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Review

The Critical Role of Artificial Intelligence in Optimizing Electrochemical Processes for Water and Wastewater Remediation: A State-of-the-Art Review

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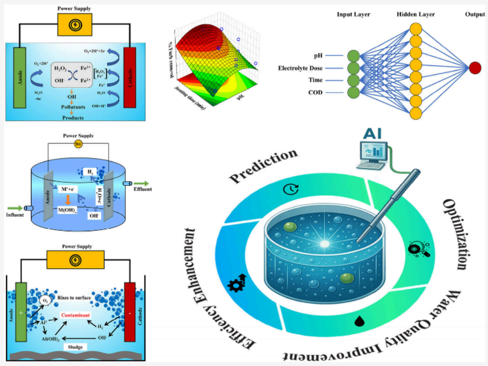
ABSTRACT: Artificial intelligence (AI) is transforming electrochemical water and wastewater treatment by enhancing efficiency, predictive accuracy, and process control. However, a comprehensive evaluation of AI models in optimizing electrochemical processes for pollutant removal is still lacking. This review addresses this gap by systematically analyzing AI applications in electrocoagulation (EC), electrooxidation (EO), electro-Fenton (EF), and electrodialysis (ED). Focusing on key advances and parameter optimization, it highlights how AI-driven models improve removal efficiency by capturing complex nonlinear interactions among variables such as current density, pH, electrode material, electrolyte composition, and pollutant concentration. Recent studies have notably shown that artificial neural networks (ANNs) and adaptive neuro-fuzzy inference systems (ANFIS) have achieved R^2 values above 0.99 in EC and EO processes, outperforming traditional models. Hybrid AI approaches like ANN-GA and ANFIS-ACO have further optimized catalyst dosage and ion migration in EF and ED. While AI has demonstrated remarkable potential, challenges such as limited data availability, model interpretability, and real-world implementation remain significant obstacles. Integrating AI with mechanistic modeling and real-time monitoring may overcome these barriers and enable autonomous, energy-efficient treatment systems. This Perspective offers critical insights into current progress and future opportunities, underscoring the role of intelligent optimization in advancing sustainable and scalable electrochemical water treatment technologies.



■ INTRODUCTION

Access to clean drinking water remains one of the most pressing challenges of our time and is a key priority under the United Nations Sustainable Development Goals (SDGs). At the same time, water pollution, driven by rapid industrialization and population growth, has become a significant environmental issue.[1] Addressing both concerns, wastewater treatment, and reuse provide a promising solution. Over the past few decades, significant advancements have been achieved in developing innovative, efficient, and cost-effective methods to eliminate contaminants from wastewater. Additionally, the use of optimization and modeling tools to evaluate performance and enhance efficiency has gained considerable momentum in recent years.[2]

Artificial intelligence (AI) is a computer-based system designed to emulate human intelligence, including the ability to acquire knowledge, make assessments, and make decisions autonomously. This rapidly evolving technology has become a ubiquitous tool in numerous fields, including the optimization of wastewater treatment processes.[3] These models can "learn" from a set of experimental data without prior knowledge of the physical and chemical laws governing the system, making them suitable for systems with nonlinearities and complex behavior. Recently, these models have proven to be valuable in water and wastewater treatment research, with successful applications in process design, water quality monitoring, parameter optimization, and performance prediction.[4]

The integration of AI into water and wastewater treatment processes offers numerous environmental benefits. For example, AI enhances the removal efficiency of pollutants, thus improving water quality and reducing environmental contamination.[5] It facilitates global sustainability efforts by enhancing efficiency and reducing the resource intensity of treatment processes. By optimizing the use of chemicals in the treatment processes, AI also helps to minimize the environmental footprint and operational costs. Furthermore, AI-driven optimizations can lead to significant energy savings by ensuring that the treatment processes run in their most efficient settings

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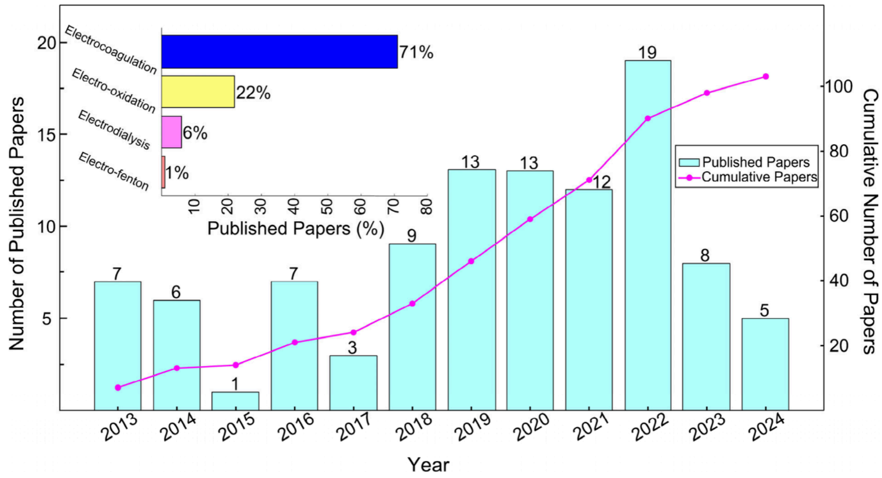

Figure 1. Bibliometric analysis on the application of AI techniques for modeling and/or optimization of electrochemical processes in water and wastewater treatment. The main chart displays the annual number and cumulative total of related publications from 2013 to 2024 (data extracted as of December 31, 2024). The inset chart illustrates the percentage distribution of AI-based studies across different electrochemical processes, highlighting the dominance of electrocoagulation, followed by electrooxidation, electrodialysis, and electro-Fenton.

without unnecessary energy expenditure. Water industries are increasingly investing in AI, with market projections estimating this investment to reach \$6.3 billion by 2030. AI has the potential to reduce operational costs by 20–30% by optimizing chemical usage in water treatment processes. Its straightforward implementation, flexibility, adaptability, and simple design make AI a valuable tool for streamlining and enhancing water treatment operations.[6,7] Until now, AI models such as ANNs, ANFIS, and support vector machines (SVMs) have been extensively used to model and optimize electrochemical processes, particularly in water and wastewater treatment.[8] Figure 1 illustrates the number of publications on the application of AI techniques for modeling and optimization of electrochemical processes for water and wastewater treatment over the recent decade. Published data demonstrate that AI models offer a robust framework for optimizing electrochemical processes, leading to improved energy efficiency, fewer experimental procedures, better parameter identification, and both economic and environmental benefits. The noticeable rise in publications after 2018, along with the cumulative trend, reflects the growing research attention and expanding adoption of AI-driven strategies in this field.

To the best of our knowledge, no comprehensive review to date has specifically examined the application of AI models in electrochemical processes for treating water and wastewater contaminated with diverse pollutants. Addressing this gap, this review analyzes the evolution of research in this area by evaluating annual publication trends and the distribution of AI applications across key electrochemical technologies. As shown in Figure 1, the inset chart highlights that EC is by far the most commonly studied process, followed by EO, ED, and EF. Rather than aiming to exhaustively review all AI and smart technologies in electrochemical water treatment, this study focuses on key findings in the literature, explores how AI integration enhances process efficiency and control, and identifies the main parameters influencing its performance. Lastly, the review outlines future perspectives and research challenges to support the continued development of AI-driven modeling in this field.

APPLICATION OF AI TECHNIQUES IN ELECTROCHEMICAL PROCESSES

Literature highlights the increasing role of AI techniques in electrochemical processes for water and wastewater treatment. AI enhances process modeling, optimization, and predictive accuracy, replacing traditional statistical approaches like response surface methodology (RSM) with advanced models such as genetic algorithms (GA), ANN, and ANFIS. As can be seen in Figure 2, the majority of the existing literature in this field has focused on RSM, with the objective of optimizing the electrocoagulation process. In recent years, there has been a notable shift toward the utilization of ANN and sophisticated AI techniques. Several studies that have explored the use of AI for modeling electrochemical water and wastewater treatment are summarized in Table S2. In the compilation of Table S2, only peer-reviewed scientific publications that are indexed in the Web of Science and Scopus databases were considered.

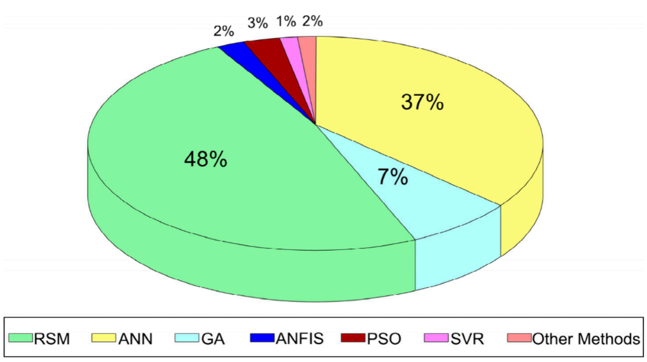

Figure 2. Distribution of the publication on the use of AI for modeling and/or optimization of electrochemical processes for water and wastewater treatment based on the applied computational approach.

AI FOR PROCESS PARAMETER OPTIMIZATION

Optimizing process parameters is crucial for enhancing the efficiency and cost-effectiveness of electrochemical treatments. Traditional one-factor-at-a-time (OFAT) approaches are time-consuming and fail to capture complex interactions between

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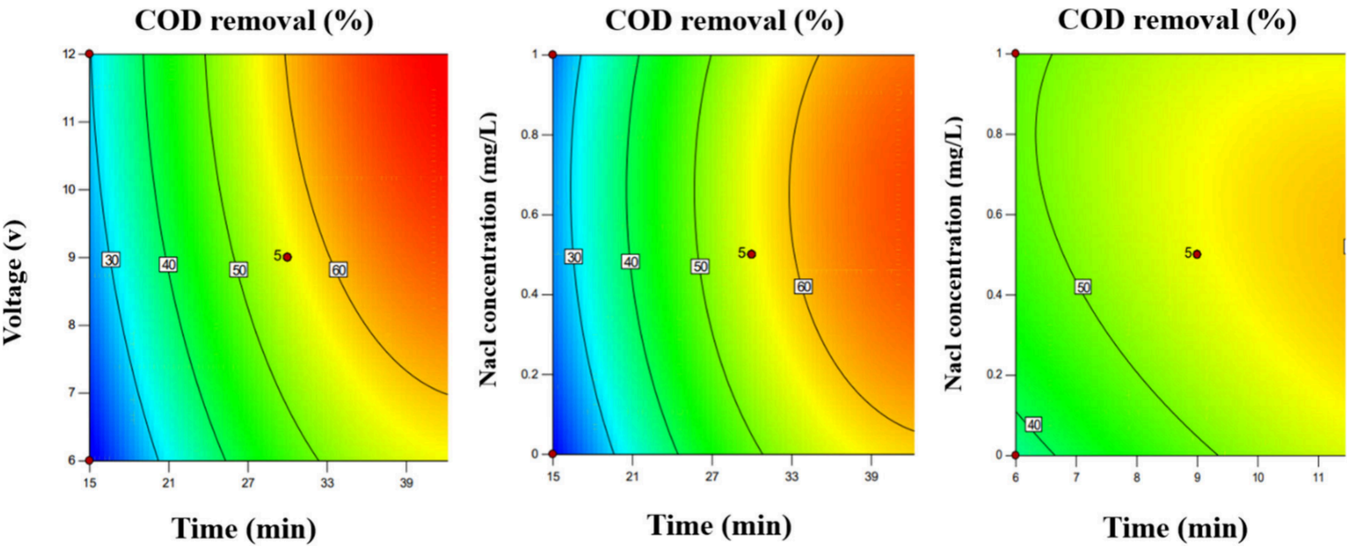

Figure 3. Interaction effects of the time, voltage, and electrolyte concentration on the EC process. Modified after Murdani et al.[9] IOP Conference Series, licensed under CC BY 3.0.

multiple variables. In contrast, AI-driven techniques, such as RSM and ANN described in detail in the Supporting Information, enable systematic optimization by modeling nonlinear relationships and predicting optimal conditions with high accuracy. These methods not only reduce the experimental workload but also enhance the treatment efficiency by identifying key operational factors that influence pollutant removal.

Murdani et al.[9] optimized hospital wastewater treatment using RSM with a Box-Behnken Design (BBD), identifying voltage (6−12 V), contact time (15−45 min), and electrolyte concentration (0−1M) as key factors in COD reduction. Optimal conditions (12 V, 45 min, and 1 M electrolyte) resulted in an 89% COD removal. BBD, with 17 runs and 5 center point repetitions, offered reliable modeling with fewer experiments than full factorial designs. A high R^2 of 0.9945 indicated excellent model fit. The quadratic model, capturing nonlinear relationships, explained 98.75% of the COD reduction variability. Predicted and actual COD removals (70.53% and 69.51%, respectively) closely matched, confirming the model reliability. On the other hand, RSM's quadratic model could not fully capture process complexity. For instance, increasing the voltage beyond 12 V or electrolyte concentration beyond 0.5 M did not enhance COD removal, indicating saturation effects. RSM also overlooked practical issues, such as electrode passivation and increased energy demand at higher voltages. These findings suggest that RSM is valuable for optimizing operational parameters but should be complemented with mechanistic studies for a more complete understanding of electrocoagulation (Figure 3).

Rumky et al.[10] investigated the impact of anode characteristics in electrochemical treatment, including material type and surface area, on the removal of COD, dissolved organic carbon (DOC), and color in wastewater treatment plants. By incorporating multiple linear regression (MLR) modeling, they identified key parameters—such as electrode spacing, system pH, reactor volume, current density, and voltage—that influenced electro-oxidation efficiency. Their findings revealed that COD and color removal were primarily dependent on reaction time, while DOC removal was strongly correlated with the reactor volume, highlighting the need for process-specific optimization. MLR modeling proved to be effective in identifying critical operational variables, aiding in the optimization of EO for enhanced performance. In a related study, Foroughi et al.[11] employed a three-dimensional electrochemical system for the treatment of tetracycline (TC)-containing wastewater, using a least-squares SVM (LS-SVM) model to predict treatment efficiency. Under optimal conditions (TC of 84 mg/L, pH 4.8, and current density of 15.72 mA/cm^2), the model predicted 90.42 ± 2.3% removal efficiency, closely matching experimental results.[11]

In contrast, Radwan et al.[12] modeled EF treatment of phenolic wastewater using a more complex 4−20−1 ANN with inputs including initial phenol concentration (50−200 mg/L), time (0−120 min), current intensity (400−900 mA), and Fe^{2+} dose (0−20 mg/L), and one output for removal efficiency (Figure 4a). Using 112 experiments (60 for training, 20 for validation, and 32 for testing), the model achieved high accuracy (R^2 = 0.9742) and low error (Se/Sy = 0.16) (Figure 4b). This model outperformed the simpler ANN used by Mirsoleimani et al.,[13] and Radwan et al.'s model benefited from a larger data set and more complex architecture, improving accuracy and error minimization.[12] The study also highlighted the effectiveness of a 5−8−1 ANN in predicting the Endosulfan removal efficiency. The model captured nonlinear relationships between parameters, such as increased removal efficiency from 66.6% to 84.57% as electrolysis time increased from 15 to 60 min, and from 74.6% to 92.6% as current density rose from 2.5 to 12 mA/cm^2. The ANN maintained high predictive accuracy even at an initial Endosulfan concentration of 50 mg/L. By learning complex patterns, the ANN significantly reduced the need for extensive experimental trials, saving time and resources. Nevertheless, the ANN did not provide insight into the underlying electrochemical mechanisms. For example, it failed to fully capture diminishing returns in removal efficiency beyond 12 mA/cm^2 or saturation effects above 50 mg/L Endosulfan, where coagulant efficiency declined despite ANN predictions suggesting continued improvement. The model's accuracy was also highly dependent on the quality and comprehensiveness of training data. Moreover, ANN optimization did not account for operational issues such as electrode passivation or sludge variability, which

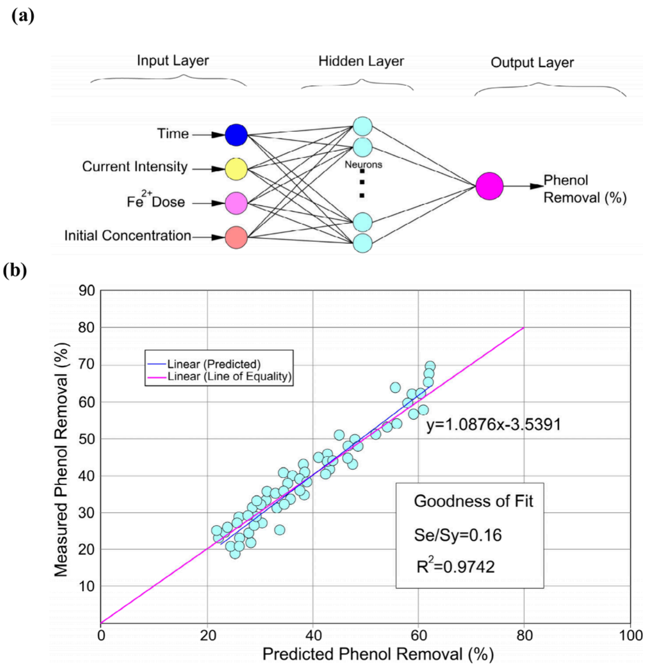

Figure 4. ANN modeling in the EF process of phenolic treatment. (a) ANN structure. (b) Measured versus predicted phenol removal percent. Modified after Radwan et al.[14] Copyright 2018 Elsevier B.V.

affect long-term performance. Despite these drawbacks, the 5–8–1 ANN proved valuable for optimizing electrocoagulation processes, offering a strong predictive performance with reduced experimental demands.

AI IN ENHANCING POLLUTANT REMOVAL EFFICIENCY

Optimizing removal efficiency is essential for improving the effectiveness and sustainability of electrochemical treatment processes. Traditional methods struggle to capture the nonlinear interactions among key parameters, limiting their ability to maximize pollutant degradation. In contrast, AI-driven techniques, such as ANN, ANFISs, and RSM, enable systematic optimization by accurately modeling the reaction dynamics and predicting optimal conditions. These approaches enhance pollutant removal by optimizing factors such as current density, pH, electrolysis time, and catalyst dosage, leading to improved coagulant generation and radical formation.

Mirsoleimani et al.[13] utilized a 5–8–1 ANN architecture to model endosulfan removal through EC with aluminum electrodes. As shown in Figure 5, their model included five input parameters: electrolysis time (0–60 min), current density (2.51–12 mA/cm^2), pH (2–10), solution conductivity (2.62–7.71 mS/cm), and initial endosulfan concentration (10–80 mg/L), with one output for removal efficiency. Their data set consisted of 70 experiments, split into 50 for training and 20 for testing, achieving a high predictive accuracy (R^2 = 0.976). However, the model's reliance on limited data and simple network architecture raises questions about its robustness for broader applications.

Mohammadi et al.[15] conducted a study that developed an improved electro-Fenton (EF) process for degrading carbamazepine (CBZ) using Fe@Fe_2O_3 nanowires as a catalyst, optimized through ANFIS combined with ant colony optimization (ACO). The ACO algorithm, applied over 200

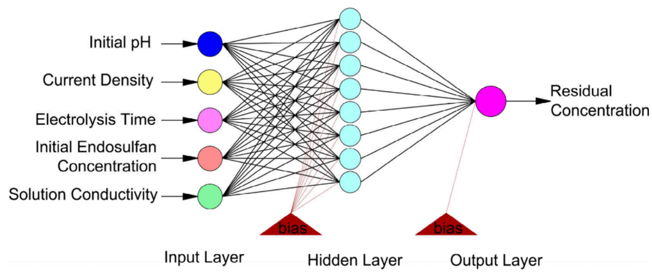

Figure 5. ANN optimized structure for the electrocoagulation process. Modified after Mirsoleimani et al.[13] Copyright 2015 American Chemical Society.

training epochs, fine-tuned the ANFIS model using Gaussian membership functions, resulting in high predictive accuracy (R^2 = 0.9988) between the predicted and experimental data. The model identified optimal conditions for 91.2% CBZ removal at pH 4, 1050 mg/L Fe@Fe_2O_3, 5.14 mA/cm^2 current density, and 45 min contact time (Figure 6). At pH 4, $^{\bullet}OH$ radical generation is maximized via H_2O_2 decomposition, while higher pH values hinder efficiency due to H_2O_2 breakdown and catalyst deactivation, in line with findings by Jiang et al.[16] The model also optimized the Fe@Fe_2O_3 dosage to ensure effective use of active sites. As shown in Figure 6a, the optimal current range (0.1–0.2 A) enhances H_2O_2 generation, Fe^{3+} reduction, and CBZ adsorption, supporting results by Panizza and Cerisola.[17] Extended contact time (20–50 min) further improves removal through greater $^{\bullet}OH$ production and Fe^{2+}–H_2O_2 interaction (Figure 6b), consistent with Mohammadi et al.[18] for ibuprofen and naproxen. The nanowires aid in iron ion release and oxygen interaction, enhancing radical formation. However, excessive nanowires may lower Fe^{2+} availability, reducing efficiency—a limitation addressed through external Fe^{2+} addition.[19] As shown in Figure 6c, CBZ concentration affects efficiency; at higher concentrations (7–15 mg/L), removal decreases due to radical competition and byproduct accumulation, aligning with Hou et al.[20] This trend emphasizes the need for balancing radical availability with pollutant load to maintain degradation performance. Overall, the study confirms the high efficacy of Fe@Fe_2O_3 nanowires, particularly when guided by intelligent modeling, for pharmaceutical pollutant removal in EF systems.

ANFIS, RSM, and ANN models were applied to optimize aluminum-based electrocoagulation (EC) for Cephalexin (CEX) removal.[21] RSM with CCD analyzed variable dependencies, while ANN and ANFIS predicted experimental outcomes, with ANFIS achieving the highest accuracy (R^2 = 0.99). Figure 7 shows the influence of key parameters (initial CEX concentration, electrolysis time, pH, and electrode type) on the removal efficiency. Electrolysis time had the greatest impact, followed by pH. Maximum removal occurred at 35 mg/L CEX and pH 7, while higher concentrations (55 mg/L) and alkaline pH reduced efficiency. Insulated electrodes enhanced isopotential regulation and maintained uniform current density, increasing Al^{3+} generation and contaminant trapping. As shown in Figure 7a and c, removal peaked at 35 mg/L and pH 7, with reduced efficiency at higher concentrations and alkaline pH. Even at 15 mg/L, acidic conditions resulted in better removal than alkaline ones. Electrolysis time and pH significantly influenced Al^{3+} production and floc formation ($Al(OH)_3$), with optimal removal at 31 min and neutral pH (Figure 7b and d). Beyond

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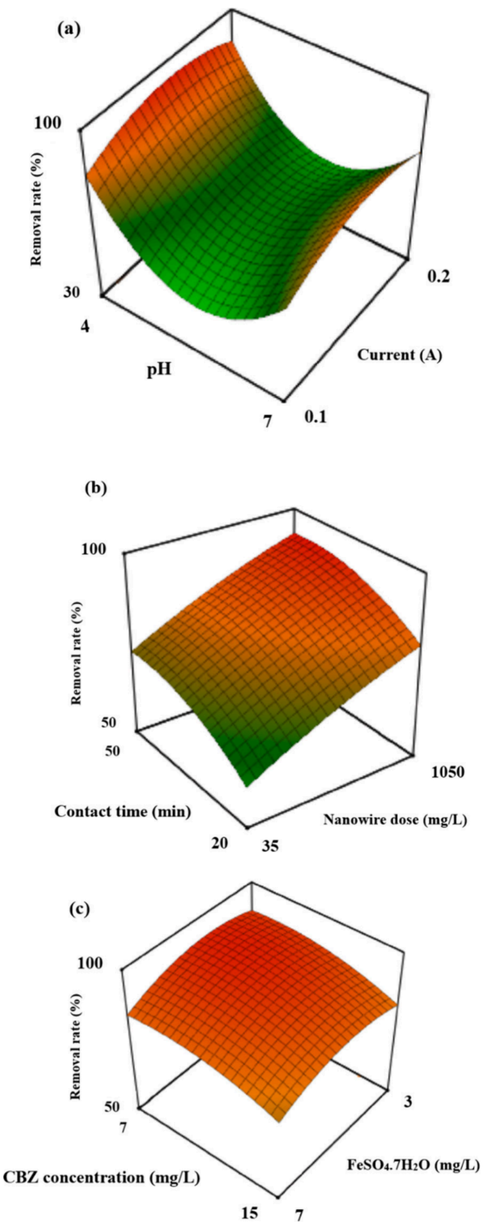

Figure 6. 3D surface plots of EF process using Fe@Fe_2O_3 nanowires for the degradation of CBZ illustrating the interaction effects between (a) pH and current, (b) contact time and nanowire Dose and (c) CBZ and $FeSO_4 \cdot 7H_2O$ concentration. Reproduced from Mohammadi et al.,[15] Copyright 2024 Elsevier B.V.

30 min, efficiency plateaued, indicating optimal treatment duration. Insulated electrodes consistently outperformed uninsulated ones by boosting OH^- generation and minimizing electron loss. Removal efficiency increased with a CEX concentration up to 35 mg/L and then reduced; similarly, it improved near neutral pH but dropped at pH 11, with insulated electrodes remaining more effective. Among the three models, ANFIS provided the most accurate predictions across 80 data points, outperforming both the RSM and ANN in modeling CEX removal.

AI-driven models such as ANN, ANFIS, and RSM allow for precise control over factors such as current density, electrolysis time, pH, and catalyst dosage, leading to higher degradation rates of contaminants. These models effectively predict pollutant removal trends and capture nonlinear interactions, enabling enhanced hydroxyl radical (·OH) production in advanced oxidation processes and optimized coagulant formation in electrocoagulation. Hybrid AI approaches, such as ANFIS-ACO, further refine optimization, achieving near-complete removal of pharmaceuticals and organic pollutants while reducing chemical and energy consumption. Additionally, AI-driven insights help mitigate process inefficiencies, such as radical scavenging, catalyst deactivation, and diminishing returns at high pollutant concentrations. Despite their advantages, these models require extensive experimental data for training and may not fully account for long-term operational challenges.

■ AI-GUIDED ELECTRODE AND CATALYST SELECTION

The effectiveness of electrochemical treatment processes largely depends on the electrode materials and catalysts, which influence reaction kinetics, pollutant degradation, and energy efficiency. Electrode composition, surface characteristics, and catalyst stability directly affect the formation of reactive species and the overall system performance. Traditionally, optimizing these components requires extensive experimentation, but AI-based models such as ANNs, SVR, and RSM now offer more efficient alternatives. These tools help predict optimal synthesis and operational conditions, enhancing material design, and improving contaminant removal and sustainability.

Yu et al.[22] explored photoelectrocatalytic degradation of norfloxacin using a Ti/SnO_2−Sb anode, which significantly enhanced total organic carbon (TOC) removal through synergistic oxidation. They employed a back-propagation ANN with parameter optimization (BP-ANN-P), using four input variables: initial norfloxacin concentration, pH, current density, and reaction time. The BP-ANN-P model outperformed conventional first-order kinetic models, achieving R^2 improvements ranging from 1.619 to 127.137 times. Even at a high norfloxacin concentration of 200 mg/L, where R^2 slightly declined due to sample size limitations (n = 86), the model still achieved R^2 = 0.969, surpassing the first-order model's R^2 = 0.922. When tested against five randomly selected data points, BP-ANN-P consistently showed a superior predictive accuracy. Comparative assessments of current density and initial pH also reinforced its higher precision, demonstrating BP-ANN-P's adaptability and robustness in predicting TOC removal across diverse operational settings.

Khan et al.[23] applied MLR, SVR, and ANN to optimize the electrochemical degradation of Navy Blue (NB) dye using Si/BDD electrodes. The key factors examined were current density, electrolyte concentration, and treatment time. All models were statistically significant (F = 99.86, p < 0.0001) with R^2, R_adj^2, and R_pred^2 > 0.90, indicating strong predictive performance. MLR identified current density as the most influential variable—dye removal increased from ∼20% to ∼ 80% as current density rose from 5 to 16 mA cm^{-2}, before plateauing due to mass transport limitations and oxygen evolution reactions. Additionally, increasing electrolyte concentration from 0.02 to 0.056 M enhanced NB removal from 60% to 88% within 30 min, due to the formation of powerful oxidants such as $SO_4^{\bullet-}$ and $S_2O_8^{2-}$, as given in eqs (S10−S12). Synergistic effects were observed when optimizing multiple parameters simultaneously.

At 15 mA cm^{-2}, 0.056 M electrolyte, pH 6.5, and 20 min, dye removal reached ∼ 90% due to enhanced $^{\bullet}$OH radical

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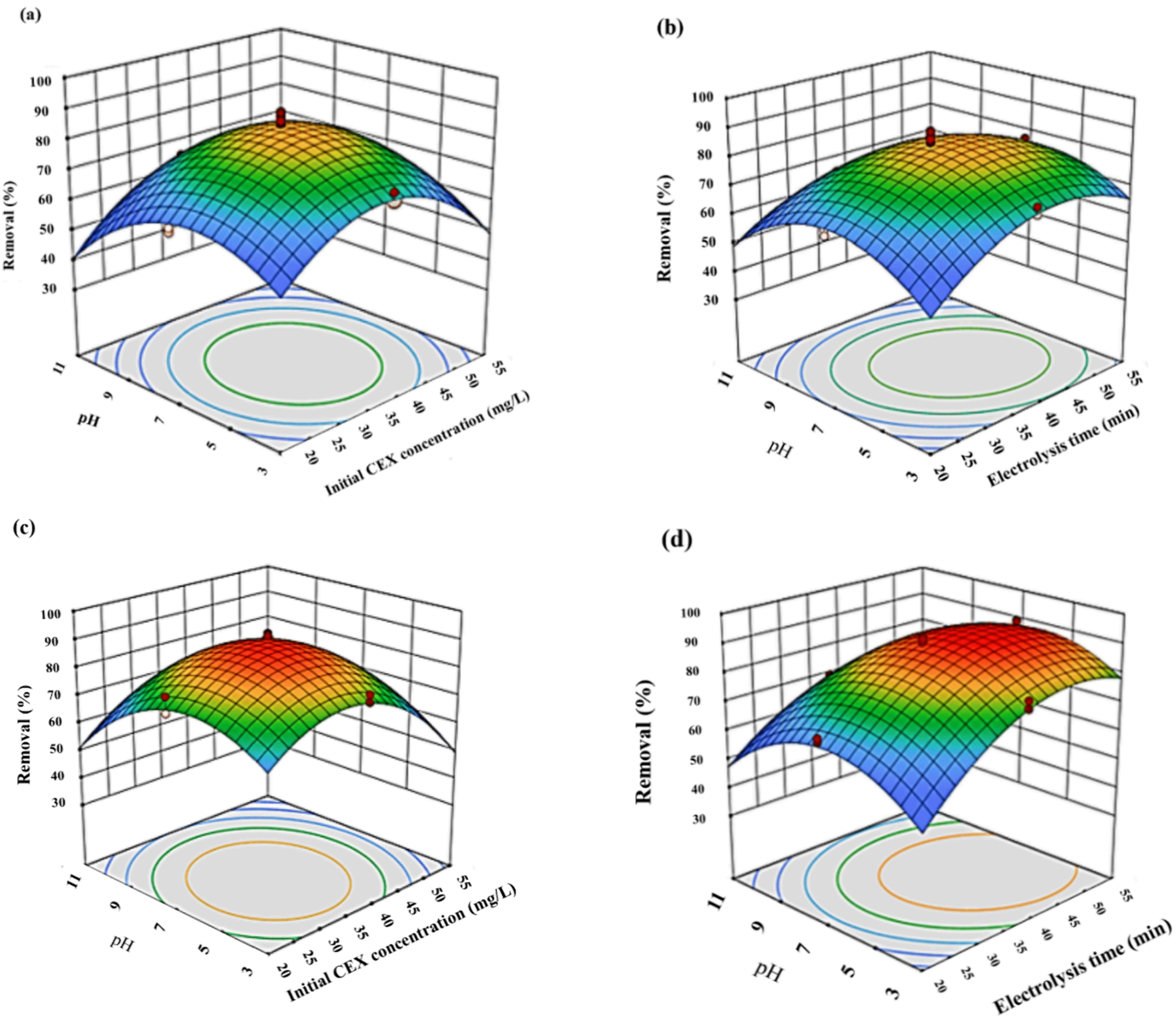

Figure 7. Response surface plot illustrating the effect of operating parameters on the CEX removal efficiency (a,b) noninsulated electrode, (c,d) insulated electrode, (a,c) interaction between pH and initial CEX concentration, (c,d) pH and electrolysis time. Reproduced from Arab et al.,[21] Elsevier, licensed under CC BY 4.0.

generation (eqs (S13 and S14)). However, efficiency declined at higher current densities due to competing reactions like oxygen evolution (eq S15) and •OH dimerization into H_2O_2 (eq (S16)). These trends aligned with Bristo et al.,[24] who reported COD removal improvements with Si/BDD and Nb/BDD electrodes from 76 to 75% to 85−83% at 30 and 60 mA cm^{-2}, respectively driven by increased •OH production via anodic water oxidation. However, excessive current led to waste reactions, reducing efficiency by recombination and •OH dimerization (eqs (S13−S16)). The rate of dye degradation increased with longer electrolysis times. This is attributed to the more extensive oxidation of organic molecules over time, consistent with findings by Melo da Silva et al.[25] Additionally, at a maximum current density of 20 mA cm^{-2}, increasing the electrolyte concentration from 0.021 to 0.056 M resulted in an improvement in dye removal efficiency from 60% to 100%. The influence of current density on dye degradation showed that increasing the current density from 8 to 20 mA cm^{-2} enhanced dye degradation from 20% to 100% at an electrolyte concentration of 0.056 M Na_2SO_4. Similarly, increasing the current density from 6 mA cm^{-2} to 16 mA cm^{-2} across all tested pH values improved NB dye removal from 30% to 80%, indicating that pH had a negligible impact on the process.

Ganthavee et al.[26] investigated a 3D electrochemical system utilizing graphite intercalation compound particle electrodes for the removal of methyl orange (MO) dye from textile wastewater. Under optimized conditions determined as current density of 15 mA/cm^2, 30 min electrolysis, and 50 mg/L initial MO concentration, the system achieved 98% removal efficiency, 3.62 kWh/kg energy consumption, and 79.53% current efficiency. To optimize process parameters, AI models including ANN, SVM, and random forests (RF) were employed. Among them, ANN exhibited the highest predictive accuracy (R^2 = 0.992), outperforming RF, SVM, and multiple regression models with the lowest error deviation. Monte Carlo simulation was also applied to assess process uncertainty, enabling robust analysis of parameter variability. ANN optimization significantly reduced the need for experimental trials, improved current efficiency (79.53%), and minimized energy loss, thereby enhancing sustainability and cost-effectiveness. However, ANN and RF are black-box models, offering limited insight into underlying mechanisms. Monte Carlo analysis, while effective, demanded substantial data sets and significant computational resources. ANN performance was also influenced by data quality and was prone to overfitting with imbalanced data sets. Although RF and SVM performed slightly less accurately, they contributed to the robustness of the overall modeling approach. A key limitation was that ANN did not consider long-term factors such as electrode passivation and side reactions, which could affect treatment efficiency over time. The study's findings indicated that while AI and ML models substantially improve process optimization, their integration with mechanistic models would enhance reliability and support better long-term system performance.[26]

A study by Mandal et al.[27] focused on synthesizing and optimizing a graphite/PbO_2 anode for landfill leachate treatment using RSM and ANN modeling. The research evaluated the effects of current intensity, $Pb(NO_3)_2$ concentration, HNO_3 concentration, and temperature on the electrode's oxidation efficiency. RSM developed a quadratic model for predicting COD removal, achieving high predictive accuracy (R^2 = 0.9632). Increasing current intensity and temperature improved performance, while excessive $Pb(NO_3)_2$ concentration reduced it. Optimal conditions (0.64 A, 0.16 mol/L $Pb(NO_3)_2$, 0.16 mol/L HNO_3, and 76.98 °C) yielded a PbO_2 anode that removed 79 ± 1.7% COD in 8 h. ANN modeling provided even higher accuracy (R = 0.99519), and sensitivity analysis identified the current intensity (47.94%) as the most influential parameter, followed by the $Pb(NO_3)_2$ concentration (20.84%), HNO_3 concentration (16.34%), and temperature (14.89%). XRD and SEM analyses confirmed that higher current promoted the formation of electrochemically superior β-PbO_2.[27]

AI-driven techniques have notably advanced electrode and catalyst design by optimizing parameters like composition, morphology, and operational conditions, improving pollutant removal and energy efficiency. These methods enhance the understanding of oxidation mechanisms and reactive species formation, such as hydroxyl and sulfate radicals. While AI aids in optimization and sensitivity analysis, it requires large data sets and lacks mechanistic interpretability, necessitating integration with electrochemical modeling for a more complete understanding of reaction pathways and electrode durability.

■ AI IN ION SEPARATION AND RESOURCE RECOVERY

Sadrzadeh et al.[28] developed a feedforward ANN model (4:6:2:1 architecture) using 81 experimental data points to optimize Pb^{2+} removal via ED, predicting separation percentage (SP) and current efficiency (CE) based on feed concentration, temperature, flow rate, and voltage (Figure 8). The multilayer perceptron (MLP) ANN, trained with the Levenberg–Marquardt algorithm, achieved high predictive accuracy (R^2 > 0.99), identifying an optimal SP of 83.22% at 1000 ppm, 30 V, and 333.15 K, and peak CE of 65.81% at 1000 ppm, 20 V, and 1.2 mL/s. The ANN effectively modeled nonlinear interactions and reduced the need for extensive experimental trials, outperforming traditional models in prediction accuracy. However, it had limitations, including a lack of explicit equations, limited interpretability, inability to model saturation effects, and high computational complexity in hidden layer optimization. Furthermore, Sadrzadeh et al. showed that increasing temperature, feed concentration, and voltage improves SP by enhancing conductivity and ion transport—higher temperatures and concentrations reduce solution resistance, while increased voltage boosts ion migration. Yet, excessive voltage can cause energy waste, membrane damage, pH shifts, and concentration polarization, ultimately hindering separation. Similarly, Min et al.[29] reported over 99% Cu and Ni removal at 12 V after 25 min, but further voltage increases led to polarization effects and metal precipitation, reducing efficiency. Conversely, higher flow rates lowered SP due to shortened ion residence time, limiting migration through membranes.

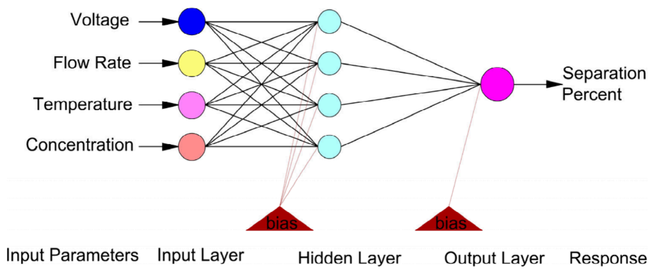

Figure 8. Structure of the ANN modeling for removing Pb^{2+} ions using ED. Modified after Sadrzadeh et al.[28] Copyright 2009 Elsevier B.V.

Zoungrana and Çakmakci[30] found that excessive flow rates decreased power density, negatively impacting separation performance in ED. Their study also showed that while increases in cell voltage and temperature initially improved CE, further increases led to energy inefficiencies and hindered ion migration. Feed concentration exhibited a nonlinear effect on CE, with diminishing returns at higher levels. The researchers emphasized the importance of balancing operational parameters—low temperature, voltage, and concentration reduced energy consumption but lower separation percentage (SP), whereas higher values improved SP but significantly increased power demand. Optimal conditions (20 V, 313.15 K, 1000 mg L^{-1} concentration, and 1.2 mL s^{-1} flow rate) achieved the best balance between SP and CE, suggesting a practical and scalable ED strategy. Under slightly more aggressive conditions (30 V, 333.15 K, 1000 mg L^{-1}), Pb^{2+} removal reached approximately 83%, confirming the effectiveness of well-optimized ED systems for wastewater treatment.

■ AI IN HYBRID ELECTROCHEMICAL TREATMENTS

The efficiency of ion separation and resource recovery in electrochemical processes depends on multiple interacting parameters such as voltage, temperature, flow rate, and feed concentration. Traditional optimization methods struggle to account for the nonlinear relationships among these factors, often requiring extensive experimental trials. AI-driven approaches, particularly ANN, have emerged as powerful tools for predicting and optimizing separation efficiency and current efficiency in ED and related processes. These models enhance ion migration predictions, reduce energy consumption, and minimize experimental workload, while improving scalability. By accurately modeling ion transport dynamics and identifying optimal operating conditions, AI-driven methods contribute to more efficient and cost-effective resource recovery from wastewater streams.

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A study by Zare et al.[31] investigated EF process for ciprofloxacin (CIP) degradation using $Fe@Fe_2O_3$ core−shell nanoparticles and a Ti/RuO_2 anode. Under optimized conditions (pH 8.83, 14.80 min reaction time, 19.19 mA/cm^2 current density, 15.13 mg/L pollutant concentration, and 199.03 mg/L catalyst dosage), they achieved 100% CIP removal and 45% TOC removal. Using RSM with CCD, they identified the current density as the most influential parameter (F = 191.90), followed by reaction time (F = 31.12), pollutant concentration (F = 25.19), and catalyst dosage (F = 5.79). The RSM model showed strong predictive capability (R^2 = 0.918) and a good fit (p > 0.05). The catalyst contributed to both adsorption and electrochemical oxidation with up to 35.89% CIP removal via adsorption at pH 5. Extended mineralization led to 70.23% TOC removal after 120 min. Biodegradability improved significantly (BOD_5/TOC from 0.68 to 0.98), and toxicity (plant stem growth inhibition) decreased from 74% to 25% post-treatment. The Ti/RuO_2 anode remained effective over 10 cycles, while $Fe@Fe_2O_3$ retained performance for four cycles before declining due to active site saturation.[31]

AI-based modeling has significantly improved the optimization of ion separation and resource recovery by providing accurate predictions of separation efficiency and current efficiency under varying operational conditions. By capturing complex nonlinear interactions among voltage, temperature, flow rate, and feed concentration, AI models enhance process efficiency while minimizing energy consumption and membrane fouling risks. These techniques facilitate precise control over ED performance, ensuring high removal rates of heavy metals and valuable ions from wastewater. However, despite their predictive accuracy, AI models, such as ANN, have limitations, including data dependency, computational complexity, and reduced interpretability compared to those of mechanistic models. Additionally, challenges such as concentration polarization, excessive energy demand at high voltages, and ion migration inefficiencies still require careful parameter balancing.

COMPARATIVE PERFORMANCE OF AI MODELS IN ELECTROCHEMICAL TREATMENT

The application of AI in electrochemical treatment has led to significant advancements in process optimization, yet the effectiveness of different AI models varies depending on system complexity, data set size, and operational conditions. Traditional models, such as RSM, provide structured statistical frameworks for parameter optimization but struggle with highly nonlinear interactions and inverse calculations. ANN offers improved predictive accuracy by capturing complex relationships, but they require extensive training data sets and suffer from interpretability challenges. Hybrid AI approaches, such as GA-ANNs (GANN) and particle swarm optimization-ANN (PSO-ANN), have further enhanced optimization by refining weight adjustments and improving model generalization. SVR and ANFIS balance interpretability and accuracy, making them suitable for specific applications. Comparing these models provides critical insights into their strengths, limitations, and suitability for electrochemical wastewater treatment, guiding the selection of the most effective approach for different treatment scenarios.

Conventional neural networks (CNNs) struggle with reverse calculations and often lack reliability when trained on small data sets for complex multivariable simulations. To overcome these limitations, Yang et al.[32] addressed the limitations of CNNs in reverse calculations and small data set reliability by integrating GA with feedforward neural networks (FFNN) for optimizing electrochemical degradation of COD and total nitrogen (TN). The GA optimized FFNN weights and biases, improving model stability, generalization, and reducing dependence on large data sets. The hybrid GANN model was validated using two DOE data sets: Taguchi Orthogonal Array (25 samples) and BBD (54 samples). GANN outperformed both FFNN and RSM, achieving 93.6% COD and 62.8% TN removal thanks to its ability to bypass predefined search constraints. While traditional NNs capture nonlinearity, they struggle with inverse prediction and suffer from instability with limited data due to random weight initialization. RSM, although useful, assumes quadratic relations, limiting its precision in complex systems. GANN demonstrated high predictive accuracy (R^2 = 0.946 for COD, 0.874 for TN) but required significant computational effort and tuning. Sensitivity analysis identified the chloride ion concentration, current density, and initial pH as key factors in COD removal, while the electrolyte concentration was most critical for TN degradation. Chloride ions enhanced reactive chlorine species formation and solution conductivity, while hydroxyl radicals ($^{\bullet}OH$), more effective in acidic conditions (2.85 V vs 2.02 V in alkaline), played a major role in COD oxidation.

Phan et al.[33] showed that lower pH significantly improves COD removal, achieving 39.57−83.1% at pH 5, compared to 39.56−83.00% at pH 7.45 and just 6.10−50.68% at pH 10. This confirms acidic to neutral conditions (pH 3−8) represented by eq S17 in the SI favor HOCl formation—a stronger oxidant than ClO^-, which dominates above pH 8 (eq (S18))—thus enhancing COD degradation. The study also reinforced that GANN provides higher predictive accuracy for COD removal than TN, as its optimization was primarily COD-focused. Integrating GA with GANN significantly improved performance (R^2 = 0.946, RMSE = 0.022, MAE = 0.019, SSE = 0.027, F = 1.48, p = 0.49), outperforming traditional neural networks (R^2 = 0.863, RMSE = 0.042, MAE = 0.034, SSE = 0.095, F = 0.84, p = 0.34) using Data set-β. GANN achieved a maximum COD removal of 93.6%, compared to 90.2% (RSM), 89.8% (GANN on Data set-α), and 82.2% (Taguchi OA). For TN, it reached 62.8% removal, outperforming RSM (58.5%), GANN on Data set-α (57.7%), and Taguchi OA (54.7%). GANN also converged faster (200 vs 400 epochs for FFNN) with lower training MSE. Notably, GANN trained on smaller Data set-α performed similarly to RSM trained on the larger Data set-β (R^2 = 0.85−0.90 for COD). Sensitivity analysis confirmed that chloride ion concentration, current density, and initial pH were most influential for COD removal, while electrolyte concentration had the greatest effect on TN. The GA-GANN integration enhanced prediction accuracy, reduced data needs, and increased efficiency, making it a powerful tool for optimizing electrochemical wastewater treatment.[33]

Gholami Shirkoohi et al.[34] optimized SVR and ANN models using GA and particle swarm optimization (PSO) to predict phosphate removal efficiency in EC (Figure 9). They used Monte Carlo cross-validation with 10 subsets (62 data points each), splitting them into 42 training, 10 validation, and 10 testing sets. Among all models, the PSO-ANN model achieved the best performance with R^2 = 0.981, MSE = 7.201, and MAPE = 2.022, outperforming other combinations. AI-driven optimization indicated that phosphate removal improves at lower pH and lower initial phosphate concentrations, while

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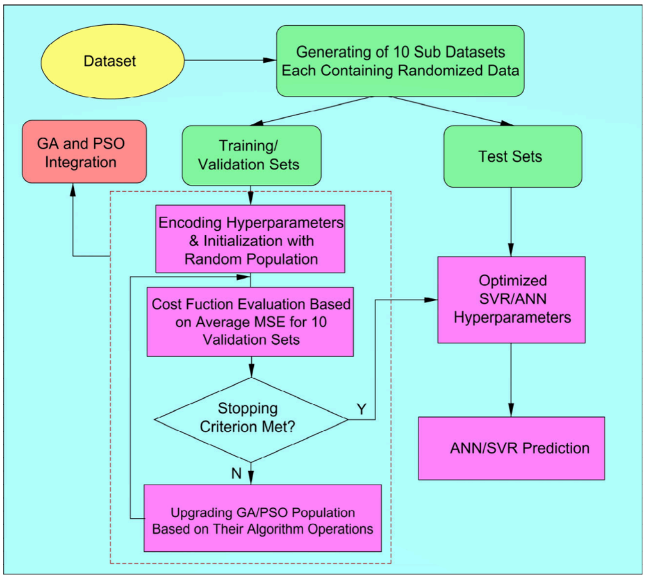

Figure 9. Flowchart of the GA and PSO approaches for predicting phosphate removal efficiency from wastewater using the EC process. Modified after Gholami Shirkoohi et al.,[34] Copyright 2022 Elsevier B.V.

higher current intensities and longer treatment times enhance the removal efficiency. Mechanistically, at low pH, Al^{3+} and Fe^{2+} ions formed $Al(OH)_3$ and $Fe(OH)_2$, which precipitated phosphate as $AlPO_4$ and $FePO_4$. Acidic conditions also promoted Al^{3+} and Fe^{3+} precipitation, whereas high pH levels led to the formation of soluble species like $Al(OH)_4^-$ and $Fe(OH)_4^-$, reducing coagulation effectiveness. Increasing the current intensity accelerated electrode dissolution, generating more coagulants, while longer treatment durations support floc formation and sedimentation. However, AI-based models have limitations. They depend on data rather than on chemical or physical mechanisms. ANN models require large data sets, and SVR models are sensitive to hyper-parameter selection, which limits their adaptability to real wastewater, electrode passivation, and pH variability. Moreover, AI models generally ignore side effects, such as electrode fouling, gas evolution, and secondary reactions, necessitating experimental validation. Additionally, the computational time required was high (GA-ANN: 4897s, PSO-ANN: 4851s), posing challenges for real-time industrial implementation. The study concluded that while AI models significantly improve predictive accuracy and process optimization, their integration with mechanistic models is essential for enhancing reliability and enabling practical applications in wastewater treatment.[34]

Igwegbe et al.[35] compared RSM, ANN, and ANFIS for modeling and optimizing electrocoagulation-flocculation (ECF) in aquaculture effluent treatment using aluminum electrodes. All three models enhanced process understanding, prediction accuracy, and efficiency while reducing experimental costs. RSM offered clear mathematical relationships for parameter interactions but was limited by its assumption of quadratic behavior. ANN could model complex nonlinear interactions with higher accuracy than RSM but required large data sets and significant computational resources. ANFIS, which integrates ANN and fuzzy logic, achieved the highest accuracy (R^2 = 0.9990), outperforming ANN (R^2 = 0.9807) and RSM (R^2 = 0.9790). When combined with GA optimization, ANFIS-GA achieved 98.98% turbidity removal, exceeding the turbidity removal of ANN-GA (97.81%) and RSM (96.01%). Optimal ANFIS-GA conditions were pH 4, 3 A current, 7.2 min electrolysis, 23 min settling time, and 43.8 °C, with pH (81%) and current (11.8%) identified as the most influential variables. While ANFIS offered superior predictive performance, it required expert tuning of fuzzy membership functions and was computationally intensive. RSM was less effective at extreme parameter values, and the ANN, despite its accuracy, lacked transparency. The study highlighted the trade-offs between accuracy, interpretability, and complexity in selecting models for ECF process optimization.[35]

Khan et al.[36] optimized electrochemical oxidation for Synozol Red dye removal using both RSM and ANN. A filter press flow cell equipped with a $Ti/RuO_{0.3}TiO_{0.7}O_2$ anode and a stainless steel cathode was employed. ANN, trained via back-propagation and gradient descent, featured four input neurons, 15 hidden neurons, and one output node (color removal efficiency), achieving a high accuracy (R^2 = 0.99). The data set was divided into 70% training, 15% testing, and 15% validation (Figure 10). RSM yielded a slightly lower R^2 = 0.954. ANN

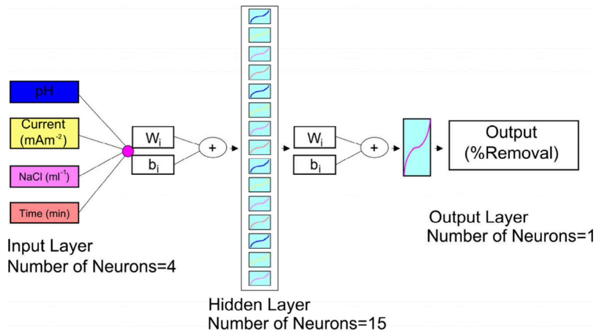

Figure 10. ANN modeling and three-layer feed forward model on the decolorization efficiency of Synozol Red dye. Modified after Khan et al.[36] Copyright 2020 Elsevier B.V.

effectively captured nonlinear relationships and predicted optimal conditions: pH 2.95, current density 5.95 mA/cm^2, NaCl concentration 0.075 M, and electrolysis time 29.83 min, achieving 98.6% dye removal, compared to RSM's 97.7% under similar conditions. RSM provided interpretability, showing pH as the most influential factor (61.03%), followed by current density (17.29%), NaCl (12.7%), and electrolysis time (8.98%). However, its quadratic assumption limited detection of nonlinear behaviors such as a plateau in NaCl efficiency beyond 0.08 M. ANN, while more accurate, lacked transparency and required higher computational effort. Sensitivity analysis via Pareto (RSM) and Garson (ANN) methods both identified pH as the dominant parameter (Figure 11a, b). Experimental validation confirmed 97.1% removal at pH 3, current density of 5.88 mA/cm^2, NaCl of 0.08 M, and 29.5 min electrolysis time. 3D surface plots showed enhanced degradation at pH 3. The Ti/RuO_2-TiO_2 anode enabled Cl^- oxidation, producing Cl_2, HOCl, and ClO^-, with HOCl (1.49 V vs SHE) prevailing at pH < 7.5. Increasing pH converted HOCl to ClO^- (0.89 V), and eventually to less effective oxidants like ClO_3^- and ClO_4^-, reducing treatment efficiency (eqs S19−S24).[37] Lowering pH from 5 to 3 increased removal from 40% to >95% at 10 mA/cm^2, and from 25% to 85% with 0.1 M NaCl. High pH hindered removal due to dye adsorption blocking the electrode sites. Similar trends were reported by Xia et al.[38] using PbO_2

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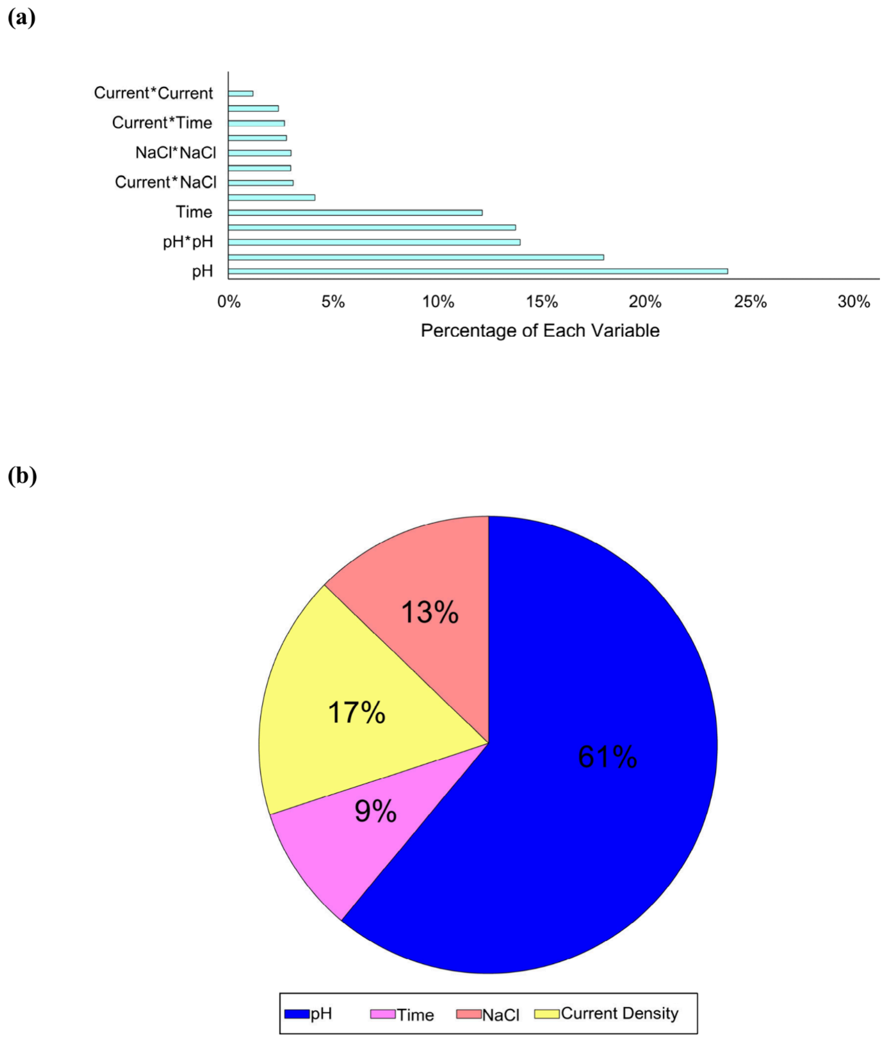

Figure 11. (a) Pareto chart and (b) sensitivity analysis of the relative importance of RSM and ANN input variables on the decolorization efficiency of the Synozol Red dye. Modified after Khan et al.[36] Copyright 2020 Elsevier B.V.

electrodes. Higher current density (0.178−9.8 mA/cm^2) improved removal, with 100% degradation at pH 3 and 60% at 0.1 M NaCl. Electrolysis time up to 35 min enhanced removal from 20% to 70%, supported by da Silva et al.[25] An optimal current density of 5.88 mA/cm^2 balanced the efficiency and energy use. Increasing NaCl from 0.0017 to 0.1 M at pH 3 improved removal from 35% to 95%. At 8 mA/cm^2, degradation rose from 20% to 60%, attributed to greater Cl^- availability, improved conductivity, and lower energy demand. Reactive species (Cl_2, HOCl, ClO^-) drove dye degradation, consistent with findings by Santos et al.[39]

Shirkoohi et al.[40] compared adaptive neuro-fuzzy inference system (ANFIS) and central composite design (CCD) for predicting caffeine removal efficiency and energy consumption in an electrochemical oxidation (EO) process. ANFIS, built with five layers and trained using Fuzzy C-Means clustering to avoid overfitting, used 75% of the data for training and 12.5% for validation and testing. CCD used a 40-run matrix to assess the influence of electrolysis time, current intensity, initial caffeine concentration, and anode type. ANFIS outperformed CCD with a higher predictive accuracy: R^2 = 0.993 for caffeine removal and R^2 = 0.976 for energy consumption, predicting optimal values of 95.24% removal and 0.91 Wh/mg energy use. While CCD provided interpretable statistical models and insights into variable interactions, its quadratic assumptions limited its ability to model nonlinear saturation effects, particularly at high currents (>0.7 A). ANFIS effectively modeled these nonlinearities but required greater computational resources and lacked transparency in mechanistic interpretation. As shown in Figure 12a and b, RSM (CCD-based) also performed well (R^2 = 0.986 for removal, R^2 = 0.939 for energy use), but ANFIS was superior, with RMSE = 2.694 for removal and 0.261 for energy. Surface and parity plots confirmed ANFIS's better fit to experimental data, especially for energy modeling. Among the tested variables, the anode type had the greatest impact on degradation (81%), followed by electrolysis time (11.8%). Boron-doped diamond (BDD) was the most effective anode, outperforming IrO_2, graphite, and Pt, due to its high oxygen evolution potential and efficient $^{\bullet}OH$ radical generation. These results were consistent with previous findings by Cotillas et al.,[41] Indermuhle et al.,[42] and de Vidales et al.[43] Caffeine removal improved with longer electrolysis time and higher current but decreased with higher initial caffeine concentration. Using BDD, removal rose from 40% to 100% as time increased from 10 to 50 min and current from 1 to 2 A. IrO_2 did not exceed 35% removal under the same conditions. Current intensity significantly influenced electron transfer and the generation of reactive oxygen species

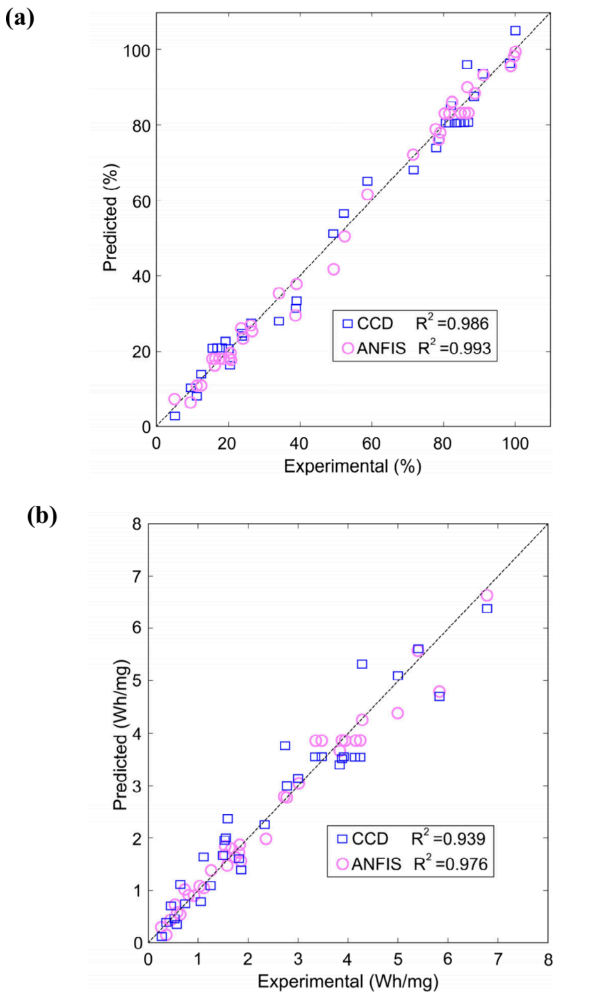

Figure 12. Surface plots of the experimental and predicted values for ANFIS and CCD models: (a) caffeine removal efficiency (%) and (b) energy consumption (Wh/mg). Modified after Gholami Shirkoohi et al.[40] Copyright 2022 Elsevier B.V.

(ROS) like $^{\bullet}OH$, $SO_4^{\bullet-}$, and HOCl.[44] For instance, Periyasamy et al.[45] also demonstrated complete florfenicol degradation in 150 min at 250 mA with BDD, emphasizing the role of current in generating secondary oxidants such as $S_2O_8^{2-}$ and $SO_4^{\bullet-}$ in sulfate-rich media (eqs S10−S12, S25).[46] Under RSM-optimized conditions—39 min electrolysis, 0.7 A current, 13 mg/L caffeine concentration, and BDD anode—93.82 ± 0.80% removal and 0.70 ± 0.02 Wh/mg energy consumption were achieved, accurately predicted by ANFIS. While RSM provided a simpler and interpretable model, ANFIS demonstrated greater precision and adaptability for modeling complex, nonlinear EO processes.

RSM remains a widely used classical statistical approach that facilitates exploration of relationships between process variables and outcomes, aiding in the identification of optimal conditions.[47] However, its effectiveness declines when applied to complex, nonlinear processes, as it relies on low-order polynomial models, which may not adequately capture intricate variable interactions.[8] In contrast, ANNs are highly capable of modeling complex chemical processes due to their ability to learn nonlinear relationships without prior assumptions about data structure. Nevertheless, ANNs are often criticized as black-box models due to their lack of interpretability, which can hinder mechanistic understanding and transparency.[48] GAs, inspired by natural selection, offer robust global optimization capabilities, effectively avoiding local minima in multidimensional search spaces. However, their computational demands increase significantly with the problem size. Other metaheuristic algorithms, such as particle swarm optimization (PSO), provide additional adaptive and nature-inspired optimization strategies, serving as valuable complements to traditional RSM and ANN models.[49] These metaheuristics enhance the optimization of electrochemical processes by addressing different challenges in efficiency, complexity, and problem specificity. Further investigation is needed into alternative experimental designs beyond CCD and BBD in RSM applications, as most studies rely on these two. Additional research is also required to evaluate RSM's feasibility for optimizing electrochemical water and wastewater treatment processes in real-world scenarios. ANN-based models offer high predictive accuracy but require large data sets and substantial computational power, limiting their practicality for real-time applications without sufficient training data. Hybrid models like GANN and PSO-ANN enhance optimization by improving parameter selection, reducing overfitting, and increasing model stability but at the cost of higher computational demands. RSM is still valuable for analyzing parameter interactions under quadratic assumptions, although it lacks flexibility for complex electrochemical systems. ANFIS and SVR strike a balance between accuracy and interpretability, making them valuable when a clear understanding of variable interactions is necessary for effective process control. Sensitivity analyses across AI models consistently identify the current density, pH, electrolyte concentration, and treatment time as critical factors influencing pollutant degradation. Despite the advancements brought by AI in improving process efficiency, challenges remain in model transparency, adaptability to real-time scenarios, and integration with fundamental mechanistic studies.

THE ROLE OF ELECTROCHEMICAL DOUBLE LAYER IN AI MODELING OF ELECTROCHEMICAL PROCESSES

The electrochemical double layer (EDL) plays a fundamental role in governing heterogeneous electrochemical reactions, particularly in the EC and advanced oxidation processes (AOPs). The EDL consists of a compact Helmholtz layer and a diffuse layer, which influence charge transfer, mass transport, and interfacial reaction kinetics. In heterogeneous reactions, the EDL modulates the adsorption of reactants onto the electrode surface, the availability of charge carriers, and the stability of intermediates, ultimately affecting the reaction efficiency.

The EDL constitutes a foundational concept in electrochemistry, playing a crucial role in electrochemical water treatment technologies. Comprising the compact Helmholtz layer and the diffuse layer, the EDL governs charge transfer, mass transport, and reaction kinetics at the electrode−electrolyte interface. These processes are critical for the efficiency of electrocatalytic reactions, particularly in environmental remediation and water treatment.[50−52] In oxidative electrochemical technologies such as electrochemical advanced oxidation processes (EAOPs), the EDL influences the formation of reactive oxygen species (ROS), pollutant adsorption, and the stability of electrogenerated intermediates. Understanding these interactions is essential for optimizing electrochemical water treatment processes, and artificial

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intelligence (AI) has emerged as a powerful tool for process modeling, parameter optimization, and predictive control.[52,53]

EDL IN ELECTROCOAGULATION PROCESS

The efficiency of the EC is critically dependent on the EDL, which governs the behavior of coagulant formation, particle interactions, and flocculation dynamics. Within the EDL, charge neutralization and coagulation occur as metal hydroxide species such as $Al(OH)_3$ and $Fe(OH)_3$ interact with suspended particles, leading to enhanced coagulation and pollutant aggregation. Additionally, electrode reactions and coagulant production are regulated by the oxidation of sacrificial anodes, which release Al^{3+} or Fe^{2+}/Fe^{3+} ions that hydrolyze into active coagulants. The compact Helmholtz layer plays a crucial role in floc aggregation, influencing particle attachment efficiency and floc size distribution.[54] The integration of AI in the EC process has revolutionized process optimization by enabling real-time monitoring, automation, and adaptive control, significantly improving pollutant removal efficiency while reducing operational costs. AI-powered machine learning (ML) models facilitate electrode material selection, identifying optimal anode compositions that enhance coagulant generation while minimizing the extent of corrosion. Additionally, AI-driven process control dynamically regulates the current density and voltage in response to fluctuations in water quality, ensuring efficient coagulant production while minimizing energy consumption. The application of predictive flocculation modeling allows AI algorithms to analyze particle aggregation trends, optimizing coagulant dosage and pH levels to maximize floc stability and improve sedimentation rates. Furthermore, AI-enhanced diagnostics can detect electrode passivation, fouling, and system inefficiencies, enabling predictive maintenance strategies that extend the operational lifespan of electrocoagulation units. By integrating AI-driven electrocoagulation with advanced oxidation processes (AOPs) and filtration systems, overall water treatment performance is enhanced, leading to more sustainable and cost-effective wastewater remediation strategies.[54]

EDL IN THE ELECTRO-FENTON PROCESS

The efficiency of the electro-Fenton (EF) process is significantly influenced by the structure and dynamics of the EDL. Within the EDL, charge interactions at the electrode interface affect the diffusion of Fe^{2+} and Fe^{3+} ions, modulating their availability for continuous radical production. Optimizing EDL properties enhances Fe^{2+} regeneration, preventing catalyst deactivation, and improving overall process stability. Additionally, the electrogeneration of H_2O_2 is heavily influenced by the EDL, as it determines the transport of oxygen molecules to the cathode surface, influencing their electrochemical reduction into hydrogen peroxide. AI-driven simulations optimize electrolyte composition, applied potential, and electrode configurations to maximize the H_2O_2 yield while minimizing energy losses. Furthermore, the compact Helmholtz layer impacts the adsorption of organic pollutants and iron species onto electrode surfaces, affecting reaction kinetics and pollutant degradation rates.[55]

AI-assisted modeling predicts optimal electrode surface modifications, enhancing the adsorption efficiency and radical generation. The integration of AI into the EF process enables real-time control and dynamic system optimization, significantly improving pollutant degradation efficiency while minimizing the reagent consumption and energy costs. AI algorithms dynamically adjust voltage, current density, and reaction conditions to maintain consistent H_2O_2 electrogeneration, preventing process inefficiencies and excessive reagent consumption. ML models optimize iron catalyst regeneration and dosing, ensuring proper Fe^{2+} levels while minimizing Fe^{3+} accumulation and sludge formation, which are common issues in EF systems. AI-assisted process control fine-tunes electrode potential and operational parameters, achieving an optimal balance between high oxidation efficiency and reduced energy input. Additionally, AI-based monitoring systems analyze real-time water quality data, dynamically adjusting pH, ionic strength, and reaction conditions to enhance pollutant degradation performance. AI-driven diagnostics track electrode degradation and iron precipitation trends, enabling predictive maintenance strategies that extend system longevity, improve efficiency, and reduce operational downtime. By incorporating AI, electro-Fenton systems become more adaptable, cost-effective, and sustainable, making them highly suitable for large-scale wastewater treatment applications.[55]

EDL IN INDIRECT ELECTROCHEMICAL OXIDATION

Indirect electrochemical oxidation (IEO) is an advanced water treatment process that utilizes electrogenerated oxidants, such as ozone (O_3), hydrogen peroxide (H_2O_2), and ferrate (Fe(VI)), to degrade pollutants. The efficiency of these oxidants is directly influenced by their formation mechanisms, stability, and reactivity, all of which are governed by electrochemical double layer (EDL) dynamics at the electrode–electrolyte interface. In ozone (O_3) formation, O_3 is produced at the anode via the oxidation of water molecules, with the EDL controlling the availability of hydroxyl ions and oxygen species that participate in ozone generation.[56] AI-driven predictive models optimize electrode material selection—such as BDD—and fine-tune voltage application to enhance ozone production while minimizing parasitic side reactions that reduce the process efficiency. In hydrogen peroxide (H_2O_2) generation, H_2O_2 is synthesized through the two-electron oxygen reduction reaction (ORR) at the cathode, where the EDL regulates proton and oxygen transport across the interface, impacting H_2O_2 yields and selectivity. ML algorithms can predict optimal pH conditions, applied potentials, and current densities, ensuring that H_2O_2 formation is maximized over competing water reduction reactions. Additionally, ferrate (Fe(VI)) electrogeneration is a promising IEO technique that involves anodic oxidation of Fe(III) to Fe(VI), a highly reactive oxidant with a strong degradation potential. The EDL modulates charge transfer kinetics and iron speciation, affecting the ferrate stability and efficiency. AI-based process optimization dynamically adjusts anodic conditions, preventing Fe(VI) degradation while reducing unwanted Fe(III)/Fe(II) cycling, which can lower the oxidation efficiency. The integration of AI into IEO processes enables real-time monitoring, adaptive process control, and predictive analytics, significantly improving oxidant generation efficiency, pollutant degradation rates, and overall energy utilization.[56]

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■ EDL IN ELECTRO-CATALYSIS AND ELECTROCHEMICAL OXIDATION

Electrocatalysis is a key technology in wastewater treatment, accelerating oxidation reactions at the electrode surface to achieve efficient pollutant degradation. The performance of EO processes is highly dependent on the EDL, which governs charge transfer kinetics, reactive oxygen species (ROS) formation, and interactions of the pollutant with electrode materials. The EDL plays a crucial role in charge transport and ROS generation, as it modulates the movement of reactive species near the electrode interface, directly influencing the formation of hydroxyl radicals (•OH) and other oxidative agents. Optimizing EDL conditions enhances radical availability, increasing pollutant mineralization efficiency and preventing undesired side reactions. Additionally, electrode material interactions are strongly influenced by the EDL, affecting the adsorption and desorption of contaminants on the electrode surface.[57] AI-assisted computational modeling enables the prediction of optimal electrode materials, such as BDD and mixed metal oxides (MMOs), to enhance the oxidation efficiency and electrode durability. Furthermore, mass transport and reaction kinetics are dictated by the compact Helmholtz layer, which influences the diffusion of pollutants to reactive sites. AI-driven multiphysics simulations optimize operational parameters, balancing mass transport limitations and charge transfer efficiency to ensure high treatment performance. The integration of AI into electrocatalysis allows for real-time process control, predictive modeling, and automated optimization, significantly improving the system efficiency and energy utilization. ML models analyze the electrode composition and surface properties, identifying materials with the highest catalytic activity and long-term stability. AI-driven adaptive control systems dynamically regulate the current density and applied voltage, optimizing ROS production while minimizing side reactions that could reduce process efficiency. Advanced AI-based simulations predict oxidation kinetics under fluctuating water quality conditions, allowing for real-time process adjustments to maintain consistent pollutant degradation. Additionally, AI optimizes power consumption by fine-tuning applied potential and reaction rates, ensuring a cost-effective operation with minimal energy waste. AI-based monitoring systems detect electrode degradation trends, enabling predictive maintenance strategies that extend the electrode lifespan and operational stability. Through these advancements, AI-enhanced electrocatalysis achieves higher degradation rates, improved energy efficiency, and greater long-term sustainability, making it an ideal technology for large-scale wastewater treatment applications.[57]

■ EDL IN INTEGRATED ELECTROCHEMICAL WATER TREATMENT PROCESSES

Integrating electrochemical advanced oxidation processes (EAOPs) with complementary treatment methods, such as membrane filtration and biological degradation, significantly enhances the pollutant removal efficiency, process stability, and overall system performance. The EDL plays a fundamental role in these hybrid treatment systems, governing charge transport, ion migration, and pollutant degradation kinetics, which are critical for optimizing system synergy. Within electrochemical oxidation and biodegradability enhancement, EAOPs generate hydroxyl radicals (•OH) and other reactive species that break down complex organic contaminants into simpler, more biodegradable intermediates. The EDL influences intermediate adsorption, oxidation efficiency, and transformation pathways, ensuring that degraded pollutants are efficiently converted to bioavailable forms for subsequent biological treatment. In membrane-electrochemical hybrid systems, the EDL regulates ion migration and fouling tendencies, which are major challenges in electrocoagulation-membrane filtration hybrids. AI-driven predictive modeling helps forecast scaling risks, optimize membrane surface charge interactions, and improve membrane lifespan, reducing operational downtime and maintenance costs. Additionally, the electrochemical removal of perfluorinated compounds (PFCs), which are persistent and difficult to degrade, is significantly influenced by the EDL's regulation of charge interactions during oxidation reactions. On BDD anodes, AI-assisted process control fine-tunes electrode potential and electrolyte composition, maximizing PFC degradation while minimizing energy consumption. The integration of AI-driven models into these hybrid systems further enhances the synergy among electrochemical oxidation, biological treatment, and membrane filtration, resulting in more energy-efficient and cost-effective solutions. AI-based dynamic process control adjusts operational parameters in real time, ensuring optimal pollutant breakdown, charge transport efficiency, and system adaptability across different treatment stages. Predictive membrane fouling mitigation utilizes AI diagnostics to forecast scaling and biofouling risks, allowing for optimized membrane cleaning cycles and an extended operational lifespan. Additionally, AI-powered oxidation pathway prediction refines reaction kinetics analysis, determining ideal oxidation conditions that maximize the level of pollutant degradation while preventing excessive byproduct formation. AI-driven energy optimization algorithms regulate power distribution across treatment trains, ensuring minimal energy wastage while maximizing pollutant removal efficiency. Furthermore, AI-based adaptive response mechanisms dynamically adjust treatment conditions based on real-time influent water quality data, ensuring consistent system performance under varying wastewater compositions. Through these AI-enhanced hybrid treatment strategies, electrochemical wastewater treatment becomes more efficient, sustainable, and adaptable, making it a viable large-scale solution for industrial and municipal applications.[58]

■ AI-DRIVEN OPTIMIZATION FOR HYBRID AND SEQUENTIAL ELECTROCHEMICAL PROCESSES

Artificial intelligence plays a crucial role in enhancing hybrid and sequential electrochemical treatments by enabling real-time process adjustments, predictive modeling, and optimization of operational parameters. One key AI-driven strategy is dynamic process control, where ML algorithms continuously adjust the current density, pH, and electrode spacing to maintain optimal EDL conditions, ensuring efficient charge transfer and pollutant aggregation. Additionally, AI-based simulations aid in electrode material selection, identifying the most suitable compositions for hybrid treatments while balancing factors, such as conductivity, oxidation potential, stability, and cost-effectiveness. AI also optimizes sequential treatment steps, determining the most effective order of electrocoagulation, electrochemical oxidation, and advanced oxidation processes to maximize contaminant removal, while minimizing energy consumption and operational costs. Furthermore, predictive maintenance algorithms analyze

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system performance trends, anticipate electrode passivation, and suggest maintenance schedules, reducing downtime and extending the equipment lifespan. Another major advantage of AI integration is energy efficiency enhancement, where AI-driven optimization minimizes power consumption while maintaining high degradation efficiency, making these treatments more cost-effective and scalable for industrial applications. By incorporation of AI into hybrid and sequential electrochemical treatments, superior pollutant degradation rates, enhanced energy efficiency, and lower operational expenses can be achieved. These advancements significantly improve the feasibility of electrochemical water treatment systems for large-scale applications, particularly in addressing complex wastewater streams containing persistent and biorefractory contaminants.[59]

AI INTEGRATION IN ELECTROCHEMICAL WATER TREATMENT TECHNOLOGIES

Modular, decentralized electrochemical water treatment technologies are highly significant due to their adaptability, energy efficiency, and effectiveness in contaminant removal. These systems offer flexibility in deployment, making them suitable for both urban and remote areas while minimizing energy consumption and operational costs. Their ability to integrate with renewable energy sources and smart monitoring systems further enhances sustainability and real-time process optimization, ensuring efficient pollutant removal across diverse water treatment applications. AI-enhanced electrochemical models improve the predictive accuracy of the oxidation efficiency under various operating conditions, leading to more sustainable and cost-effective electrochemical treatments. AI-based models play a crucial role in multiple electrochemical water treatment techniques by facilitating parameter tuning, predictive analytics, and adaptive process control.[59]

In the EC process, AI models simulate EDL dynamics, predicting optimal electrode material, current density, and ionic strength required for efficient charge neutralization and floc formation. Reinforcement learning techniques dynamically adjust operating parameters, ensuring long-term system efficiency while mitigating electrode passivation.[60] In electroflotation, AI-powered computational fluid dynamics (CFD) simulations optimize bubble size distribution, surface charge interactions, and hydrodynamic flow, maximizing the pollutant flotation efficiency. ML models refine electrode spacing and gas production rates, reducing energy consumption while maintaining high pollutant separation performance.[39] For indirect electrochemical oxidation, AI-driven process modeling optimizes electrode material selection and reaction kinetics, improving the generation of reactive oxygen species (ROS) such as hydroxyl radicals, ozone, and ferrate. AI-based adaptive control systems fine-tune voltage application and electrolyte composition, balancing oxidation efficiency with energy demands.[17] In electro-Fenton and photo-Fenton processes, AI-enhanced reaction simulations predict iron cycling dynamics (Fe^{2+}/Fe^{3+} regeneration), optimize hydrogen peroxide dosing, and enhance hydroxyl radical production, ensuring high pollutant degradation rates while reducing unnecessary chemical consumption. Adaptive AI models control UV/solar irradiation intensity, dynamically adjusting radical formation in response to real-time environmental conditions to optimize degradation efficiency.[55] Additionally, in photoelectrocatalysis, AI optimizes photoanode material selection, such as titanium dioxide (TiO_2), tungsten trioxide (WO_3), and zinc oxide (ZnO), while fine-tuning bias potential applications to enhance electron−hole separation efficiency and reduce recombination losses. Through ML integration, solar-driven electrochemical systems dynamically adjust light intensity and reaction kinetics, ensuring continuous pollutant degradation while minimizing operational costs. By leveraging AI-assisted predictive modeling, real-time control, and adaptive learning, electrochemical water treatment systems become more efficient, cost-effective, and scalable, supporting the transition to sustainable and decentralized water treatment solutions.[61]

ADVANTAGES AND DISADVANTAGES OF AI IN ELECTROCHEMICAL PROCESSES

AI has significantly improved electrochemical water treatment processes by enhancing process control, predictive modeling, and system efficiency; however, its implementation also presents challenges. Among its advantages, AI enables real-time optimization, dynamically adjusting parameters such as current density, pH, and electrolyte composition to improve the process efficiency. It also supports predictive maintenance, allowing ML models to anticipate electrode degradation and scaling, thereby reducing the downtime. Additionally, AI enhances energy efficiency by optimizing power consumption, balancing applied potential and reaction kinetics, and lowering operational costs. It further improves the pollutant removal efficiency by predicting the most effective electrode materials and reaction conditions, leading to higher degradation rates. Moreover, automated process control ensures a stable treatment performance by continuously monitoring and adapting to fluctuations in wastewater composition. Despite these benefits, AI integration faces several challenges. One major drawback is its high initial cost, as implementing AI-driven systems requires specialized software, sensors, and computational resources. Additionally, AI introduces complexity in data interpretation, demanding large data sets and continuous updates to maintain predictive accuracy. Integration challenges also arise when retrofitting AI into existing electrochemical systems, often requiring hardware and software modifications. Furthermore, AI models, while effective in optimizing process conditions, have a limited mechanistic understanding of the electrochemical reaction pathways. Lastly, AI-driven systems depend on high-quality real-time data, which may not always be available in all treatment facilities. Despite these limitations, AI continues to advance electrochemical water treatment, offering significant potential for improving efficiency and sustainability.[62]

INFLUENCE OF EDL ON REACTIONS AND AI-DRIVEN OPTIMIZATION

The EDL is critical to heterogeneous electrochemical reactions, governing charge transfer, mass transport, and interfacial kinetics. It comprises the inner Helmholtz plane (IHP), outer Helmholtz plane (OHP), diffuse layer, and bulk electrolyte, with each region playing distinct roles in adsorption, electric field regulation, and potential distribution (Figure 13). Optimization of the EDL structure enhances reaction efficiency and selectivity, which is vital for technologies like electrocatalysis, EC, and energy storage.[63−66]

Recent developments have reshaped our understanding of the EDL behavior. Wu and Qi challenged the classical Stern model by studying localized high-concentration electrolytes

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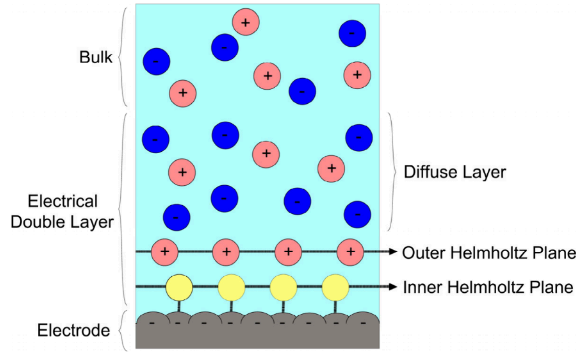

Figure 13. Schematic of the electrical double layer. Modified after Dunwell et al.[64] Copyright 2018 Elsevier B.V.

(LHCEs), which form micelle-like salt-solvent clusters distinct from diluent regions. Their DFT and MD simulations showed how this heterogeneity influences Li+ distribution, charge screening, and SEI stability.[67] Zhu et al. emphasized the impact of interfacial water structure on the hydrogen evolution reaction (HER), noting that hydrogen bond networks and Na^+ hydration alter charge screening and drive kinetic pH effects in alkaline media.[68]

Cao and Wu[69] focused on the EDL in electrochemical double-layer capacitors (EDLCs), highlighting how the Helmholtz and Stern models explain ion adsorption in porous carbon electrodes. They concluded that the electrolyte concentration and ion mobility are key to optimizing energy storage. Kelly et al.[66] took an innovative approach by converting interfacial charge dynamics into audible signals using a relaxation oscillator circuit, linking EDL changes to applied potential, electrode material, and reaction kinetics.

Several studies have addressed environmental and pH-dependent influences on the EDL structure. Liu et al. demonstrated how pH variations restructure the EDL, affecting PCET mechanisms in HER, ORR, and CO_2RR by altering mass transport and surface adsorption.[70] Dong et al.[63] investigated Fe_2O_3 electrodes and showed that specific ion adsorption in the IHP governs charge storage performance in energy systems.

Advanced techniques have enabled the direct visualization of EDL behavior. Favaro et al.[65] used ambient pressure X-ray photoelectron spectroscopy (APXPS) to validate the potential of zero charge (PZC) and map the potential drop across the EDL. Shin et al.[71] applied first-principles simulations to reveal how ion electrosorption and structural transitions influence EDL capacitance and CO_2 reduction.

The adsorption of target contaminants onto the electrode surface is influenced by EDL characteristics, particularly its charge distribution. A strong electrostatic interaction between the charged surface and pollutant molecules improves the efficiency of charge-driven reactions, facilitating pollutant degradation or coagulation. Li et al. critiqued the limitations of the traditional Gouy−Chapman−Stern (GCS) model and proposed an integrated ab initio−continuum approach to better capture complex electrolyte effects, including pH, ion identity, and hydrogen bonding.[72] Swift et al.[73] extended EDL modeling to solid-state batteries, describing EDLs as space-charge layers driven by point defects, not solvated ions, and presenting a DFT-based continuum model for all-solid-state interfaces.

Finally, AI has greatly accelerated EDL-related research. Kim et al.[74] developed ML models trained on DFT data to identify catalysts for nitrogen electro-reduction, dramatically speeding up the discovery of optimal adsorption surfaces. Zhou et al.[75] introduced ML potentials to simulate electrochemical interfaces with high accuracy, while Dufils et al.[76] launched PiNNwall, a powerful ML-integrated simulation tool for modeling polarized oxide surfaces in energy storage contexts.

POTENTIAL CHALLENGES AND CONCLUSION

The integration of AI into electrochemical water treatment significantly improved process efficiency, predictive accuracy, and operational control. However, scaling up these AI-driven electrochemical processes from laboratory experiments to full-scale industrial applications presents significant challenges. These include material costs, energy consumption, process optimization, data availability, and integration with the existing water treatment infrastructure. The selection and synthesis of electrode materials, such as BDD and MMOs, play crucial roles in determining treatment efficiency. However, their high production costs and limited scalability remain obstacles to widespread adoption. AI can address these issues by predicting material performance, optimizing synthesis processes, and identifying cost-effective alternatives, reducing dependence on expensive materials, and improving electrode longevity.

Economic feasibility is another critical factor in implementing an AI-driven electrochemical water treatment. AI can facilitate cost analysis by integrating real-time data on operational expenses, capital investments, and maintenance costs, helping to optimize treatment processes while minimizing financial burdens. Predictive maintenance, enabled by ML models, can anticipate equipment failures and reduce unplanned downtime, thereby improving the system reliability and cost-effectiveness. Furthermore, AI-driven decision-making can assess the integration of renewable energy sources, such as solar and wind power, into electrochemical treatment systems, reducing energy costs and environmental impact.

Energy consumption is a major limitation of electrochemical processes, particularly for industrial-scale applications. AI-driven optimization techniques, such as reinforcement learning and GAs, can dynamically adjust key operational parameters including voltage, current density, and treatment time based on real-time system performance, reducing unnecessary energy usage while maintaining high pollutant removal efficiency. AI can also model energy demands under different operational scenarios, allowing for better planning and integration of sustainable energy sources to enhance the overall efficiency.

Despite these benefits, AI-based models face challenges related to data dependency, computational complexity, and generalization under different treatment conditions. AI models require extensive, high-quality data sets to ensure reliable predictions. However, data availability remains a limitation, particularly for highly complex electrochemical processes that involve multiple interacting parameters. Overfitting and model bias are potential risks when AI is trained on limited data sets, leading to reduced predictive accuracy in new scenarios. Addressing these issues requires robust training data sets, improved feature selection, and hybrid AI approaches that combine data-driven models with mechanistic simulations for enhanced accuracy and interpretability.

Selecting the appropriate ML algorithm tailored to electrochemical pollutant treatment is crucial for optimizing the performance. Algorithms such as ANN, SVM, RF, and extreme

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gradient boosting (XGBoost) offer unique advantages in handling high-dimensional data, noise robustness, and nonlinear relationship modeling. In pollutant degradation processes, RF and SVM demonstrate strong predictive capabilities for complex data sets, while ANN excels in recognizing patterns and modeling nonlinearity. Integrating hybrid approaches, such as RF-XGBoost-ANN, enhances predictive accuracy while minimizing computational error. Future ML advancements in electrochemical oxidation should focus on improving model interpretability, autonomous real-time data processing, and energy-efficient learning techniques. The development of AutoML frameworks can streamline model selection and hyper-parameter tuning, further optimizing AI-driven electrochemical water treatment.

Despite these challenges, AI holds immense potential for revolutionizing electrochemical water treatment. By enhancing process efficiency, reducing operational costs, and enabling predictive control, AI-driven techniques can significantly improve water quality management. Future research should focus on refining AI integration in electrochemical processes, bridging the gap between theoretical models and real-world applications and ensuring AI-driven solutions are adaptable across different water treatment scenarios. Additionally, advancements in real-time monitoring, sensor integration, and automated decision-making will further enhance the feasibility of AI in industrial-scale electrochemical water treatment. As AI technology continues to evolve, its synergy with electrochemical treatment methods is expected to drive the development of more sustainable, cost-effective, and scalable water purification strategies, addressing global water pollution challenges and contributing to long-term environmental sustainability. Furthermore, incorporating AI into real-time autonomous systems could lead to a paradigm shift in water treatment, enabling smart, self-optimizing treatment plants that dynamically adjust operational conditions for maximum efficiency. The fusion of AI with electrochemical processes is not just an incremental improvement but a transformative step toward a future where water treatment is more intelligent, adaptive, and efficient on a global scale.

■ ASSOCIATED CONTENT

Data Availability Statement

The authors confirm that the data supporting the findings of this study are available within the article.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestwater.5c00238.

> Description of most popular artificial intelligence techniques used in water and wastewater treatment, categorization of AI techniques; Table S1: application, advantages, and limitations of commonly used AI techniques; Table S2: electrochemical applications optimized AI techniques used in various water matrix and wastewaters; and additional references (PDF).

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Notes

The authors declare no competing financial interest.

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