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Artificial neural networks applied to photo-Fenton process: An innovative approach to wastewater treatment

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Abstract

Artificial intelligence (AI) is a revolutionizing problem-solver across various domains, including scientific research. Its application to chemical processes holds remarkable potential for rapid optimization of protocols and methods. A notable application of AI is in the photo-Fenton degradation of organic compounds. Despite the high novelty and recent surge of interest in this area, a comprehensive synthesis of existing literature on AI applications in the photo-Fenton process is lacking. This review aims to bridge this gap by providing an in-depth summary of the state-of-the-art use of artificial neural networks (ANN) in the photo-Fenton process, with the goal of aiding researchers in the water treatment field to identify the most crucial and relevant variables. It examines the types and architectures of ANNs, input and output variables, and the efficiency of these networks. The findings reveal a rapidly expanding field with increasing publications highlighting AI's potential to optimize the photo-Fenton process. This review also discusses the benefits and drawbacks of using ANNs, emphasizing the need for further research to advance this promising area.

Keywords: Artificial neural networks; Degradation; Machine learning; Optimization; Persistent organic pollutants; Wastewater

1. Introduction

1.1. Photo-Fenton process

Advanced oxidation processes (AOPs) have proven highly effective in the degradation of recalcitrant organic contaminants in wastewater (Wang and Wang, 2021). These processes are based on the generation of highly reactive radicals, such as hydroxyl, superoxide, or singlet oxygen radicals, which play a fundamental role in the oxidation of contaminants (Li and Cheng, 2021; Tolba et al., 2019; Wang and Wang, 2020, 2021). These radicals exhibit different oxidation potentials and reaction mechanisms that make them effective in degrading a wide range of persistent organic pollutants (Wang and Wang, 2020, 2021).

One of the most widely adopted AOPs is the Fenton process, which relies on the reaction between hydrogen peroxide (H_2O_2) and Fe^{2+} ions to generate highly reactive hydroxyl radicals ($\cdot OH$) (Wang and Wang, 2020; Bhaskar et al., 2024). However, the Fenton process is limited by the challenging regeneration of Fe^{2+}, which becomes the rate-determining step of the whole process. An enhancement of the Fenton process is the photo-Fenton process, in which ultraviolet (UV) or visible light is used to promote the photoreduction of Fe^{3+} to Fe^{2+}, significantly improving degradation kinetics and reducing treatment time (Li and Cheng, 2021). Furthermore, the photo-Fenton process can be efficiently operated under visible light, allowing the use of solar light as a more sustainable radiation source compared to other advanced oxidation systems (Lumbaque et al., 2019). Therefore, the photo-Fenton process has been successfully applied in removing a wide range

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of persistent contaminants using both homogeneous and heterogeneous iron catalysts (Cabrera Reina et al., 2020; Hosseinzadeh et al., 2021; Lumbaque et al., 2019; Palma et al., 2018).

The optimization of AOPs, such as the photo-Fenton process, necessitates the simultaneous consideration of several parameters, such as initial contaminant concentration, hydrogen peroxide dosage, iron concentration, pH, and radiation intensity (Hosseinzadeh et al., 2021; Santana et al., 2021; Sarı et al., 2023; Talwar et al., 2019; Tolba et al., 2019). Furthermore, despite their effectiveness in degrading organic contaminants, AOPs can also generate toxic by-products. Several studies have identified the formation of oxidation by-products, such as carboxylic acids and halogenated compounds, which pose environmental risks (Wang and Wang, 2020, 2021). Therefore, assessing the formation and toxicity of these degradation by-products is crucial during the design and implementation of the photo-Fenton process. Additionally, identifying the reactive species involved in contaminant degradation and understanding the factors influencing their generation are essential for optimizing these processes and ensure their safe and effective applications (Wang and Wang, 2020, 2021).

The inherent complexity of AOPs necessitates the use of sophisticated tools for their monitoring and optimization. Using the one-at-a-time approach to manage multiple variables can be cumbersome and costly, involving extensive experimentation, associated costs, and waste generation. In recent years, the development of prediction models based on artificial neural networks (ANNs) and machine learning (ML) has been instrumental in enhancing the monitoring of AOPs (Hosseinzadeh et al., 2021; Santana et al., 2021; Sarı et al., 2023; Talwar et al., 2019; Tolba et al., 2019). A recent review (Dhamorikar et al., 2024) has highlighted the potential of ANNs to optimize AOPs with high precision, thereby reducing both the economic and environmental impacts of these reactions.

These prediction models take advantage of the capacity of ANNs to learn and recognize complex patterns in data by capturing the nonlinear relationships between variables and efficiency responses. This capability allows them to overcome the limitations of traditional models based on mathematical equations (Hosseinzadeh et al., 2021; Krogh, 2008). ML facilitates the integration of multiple input variables into unified prediction models for the photo-Fenton process. Notably, appropriately selecting input variables is crucial for developing reliable and robust prediction models. Therefore, a careful analysis of input variables is necessary when developing prediction models for the photo-Fenton process (Hosseinzadeh et al., 2021; Santana et al., 2021; Sarı et al., 2023; Talwar et al., 2019; Tolba et al., 2019). These models can predict the removal efficiency of organic contaminants and optimize operating conditions, thereby enhancing the efficiency and profitability of the photo-Fenton process (Hosseinzadeh et al., 2021; Sarı et al., 2023; Talwar et al., 2019; Tolba et al., 2019).

1.2. Application of artificial intelligence in wastewater treatment

The implementation of artificial intelligence (AI) in wastewater treatment has been extensively explored in the literature, with a significant increase in AI-related publications on wastewater treatment and management (Wang et al., 2023).

As wastewater is a complex matrix containing multiple pollutants and compounds, its optimal degradation is an important issue in wastewater treatment. The selection and optimization of relevant variables are crucial for proper management of wastewater samples. AI has been applied to systematically analyze compounds targeted for removal and optimization of the entire treatment processes (Krovvidy et al., 1991; Wang et al., 2023). Given that the composition of both the initial wastewater and effluent from wastewater treatment facilities should be considered, AI-optimized protocols have been developed to predict effluent composition, and AI has been successfully employed to predict key parameters such as suspended solids, nitrogen, and phosphorus (Khatri et al., 2019, 2023).

Recent reviews have comprehensively analyzed the literature on wastewater treatment, focusing on aspects such as water quality determination and treatment (Safeer et al., 2022; Zhao et al., 2020). These studies have emphasized the necessity of thoroughly investigating experimental variables to effectively implement AI tools in wastewater experiments (Zhao et al., 2020). Building on this foundation, this review aims to highlight specific applications of AI in optimizing the photo-Fenton process. The objective is to explore how AI can enhance research in terms of degradation efficiency and time/cost saving.

Regarding AI applications in wastewater treatment techniques, two relevant review articles were identified (Senthil Rathi et al., 2024; Wang et al., 2023). Senthil Rathi et al. (2024) reviewed AI applications across various wastewater treatment methods, including adsorption, coagulation, electrochemical processes, AOPs, and biological wastewater treatment, but did not specifically address Fenton and photo-Fenton processes. Wang et al. (2023) summarized commonly used AI models and their applications in wastewater treatment, covering water quality monitoring, laboratory-scale research, and process design. Both review articles introduced the used AI models and their purposes, advantages, and disadvantages, and comprehensively reviewed the inputs, outputs, objectives, and primary findings of specifically AI applications. However, neither review specifically focused on Fenton and photo-Fenton processes.

This comprehensive review explores the significant advancements in ANNs for contaminant degradation, with specific emphasis on their practical applications in optimizing the photo-Fenton process and the predictive potential of these models. While these ANN models demonstrate substantial potential, it is essential to optimize specific parameters, such as the number of neurons, activation functions, and other relevant factors. Additionally, this review evaluates the effectiveness of ANNs through rigorous comparisons with mathematical response surface models, yielding encouraging results that highlight the potential of ANNs to significantly enhance the efficiency of the photo-Fenton process, sometime outperforming traditional response surface models.

Given the complexity of new technologies like AI, recently studies have emphasized the importance of multidisciplinary collaboration among experts to effectively implement AI in chemical processes (Wang et al., 2023). This review aims to compile, synthesize, and summarize the available information on the application of AI to the degradation of organic compounds using photo-Fenton and similar processes.

2. Types of artificial neural networks

There are many available AI tools, such as support vector machines (SVM), random forests, and ANNs. ANNs have demonstrated their great potential in water treatment management (Narayanan et al., 2024) and are readily available in many software, enabling their application by non-experts. Thus, this review specifically focuses on ANNs with their application to the optimization of the photo-Fenton process.

ANNs are computational models inspired by human brain's structure, renowned for their learning capabilities. ANNs can acquire knowledge from data and recognize complex patterns, making them suitable for applications such as data prediction and classification (Krogh, 2008).

In the context of the photo-Fenton process, several types of ANNs are particularly relevant: the feed-forward multi-layer perceptron (MLP or FFN) (Amorim et al., 2020; de Moraes et al., 2021; Joy et al., 2022; Singa et al., 2023; Talwar et al., 2019), cascade-forward net (CFN) (Joy et al., 2022), feed-forward backpropagation (FFBP) (Tolba et al., 2019), and radial basis function network (RBF) (Amorim et al., 2020). The properties and structures of these ANNs have been extensively covered in the literature. Given the scope of this review, readers can consult the cited papers for further details. This review only provides a brief description.

MLP models are among the simplest neural networks. They transmit information sequentially from the input layer to the output layer through intermediate layers, known as hidden layers. Each layer contains a specified number of neurons, and both the number of layers and the number of neurons per layer affect the final output. In contrast, CFN models utilize input data for all subsequent layers, not just the immediately adjacent ones. These networks typically use linear or sigmoid activation functions and are commonly employed in classification processes (Moshkbar-Bakhshayesh, 2019). FFBP networks are distinguished by their error backpropagation process. The error between predicted results and actual training data is calculated and back-propagated to adjust neuron weights, minimizing error and enhancing accuracy (Shaik et al., 2020). This process makes FFBP networks more sophisticated and computationally intensive. RBF ANNs organize neurons into layers, where each hidden-layer neuron is associated with a radial basis function. These functions focus on specific points in the input space and are activated based on the distance between input data and their associated centers. In an RBF ANN, the input layer receives input data and propagates them to the hidden layer, where distances between data and the centers of radial basis functions are calculated. The outputs of these functions are then transmitted to the output layer for classification or regression (Bugshan et al., 2022).

When working with these computational models, several key parameters must be considered, such as architecture, which refers to the number of layers and neurons in each layer (Amorim et al., 2020; de Moraes et al., 2021; Joy et al., 2022; Singa et al., 2023; Talwar et al., 2019; Tolba et al., 2019), training methods (Amorim et al., 2020; de Moraes et al., 2021; Joy et al., 2022), transfer and activation functions (Amorim et al., 2020; de Moraes et al., 2021; Joy et al., 2022; Tolba et al., 2019), and epochs (de Moraes et al., 2021; Joy et al., 2022; Talwar et al., 2019; Tolba et al., 2019). Moreover, ANNs can be combined with other algorithms to generate more complex and efficient models (Joy et al., 2022; Zhao et al., 2023). Fig. 1 illustrates the different types of ANNs and highlights the important parameters requiring optimization.

As shown in Fig. 1, the application of ANNs in optimizing the photo-Fenton process is a complex task involving many different variables, including both experimental and computational variables. Chemically, variables such as pH, iron concentration, and radiation intensity are crucial. Computationally, considerations include ANN architecture and functioning, such as transfer functions, input and output variables, and the numbers of neurons and layers. As AI continues to grow, this review aims to synthesize existing literature on the use of ANNs in the photo-Fenton process, providing researchers with up-to-date information to inform their experiments. The following sections systematically present the relevant references, highlighting the experimental and computational values studied in each case and identifying which approaches yielded best results.

3. Application of ANNs in photo-Fenton process for water treatment

Over the past two decades, the photo-Fenton process has emerged as a promising technique for degrading persistent organic compounds in contaminated water. This process relies on the interaction between UV/visible radiation, hydrogen peroxide, and iron(II) to generate hydroxyl radicals that react with organic matter. The literature highlights the use of ANNs for predicting and optimizing this process, often in comparison with other techniques. This section aims to concisely summarize the most significant contributions in this field, providing readers with a comprehensive overview to facilitate future advancements.

When using ANNs to optimize chemical processes, the underlying rationale is to predict the most favorable scenario that yields optimal outcomes based on a limited number of experimental trials. In the context of this article, optimizing the degradation of organic compounds involves predicting the concentration of reagents that would achieve the highest degradation percentage. Two primary considerations arise: (1) the design of the ANN itself, including its architecture, the number of neurons and layers, and the type of ANNs employed; and (2) validation of the results from the ANN employed. These considerations can be achieved by statistically comparing the actual outcomes with the predicted ones, enabling the selection of the most effective ANN model. This section is organized following the same logical sequence. Table 1 presents a compilation of applications of ANNs in optimizing the photo-Fenton process, highlighting the specific parameters optimized in various studies from the literature.

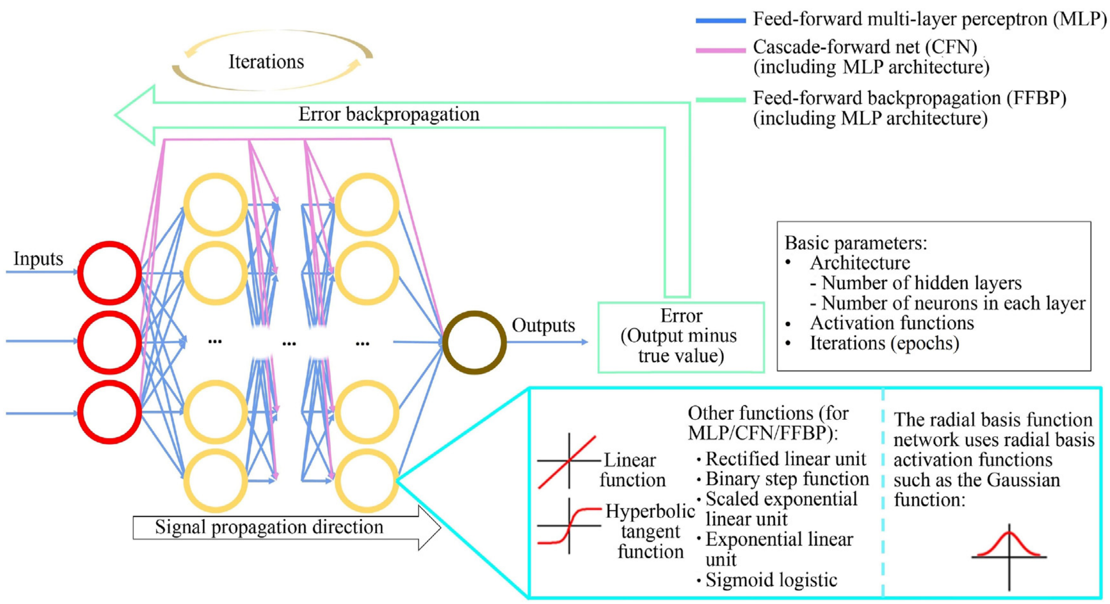

Fig. 1. Schematic representation of main ANN architectures used for photo-Fenton process optimization and key parameters for model development.

Table 2 provides a concise summary of the statistical results, focusing on the correlation between predicted results and experimentally obtained real values. Additionally, it includes results from other methodologies to provide a broader perspective by comparing them to conventional techniques.

Singa et al. (2023) employed an MLP neuron network to quantify the degradation of polycyclic aromatic hydrocarbons (PAHs) and the removal of chemical oxygen demand (COD). Singa et al. (2023) also used a central composite design factorial, allowing for a comparison between the predictive capabilities of two methods. The results showed that the ANN achieved high correlations, with the determination coefficient (R^2) values exceeding 0.86, which were calculated between the measured and its predicted values. However, the response surface methodology (RSM) using the central composite design (CCD) yielded higher determination coefficients, particularly for COD removal, with an R^2 of 0.930. Although CCD-RSM provided a slightly higher R^2 for the degradation of PAHs, the ANN obtained a lower root mean square error (RMSE) of 2.392%, compared to 3.019% for CCD-RSM. Notably, the neural network used in Singa et al. (2023) did not explicitly detail a parameter optimization process for training, making it challenging to identify the optimized parameters and their ranges.

Sabour and Amiri (2017) compared the performances of ANNs and RSM in the Fenton process and found that ANNs outperformed RSM in predicting mass content ratio and mass removal efficiency, achieving R^2 values between 0.954 and 0.968, with an RMSE value of 1.86% and absolute errors ranging from 2.56% to 3.66%. The authors suggested combining RSM and ANNs to enhance predictive capacity by feeding RSMs results into ANNs.

Joy et al. (2022) developed models for estimate optimal performances in terms of total organic carbon (TOC), COD, and residual iron (Fe^{2+}) removal. Their work included parameter optimization for neural network development and selection of the best-fitting mode. After comparing the results from MLP and CFN networks, CFN was identified as the top performer, with R^2 values of 0.890 9, 0.947 1, and 0.890 4 for TOC, COD, and residual iron removal, respectively. However, the CCD-RSM model achieved higher R^2 values of 0.901 4, 0.989 7, and 0.989 4, thus outperforming ANNs. This study further used the non-dominated sorting genetic algorithm III (NSGA-III) to optimize both machine learning regression models and RSM and incorporated equations from these techniques as objective functions to calculate optimal performance for the parameters under investigation, with 80% of the dataset for training and 20% for validation. The results predicted by CFN-NSGA-III showed satisfactory agreement with the experimental findings.

Similarly, Shokry et al. (2018) predicted TOC values in paracetamol degradation using a photo-Fenton batch reactor. They evaluated the predictive capabilities of various nonlinear data-driven modeling tools, such as ordinary Kriging (OK), support vector regression (SVR), Gaussian process (GP)-regression (GPR), and ANNs. Specifically, they employed an MLP network with one hidden layer containing nine neurons. The input data for all modeling tools consisted of initial TOC, temperature, and redox potential values, as well as online measurements of temperature and redox potential. OK and GPR demonstrated superior average performance in terms of prediction accuracy and were noted for their flexibility and robustness during training. Furthermore, OK and GPR metamodels offered the significant advantage of estimating prediction errors. In contrast, ANNs required substantial effort in testing to select the optimal configuration, including the number of layers, the number of neurons per layer, and training algorithms, making their use less convenient compared to OK and GPR.

Turkes et al. (2024) investigated COD removal in textile wastewater samples and compared various optimization methods such as Box–Benhken design, ANNs, SVR, decision trees, and random forests. They employed an FFBP network

Table 1
Different types of ANNs employed for photo-Fenton process and relevant parameters regarding their optimization and structure.

Reference	ANN type	Software	Evaluated parameters	Parameter values
Tolba et al. (2019)	FFBP	Not provided (optimization)	Number of neurons (N) Architecture Epochs Activation function Training/validation/testing (%)	4−14 5−N−1 ≤ 8 000 tansig 70/15/15
Santana et al. (2021)	MLP	Statistica 10.0 (optimization)	Architecture Activation functions Algorithm Epochs Iterations Training/validation/testing (%)	3−10−2 Identity, logistic, hyperbolic, tangential sinusoidal, and exponential Broyden−Fletcher−Goldfarb−Shanno (BFGS) 200 2 000 70/15/15
Talwar et al. (2019)	MLP	Matlab (optimization)	Number of neurons (N) Architecture Epochs Training/validation/testing (%)	2−11 6−N−1 987 70/15/15
Singa et al. (2023)	MLP	Not provided (no optimization)	Architecture Alpha values Random state	5−7−4−1 and 5−5−3−1 0.000 1 and 0.000 066 70 and 75
Joy et al. (2022)	MLP and CFN	Matlab (optimization)	Number of neurons (N) Architecture Training algorithms Activation functions Training/validation (%)	1−20 5−N−1 Evenberg−Marquardt, gradient descent with variable learning rate, Bayesian regularization, and scaled conjugate gradient Sigmoidal tangent and sigmoidal logarithmic 80/20
Amorim et al. (2020)	MLP and RBF	Statistica (optimization)	Architecture Activation functions Training algorithm Training/validation/testing (%)	5−21−2 Identity, logistic, hyperbolic, exponential, sinusoidal, and hyperbolic tangential BFGS 70/15/15
de Moraes et al. (2021)	MLP	Not provided (optimization)	Architecture Training algorithm Activation functions Epochs Training/validation/testing (%)	5−6−3 BFGS 297 tanh and logistic 2 000 70/15/15
Shokry et al. (2018)	MLP	Matlab ANN toolbox	Number of hidden layers Number of neurons Training algorithm	1 9 Bayesian regularization backpropagation (trainbr)
Turkes et al. (2024)	FFBP	Matlab (2020)	Transfer functions Epochs Architecture Number of neurons (N)	tansig and logsig 1 000 3−N−1 1−10
Bouizzar and Berkani (2024)	MLP	Not provided	Training/validation/testing (%) Number of input points Epochs	75/15/15 200 455
Bassam et al. (2012)	FFBP	Matlab (optimization)	Architecture	5−7−2
Hajiahmadi et al. (2022)	FFBP	Matlab 7 (2013) ANN toolbox (optimization)	Number of neurons (N) in hidden layer Architecture Activation function Training/validation/testing (%)	2−20 5−12−1 Sigmoid 60/20/20

(continued on next page)

Table 1 (*continued*)

Reference	ANN type	Software	Evaluated parameters	Parameter values
Gholizadeh et al. (2021)	FFBP	Matlab (2013) ANN toolbox (optimization)	Number of neurons (N) Architecture Activation function	2−20 5−10−1 tansig
Mousavi et al. (2020)	MLP	Not provided (optimization)	Number of neurons (N) Architecture Training algorithm Activation functions Training/validation/testing (%)	2−10 3−8−1 Levenberg−Marquardt (LM) tansig and linear transfer function (purelin) Not specified
Sabour and Amiri (2017)	FFN	Matlab ANN toolbox	Number of layers Number of input nodes Number of neurons	3 3 5−6
Ali et al. (2024)	MLP	IBM SPSS modeler	Number of neurons Architecture Activation functions Training/validation/testing (%)	10 1−10 Sigmoid, tanh, and rectified linear unit (ReLU) Not specified

and examined properties like the number of neurons in the hidden layer (ranging from 1 to 10) and the type of transfer functions (such as the log-sigmoid transfer function (logsig) and the hyperbolic tangent sigmoid transfer function (tansig)). The results showed that the tansig function offered the best predictive capacity, with lower prediction errors. The optimal ANN topology was 3−6−1, achieving an R^2 value of 0.999 86. This ANN consisted of one hidden layer with three input variables (initial pH, iron concentration, and hydrogen peroxide concentration) and one output value representing the COD level. Under optimal conditions (pH of 3, $[Fe^{2+}]$ = 0.75 g/L, and $[H_2O_2]$ = 5 mmol/L), a total COD reduction of 87% was achieved.

Some studies have focused on the degradation of specific compounds. Talwar et al. (2019) modeled and optimized metronidazole (MTZ) degradation in a hybrid system combining photocatalysis and photo-Fenton processes using an MLP ANN coupled with a genetic algorithm. They optimized the number of neurons in the hidden layer and the number of epochs, selecting four neurons and 987 epochs, respectively. The ANN was executed using three distinct dataset groups: a training set (70%), a validation set (15%), and a testing set (15%). Although this study did not compare the ANN with other techniques, the correlation results across different groups obtained R^2 values greater than 0.98, indicating the accuracy of this methodology in predicting MTZ degradation. These findings align with results from similar studies (Amorim et al., 2020; de Moraes et al., 2021; Joy et al., 2022; Tolba et al., 2019).

Bouizzar and Berkani (2024) conducted a study on the degradation of tribenuron methyl (TBM), an herbicide, using a modified photo-Fenton approach where oxygen peroxide was replaced with sodium hypochlorite, purportedly as a more economical procedure. Similar to Talwar et al. (2019), this study used an MLP ANN coupled with a genetic algorithm to identify optimal conditions. The dataset was divided into training (75%), testing (15%), and validation (15%) sets, consistent with the approach of Talwar et al. (2019). The input variables included the initial concentration of TBM, NaClO, mass of iron sulfate, and initial pH, while the output variable was the total degradation percentage of TBM. The ANN model demonstrated excellent prediction capability, achieving an R^2 value of 0.999 83 for the degradation percentage of TBM.

Bassam et al. (2012) proposed a multivariate experimental design to monitor photo-Fenton treatment in a pilot solar plant for wastewater containing synthetic mixtures of pesticides. ANNs were developed to predict TOC and the initial mineralization kinetic rate constants of the reactions. The input data included experimental values for temperature, pH, hydrogen peroxide (H_2O_2) consumption, initial concentration of Fe^{2+}, and accumulated energy. An ANN architecture with seven hidden neurons yielded the best result for the photo-catalytic degradation efficiency of the photo-Fenton process, with an R^2 value exceeding 0.99 and errors no greater than 1%.

Cüce and Özçelik (2022) adopted an ML and AI perspective to model the treatment of landfill leachate using classical Fenton and photo-Fenton processes. They employed SVR, FFBP, and RBF. The performance of these ML-based modeling tools was compared with the results from the more traditional statistical approach (RSM). Their study included several aspects: (1) predicting COD removal from real solid waste landfill leachate using SVR, FFBP ANN, and RBF ANN for both classical Fenton and photo-Fenton processes; (2) comparing the results obtained from ML methods with those from RSM; (3) analyzing statistical error metrics; (4) conducting simple linear regression analysis and utilizing visual graphs for data interpretation; (5) examining the effects of pH, Fe^{2+} concentration, H_2O_2 concentration, and contact time on both Fenton processes; and (6) employing GA to optimize parameters for treating landfill leachate using both Fenton processes. The results showed that both RBF ANN and FFBP ANN exhibited superior predictive capabilities compared to RSM and other ML methods like SVR, based on both RMSE and mean absolute percentage error (MAPE) criteria.

Amorim et al. (2020) evaluated the degradation of the red 83 dye using models based on RBF and MLP. The comparison between RBF and MLP revealed that RBF was not suitable for

Table 2
Statistical results of reviewed prediction techniques in literature.

Reference	Input	Output	Method and ANN structure	RMSE	R^2	MAE (%)	MSE	Adjusted R^2
Tolba et al. (2019)	Initial analyte concentration, reaction time, pH, $[H_2O_2]$, and Fe_3O_4 concentration in nanotubes	Removal efficiency (training)	FFBP (5–11–1)		0.999 6			
		Removal efficiency (testing)	FFBP (5–11–1)		0.977 5			
		Removal efficiency (validation)	FFBP (5–11–1)		0.992 6			
Santana et al. (2021)	$[Fe^{2+}]$, $[H_2O_2]$, and reaction time	Degradation (training)	MLP (3–10–1)		0.98			
		Degradation (testing)	MLP (3–10–1)		0.96			
		Degradation (validation)	MLP (3–10–1)		0.98			
		Residual H_2O_2 concentration (training)	MLP (3–10–1)		0.99			
		Residual H_2O_2 concentration (testing)	MLP (3–10–1)		0.99			
		Residual H_2O_2 concentration (validation)	MLP (3–10–1)		0.99			
Talwar et al. (2019)	Treatment time, light intensity, surface area, area/volume ratio, pH, and $[H_2O_2]$	MTZ degradation (training)	MLP (6–4–1)		0.994 2			
		MTZ degradation (validation)	MLP (6–4–1)		0.990 4			
		MTZ degradation (testing)	MLP (6–4–1)		0.987 7			
		MTZ degradation (all)	MLP (6–4–1)		0.993 3			
Singa et al. (2023)	$[H_2O_2]$, $[Fe^{2+}]$, pH, mixing speed, and reaction time	COD removal	MLP (5–5–3–1)	4.723%	0.867	3.603		
			CCD-RSM	3.512%	0.930	2.934		
		PAH removal	MLP (5–7–4–1)	2.392%	0.941	1.803		
			CCD-RSM	3.019%	0.950	2.741		
Joy et al. (2022)	Initial TOC concentration, pH, $[H_2O_2]$, $[Fe^{2+}]$, and reaction time	TOC removal	CFN (5–5–3)	6.931 4%	0.890 9			0.870 5
			CCD-RSM	5.931 4%	0.901 4			0.988 5
		COD removal	CFN (5–5–3)	9.304 1%	0.947 1			0.924 3
			CCD-RSM	7.304 1%	0.989 7			0.984 1
		Residual Fe^{2+}	CFN (5–5–3)	3.971 2 mg/L	0.890 4			0.864 2
			CCD-RSM	2.371 2 mg/L	0.989 4			0.984 8
Amorim et al. (2020)	Initial dye concentration, reaction time, $[H_2O_2]$, [Fe], and pH	Signal at 288 nm	MLP (5–21–2)		> 0.98			
		Signal at 544 nm	MLP (5–21–2)		> 0.97			
de Moraes et al. (2021)	$[H_2O_2]$, [Fe], pH, reaction time, and photon emission	Signal at 254 nm (training)	MLP (5–6–3)		0.982 3			
		Signal at 254 nm (testing)	MLP (5–6–3)		0.953 2			
		Signal at 254 nm (validation)	MLP (5–6–3)		0.963 7			
		Signal at 307 nm (training)	MLP (5–6–3)		0.994 1			
		Signal at 307 nm (testing)	MLP (5–6–3)		0.994 1			
		Signal at 307 nm (validation)	MLP (5–6–3)		0.985 8			
		Signal at 548 nm (training)	MLP (5–6–3)		0.992 4			
		Signal at 548 nm (testing)	MLP (5–6–3)		0.987 0			
		Signal at 548 nm (validation)	MLP (5–6–3)		0.987 0			

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Table 2 (*continued*)

Reference	Input	Output	Method and ANN structure	RMSE	R^2	MAE (%)	MSE	Adjusted R^2
Shokry et al. (2018)	Initial TOC concentration, temperature, redox potential, online measured temperature, and redox potential	TOC removal	MLP	3.84%	0.990 1			
Turkes et al. (2024)	$[Fe^{2+}]$, $[H_2O_2]$, and pH	COD (training)	FFBP (logsig; 3–6–1)		0.999 86			
		COD (validation)	FFBP (logsig; 3–6–1)		1			
		COD (testing)	FFBP (logsig; 3–6–1)		0.994 33		0.004 16	
		COD (training)	FFBP (tansig; 3–7–1)		0.956 90			
		COD (validation)	FFBP (tansig; 3–7–1)		1			
		COD (testing)	FFBP (tansig; 3–7–1)		0.980 85		0.977 70	
Bouizzar and Berkani (2024)	Mass of $FeSO_4$, TBM concentration, [NaClO], and pH	Degradation percentage	MLP (not specified)		0.999 83		6.6×10^{-8}	
Bassam et al. (2012)	Temperature, pH, H_2O_2 consumption, $[Fe^{2+}]$, and accumulated energy	TOC removal and initial mineralization kinetic rate constant	FFN (5–7–2)		> 0.99	1		
Cüce and Özçelik (2022)	$[Fe^{2+}]$, $[H_2O_2]$, pH, and contact time	COD removal	FFBP	2.2%		2.0		
			RBF	1.6%		2.0		
			SVR	5.2%–6.9%		5.41–6.80		
Hajiahmadi et al. (2022)	Initial analyte (paclitaxel) concentration, initial pyrite concentration, pH, current intensity, and reaction time	Removal efficiency (training)	FFBP (5–12–1)		0.999 7			
		Removal efficiency (testing)	FFBP (5–12–1)		0.938 8			
		Removal efficiency (validation)	FFBP (5–12–1)		0.991 1			
Gholizadeh et al. (2021)	Time, initial PHP concentration, pH, current intensity and surface ratio	PHP degradation efficiency (training)	FFBP (5–10–1)		0.981 6			
		PHP degradation efficiency (testing)	FFBP (5–10–1)		0.945 2			
		PHP degradation efficiency (validation)	FFBP (5–10–1)		0.959 6			
Mousavi et al. (2020)	$[Fe^{2+}]$, $[H_2O_2]$, and reaction time	MB removal (training)	MLP (3–8–1)					
		MB removal (testing)	MLP (3–8–1)					
		MB removal (validation)	MLP (3–8–1)					
Sabour and Amiri (2017)	pH, Fe^{2+} dosage, and $[H_2O_2]/[Fe^{2+}]$	Mass content ratio and mass removal efficiency	FFN	1.74%–2.27%	0.954–0.968	2.25–3.66		
			RSM					
Ali et al. (2024)	Initial phenol concentration, pH, H_2O_2, flow, $[Fe^{3+}]$, TiO_2 mass, light intensity, and contact time	Phenol (360 nm)	MLP (1–10)		0.916			
			Regression model		0.287			
			Generalized linear model		0.287			
			CHAID		0.233			
			Linear model		0.070			
			Random forest classification		0			

this prediction task. The optimized ANN was an MLP with a 5−21−2 architecture, which provided the predicted absorbance of the dye after treatment at wavelengths of 288 nm and 544 nm. The reported R^2 values ranged between 0.98 and 0.97.

de Moraes et al. (2021) yielded similar findings on the degradation of acid violet 17. They utilized an MLP neural network model with a 5−6−3 architecture, incorporating hyperbolic tangent (tanh) and logistic activation functions and the BFGS 297 algorithm. The model established a strong correlation between predicted and experimental values, achieving R^2 values above 0.95. Consistent with these results, Tolba et al. (2019) investigated the degradation of methylene blue (MB) using an FFBP neural network model. After optimizing the number of neurons, the model employed a 5−11−1 architecture. The results demonstrated a high predictive accuracy for removal efficiency, with R^2 values exceeding 0.97.

Several studies have adopted a similar structure to that of Tolba et al. (2019) in the context of environmental degradation analysis. Notably, Hajiahmadi et al. (2022) and Gholizadeh et al. (2021) employed neural network architectures with five input parameters and one output variable, along with 12 and ten neurons in the hidden layer, respectively. In both studies, four common input parameters were reaction time, initial pH, applied current, and initial contaminant concentration. The studied contaminants were paclitaxel (Hajiahmadi et al., 2022) and phenazopyridine (PHP) (Gholizadeh et al., 2021). The fifth parameter differed between both studies, the concentration of pyrite in Hajiahmadi et al. (2022) and the ratio of magnetite nanoparticles to activated carbon surface area in the utilized electrode in Gholizadeh et al. (2021). Both studies achieved notable results, with R^2 values exceeding 0.93, demonstrating comparable predictive capabilities.

Several studies have employed ANNs with a reduced number of input parameters. Notably, the works of Mousavi et al. (2020) and Santana et al. (2021) stand out for using only three input parameters, making them interesting cases for evaluating the efficiency of ANNs with minimal input information. Both studies employed the same input data: iron concentration, H_2O_2 concentration, and reaction time, with the output representing the degradation level. However, Santana et al. (2021) employed a dual-output architecture, where the second output parameter represented the residual concentration of H_2O_2. Mousavi et al. (2020) demonstrated a strong correlation between predicted and actual values, although specific numerical data were not provided. In contrast, Santana et al. (2021) achieved remarkable results, with R^2 values exceeding 0.96, and particularly impressive results for the residual H_2O_2 concentration, with R^2 values of 0.99.

Ali et al. (2024) studied the removal of phenol in wastewater using photo-Fenton reagents and assessed treatment effectiveness based on the reaction rate. Laboratory data were submitted to various ML classifiers, and models were trained accordingly. A deep neural network with ten nodes in one hidden layer achieved an accuracy of 97.2%. The top three predictors for different ML models were identified: (1) deep neural network: phenol (29%), pH (23%), and Fe^{3+} (15%); (2) generalized linear model: TiO_2 (23%), pH (22%), and phenol (20%); (3) linear regression: Fe^{3+} (29%), H_2O_2 (18%), and TiO_2 (16%); (4) random trees: pH (exceeding 60%), light intensity (approximately 16%), and phenol (approximately 16%); (5) ensemble model: phenol (21%), light intensity (17%), and Fe^{3+} (17%); and (5) chi-squared automatic interaction detection (CHAID): phenol (50%), light intensity (28%), and pH (22%).

4. General trends in ANN application to photo-Fenton process

A detailed analysis of the aforementioned articles reveals several trends in the application of ANNs to the photo-Fenton process. MLP is the most commonly used type of ANNs, employed in approximately 67% of the reviewed studies (Ali et al., 2024; Bouizzar and Berkani, 2024; de Moraes et al., 2021; Mousavi et al., 2020; Santana et al., 2021; Shokry et al., 2018; Singa et al., 2023; Talwar et al., 2019). FFBP is the second most applied (Bassam et al., 2012; Gholizadeh et al., 2021; Hajiahmadi et al., 2022; Turkes et al., 2024). RBF has limited applications, with only a few instances noted, such as Amorim et al. (2020).

In the development and optimization of ANNs for the photo-Fenton process, the software used by researchers is an important aspect. Notably, Matlab is the most commonly mentioned software, being used in approximately half of the reviewed articles (Bassam et al., 2012; Gholizadeh et al., 2021; Hajiahmadi et al., 2022; Joy et al., 2022; Sabour and Amiri, 2017; Shokry et al., 2018; Talwar et al., 2019; Turkes et al., 2024). However, the specific role of programming software in ANN development and optimization is generally underemphasized. It is crucial to highlight this aspect in future studies, as the choice of software significantly impacts in the development and implementation of ANNs.

Table 1 reveals a diverse and heterogeneous perspective on the architecture and characteristics of ANNs used in the photo-Fenton process. While some studies thoroughly implemented and optimized various variables, others relied on specific combinations without detailed discussion. Despite generally satisfactory predictive results, this lack of detail leads to a certain obscurity in the application of ANNs in the photo-Fenton process. Therefore, it is necessary to disclose detailed descriptions of ANN architectures to enhance reproducibility and applicability.

Table 2 highlights numerous attempts to apply AI to optimize AOPs for organic compounds. Many studies focused on COD and TOC as output variables (Bassam et al., 2012; Cüce and Özçelik, 2022; Joy et al., 2022; Shokry et al., 2018; Singa et al., 2023), as they are key parameters in wastewater management. Other articles used ANNs to predict the final concentration or degradation percentage of target molecules (Ali et al., 2024; de Moraes et al., 2021; Gholizadeh et al., 2021; Singa et al., 2023; Talwar et al., 2019), often organic compounds found in wastewater such as phenol (Ali et al., 2024), dyes (Amorim et al., 2020; de Moraes et al., 2021), TBM (Bouizzar and Berkani, 2024), PAHs (Singa et al., 2023), and PHP (Gholizadeh et al., 2021). A third group of studies used analytical signals like absorbance as the output, which is common in degradation processes where residual absorbance indicates the amount of non-degraded analyte. This analysis demonstrates the versatility of AI tools, which can adapt to the needs of most study cases.

In the context of the photo-Fenton process, three primary chemical variables are systematically used as inputs: oxidant concentration, iron concentration, and pH. These variables are crucial in the chemical pathway of the photo-Fenton process and are included in most studies. This approach is straightforward, relying on just three original values to predict the final output, which can be advantageous as it minimizes the number of experimental data points needed. Nevertheless, it also limits the pool of variables considered. Therefore, some studies incorporated additional input variables to enhance predictive capabilities, including the concentration of target molecules (Ali et al., 2024; Amorim et al., 2020; Bouizzar and Berkani, 2024; Hajiahmadi et al., 2022), reaction time (Ali et al., 2024; de Moraes et al., 2021; Mousavi et al., 2020; Santana et al., 2021), and more specific parameters related to the experimental setup, such as applied current or surface area (Talwar et al., 2019).

The fitness of predicted data compared to real values is typically assessed using the determination coefficient (R^2) and error estimation methods. Correlation analysis is useful for determining whether predicted and real data share similar trends. However, it is less common for authors to visually plot their results (i.e., predictions versus observations), which is crucial for confirming a linear correlation between both the two datasets. A high R^2 value close to 1 does not necessarily imply a good fit. To further validate the correctness of predicted data, employing fitness equations can be beneficial. This involves checking whether the intercept is statistically zero and the slope is one. Regarding error estimation, there was no consistent trend in how authors reported their results. Different error metrics, such as RMSE, mean absolute error (MAE), and mean square error (MSE), were used interchangeably.

Notably, when optimizing ANNs for specific applications, authors sometimes found that one option yielded a better correlation (R^2), while another option provided a lower prediction error. This can lead to somewhat arbitrary selection of optimal conditions. To address this, it is advisable to base the final selection on a compromise between both sets of values, potentially prioritizing a lower error over a higher R^2, as the latter can be affected by extreme values. In such cases, visualizing the data becomes crucial for making evidence-based decisions.

5. Conclusions

The application of ANNs in the photo-Fenton process for degrading organic compounds in wastewater samples has yielded promising results. ANNs effectively model the complex relationships among variables involved in these processes. Nevertheless, significant differences in outcomes are observed depending on the parameters and methodologies used. Some studies have indicated that models based on CCD-RSM achieve higher correlations than neural networks. This suggests that while ANNs are generally effective, they should not be the default choice without exploring other optimization approaches, such as surface response plots.

AI tools like ANNs are highly suitable for water research community, particularly in processes like the photo-Fenton reaction. By implementing ANNs with prior data, researchers can predict outcomes based on initial conditions, thereby optimizing processes more efficiently. This approach saves resources, reagents, energy, and time. Additionally, ANNs can provide information about leftover reagent concentrations, such as H_2O_2, allowing for better preparation and handling.

Declaration of competing interest

The authors declare no conflicts of interest.

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