activation overpotential (η_{act}) directly characterizes the kinetic resistance of the electrode reactions, following the Tafel relationship. It is governed by the kinetics of the oxygen reduction reaction (ORR) and hydrogen oxidation reaction (HOR), rendering it highly sensitive to the state of the catalyst layer. Its increase is primarily driven by the loss of the electrochemically active surface area (ECSA) and a decrease in the intrinsic activity of the remaining active sites. This increase is modeled through three time-dependent contributions. First, the principal mechanism involves platinum catalyst surface oxidation during PEMFC operation. Platinum particles undergo oxidation in the reaction environ-

ment, thereby reducing the proportion of active sites within the catalyst's electrochemically active surface area (ECSA). This process is modeled using a linear function, reflecting the approximately linear accumulation of voltage loss over time [24]:

$$V_{\text{PtO}}(t) = V_{\text{PtO},0} + V_{\text{PtO},1}t \quad (4)$$

Here, $V_{\text{PtO},0}$ represents the initial loss, and $V_{\text{PtO},1}$ quantifies the rate of voltage decay due to progressive catalyst oxidation.

Under potential cycling, the electrochemical active surface area (ECSA) decays through mechanisms extending beyond surface oxidation, including platinum nanoparticle dissolution, Ostwald ripening, and particle agglomeration [25]. Additional degradation mechanisms, such as carbon support corrosion and ionomer degradation, can result in catalyst layer detachment. This decay typically exhibits a two-stage trend: an initial rapid drop, attributed to the dissolution and agglomeration of small particles, followed by a slower phase involving the evolution of larger, more stable particles. Therefore, the time-dependent effective active area, $A(t)$, is represented by a semi-empirical logarithmic decay model [24]:

$$A(t) = A_0 - A_1 \ln(A_2 t + 1) \quad (5)$$

The cross-current density i_{loss} arises from side reactions induced by hydrogen and oxygen crossover through the membrane. Its exponential increase is attributed to membrane pore expansion, a form of physical degradation. These parasitic reactions occupy active sites on the platinum surface, thereby reducing the number of sites effectively available for the main hydrogen oxidation and oxygen reduction reactions. Consequently, a higher overpotential is required to sustain the main reaction rate, manifesting as an increase in the activation overpotential [25]:

$$i_{\text{loss}}(t) = i_{\text{loss},0} \exp(i_{\text{loss},1} t) \quad (6)$$

As i_{loss} increases, the argument of the logarithmic term in the activation overpotential equation grows, resulting in a higher apparent activation loss.

The ohmic overpotential (η_{ohm}) is directly proportional to the total resistance, which comprises the protonic resistance of the membrane and the electronic resistances of other components. Among these, the membrane resistance (R) is the dominant and most variable contributor during aging [24]. Its increase stems from two synergistic pathways:

(i) Chemical degradation: Radicals (e.g., $\cdot\text{OH}$, $\cdot\text{OOH}$) generated during catalyst surface reactions or via gas crossover attack the polymer backbone and sulfonic acid groups, degrading the ion exchange capacity [26].

(ii) Mechanical degradation: Repetitive hydration/dehydration cycles and thermal stress induce membrane thinning, crack formation, and a loss of mechanical integrity, thereby increasing the resistance [27].

These processes often exhibit self-accelerating behavior because the degradation products can catalyze further radical generation. Therefore, the time-dependent membrane resistance, $R(t)$, is modeled using an exponential growth function [28]:

$$R(t) = R_0 \exp(R_1 t) \quad (7)$$

The parameter R_1 encapsulates the effective rate constant for the combined chemical and mechanical degradation processes that diminish ionic conductivity.

Mass transport losses, which become significant at high current densities, tend to intensify over time. The key aging mechanisms involve the restructuring and fouling of the porous media within the gas diffusion layer (GDL) and catalyst layer (CL) [29,30].

Pores in the GDL can be blocked by liquid water accumulation, carbon corrosion products, and leached metal ions. Meanwhile, within the CL, Pt agglomeration, ionomer redistribution, and structural collapse increase tortuosity and reduce porosity [25]. These changes collectively impede the transport of reactants to the active sites and the removal of product water, thereby lowering the effective limiting current density. These effects are empirically captured by the parameters m and n in the concentration overpotential term, which relate to the mass transport resistance and the limiting current density, respectively. Their time evolution is modeled using exponential functions, reflecting the progressive nature of pore blockage and structural compaction [28]:

$$m(t) = m_0 \exp(m_1 t) \quad (8)$$

$$n(t) = n_0 \exp(n_1 t) \quad (9)$$

Ultimately, incorporating the aforementioned ageing degradation terms into the voltage equation yields the dynamic ageing model:

$$V(t) = E_{\text{rev}} - V_{\text{PtO}}(t) - \frac{RT}{2\alpha_a F} \ln\left(\frac{i(t) + i_{\text{loss}}(t)}{i_{0,a}}\right) - \frac{RT}{4\alpha_c F} \ln\left(\frac{i(t) + i'_{\text{loss}}(t)}{i_{0,c}}\right) - i(t)R(t) - m(t) \left[e^{n(t)i(t)} - 1 \right] \quad (10)$$

A critical model assumption is that the initial exchange current densities ($i_{0,a}$ and $i_{0,c}$) remain constant. The model thus focuses on capturing how structural and compositional changes modulate the effective reaction kinetics. The complete set of parameters to be identified, denoted as X_{PEMFC} , thus encompasses both static electrochemical parameters and dynamic degradation coefficients:

$$X_{\text{PEMFC}} = [V_{\text{PtO},0}, V_{\text{PtO},1}, A_0, A_1, A_2, i_{\text{loss},0}, i_{\text{loss},1}, i_{0,a}, i_{0,c}, R_0, R_1, m_0, m_1, n_0, n_1] \quad (11)$$

Identifying the 15 parameters in X_{PEMFC} from experimental polarization curves obtained at multiple aging stages constitutes a high-dimensional, nonlinear, and potentially ill-posed optimization problem [6]. The optimization objective is to find the parameter set that minimizes the discrepancy between the model predictions and the experimental data across the entire aging timeline. The root mean square error (RMSE) is adopted as the objective function for this purpose. It is a standard metric that penalizes large deviations and provides a voltage-based error measure, facilitating direct comparison across studies [25]. The RMSE, which quantifies the model's predictive accuracy, is calculated as follows:

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (V_{\text{exp},i} - V_{\text{model},i})^2} \quad (12)$$

where N denotes the number of experimental data points; $V_{\text{exp},i}$ represents the experimentally measured voltage value at the i current density; and $V_{\text{model},i}$ indicates the model-predicted voltage value at the i current density.

The RMSE metric imposes a greater penalty on larger errors. This characteristic aids the optimization algorithm in seeking a globally optimal fit across the entire polarization curve while preventing substantial local deviations. As the RMSE shares the same unit as the measured voltage, its physical significance is intuitively clear: it represents the average magnitude of deviation between the model predictions and the experimental measurements [26]. Widely adopted in PEMFC parameter identification studies, the use of RMSE facilitates direct comparison with findings reported in the literature.

By minimizing the RMSE, the optimization algorithm iteratively adjusts the unknown parameters in the model until the deviation between the predicted and the experimentally measured aging polarization curves is minimized [31]. This process enables the effective identification of the activation-, ohmic-, and concentration-overpotential-related parameters that evolve during the aging process.

3. Adaptive Optimization Algorithm Developing

Conventional differential evolution (DE) algorithms are prone to convergence to local optima when addressing the optimization challenges inherent in fuel cell polarization curve parameter fitting. These challenges include strong nonlinearity, multi-parameter coupling, and complex constraints [6]. This limitation stems from an imbalance between exploration and exploitation, a lack of awareness of the population state, and limited diversity in search strategies. To overcome these limitations, this paper proposes a Threat-Response Adaptive Differential Evolution (TRADE) algorithm, grounded in the “perception–decision–response” intelligent paradigm [32]. The algorithm constructs a threat–response theoretical framework. This framework enables the real-time perception of potential threats to the population state—such as declining diversity or convergence stagnation—and dynamically activates corresponding three-tier response strategies. This mechanism achieves adaptive regulation of the search behavior.

3.1. Threat Response Theoretical Framework

The core concept of the threat–response theory is to endow optimization algorithms with a closed-loop control capability, akin to that of intelligent systems. This capability enables the autonomous adjustment of search strategies based on feedback from the algorithm’s internal state. This framework consists of three core components: threat perception, strategy decision-making, and strategy execution. The mapping of this framework onto the optimization process is illustrated in Figure 1.

3.2. Three-Tier Threat-Response Mapping Model

To address the complex optimization challenges in PEMFC parameter identification, a three-tier threat–response mapping model is established. This model defines three levels of threat—low, medium, and high—based on the population state. These threat levels are mapped to three corresponding strategies: refined response, evasive response, and aggressive response. This mapping enables the algorithm to dynamically adjust its search behavior in response to perceived threats, such as diversity loss or convergence stagnation. Specifically:

- Under low-threat conditions, the algorithm focuses on intensive exploitation around current promising solutions via an elite-guided refined search to enhance convergence accuracy.
- Under medium-threat conditions, it balances local exploitation and global exploration by combining individual historical best positions with random differential vectors, thereby proactively avoiding premature convergence.
- Under high-threat conditions, the algorithm employs strong perturbation mutations based on population statistics and a historical archive, executing a breakthrough–restart mechanism to escape local optima.

This structured state–threat–response mapping mechanism enables the TRADE algorithm to autonomously adapt to different optimization stages. It thereby achieves a fundamental transition from parameter self-adaptation to the self-adaptation of behavioral strategies.

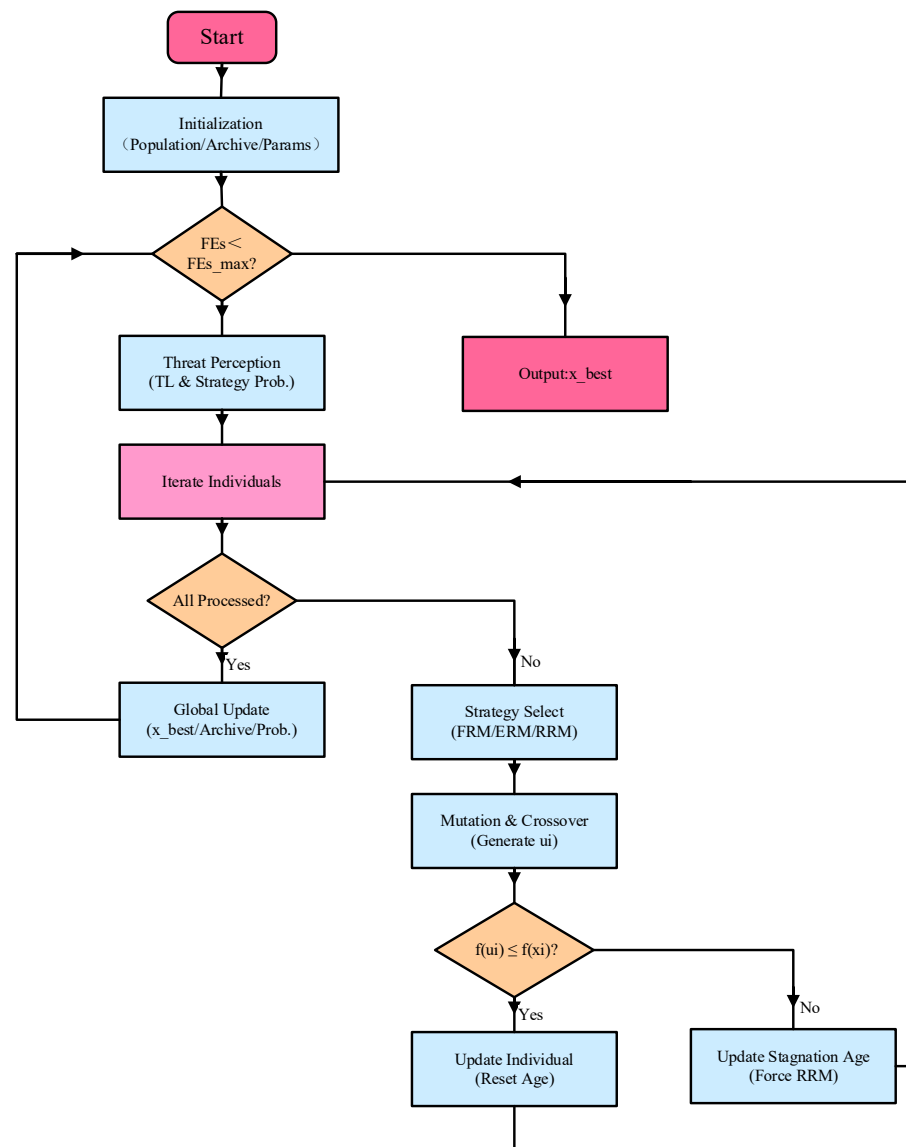


Figure 1. Flowchart of the TRADE algorithm.

3.3. Multi-Dimensional Threat Perceiver

Built upon this framework, the TRADE algorithm incorporates three core modules: a Multi-dimensional Threat Perceiver, a Three-tier Response Strategy Library, and an Adaptive Strategy Decision-maker. These modules are responsible for state perception, strategy execution, and strategy decision-making, respectively. They work synergistically to form a complete adaptive optimization mechanism.

Population clustering serves as a measure of diversity. The clustering threat index, denoted as I_c , is defined as the average Euclidean distance between each individual and the centroid of the population. Here, I_{c_max} denotes the maximum observed value of the clustering index, typically recorded at the initial iteration. After normalization to the range $[0, 1]$, a higher value of D_t' indicates a greater perceived threat to population diversity.

$$I_c = \frac{1}{NP \cdot D} \sum_{i=1}^{NP} \|x_i - \bar{x}\| \quad (13)$$

$$D_t' = 1 - \frac{I_c}{I_{c_max}} \quad (14)$$

Evolutionary Stagnation Threat Index I_s used to assess convergence trends, values approaching 1 indicate severe evolutionary stagnation.

$$I_s = \exp\left(-\frac{\Delta f_{\text{best}}}{L \cdot \epsilon}\right) \quad (15)$$

where Δf_{best} denotes the average improvement in optimal fitness over the most recent L generations; L represents the sliding window size; ϵ is the tolerance threshold (preventing the denominator from becoming zero).

The fitness distribution equilibrium threat index I_d reflects the algorithm's search vitality. It is defined as the ratio of the fitness range to the mean deviation:

$$I_d = \frac{f_{\text{worst}} - f_{\text{best}}}{\frac{1}{NP} \sum_{i=1}^{NP} |f_i - \bar{f}| + \delta} \quad (16)$$

where f_{best} , f_{worst} denote the optimal and worst fitness values of the current population, respectively, $\bar{f}$ represents the fitness mean, and δ denotes the minimum value (to prevent a zero denominator).

The comprehensive threat level TL maps the standardized indices into fuzzy values via a triangular membership function. Subsequently, weights w_1 , w_2 , and w_3 are assigned based on the importance of each indicator (satisfying $w_1 + w_2 + w_3 = 1$). The weighted average method calculates the comprehensive threat level, which is then categorized into low, medium, and high levels according to a threshold:

The comprehensive threat level, denoted as TL, is derived by fusing the standardized indices through a triangular membership function to obtain fuzzy values. Subsequently, weights w_1 , w_2 , and w_3 are assigned to the respective indices based on their perceived importance, subject to the constraint $w_1 + w_2 + w_3 = 1$. The comprehensive threat level TL is then calculated using a weighted average. This TL value is finally categorized into low, medium, or high levels based on predefined thresholds.

$$TL = \frac{w_1 \tilde{D}_t + w_2 \tilde{I}_s + w_3 \tilde{I}_d}{w_1 + w_2 + w_3} \quad (17)$$

Corresponding to these threat levels, three distinct mutation operators are designed, forming a three-tier response strategy library:

For a low-threat state, the Fine-grained Response Mutation (FRM) strategy emphasizes intensive exploitation. It incorporates guidance from elite individuals to direct the search while maintaining limited exploration capability via small Gaussian perturbations. Here, x_{elite} denotes an individual randomly selected from an elite archive, which comprises the top 10% of individuals with the best fitness. This guides the search toward promising regions of the solution space.

$$v_i = x_{\text{elite}} + F_1(x_a - x_b) + \eta N(0, \sigma^2 I) \quad (18)$$

For medium threat states, the Evasive Response Mutation (ERM) strategy centers on "evasion-exploration, integrating individual historical optimal guidance with random differential perturbations to balance local exploitation and global exploration. Here, x_{pbest} denotes the historical optimal solution of the individual, guiding individuals towards their own optimal direction.

For a medium-threat state, the Evasive Response Mutation (ERM) strategy focuses on balanced exploration–exploitation. It integrates guidance from an individual's historical best position with random differential perturbations to navigate between local refinement

and global search. Here, x_{pbest} represents the personal historical best solution of the i -th individual, providing a directional bias toward its locally optimal region.

$$v_i = x_i + F_m(x_{pbest} - x_i) + F_m(x_r - x_s) \quad (19)$$

For high-threat states, the Radical Response Mutation (RRM) strategy adopts a “breakthrough-restart” approach. By incorporating population statistics and historical archive data, it generates disruptive perturbations. In the formula, $\bar{x}$ represents the current population mean, providing a global position reference; x_{r1} , x_{r2} represents randomly selected distinct individuals; ζ denotes the archive perturbation coefficient; x_{arch1} , x_{arch2} indicates an individual randomly selected from an external archive set (preserving historically high-quality and diverse individuals).

$$v_i = \bar{x} + F_h(x_{r1} - x_{r2}) + \zeta(x_{arch1} - x_{arch2}) \quad (20)$$

3.4. Adaptive Strategy Decision-Maker

The core function of the Adaptive Strategy Decision-maker is to dynamically allocate response strategies by integrating the comprehensive threat level with individual performance metrics. This achieves a two-layer decision-making paradigm that combines global regulation and local adaptation. This module employs a global–local two-tier decision mechanism, which operates as follows:

Global Decision: Based on the current generation’s comprehensive threat level TL , determine the baseline allocation probabilities ($P_{base} = [p_f, p_e, p_r]$) for the three response strategies: FRM, ERM, and RRM. In this study, when threat is low ($p_f = 0.7, p_e = 0.2, p_r = 0.1$), the refinement strategy is prioritized; when threat is moderate ($p_f = 0.3, p_e = 0.6, p_r = 0.1$), the evasion strategy is prioritized; when threat is high ($p_f = 0.1, p_e = 0.2, p_r = 0.7$), the radical strategy is prioritized.

Local Decision: Fine-tune the base probabilities based on each individual’s differentiated performance. Two individual characteristic metrics are introduced: relative fitness ranking ($rank_i$), reflecting current performance (higher ranking indicates superior performance), and historical improvement stagnation index (age_i), indicating whether an individual has stagnated (higher values denote more severe stagnation). The fine-tuning formula is shown in the following equations:

$$p_f' = p_f + \alpha \frac{NP - rank_i}{NP} \quad (21)$$

$$p_r' = p_r + \beta \frac{age_i}{age_{max}} \quad (22)$$

$$p_e' = 1 - p_f' - p_r' \quad (23)$$

Here, α and β are adjustment coefficients (typically in the range [0.1, 0.3]), and age_{max} is the predefined maximum stagnation threshold. Finally, the adjusted probabilities (p_f', p_e', p_r') are normalized. Each individual then selects a mutation operator for the current iteration from the three available strategies using a roulette wheel selection mechanism based on these normalized probabilities.

3.5. Algorithm Summary and Discussion

The core of the TRADE algorithm lies in its intelligent closed-loop control paradigm of “perception–decision–response.” Algorithm 1 summarizes its key procedural steps.

Algorithm 1: Core Procedure of the TRADE Algorithm

Input: Experimental polarization curve data D_{exp} , parameter bounds [LB, UB]
Output: Optimal parameter set X_{best} , minimum RMSE f_{best}

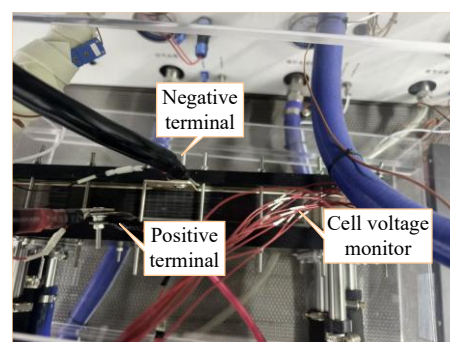
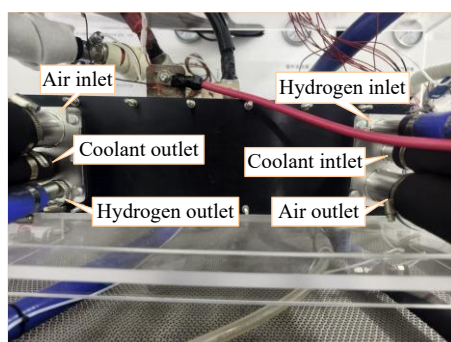
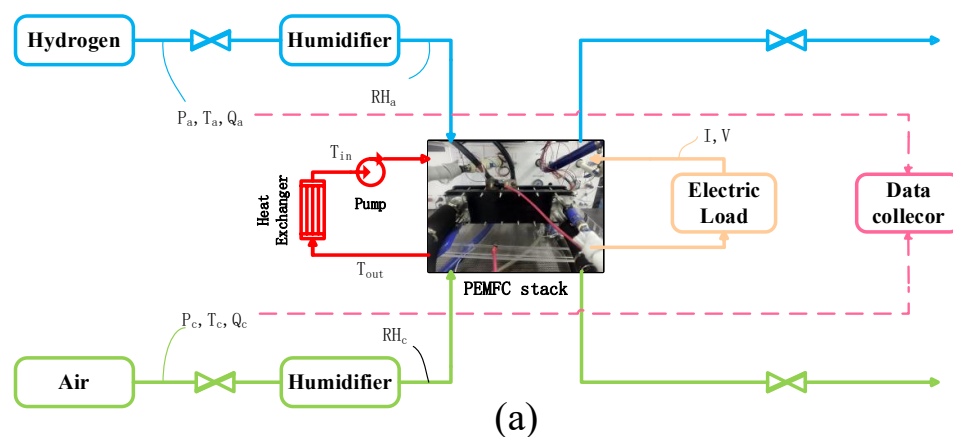
- 1: Initialize population P , evaluate fitness $f(x) = \text{RMSE}(x, D_{\text{exp}})$.
- 2: while not reaching termination criterion do
- 3: //1. Multi-dimensional Threat Perception
- 4: Calculate threat indices I_c, I_s, I_d using Equations (13), (15) and (16).
- 5: Compute Comprehensive Threat Level TL using Equation (17).
- 6: Determine base strategy probabilities $P_{\text{base}} = [p_f, p_e, p_r]$ based on TL .
- 7: //2. Adaptive Strategy Decision
- 8: for each individual i in population do
- 9: Adjust individual strategy probability p_i' based on rank_i and age_i using Equations (21)–(23).
- 10: Select a mutation strategy (FRM/ERM/RRM) via roulette wheel selection according to p_i' .
- 11: //3. Response Execution
- 12: Generate trial vector v_i using the selected strategy (Equations (18)–(20)).
- 13: Perform crossover and boundary check to obtain u_i .
- 14: Select the better one between x_i and u_i for the next generation.
- 15: Update elite set, historical archive, and global best solution ($X_{\text{best}}, f_{\text{best}}$).
- 16: return $X_{\text{best}}, f_{\text{best}}$

The process begins with a multi-dimensional threat perception of the population state (Equations (13)–(16)), followed by the calculation of a comprehensive threat level TL (Equation (17)). Subsequently, the adaptive decision-making mechanism dynamically assigns each individual a refined, exploratory, or radical mutation strategy (Equations (18)–(20)) based on the global TL and the individual's historical performance (Equations (21)–(23)). This dynamic adjustment enables the algorithm to precisely balance exploration and exploitation across different optimization phases, thereby achieving an efficient and robust search in complex parameter spaces.

The TRADE algorithm facilitates a paradigm shift from parameter-level self-adaptation to behavioral-strategy self-adaptation. By establishing a quantifiable threat-perception framework and a hierarchical response mechanism, it significantly enhances both robustness and solution accuracy in complex optimization tasks. It should be noted that the threat-perception module introduces additional computational overhead, which necessitates efficiency trade-offs for problems with extremely high evaluation costs. Future work could explore lightweight threat metrics or asynchronous update mechanisms to further improve the algorithm's computational efficiency.

4. Results

Ageing tests were conducted on a commercial 3 kW PEMFC stack. The membrane electrode assembly (MEA) consisted of a Pt/C catalyst with a loading of $0.4 \text{ mg} \cdot \text{cm}^{-2}$ on both the anode and cathode, and a perfluorosulfonic acid (PFSA) membrane with a thickness of $25 \text{ } \mu\text{m}$. During polarization curve acquisition, the operating conditions were strictly controlled as follows: cell temperature, $70 \text{ } ^\circ\text{C}$; backpressure, 1.1 bar for both anode and cathode; relative humidity (RH), 80% (anode) and 80% (cathode). The hydrogen and air stoichiometries were set to 1.5 and 2.0, respectively. Polarization curves were measured by recording the steady-state voltage at each discrete current density. The structural diagram of the PEMFC is shown in Figure 2a, and the physical photograph is shown in Figure 2b. The key material properties and operational parameters are summarized in Table 2.



(b)

Figure 2. (a) Schematic diagram of test station, (b) stack used in the experiment.

Table 2. PEMFC stack material specifications and test conditions.

Parameter	Value	Unit
Catalyst (Anode/Cathode)	Pt/C (0.4/0.4)	mg·cm ^{−2}
Membrane type	PFSA	-
Membrane thickness	25	μm
Active area	220	cm ²
Operating temperature	70	°C
Anode/Cathode backpressure	1.1/1.1	bar
Anode RH/Cathode RH	80%/80%	-
H ₂ stoichiometry	1.5	-
Air stoichiometry	2.0	-
Current range	0–198	A

All polarization curve measurements and durability tests were performed on an integrated fuel cell test station. High-purity hydrogen (99.999%) and compressed air served as the reactant gases. The flow rates of both gases were precisely controlled using Alicat MC-Series mass flow controllers to maintain the specified stoichiometric ratios. Cell voltage and current were controlled and measured with a Faith FT6430N electronic load. The stack temperature was maintained at 70 °C by a Kaideli PCB industrial chiller. Reactant humidification was achieved using a Hetai Basic-Q30 pure water humidification system. For detailed electrochemical characterization, a ZAHNER ZENNIUM PRO electrochemical workstation was used. All experiments were conducted on a custom-built 24-cell PEMFC stack with a rated power of 3 kW.

4.1. Experimental Setup and Performance Metrics

All algorithms were configured with a population size of 30 and a maximum of 2000 iterations. To ensure statistical reliability, each algorithm was independently executed 10 times. The algorithms selected for comparison were Particle Swarm Optimization (PSO), Beluga Whale Optimization (BWO), standard Differential Evolution (DE), and the proposed TRADE. The lower and upper bounds for each parameter to be identified are provided in Table 3 [22]. The performance was evaluated using the root mean square error (RMSE), mean absolute error (MAE), and standard deviation (SD).

Table 3. PEMFC Data Sheet.

	$V_{\text{PtO},0}$	$i_{\text{loss},0}$	$i_{0,a}$	$i_{0,c}$	R_0	m_0	n_0
Max bound	0.35 V	0.015 A/cm ²	0.01 A/cm ²	0.01 A/cm ²	0.35 $\Omega \cdot \text{cm}^2$	0.5	0.25
Min bound	0.15 V	0.001 A/cm ²	0.005 A/cm ²	0.005 A/cm ²	0.05 $\Omega \cdot \text{cm}^2$	0.001	0.05
	$V_{\text{PtO},1}$	$i_{\text{loss},1}$	A_1	A_2	R_1	m_1	n_1
Max bound	$1 \times 10^{-5} \text{ h}^{-1}$	$2 \times 10^{-3} \text{ h}^{-1}$	0.15 cm ²	0.01 h ⁻¹	$2 \times 10^{-3} \text{ h}^{-1}$	0.01	1×10^{-3}
Min bound	$2 \times 10^{-3} \text{ h}^{-1}$	$1 \times 10^{-5} \text{ h}^{-1}$	0.01 cm ²	$1 \times 10^{-4} \text{ h}^{-1}$	$1 \times 10^{-5} \text{ h}^{-1}$	1×10^{-4}	1×10^{-5}

4.2. Parameter Identification Results

4.2.1. Optimal Parameter Set

After ten independent runs, the TRADE algorithm converged to a unified parameter set (Table 4) that effectively captures the key aging mechanisms. Critically, this single parameter set accurately reproduces the four polarization curves measured at 100, 400, 700, and 1000 h of operation. This demonstrates the model's capability to quantitatively characterize the evolution of aging throughout the entire test cycle.

Table 4. Optimal Parameter Set.

	$V_{\text{PtO},0}$	$i_{\text{loss},0}$	$i_{0,a}$	$i_{0,c}$	R_0	m_0	n_0
Value	0.2437	0.00259	0.00837	0.00834	0.1002	0.0165	0.2499
	$V_{\text{PtO},1}$	$i_{\text{loss},1}$	A_1	A_2	R_1	m_1	n_1
Value	2.66×10^{-5}	1.003×10^{-3}	0.02079	0.00827	1.04×10^{-5}	2.49×10^{-4}	1.005×10^{-3}

4.2.2. Statistical Comparison of Algorithm Performance

Table 5 presents the statistical results of key performance metrics from 10 runs of each algorithm. The TRADE algorithm achieved optimal performance across all key metrics. It attained an average RMSE of 0.00705 V, which is approximately 41.7% lower than that of the second-best performer, the DE algorithm (0.0121 V). Its average MAE was 0.004603 V, about 31.5% lower than the DE algorithm's 0.006724 V. Furthermore, its standard deviation was only 0.0008 V, indicating superior stability. In terms of fitting efficiency, the TRADE algorithm also demonstrated excellent precision and robustness.

Regarding extreme value performance, the TRADE algorithm achieved a minimum RMSE of 0.00675 V and a maximum of 0.0082 V. This fluctuation range was substantially narrower than those of the other algorithms. By contrast, the other algorithms exhibited average RMSE values above 0.0121 V and standard deviations exceeding 0.0015 V, indicating comparatively poorer stability across multiple runs. Notably, when fitting the end-of-life aging curves, the TRADE algorithm maintained high accuracy, while the comparison algorithms exhibited significantly higher RMSE values. This further underscores TRADE's superior capability in modeling aging degradation mechanisms.

Table 5. Statistical Comparison of Performance Metrics for Each Algorithm.

Algorithm	Best RMSE (V)	Worst RMSE (V)	Average RMSE (V)	MAE (V)	SD (V)	R ²
TRADE	0.00675	0.0082	0.00705	0.004603	0.0008	0.9986
DE	0.00997	0.0132	0.0121	0.006724	0.0015	0.9951
BWO	0.0117	0.0182	0.0154	0.009206	0.0018	0.9923
PSO	0.0142	0.0172	0.0164	0.011834	0.0021	0.9905

4.2.3. Physical Interpretation and Validation of Identified Aging Parameters

The physical relevance of the aging rate parameters identified by the TRADE algorithm (Table 3) was validated by comparing their orders of magnitude and temporal trends with values reported in the existing literature. The dominant degradation mechanism, Pt catalyst oxidation, was associated with a linear voltage decay rate of $V_{\text{PtO},1} = 2.66 \times 10^{-5} \text{ V} \cdot \text{h}^{-1}$. This value is consistent in order of magnitude with findings from literature focusing on catalyst degradation, which also reports a voltage decay rate associated with Pt oxide formation (V_{PtO}) of $6.75 \times 10^{-3} \text{ V} \cdot \text{h}^{-1}$ for a stack operating at 55°C , a low membrane resistance degradation rate of $2.11 \times 10^{-6} \Omega \cdot \text{cm}^2 \cdot \text{h}^{-1}$, and an m_1 value of $4.29 \times 10^{-4} \text{ h}^{-1}$ linked to catalyst layer restructuring [22]. The slightly lower rate observed in this study can be attributed to the higher operating temperature (70°C) which may mitigate certain kinetic losses-and to differences in catalyst specifications.

Furthermore, the membrane resistance growth rate identified by the TRADE algorithm ($R_1 = 1.04 \times 10^{-5} \Omega \cdot \text{cm}^2 \cdot \text{h}^{-1}$) is extremely low. This aligns with observations of stable membrane performance under well-humidified conditions and is comparable to the low degradation rates noted in the aforementioned literature. The increase in the mass transfer resistance parameters ($m_1 = 2.49 \times 10^{-4} \text{ h}^{-1}$, $n_1 = 1.00 \times 10^{-3} \text{ h}^{-1}$) also falls within a physically plausible range for long-term operation, consistent with the catalyst layer restructuring-related trends reported previously.

While direct numerical agreement is not expected due to differences in materials and test protocols, the consistent orders of magnitude and corroborated dominance of the Pt oxidation mechanism confirm that the parameter set identified by the synergistic model and TRADE algorithm is physically meaningful and captures the essential aging dynamics of PEMFCs.

4.3. Analysis of Polarisation Curve Fitting Performance

Comparison of Fitting Across the Entire Ageing Cycle

As shown in the Figure 3, the polarization curves predicted by the TRADE algorithm show close agreement with the measured data across the entire current density range. The predictions accurately capture the aging-induced increase in activation overpotential at low current densities, closely follow the progressive change in ohmic losses at medium current densities, and faithfully reproduce the gradual intensification of concentration overpotential at high current densities.

Analysis of the polarization curves at 100, 400, 700, and 1000 h confirms the close alignment between the predicted and measured curves across all aging stages. At 100 h, the predicted curve accurately captures the aging-induced increase in activation overpotential at low current densities, with the predicted and measured trajectories nearly overlapping. After 400 h of aging, the model continues to closely match the progressive change in ohmic losses in the medium current density region, showing no degradation in fit quality with time. At 700 h, the predicted curve faithfully reproduces the gradual intensification of concentration overpotential at high current densities, exhibiting decay trends that are highly synchronized with the measured data. Even after 1000 h of aging, despite increased

dispersion in the measured data points, the TRADE prediction curve remains closely aligned with the overall measured trend, successfully capturing the aging characteristics of all overpotential types across the entire current density range.

Across all four aging stages, the model demonstrates consistently superior fitting performance in the medium current density region. This consistently high accuracy underscores TRADE's capability to precisely model the coupled evolution of multiple overpotential types throughout the entire fuel cell aging cycle.

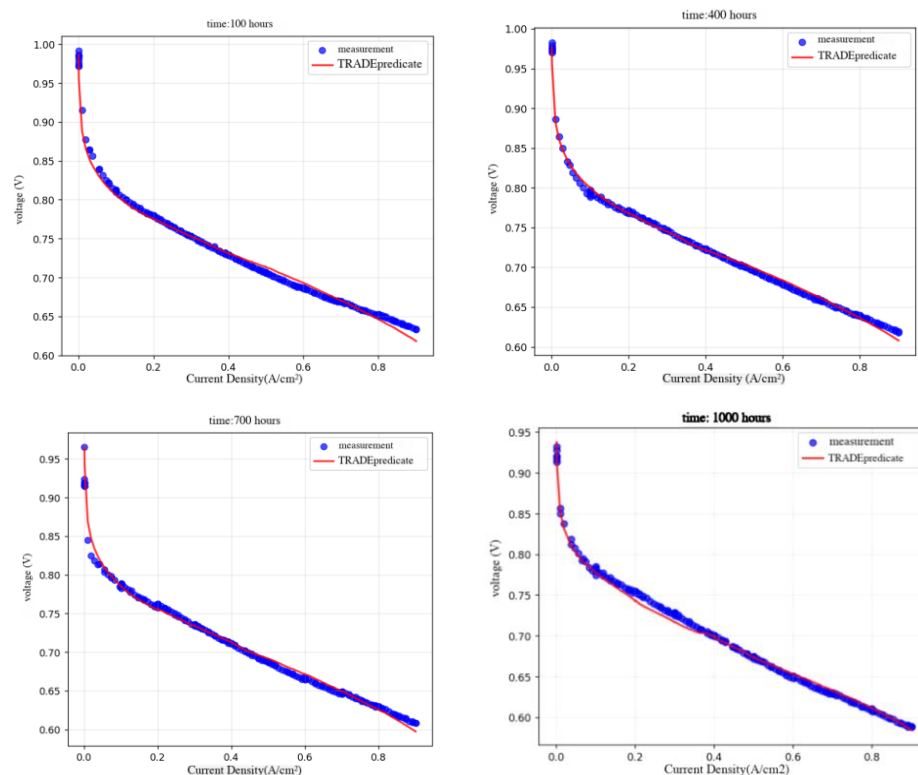


Figure 3. Calculated and measured V-I curves for PEMFC stacks.

4.4. Sensitivity Analysis of Ageing Degradation Mechanisms

The contribution of different voltage loss mechanisms—activation, ohmic, and concentration overpotential—to PEMFC performance degradation varies significantly with operating time and conditions [7,22]. This section quantifies the sensitivity of each degradation mechanism across both time and current density. The analysis utilizes experimental aging data obtained at 100, 400, 700, and 1000 h. This analysis combines decay-area plots with overpotentiating contribution diagrams to identify the dominant degradation sources, thereby providing targeted insights for lifetime optimization.

4.4.1. Analytical Dimensions and Evaluation Metrics

The sensitivity of each mechanism is quantified by its contribution ratio to the total voltage loss. Four characteristic time points are analyzed: 100 h (early stage), 400 h (mid-stage), 700 h (mid-to-late stage), and 1000 h (end-of-life stage). Four representative current densities are examined: $0.05 \text{ A} \cdot \text{cm}^{-2}$ (low load), $0.3 \text{ A} \cdot \text{cm}^{-2}$ (medium-low load), $0.6 \text{ A} \cdot \text{cm}^{-2}$ (medium-high load), and $0.9 \text{ A} \cdot \text{cm}^{-2}$ (high load).

4.4.2. Sensitivity Evolution in the Time Dimension

Throughout the aging cycle, the activation overpotential—primarily driven by platinum catalyst oxidation—was the dominant contributor to performance degradation. Its contribution increased steadily from 74.6% at 100 h to 75.5% at 1000 h, consistently exceed-

ing the sum of all other degradation factors by a factor greater than 2.9. This dominance stems from the significant initial voltage drop (0.2437 V) and the sustained linear accumulation rate (approximately $2.66 \times 10^{-5} \text{ h}^{-1}$) associated with the loss of electrochemically active surface area due to platinum oxidation.

Against this backdrop of dominant activation loss, the contributions from other degradation mechanisms exhibit distinct phase-dependent characteristics. During the initial and mid-term aging periods (up to 700 h), the combined contribution of ohmic and concentration overpotentials remained below 6.5%, exerting a negligible influence on overall performance decay. However, under sustained high-current-density operation (e.g., $0.9 \text{ A} \cdot \text{cm}^{-2}$) for 1000 h, the concentration overpotential increases significantly. This increase is driven by the continuous accumulation of mass-transfer resistance, as captured by the growth of parameters m and n (with rates of $2.49 \times 10^{-4} \text{ h}^{-1}$ and $1.00 \times 10^{-3} \text{ h}^{-1}$, respectively). Under these conditions, the concentration overpotential accounts for 33% of the non-activation decay, making it a significant secondary degradation source. This trend is visually summarized in Figure 4, which illustrates the voltage retention rate decline over time.

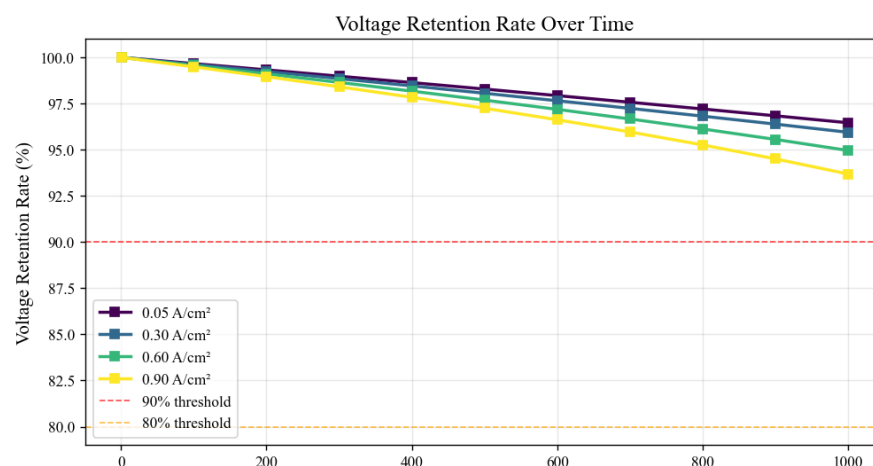


Figure 4. Voltage retention rate over time.

4.4.3. Sensitivity Distribution in the Current Density Dimension

At a given ageing stage (e.g., 1000 h), the dominant role of the activation overpotential (including degradation from platinum catalyst oxidation) remains unchanged with varying current densities, consistently contributing over 70% (with an absolute decay of approximately 0.27 V). However, current density significantly influences the internal composition of the secondary decay components. As depicted in Figure 5, the contributions of individual degradation mechanisms to total voltage loss vary notably with current density.

At low current densities ($0.05 \text{ A} \cdot \text{cm}^{-2}$), performance decay is dominated by the kinetic loss component of the activation overpotential (96.0%). Contributions from ohmic and concentration overpotentials are negligible, together constituting less than 4%. As the current density increases, the influence of the concentration overpotential rises sharply. Under high-load conditions ($0.9 \text{ A} \cdot \text{cm}^{-2}$), the concentration overpotential accounts for 33% of the total non-activation decay, forming a core degradation component alongside the remaining activation losses (65%). This finding indicates that under high-load, long-term operating conditions, degradation of the mass transfer pathways—reflected in the increase in parameters m_1 and n_1 —emerges as a critical optimization target, alongside the continuous monitoring of catalyst activity decay.

Figure 6 illustrates the progressive shifts in the contributions of activation, ohmic, and concentration overpotentials throughout the entire aging period, highlighting the dynamic interplay between these degradation pathways over time.

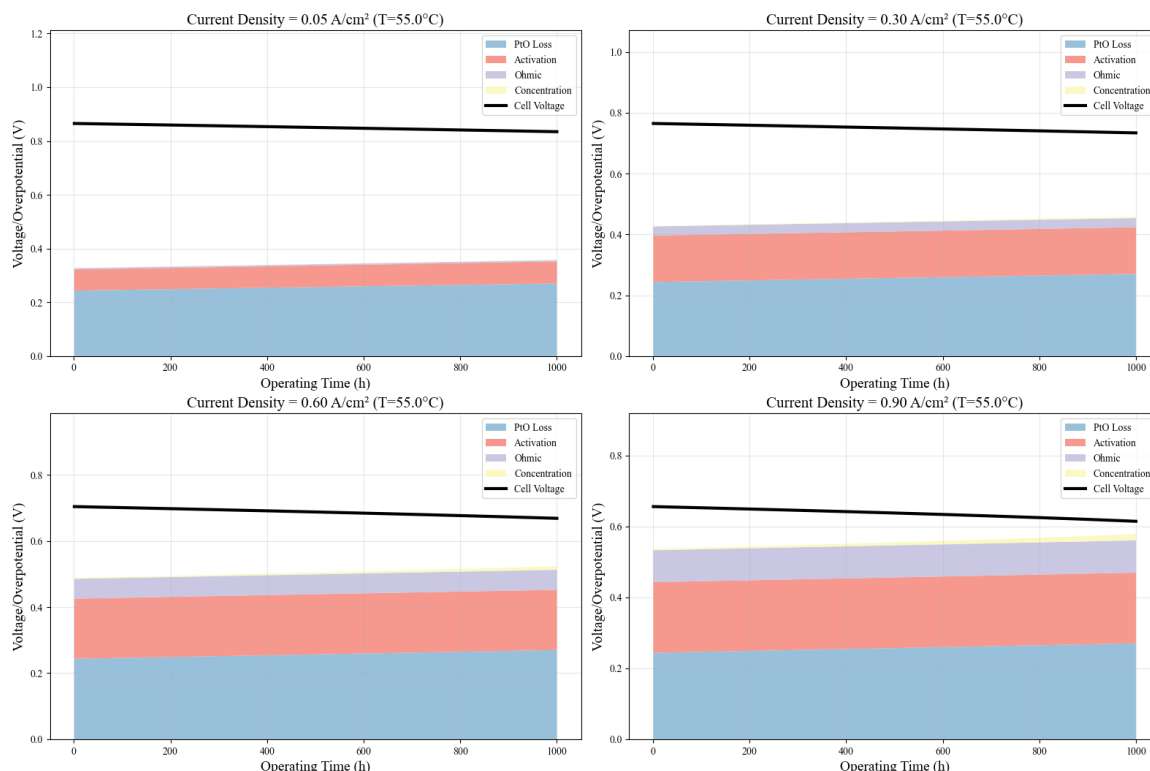


Figure 5. Contribution of individual degradation mechanisms to total voltage loss under varying.

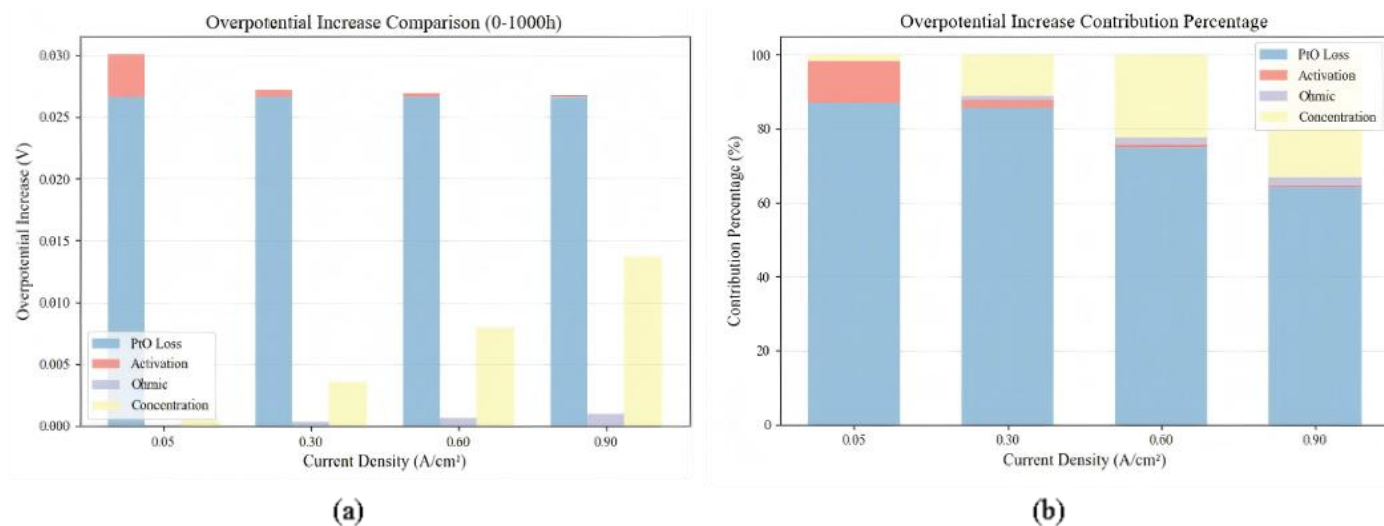


Figure 6. Evolution of overpotential contributions across the ageing cycle (0–1000 h): (a) Overpotential Increase Comparison (0–1000 h), (b) Overpotential Increase Contribution Percentage.

5. Discussion

This study addresses the challenge of identifying aging degradation parameters in PEMFCs. A multi-mechanism synergistic dynamic aging model is developed, and the Threat Response Adaptive Differential Evolution (TRADE) algorithm is proposed for parameter identification. The TRADE algorithm demonstrated superior parameter identification performance. It achieved an average RMSE of 0.00705 V over the entire aging cycle (100–1000 h), representing a 41.7% reduction compared to the standard Differential

Evolution (DE) algorithm. Even at the end of the aging test (1000 h), it maintained a low fitting error (0.0231 V), demonstrating robust and precise performance within the complex, nonlinear parameter space.

The developed model, which incorporates specific aging attenuation terms, achieves precise characterization of the synergistic decay among activation, ohmic, and concentration overpotentials. It quantifies key aging mechanisms—including platinum catalyst oxidation and electrochemical active area loss—thereby providing a quantitative tool for aging mechanism analysis. Sensitivity analysis confirms that degradation due to platinum catalyst oxidation is the dominant decay source throughout the entire cycle and across the current density range (contribution $\geq 70\%$). In contrast, under long-term, high-current-density conditions, the contribution of concentration overpotential rises to 33% of the non-activation decay. These insights provide targeted guidance for catalyst optimization and mass transfer system improvement.

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Nomenclature

Parameter	Description	Unit
A_1	Active surface area aging parameter	cm^2
A_2	Active surface area aging parameter	h^{-1}
A_0	Active surface area at $t = 0$	cm^2
E_{rev}	Reversible open-circuit voltage	V
F	Faraday constant	$96,485 \text{ C} \cdot \text{mol}^{-1}$
i	Current density	$\text{A} \cdot \text{cm}^{-2}$
$i_{\text{loss},1}$	Aging parameter of i_{loss}	$[\text{h}^{-1}]$
$i_{\text{loss},0}$	Initial parameter of i_{loss}	$[\text{A} \cdot \text{cm}^{-2}]$
i_{loss}	Internal current density	$[\text{A} \cdot \text{cm}^{-2}]$
$i_{0,a}$	Exchange current density at the anode	$[\text{A} \cdot \text{cm}^{-2}]$
$i_{0,c}$	Exchange current density at the cathode	$[\text{A} \cdot \text{cm}^{-2}]$
m	Empirical parameter in concentration loss equation	[V]
m_1	Aging parameter of m	$[\text{h}^{-1}]$
m_0	Initial parameter of m	[V]
n	Empirical parameter in concentration loss equation	$[\text{cm}^2 \cdot \text{A}^{-1}]$
n_1	Aging parameter of n	$[\text{h}^{-1}]$
n_0	Initial parameter of n	$[\text{cm}^2 / \text{A}]$

p	Pressure	[pa]
P_{H_2}	the partial pressure of H_2	[atm]
P_{O_2}	the partial pressure of O_2	[atm]
R	Idea gas constant	[8.314 J·mol ⁻¹ ·K ⁻¹]
R	Ohmic resistance	[ohm·cm ²]
R_1	Aging parameter of R	[h ⁻¹]
RH	Relative humidity	[%]
R_0	Initial parameter of R	[ohm·cm ²]
T	Temperature	[K]
$V_{PtO,1}$	Aging parameter of V_{PtO}	[h ⁻¹]
$V_{PtO,0}$	Initial parameter of V_{PtO}	[V]
V_{PtO}	Platinum oxide overvoltage	[V]
α_a	Charge transfer coefficient at the anode	0.5
α_c	Charge transfer coefficient at the cathode	0.5
NP, D	Population size, Problem dimension	-
$x_i, \bar{x}$	Individual vector, Population mean vector	-
I_{c_max}	Initial clustering index	-
L, ε	Sliding window size, Tolerance threshold	-
δ	Small value to prevent zero denominator	-
F_l, η, σ	Low-threat scaling factor, Gaussian perturbation parameters	-
F_m	Medium-threat scaling factor	-
F_h, ζ	High-threat scaling factor, Archive perturbation coefficient	-
α, β, age_{max}	Adjustment coefficients, Maximum stagnation threshold	-

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