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Optimization strategy to improve the removal efficiency of commercial herbicides using a multivariable inverse artificial neural network adapted with particle swarm optimization

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ABSTRACT

The present work is focused on the implementation of a new optimization strategy using a multi-variable inverse artificial neural network (ANNim) to increase the removal efficiency of commercial herbicides in a sonophotocatalysis process. This research contributes significantly for the removal of pollutants in aquatic ecosystems, reducing the chemical oxygen demand (COD). To carry out the strategy, it is necessary to develop an artificial neural network (ANN) model considering the multiple input variables of the process. The ANN model obtained satisfactory results, showing a coefficient of determination (R^2) of 0.9723 and a root mean square error equal to 0.0414. The training data was fitted with a Levenberg–Marquardt algorithm with a hyperbolic tangent sigmoid function in the hidden layer. Subsequently, an objective function is proposed using the coefficients generated by the ANN model to minimize the COD value. For the determination of optimal variables, this work adapted particle swarm optimization (PSO), obtaining the ANNim-PSO computational strategy. The hybridization of the ANNim model with the PSO algorithm was necessary to determine the optimal parameters in the shortest possible time, improving the rate of removal of the active ingredients of herbicides compared to other degradation methods. The results showed that by optimizing one variable at a time in a specific experimental test, it is possible to increase the removal efficiency of commercial herbicides from 84.1% to 100% due to the effect of the TiO_2 catalyst (250 mg/L) in 55 min. However, optimizing more than one variable at the same time, the elimination of commercial herbicides was achieved in less time, reaching 100% due to the combined effect of pH (5), TiO_2 (250 mg/L) and K_2SO_4 (3 mM) catalysts in 5 min. Finally, the optimal parameters imply a total removal of the active ingredients of commercial herbicides in a considerably short time due to the increase in the superficial concentrations, obtaining a better absorption of the energy produced by the effect of pH and the TiO_2, the deposition of K_2SO_4, and the effective combination of ultrasound with the photocatalysis process.

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Keywords: Multivariable inverse artificial neural network; Sonophotocatalysis; Commercial herbicides; Chemical oxygen demand removal; Water treatment

1. Introduction

In agriculture, the application of fertilizers and pesticides has improved the quality of the production of various crops, promoting plant growth and pest control. On the other hand, the application of herbicides has facilitated the work of the farmers in the cleaning of weeds in the sowing fields. However, in compliance with the demands of quality and competence during the sale of the products, the farmers abuse the use of these agrochemicals. Among the negative effects caused by the abuse of agrochemicals, are: significant changes in the composition and reduction of specific species of flora, pollution to the urban environment, toxicity in mammals affecting the endocrine systems, reproductive, neural, and immune liver, groundwater contamination [1–4]. Based on the negative effects mentioned, groundwater contamination is one of the most alarming. This is due to the direct impact that occurs in the food chain between flora and fauna, as well as the population.

Among the commercial herbicides, there are those of the selective type focused on eliminating weeds in a specific way, such as: the alazine (active ingredient: alachlor, atrazine) and gesaprim (active ingredient: atrazine). However, these commercial herbicides are often improperly applied by farmers, especially during the rainy season. For this reason, it is imperative to propose a method to remove the active ingredients that cause toxic effects in the water. In the literature, various methodologies have been reported to treat the degradation of these pollutants, as presented: Bahena et al. [5] applied to sonophotocatalytic process to remove the herbicides alazine and gesaprim using aqueous suspensions of TiO_2 under UV light. The results show high performance in decomposition, improving the photodegradation with the use of ultrasound, achieving the degradation of the active component in more than 90% for gesaprim and 100% for alazine after 150 min. Jiménez et al. [6] evaluated through solar photo-Fenton the contaminants of a mixture of herbicides (hierbamina and gesaprim) in wastewater using a compound parabolic collector. In the experimental tests, satisfactory results were found applying 10 mg/L of iron (Fe), obtaining the removal of the active ingredients with a mineralization of at least 60% of the original total organic carbon (TOC). Garza-Campos et al. [7] studied the coupling of solar photoelectro-Fenton with boron-doped diamond (BDD) and solar heterogeneous photocatalysis for the mineralization of an atrazine solution (prepared from a commercial herbicide). The results showed to obtain 80% mineralization of atrazine after 300 min. Yu et al. [8] employed mycelial pellets of *Aspergillus niger* Y3 to immobilize ZXY-2, forming a self-immobilized biomixture (SIB) to remove atrazine. The results showed that using the biomixture was possible to remove atrazine efficiently after 8 h under specific conditions of temperature, pH and initial concentration.

In the previously described works dedicated to the removal of active ingredients from commercial herbicides, the sonophotocatalysis method excels at obtaining a high removal performance of alazine and gesaprim. The operation of this process is complex due to the interaction of the multiple variables involved during the photocatalytic reaction. The determination of optimal parameters through experimental tests are potentially an alternative to improve the degradation of pollutants present in aqueous treatments. Consequently, this work proposes the development of a new computational strategy to optimize the sonophotocatalysis process by minimizing the value of the chemical oxygen demand (COD). The computational strategy is based on the approach of a multivariable inverse artificial neural network (ANNim), which consists of increasing the performance of a process using the coefficients generated by an artificial neural network (ANN) [9]. ANN's models have been widely recommended to simulate non-linear behaviors related to desalination in forward osmosis processes and humidification–dehumidification systems [10,11]. Therefore, an ANN model is used to simulate the COD value of the sonophotocatalysis process. The hybridization of computational models with meta-heuristic algorithms has been an excellent combination to achieve specific objectives related to process optimization. The particle swarm optimization (PSO) algorithm has the ability to provide optimal solutions in problems that cannot be differentiable, that are not regular or homogeneous, even with noise signals or variations during time dynamics. In the area of computational methods, the PSO algorithm has contributed significant advances, such as the computation of weights in artificial neural networks, time series analysis, and optimization in finance, among others [12–15].

The potential of the PSO algorithm to solve non-linear optimization problems as non-derivable using classical mathematical methods is favorable to couple with the ANN model. The coupling of the PSO algorithm has been developed in the adjustment of the weights and bias during the training of the ANN models, with advantages, such as: minimum time consumption with less number of experiments, less control effort, and the ability to solve problems of nonlinear optimization efficiently [16]. Among the investigations that have applied an ANN model combined with relevant training algorithms oriented to the prediction of desalination processes, there are those published by: Mahadeva et al. [17] implemented an artificial neural network to predict the production of a desalination plant based on reverse osmosis. The combination of the softmax-purelin training function in the hidden and output layers were effective. In the results, the ANN model obtained superior simulation values (R^2 = 99.4%, Error = 0.003) compared to existing models. Mahadeva et al. [18] presented a model for the performance prediction of desalination plants using an artificial neural network with an unconventional optimization algorithm in the adjustment of weights and bias. Six models were developed to more accurately simulate the performance of a reverse osmosis desalination plant, differently dividing the database for the ANN model learning process. In the results, a hybrid model of grey wolf optimizer based ANN (GWO-ANN) produced superior results

(R^2 = 98.9%, Error = 0.007) compared to other algorithms. In addition to these investigations, there are the hybridizations of the ANN-PSO model developed to predict processes of water treatment desalination plants. Specifically, predicting the performance of the reverse osmosis process has been favored with the ANN-PSO technique, which generates greater precision than existing ANN models (with conventional algorithms) because the PSO algorithm has turned out to be more suitable for searching for optimal solutions during the learning [16,19].

The advantages of optimizing desalination processes have been relevant, from the determination of optimal parameters to the reduction of operating costs, as reported by: Pai et al. [20] applied an artificial neural network for the design and optimization of a cyclic adsorption process through the proposal of two separate multi-objective functions to maximize purity and recovery, and to maximize productivity and purity. Zhang et al. [21] optimized a hybrid system of a novel desalination plant using a multi-objective particle swarm algorithm. The results show that the inlet air humidity ratio or seawater temperature increases the performance of the system. Mahadeva et al. [22] developed multiple computational models for the prediction and optimization of the membrane performance of the seawater reverse osmosis desalination plants. The authors conclude that particle swarm optimization assisted artificial neural networks obtained the best performance. Specifically, it is possible to find in the literature some research works focused directly on the optimization of processes to degrade herbicides, such as those developed by: Trovó et al. [23] described the optimization of a photo-Fenton process for the degradation of the herbicide paraquat. In the results, it was shown that by obtaining optimal concentrations of Fe and H_2O_2 the removal of the herbicide was increased. Zazou et al. [24] studied the comparison of the electrochemical oxidation of an herbicide proposing a parametric optimization. The results showed that the optimization of some experimental parameters was essential to generate a higher degradation efficiency. Dargahi et al. [25] analyzed experiments based on the central composite design to optimize parameters of an electrocatalytic process in the degradation of an herbicide. In the results, high COD and TOC removal efficiencies were generated using optimal conditions. Hamzaoui et al. [26] developed an ANNi model to optimize the reaction time of the sonophotocatalysis process, applying the same database as the present work. However, due to the limitation of optimizing one variable at a time, it was not feasible to demonstrate a significant improvement in the removal of herbicides. For this reason, the current work considers the determination of multiple variables at the same time.

Finally, this work aims to increase the degradation efficiency of commercial herbicides in the shortest possible time by optimizing a sonophotocatalysis process using a multivariable inverse artificial neural network (ANNim). The COD parameter was chosen as the output value to model and minimize in the optimization due to its property to indicate the removal of active ingredients during an aqueous treatment. The input variables used to train the ANN model are: reaction time, pH, initial concentration of herbicide, pollutant, ultrasound, ultraviolet radiation, catalysts (TiO_2, K_2SO_4) and solar radiation. The optimization variabes are pH, initial concentration of TiO_2, and K_2SO_4, established because they can be manipulated when starting the process operation. The new objective function is proposed in order to find more than one optimal parameter at the same time. To solve the multivariable function, a particle swarm optimization (PSO) algorithm was adopted to obtain a response in the shortest possible time.

In summary, the main contributions of this paper are:

- Propose a new optimization strategy capable of improving the degradation performance of a sonophotocatalysis process through the integration of computational models.
- Determine the combination of optimal parameters that minimize the chemical oxygen demand by developing a multivariable objective function.
- Achieve through the ANNim-PSO model the removal of 100% of the active ingredients of commercial herbicides in the shortest possible time concerning other methods of contaminant degradation in aqueous treatment.

2. Experimental

2.1. Material and sample preparation

Alazina (30/18 LM), consisting of alachlor, atrazine and formulation agents as active ingredients. Gesaprim (90 GDA) composed of atrazine and formulating agents. Herbicides were obtained through direct purchase with Syngenta Crop Protection Inc., (USA). Pure compounds of atrazine and alachlor were high-performance liquid chromatography (HPLC) grade (99.9%) and purchased from Sigma-Aldrich. TiO_2 (Degussa P25) and H_2SO_4 were analytical grade (Sigma-Aldrich). The chemicals were applied as purchased, without further purification. The distilled water was supplied by the company Baxter México S.A.

2.2. Ultrasound photodegradation procedure

This section summarizes the experimental procedure (the detailed information is described by the study of Bahena et al. [5]). The experimental photodegradation tests were carried out for each herbicide using a chemical reactor recirculating a volume of 250 mL and a flow rate of 5.63 L/min. The diagram of the photodegradation process integrating a jacketed ultrasonic cell (150 cm^3) is shown in Fig. 1. The photochemical reactor has an ultrasonic probe (500 W, 20 kHz, Cole-Parmer), which is controlled by temperature with the recirculation of water. The process also contains a polypropylene flow centrifugal pump (Cole-Parmer) and an ultraviolet lamp, which is 44 cm long and 3 cm in diameter (15 W, 352 nm, Cole-Parmer). A stream of oxygen was supplied to the experimental sample by the ultrasonic cell. The experimental samples were extracted at different degradation time intervals to analyze the concentration of atrazine and alachlor through HPLC. To control the experimental tests, it was necessary to carry out sampling, removing less than 10% of the total volume and filtering according to what was collected before the analysis. The chemical oxygen demand (COD) was determined using standard methods and tubes within the concentration

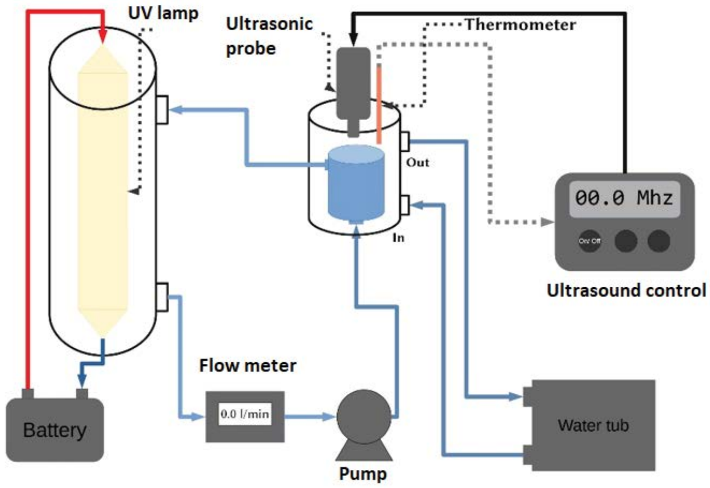


Fig. 1. General diagram of the photochemical reactor configured for the removal of herbicides using ultrasound.

Table 1
Experimental parameters used for the degradation of herbicides by a sonophotocatalysis process

N°	Input layer variable	Abbreviation	Execution interval
1	Reaction time (min)	Time	0–480
2	pH	pH	1–5
3	Initial concentration of herbicide	mM	0.1540–0.3090
4	Pollutant	Contaminant	1–9
5	Ultrasound (kHz)	Us	0–20
6	Ultraviolet radiation (nm)	UV	0–352
7	TiO_2 (mg/L)	TiO_2	0–300
8	K_2SO_4 (mM)	K_2SO_4	0–13
9	Solar radiation (W/m²)	SR	0–820

range of 0–150 mg/L and 0–20 mg/L [27]. Table 1 shows the parameters established in the experimental tests during the operation of the sonophotocatalysis process.

3. Simulation methodology using artificial neural networks

3.1. Development of ANN

The artificial neural network (ANN) is a computational tool used to simulate simple (linear) and complex (non-linear) behaviors. The most used type in the configuration of ANNs models is the multilayer perceptron with feedforward, applying one or more neurons in the hidden layer. The development of the ANN model is determined through three main layers, structured as: input layer, hidden layer and output layer. The data obtained from the experimental tests of the sonophotocatalysis process enter the input layer to generate coefficients (weights and bias) and later, transmit information (n_s) in the neurons of the hidden layer, as described in Appendix A.

Once the information has been transmitted in the hidden layer, transfer functions are applied to model the behavior internally. In this work, a hyperbolic tangent sigmoid (TANSIG) function was used, which produces an output with an interval of [–1 to 1]. In order to compare the adaptability of the data with the appropriate transfer function, the logarithmic sigmoidal (LOGSIG) function was applied, which generates an output with an interval from [0 to 1], given by Eqs. (2) and (3) in Appendix A. For the output layer, it is common to use a linear transfer function (PURELIN) to represent the simulated data (Out) by integrating the coefficients obtained during the learning process in the hidden and output layer (W_o, b_2). The network output can be represented using a TANSIG function, as follows:

$$\text{Out} = \text{PURELIN}\left(W_0 \times \left[\text{TANSIG}\left(\text{Wi}_{(s,k)} \times \text{In}_k + b_{1(s)}\right)\right] + b_2\right) \quad (1)$$

The learning process is a fundamental part of the development of the ANN model. To carry out this process effectively, the input variables were normalized in an interval of [0.1–0.9] in order to facilitate the obtaining of coefficients. The equations that represent the development of the ANN model, as well as the normalization of the data, are described in Appendix A. Fig. 2 shows a schematic of the ANN model using the experimental variables obtained from the sonophotocatalysis process.

In the training phase, the backpropagation algorithm (BP) effectively carries out the ANN model's learning process, adjusting the connections between feed-forward layers and with each layer sequential to the next. With the application of this algorithm, it is possible to minimize the error of the experimental value against the simulated value through the layers of the ANN. In this work, the experimental data were divided into training with 60%, test 20% and validation 20%. Among the backpropagation algorithms most used in ANNs models, the Levenberg–Marquardt

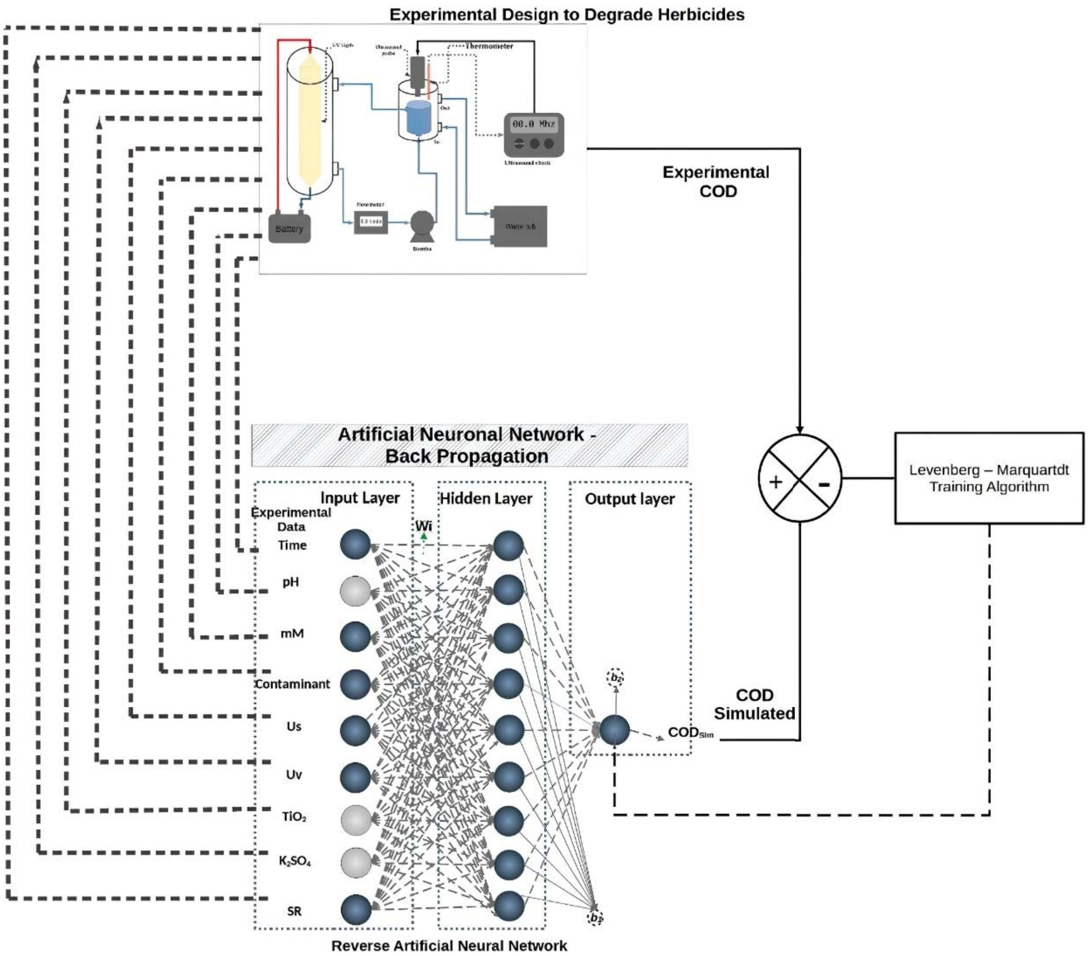


Fig. 2. Scheme of the procedure used for learning the ANN model based on the experimental variables of the sonophotocatalysis process.

optimization algorithm stands out, which has been applied to generate an excellent fit in water treatment processes and to establish the number of optimal neurons in the hidden layer in processes related to the elimination of turbidity in water [28,29]. The root mean square error (RMSE) was used to determine the minimum error between the input data and those generated by the ANN model during the learning process. Fig. 3 shows the diagram of the training cycle that the ANN follows until finding the minimum error of the simulated value and the experimental value. The development steps of the BP algorithm are based on the description provided in [30]. All the calculations applied for the development of the ANN model were performed with MATLAB R2014a (ANN toolbox).

3.2. Evaluation of the ANN model

The ANN model is validated through statistical criteria to provide certainty regarding the effectiveness of the simulation. The statistical criteria chosen were: the coefficient of determination (R^2) and the mean absolute percentage error (MAPE). These criteria corroborate the correlation of the experimental data with respect to the simulated data, as well as establish the difference between the two pairs of data. Eqs. (2) and (3) describe R^2 and MAPE, respectively.

$$R^2 = 1 - \frac{\sum_{i=n}^{n}\left(x_{\exp(i)} - y_{\mathrm{sim}(i)}\right)^2}{\sum_{i=n}^{n}\left(x_{\exp(i)} - x_{\exp(i)}\right)^2} \tag{2}$$

$$\mathrm{MAPE} = \frac{\sum_{i=1}^{n}\left|\frac{x_{\exp(i)} - y_{\mathrm{sim}(i)}}{x_{\exp(i)}}\right|}{n} \times 100(\%) \tag{3}$$

where $\bar{x} = \frac{1}{n}\sum_{i=1}^{n} x$ is the average of the value; $x_{\exp}$ is the experimental value and y_{sim} is the output value obtained by the model ANN.

The statistical criteria previously described can provide a correlation between experimental and simulated data. However, to determine the good fit of the ANN model, it is possible to apply the paired *t*-test to compare the means of two dependent variables. The paired *t*-test is defined by proposing the following hypotheses:

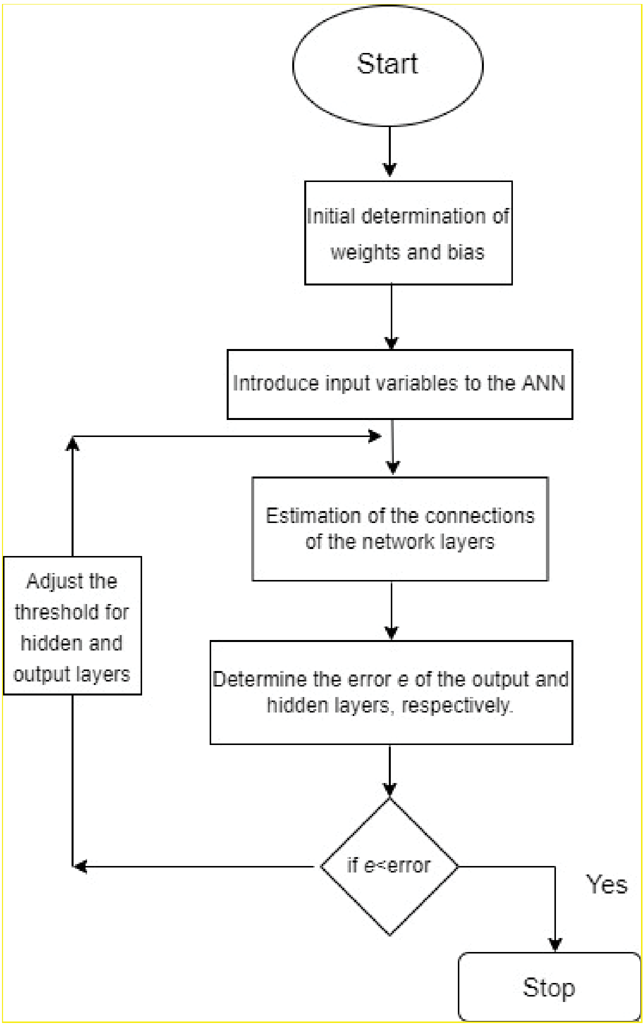


Fig. 3. Flowchart to represent the development of the back-propagation algorithm during the learning of the ANN model (based on Mahadeva et al. [30]).

The null hypothesis H_o is that the sample mean of the differences is zero. Therefore, there is no statistically significant difference between the experimental data and those obtained by the ANN model.

The alternative hypothesis H_i is that the sample mean of the differences is nonzero. Therefore, there is a statistically significant difference between the experimental data and those obtained by the ANN model.

For the null hypothesis accepted, the value obtained by Eq. (4) must be less than the critical value (T_c) obtained from tables [31].

$$t = \frac{\bar{d}}{s_{\bar{d}}} \tag{4}$$

where $s_{\bar{d}} = \sqrt{s_d^2 / n}$ and s_d is the variance of the difference between the experimental data and those obtained by the model ANN and $\bar{d}$ is the average of difference between each pair of data.

Table 2
Comparison of transfer functions and architectures to determine the best ANN model

Architecture ANN	Number of neurons	Epoch	RMSE	R^2	RMSE	R^2
			TANSIG		LOGSIG	
09-1-1	1	1,000	0.0999	0.7089	0.0945	0.789
09-2-1	2	1,000	0.0931	0.7348	0.0909	0.817
09-3-1	3	1,000	0.0888	0.7415	0.0837	0.8186
09-4-1	4	1,000	0.0659	0.7526	0.0696	0.821
09-5-1	5	1,000	0.0599	0.8073	0.0627	0.8496
09-6-1	6	1,000	0.0522	0.8325	0.0558	0.8549
09-7-1	7	1,000	0.0521	0.8856	0.0548	0.8365
09-8-1	8	1,000	0.0482	0.9568	0.0469	0.8436
09-9-1	9	1,000	0.0414	0.9723	0.0459	0.9665
09-10-1	10	1,000	0.0447	0.968	0.048	0.9630

3.3. Direct ANN model proposed to simulate the COD value

A feed-forward type ANN model was developed applying an architecture with nine neurons in the hidden layer. During the degradation of commercial herbicides, the process reached a COD value with an interval of [0.07–1], which was simulated by the ANN model. Table 2 compares different transfer functions and architecture configurations employed by the ANN model to determine the number of optimal neurons. During the learning process, finding the neuron number in the hidden layer is decisive to represent the output to be simulated. The simulation accuracy improves gradually by increasing the number of neurons in the hidden layer. The gradual exposure of the results generated by applying the neuron number from 1 to 10 in the hidden layer allows the identification of the optimal point and, in turn, the beginning of overlearning. Therefore, the optimal architecture according to the statistical criteria was 9-9-1 with a RMSE = 0.0414. Establishing the optimal number of neurons in the hidden layer has a significant role in the design of the ANNim model due to the direct relationship that exists with the precision of the simulation. This process is essential to obtain coefficients with a good fit and, in turn, provide reliable values for the development of the subsequent optimization process. The ANN model was fixed with a hidden layer, the implementation of a multilayer neural network may require more time and computational effort, degrading the speed of convergence [32–34]. This factor is critical for the effective fit of the coefficients and in the search for the optimal parameters when solving the objective function. The hyperbolic tangent transfer function demonstrated better adaptation to the data type compared to the logarithmic function.

With the optimal architecture confirmed, it is critical to verify that the partitioning of data into training, testing, and validation is adequate. Table 3 shows the results obtained by testing various percentages in the division of the data. The partition of 60% for training, 20% for testing, and 20% for validation proved to have the best impact in minimizing error and good correlation between the experimental and simulated data. Fig. 4 shows the agreement between

Table 3
Determining the best data partition for training the ANN model

Data division			Architecture ANN	Epoch	RMSE	R^2	RMSE	R^2
Train	Test	Validation			TANSIG		LOGSIG	
80%	10%	10%	09-9-1	1,000	0.0420	0.9718	0.0421	0.9713
70%	15%	15%	09-9-1	1,000	0.0440	0.9686	0.0443	0.9685
60%	20%	20%	09-9-1	1,000	0.0414	0.9723	0.0459	0.9665
50%	25%	25%	09-9-1	1,000	0.0544	0.9522	0.0505	0.9588

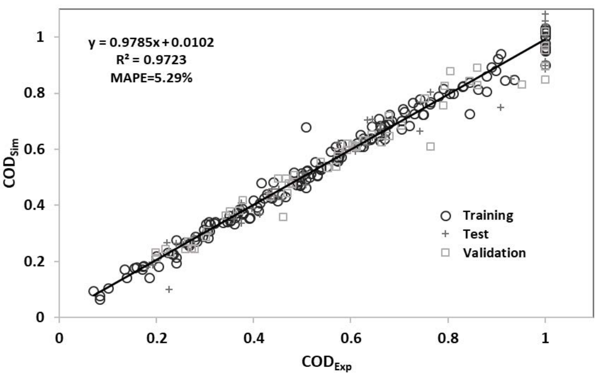


Fig. 4. Correspondence between experimental and simulated data through the coefficient of determination and mean absolute percentage error.

Table 4
Results of the paired t-test to determine the statistical significance between the experimental and simulated data

Paired t-test
t [Inflicted Eq. (4)] 0.76
T_c (Inflicted from statistical tables 95%) 1.96
Comparison 0.76 < 1.96
Null hypothesis H_0 is accepted "$\bar{x}_{\text{exp}} \approx \bar{y}_{\text{model}}$"

the experimental and simulated data with a R^2 = 0.9723 and MAPE value of 5.29%. Table 4 presents the results of the paired t-test, where the null hypothesis is accepted with a confidence level of 95%. Therefore, it is possible to conclude that the fit of the data was satisfactory, considering the mean of the experimental data and those obtained by the ANN model as similar.

Once the ANN model has been evaluated, it can be applied to represent the simulated data analytically (coupling the coefficients from Table 5), as shown by the following equation:

$$\text{COD} = \sum_{s=1}^{s}\left[W_{o_{(s)}}\left(\frac{2}{1+\exp\left(-2\left(\sum_{k=1}^{k}\left(Wi_{s,k}\times \ln_k\right)+b_{1_{(s)}}\right)\right)}\right)\right]+b_2 \tag{5}$$

where s is the neurons number in the hidden layer (s = 9), k is the neurons number in the input layer (k = 9), and $\text{Wi}_{(s,k)}$ and $b_{1_{(s)}}$ are weights and biases, respectively.

To verify the accuracy of the ANN model, Eq. (5) was programmed to simulate the COD value (as shown in Fig. 5). Four experimental tests with different operating conditions were chosen due to their effectiveness during the degradation of commercial herbicides. The good agreement between both data confirms that the ANN model is solid to continue with the optimization strategy.

Table 5
Coefficients obtained from the best ANN model

Neurons	Weights										Bias	
	Wi									W_o	b_1	b_2
S	k_1	k_2	k_3	k_4	k_5	k_6	k_7	k_8	k_9	Output layer		
1	–2.0970	0.6484	–3.2035	4.8537	–2.9265	–3.1246	1.5114	–2.7757	3.1004	2.8585	–3.5603	6.2647
2	–0.0510	–0.2693	0.8393	6.0571	6.7860	–0.5380	–0.5094	1.9449	1.5005	1.0870	–7.9669	
3	5.3752	4.3615	0.8149	4.7770	–5.2310	–1.5314	4.0720	–1.5212	–2.0809	–0.1806	–3.7015	
4	–1.0185	0.2531	–2.5121	4.8007	–1.3469	–2.0247	3.8597	2.1990	0.0730	5.3151	0.2341	
5	0.4070	–0.3006	0.2712	–0.3582	–6.6473	4.5353	3.7563	3.3936	–2.6900	–1.1609	–1.6706	
6	1.6445	–0.4368	1.1008	–5.2458	–2.5064	–5.9454	–1.2695	2.6211	0.6747	3.7514	2.6434	
7	–1.6707	–0.0584	–1.0165	1.8453	–7.6288	–0.8201	4.7612	–1.0470	1.1476	–2.0375	0.9336	
8	26.7558	–0.0881	0.1750	–0.0618	–0.1730	–0.1293	–0.2840	0.0089	0.4462	–4.3949	–0.9266	
9	1.7985	0.1848	–0.0218	0.1676	8.8390	–3.7220	–0.9209	0.3314	0.4636	–4.4213	0.4479	

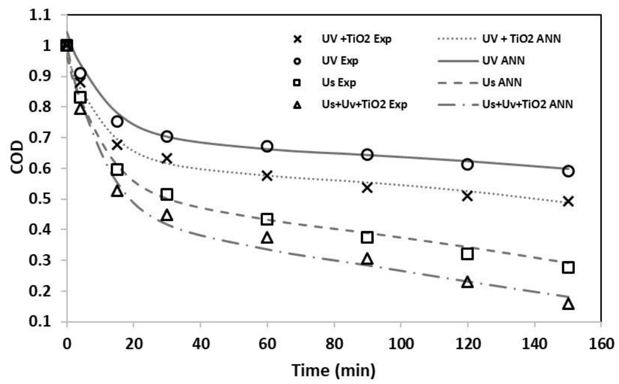
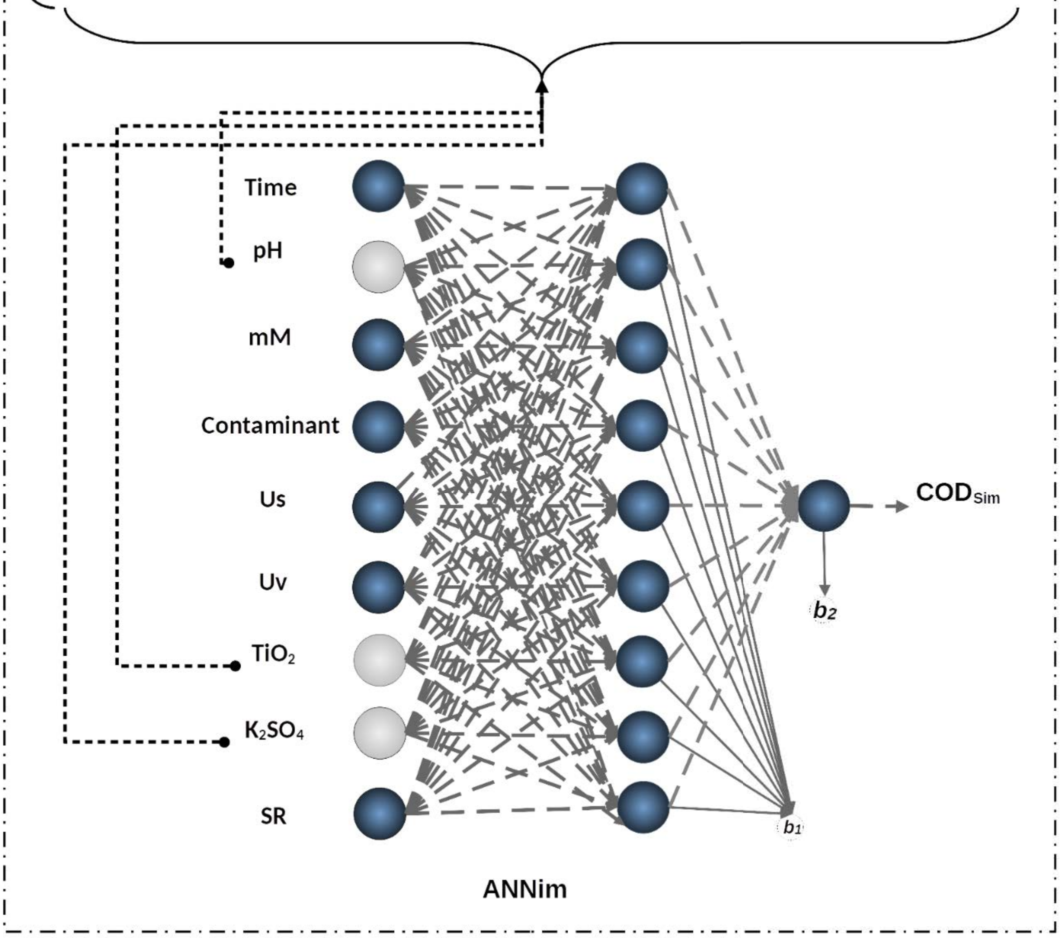


Fig. 5. Verification of the ANN model applied in four experimental tests with different operating conditions.

4. Optimization strategy methodology

4.1. Development of the ANNim model proposed to improve the sonophotocatalysis process

The development of the ANNim model consists of inverting the ANN model, previously trained in order to obtain the specified output value. This optimization strategy has been applied in thermal processes to increase the efficiency of an absorption thermal transformer, as well as to improve the performance of a solar parabolic trough collector [9,35]. The ANNim model is focused on providing a series of optimal parameters through a multivariable objective function. Fig. 6 shows that from the coefficients obtained during the training of the ANN model, the multivariable objective function is proposed. During this step, the variables to be optimized to improve the degradation process are



$$\mathrm{Fun}_{(\mathrm{pH,TiO_2,K_2SO_4})} = b2_{(j)} - \sum_{j=1}^{s}\left[\frac{2W0_{(j)}}{1+\exp(-2(Wi_{j,2}{*}\mathrm{pH} + Wi_{j,7}{*}\,\mathrm{TiO_2} + Wi_{j,8}{*}\,\mathrm{K_2SO_4}) + \sum_{k\neq 2,7,8}^{k} Wi_{j,k}{*}\ln_{(k)} + b1)}\right]$$

Fig. 6. Schematic diagram of the ANNim model proposed to increase the degradation efficiency of commercial herbicides by sonophotocatalysis.

defined. Eq. (6) describes the integration of the input variables of the sonophotocatalysis process, as well as the variables to optimize based on the desired COD.

$$\min\left(f_{\mathrm{pH},\mathrm{TiO_2},\mathrm{K_2SO_4}}\right) = -\mathrm{COD}_{\mathrm{deseado}} + \sum_{s=1}^{s}\left[W_{o(l,s)} \cdot \left(\frac{2}{1+e^{\left(-2\cdot\left(W_{i(s,2)}\cdot \mathrm{pH}_{(2)} \cdot W_{i,(s,7)} \cdot \mathrm{TiO}_{2(7)} + W_{i(s,8)} \cdot \mathrm{K_2SO}_{4(8)} + \sum_{k\neq 2,7,8}^{k} W_{i(s,k)} \cdot \mathrm{ln}_{(k)} + b1_{(s)}\right)\right)}} - 1\right)\right] + b_{2(l)} \tag{6}$$

In Eq. (6) pH, TiO_2 and K_2SO_4 were chosen as variables to optimize, because these parameters can be modified at the start of each experimental test. Another reason is the close relationship that catalysts present at different pH values with respect to the degradation of herbicides. The search restrictions applied for each variable to be optimized correspond to the minimum and maximum values used experimentally in order to contemplate all the conditions obtained from the process.

$$1 \le \mathrm{pH} \le 5 \tag{7}$$

$$0 \le \mathrm{TiO_2} \le 300 \tag{8}$$

$$0 \le \mathrm{K_2SO_4} \le 13 \tag{9}$$

In this work, the desired COD value must be reduced as a function of time to successfully reflect the increase of the removal efficiency applied to a specific experimental test. The proposed ANNim model is difficult to solve instantly by conventional algebraic methods. This is due to the number of coefficients used in each hidden layer and the number of variables to optimize. Therefore, to minimize the multivariable objective function, it is necessary to couple an approximation algorithm focused on generating a solution in the shortest possible time.

4.2. Particle swarm optimization

The swarm concept is based on the collective behavior derived from the social relationship between different animal species, such as birds, fish, bees, or ants, for example, where these individuals are taken as simple and homogeneous agents that perform basic tasks and the Interaction between these agents does not have a central control [36]. To find the global solution to the problem, the particles or agents define trajectories in the solution space, within this space, it is iteratively updated based on the equation of motion, which is described as:

$$x_i(t+1) = x_i(t) + v_i(t+1) \tag{10}$$

From Eq. (10) different parameters are defined, among them are the inertia or moment of the particle's movement. The second-term is known as the cognitive component from this it is explained that the particles return to their best positions previously found. The last of these components is the social component from this the tendency of a particle to move to the best position of the whole of the swarm is identified.

$$v_i(t+1) = \omega(t+1)v_i(t) + c_1\left(p_i - x_i(t)\right)R_i + c_2\left(g - x_i(t)\right)R_2 \tag{11}$$

In addition, the algorithm has two constants called c_1 and c_2, these are found within the real numbers, with these coefficients the magnitude of the steps of the particles is regulated in the best of the directions, taking the best personal and global mark in the gutter.

4.3. Adaptation of the PSO algorithm to solve the ANNim model

Fig. 7 shows the adaptation of the PSO algorithm to solve the multivariable objective function [Eq. (6)]. With this coupling, the best solution can be determined in a short computational time. The PSO algorithm starts by setting initial parameters for the particles corresponding to the first generation. Subsequently, these will move according to inertia and the best position within the space of possible solutions. The ANNim-PSO adaptation is efficient when the multivariable objective function is minimized as close to zero, obtaining a series of optimal parameters that satisfy the desired COD value.

The justification for coupling the PSO optimization algorithm depends on its balanced effectiveness in solving objective functions, which may contemplate a search with restrictions or specifications. On the other hand, it is possible to couple new advanced optimization algorithms such as GWO and Whale Optimization Algorithms (WOA) in future research. However, when choosing an optimization algorithm, it is necessary to consider the convergence of the solutions, the precision, and, above all, the computation time. Therefore, this research applied the PSO algorithm due to the stabilization that characterizes it between convergence and computational performance in the search for optimal parameters.

5. Results and discussion

5.1. Validation of the adaptation of the PSO algorithm to the ANNim model

The validation of the ANNim-PSO model is an important procedure in order to guarantee the correct application of the optimization strategy. The PSO algorithm is subjected to a rigorous demonstration, which consists of determining all the input variables in a specific experimental test based on the desired COD value. The statistical criterion of mean absolute error (MAE) was applied as a measure to determine the error between the experimental data and those found by the ANNim-PSO model. Table 6 shows a comparison between the experimental data and those obtained by the ANNim-PSO model, gradually establishing the total

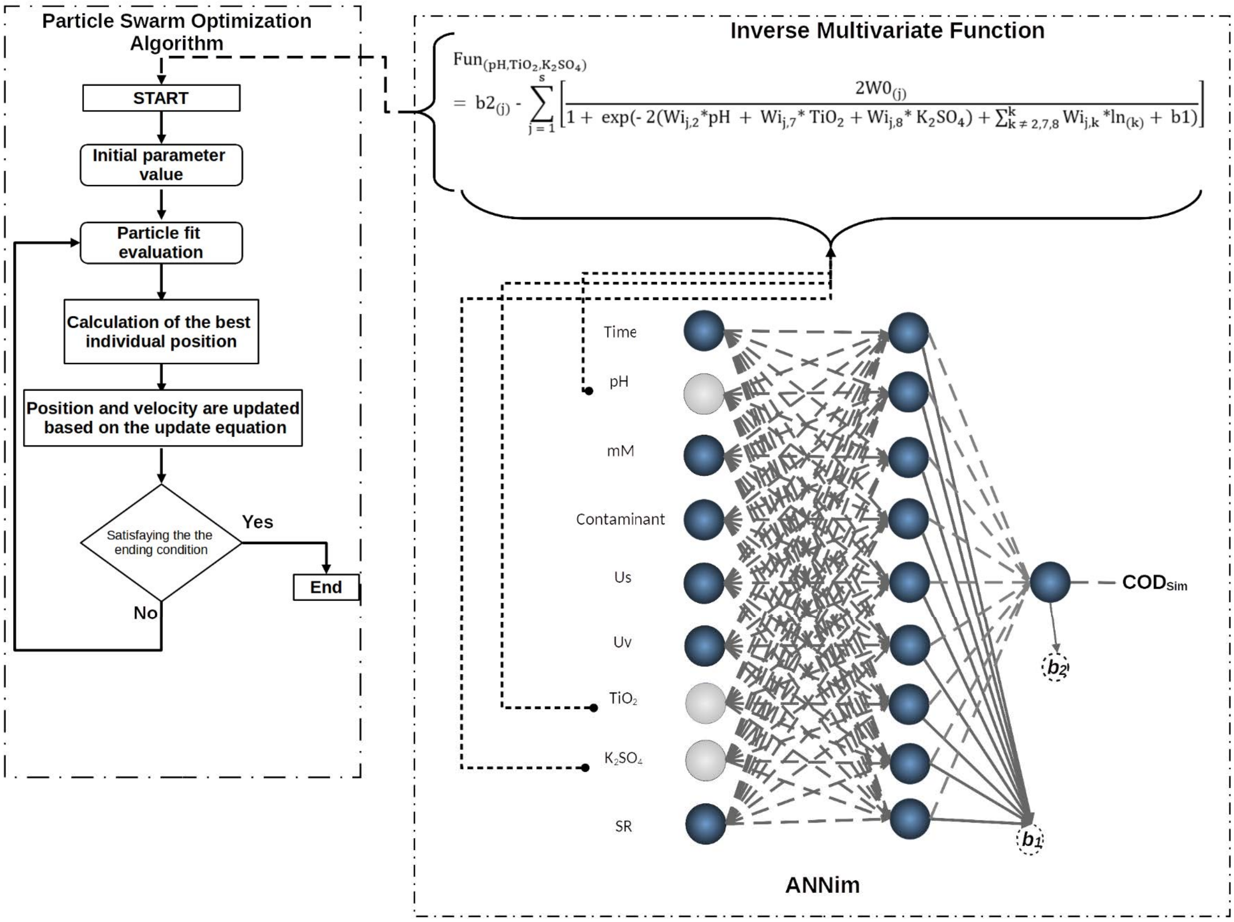


Fig. 7. Scheme of the integration of the PSO algorithm to the ANNim model to determine the multiple optimal input variables of the sonophotocatalysis process.

of the input variables of the process. The validation of the ANNim-PSO model had a maximum error and a computational time that increased according to the number of variables, reaching a value of 0.426 and 2.52 s. However, the maximum error obtained when optimizing three variables at the same time was 0.136, which is satisfactory considering the number of variables to optimize in this work. With these criteria, it is possible to corroborate the correct adaptation of the ANNim-PSO model to continue with the application focused on increasing the removal efficiency of the sonophotocatalysis process.

5.2. Optimization of one variable at a time using ANNim-PSO

From Fig. 6, the ANNim-PSO model was applied to the experimental test that obtained the minimum value of COD. Table 7 shows in detail the experimental parameters used during the removal of herbicides with the best minimization of COD. The parameters to optimize were: pH and the concentration of the catalysts (TiO_2 and K_2SO_4). These variables were chosen because they can be manipulated at the beginning of each experimental test and, furthermore, these parameters were not modified in the research's by Bahena et al. [5] and Hamzaoui et al. [26]. The application of the ANNim-PSO model was gradual in this section, optimizing one variable at a time. Fig. 8 shows the minimization of the COD value by optimizing a) the pH value, b) the initial concentration of TiO_2 and c) the initial concentration of K_2SO_4. With the results generated, it is important to highlight that each of the optimized variables improved the removal of herbicides compared to those carried out experimentally (~84% removal). The determination of the optimal initial concentration of TiO_2 achieved a removal of 100% in 55 min when using 250 mg/L as opposed to the 200 mg/L established experimentally. Subsequently, the optimal initial concentration of 3 mM by the K_2SO_4 catalyst reached a 100% removal in 115 min. Finally, the optimum value of pH = 5 achieved a removal of 88% after 150 min.

The optimum value of TiO_2 had a greater impact on the other parameters in the removal of the ingredients because the ultraviolet light provides the necessary energy on the appropriate surface of the catalyst, improving the rate of the chemical reaction. The removal of contaminants obtained from the optimal concentration of the K_2SO_4 catalyst is total, due to the stabilization it provides in the deposition during the production of anionic species resulting from degradation. The degradation of the contaminants was not complete when determining only the optimal pH value. However,

Table 6
Validation of the adaptation of the PSO algorithm to the ANNim model

Experimental data	Time	pH	Concentration	Pollutant	Ultrasound	Ultraviolet radiation	TiO_2	K_2SO_4	Solar radiation	COD_{EXP}	COD ANNim-PSO	MAE Error	Computing Time (s)
Number of variables	60	2.3	0.193	0.9	20	0	0	0	0	0.576			
1	60	–	–	–	–	–	–	–	–		0.57640	0	0.3582
2	60	2.31	–	–	–	–	–	–	–		0.57610	0.005	0.6358
3	60	1.9	0.2	–	–	–	–	–	–		0.57610	0.136	0.8964
4	60	3.333	0.154	0.8183	–	–	–	–	–		0.57600	0.288	1.139
5	60	3.2293	0.1593	0.895	20	–	–	–	–		0.57600	0.194	1.4
6	60	1.7156	0.154	0.7549	20	0.4073	–	–	–		0.57600	0.196	1.69
7	60	1.2335	0.154	0.7563	19.9954	0.1759	0.9038	–	–		0.57600	0.333	1.9552
8	60	3.3544	0.154	0.9	20	0	0.5353	0.0363	–		0.57600	0.208	2.196
9	60	3.343	0.154	0.9	20	0.8874	0.9024	0.6232	0.3352		0.57600	0.426	2.52

it overcomes the experimentally obtained degradation, allowing the molecules to absorb more energy from visible and ultraviolet light.

5.3. Multivariable optimization using ANNim-PSO

Table 8 presents the application of the ANNim-PSO model to minimize the COD value by optimizing two variables at the same time. The variables chosen were in consideration of the previous results, where the TiO_2 value achieved a total removal in the shortest possible time and the pH value in order to corroborate the activation it causes in the catalyst. The results indicate that supplying an optimum value with a pH of 5 and a concentration of the TiO_2 catalyst of 250 mg/L is feasible to obtain a removal of 100% in 27 min. This optimization with two variables produces an effect that favors conditions to increase the light absorption in the TiO_2 molecules, which allows degradation in less time.

Subsequently, three input variables of the process were optimized at the same time. Table 9 shows the increase in herbicide removal optimizing three input variables of the process by applying the ANNim-PSO model. The determination of the three optimal variables achieves a total removal in 5 min using a pH of 5, an initial concentration of TiO_2 of 250 mg/L and 3 mM of K_2SO_4. Fig. 9 shows the experimental test with the best COD reduction reported by Bahena et al. [5] and the data optimized by the ANNim-PSO model. The effect of the combination of the optimal parameters significantly improves the rate of the reaction. The absorption of energy produced by the pH and the TiO_2 accelerates the degradation. The optimal value of the K_2SO_4 concentration regulates the superficial acidity controlling the production of the resulting species and the redox capacity. In this way, the application of the optimization strategy allowed extrapolating results that were not experimentally tested, obtaining better removal values and in less time.

Table 10 shows the application of other methods of removal of commercial herbicides in harassment treatment compared to the present work. In agreement with other research works presented in Table 9, the current work obtained good results when a pH value of 5 is established in the presence of the TiO_2 and K_2SO_4 catalysts.

Finally, the fruitful findings of this research can be summarized as follows:

- The new optimization strategy for the degradation of active ingredients in commercial herbicides turned out to be adequate, minimizing the value of COD by integrating powerful computational models such as neural networks and meta-heuristic optimization algorithms (obtaining a value of MAE = 0.426 and a time of 2.52 in the validation of the hybridization of the ANNim-PSO model).
- The hybridization of the ANNim-PSO models produced an optimal combination of parameters (using a pH of 5, an initial concentration of TiO_2 of 250 mg/L and 3 mM of K_2SO_4) that allowed solving an objective function, which consisted in exploring parameters not tested during the experimental process.
- The total removal of the active ingredients of commercial herbicides was in a short time (5 min) compared to other degradation methods (especially the one reported by Cabrera et al. [41]).

Table 7
Experimental data applied to optimize the COD value using the ANNim-PSO method

Time	pH	Concentration	Pollutant	Ultrasound	Ultraviolet radiation	TiO_2	K_2SO_4	Solar radiation	COD_{EXP}	Removal (%)
0	2.3	0.193	0.9	20	352	200	0	0	1	0
4	2.3	0.193	0.9	20	352	200	0	0	0.794	20.6
15	2.3	0.193	0.9	20	352	200	0	0	0.526	47.4
30	2.3	0.193	0.9	20	352	200	0	0	0.449	55.1
60	2.3	0.193	0.9	20	352	200	0	0	0.374	62.6
90	2.3	0.193	0.9	20	352	200	0	0	0.306	69.4
120	2.3	0.193	0.9	20	352	200	0	0	0.229	77.1
150	2.3	0.193	0.9	20	352	200	0	0	0.159	84.1

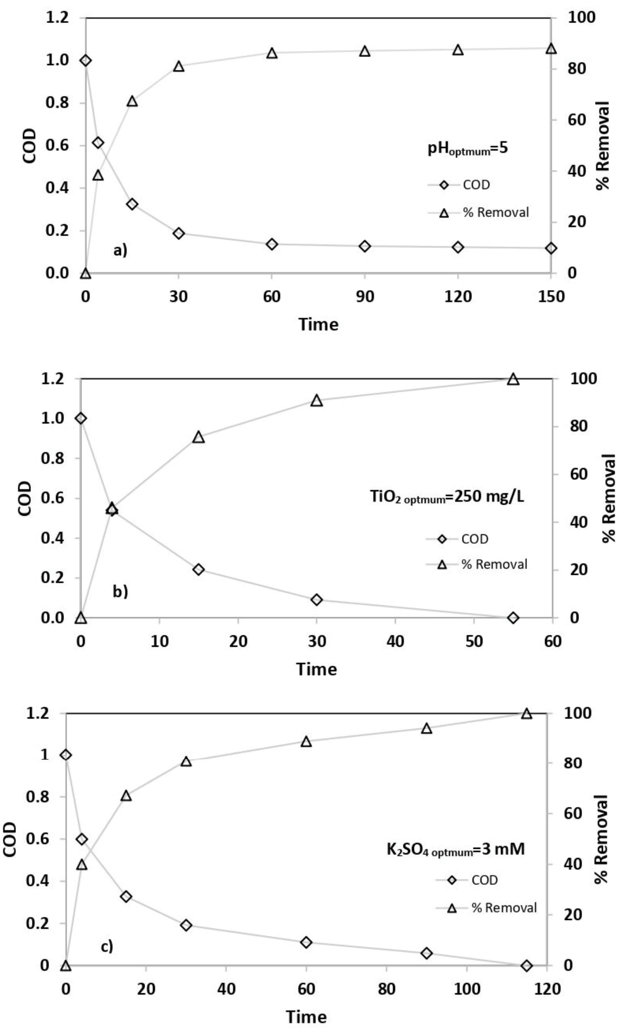


Fig. 8. Application of the ANNim-PSO model to improve herbicide removal efficiency by optimizing one variable at a time; (a) the pH value, (b) the initial concentration of TiO_2 and (c) the initial concentration of K_2SO_4.

6. Conclusion

This paper presents the optimization of a sonophotocatalysis process based on a new optimization strategy, which increases the removal efficiency of commercial herbicides in aqueous treatment. The ANNim optimization model was developed to determine the optimal input values of the process, which were: pH and concentration of catalysts (TiO_2 and K_2SO_4). The objective function was proposed as a function of reducing the COD value applied in the best test obtained experimentally. To achieve this, an ANN model was trained using the input variables of the process and as an output to simulate the COD value. Considering that the ANN model must have good precision and, in turn, generate a number of adequate coefficients to carry out the optimization strategy effectively, it was enough to train an architecture with a hidden layer. The best representation was obtained by integrating 9 neurons and applying a hyperbolic tangent sigmoid function (TANSIG) in the hidden layer, reaching an R^2 of 0.9723 and a MAPE of 5.29%. To demonstrate the good representation of the ANN model, it was concluded by means of a paired *t*-test that there is no significant difference when comparing the mean of the real and simulated data. The simulation's precision allows minimizing the COD value through the optimization strategy with strict adherence to the actual operating conditions of the sonophotocatalysis process.

The coefficients generated from the ANN model were later used for the development of the ANNim model. However, the objective function when considering multiple variables to optimize at the same time, it was necessary to adapt a particle swarm optimization (PSO) algorithm in order to search for optimal parameters in the shortest possible time. The choice of this algorithm rests on the speed with which it converges to find an optimal value. The ANNim-PSO model was applied in a specific experimental test to verify its effectiveness in the search for optimal parameters. In this step, the correct coupling between the ANNim model and the PSO algorithm was reflected, determining all the input variables of the process in a time of 2.52 s and an MAE of 0.426.

Once the good coupling of the ANNim-PSO optimization strategy had been validated, the removal efficiency was gradually increased using the best experimental test obtained during the operation of the process. Optimizing one variable at a time, the determination of the initial concentration of the TiO_2 catalyst obtained a removal efficiency of 100% in a time of 55 min, followed by the concentration of the K_2SO_4 catalyst in a time of 115 min and the pH with 88.17% removal in a time of 150 min. The specification of

Table 8
Increase in the removal efficiency of herbicides by determining two optimal values: pH and the concentration of the TiO_2 catalyst

Time	Optimal pH	Concentration	Pollutant	Ultrasound	Ultraviolet radiation	Optimal TiO_2	K_2SO_4	Solar radiation	COD ANNim-PSO	Removal (%)
0	5	0.193	0.9	20	352	250	0	0	1	0
4	5	0.193	0.9	20	352	250	0	0	0.4344	56.56
15	5	0.193	0.9	20	352	250	0	0	0.1277	87.23
27	5	0.193	0.9	20	352	250	0	0	0	100

Table 9
Increase in the removal efficiency of herbicides by determining three optimal values: pH and the concentration of catalysts (TiO_2, K_2SO_4)

Time	Optimal pH	Concentration	Pollutant	Ultrasound	Ultraviolet radiation	Optimal TiO_2	Optimal K_2SO_4	Solar radiation	COD ANNim-PSO	Removal (%)
0	5	0.193	0.9	20	352	250	3	0	1	0
1	5	0.193	0.9	20	352	250	3	0	0.19034	80.96
2	5	0.193	0.9	20	352	250	3	0	0.13859	86.14
3	5	0.193	0.9	20	352	250	3	0	0.09065	90.93
4	5	0.193	0.9	20	352	250	3	0	0.04627	95.37
5	5	0.193	0.9	20	352	250	3	0	0	100

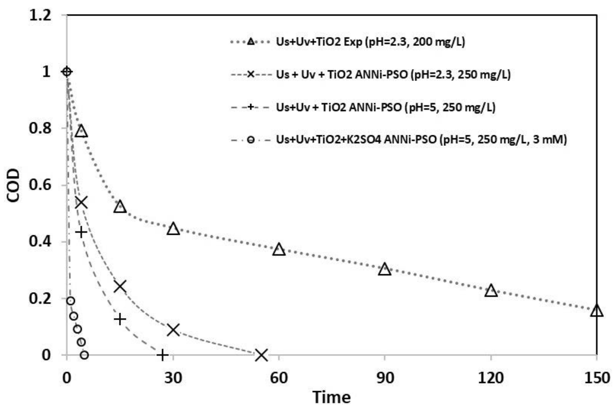


Fig. 9. Reduction of the COD value profile of commercial herbicides as a function of time applying the variation of the parameters experimentally and those obtained by the ANNim-PSO model.

the catalyst concentration TiO_2 = 250 mg/L could generate the total removal of the active ingredients of the commercial herbicide according to the ANNim-PSO model. A possible explanation for the significance of the optimal value consists of the interaction of UV light scattering in high concentrations of TiO_2, leading to a decrease in the surface area to absorb light and accelerate the reaction. The optimal presence of the catalyst concentration K_2SO_4 = 3 mM can improve the degradation of pollutants by leveling the surface acidity and redox capacity. On the other hand, the optimum value of pH = 5, indicates that the degradation is faster in acid conditions, favoring the speed of the catalytic reaction.

Optimizing two variables at the same time, 100% removal was achieved in a time of 27 min due to the effect of pH on the TiO_2 catalyst. The optimal combination of pH = 5 with a TiO_2 concentration = 250 mg/L may have points to consider on the catalyst surface, especially for the effective adsorption of the compound. In addition, it is possible to present an increase in the number of photons absorbed in UV light due to the higher concentration of the catalyst.

Optimizing three variables, 100% removal was obtained in 5 min due to the effect of pH on both catalysts (TiO_2 and K_2SO_4). The addition of the optimal concentration of K_2SO_4 = 3 mM in the combination obtained from pH = 5 and TiO_2 = 250 mg/L produced a total removal of the active ingredients, reporting the shortest time compared to other published works. This result can be justified by the impregnation of K_2SO_4 increasing the superficial concentrations of TiO_2, based on the decomposition of the atomic concentration of potassium. The deposition of K_2SO_4 on the catalyst could involve direct intervention in the exposure of active sites during the adsorption process, improving the reaction rate. Therefore, the photodegradation of the active ingredients of the commercial herbicides was forced through the optimal parameters found and conserving the effective use of ultrasound and photocatalysis.

Finally, it is important to highlight that the results obtained are the product of combinations of parameters determined by the ANNim-PSO optimization strategy and must be validated with future experiments. However, developing the optimization strategy in other processes can support the deduction of possible results by determining multiple optimal parameters to improve the operation of technologies related to desalination and water purification.

Table 10
Comparison of herbicide removal in accuse treatment applying other removal methods with respect to the sonophotocatalysis, method optimized by ANNim-PSO

Removal method reference	Herbicide removal method	Herbicide	Removal time	Particular parameters
Dugandžić et al. [37]	Photocatalytic degradation	Nicosulfuron	30 min	UV light with 315–400 nm; initial TiO_2 optimal concentration was 2 g/L; pH = 5.0
Piera et al. [38]	Catalyzed ozonation	2,4-dichlorophenoxyacetic acid	~20 min	UVA light in presence of Fe(II); pH = 2.6; $[TiO_2]$ = 2 g/dm^3; Fe(II) = 10^{-3} mol/dm^3
Abdennouri et al. [39]	Photocatalysis	2,4-dichlorophenoxy-acetic acid (2,4-D) and 2-(2,4-dichlorophenoxy) propionic acid (2,4-DP)	90 min	UV light with 365 nm; mass ratio of 3%-Pt/TiO_2
Verma et al. [40]	Photocatalysis	Isoproturon	120 min	UV light with 365 nm; TiO_2 loading 0.5 g/L; pH = 5.0
Cabrera et al. [41]	Fenton process	Diuron	10 min	pH = 2.5; 2 g of iron; 2 mmol/L H_2O_2
Present work	Sonophotocatalysis process optimized by ANNim-PSO	Alazina; Gesapim	27 min	Ultrasound source of 20 kHz; pH = 5; TiO_2 catalyst concentration of 250 mg/L
			5 min	Ultrasound source of 20 kHz; pH = 5; TiO_2 catalyst concentration of 250 mg/L ; K_2SO_4 catalyst concentration of 3 mM

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A

The development of an artificial neural network is described by following equations.

$$n_s = \mathrm{Wi}_{(s,1)} \times \ln_1 + \mathrm{Wi}_{(s,2)} \times \ln_2 + \ldots + \mathrm{Wi}_{(s,k)} \times \ln_k + b_{1(s)} \tag{A1}$$

where k represents the number of input variables, In is the input variable, s is the number of neurons in the hidden layer, Wi are weights generated between the input layer and the hidden layer and b_1 is the bias produced by each neuron applied in the hidden layer.

The transfer functions are given by the following:

$$f(x) = \mathrm{TANSIG} = \frac{2}{1+e^{-2n_s}} - 1, \quad -1 < f(x) < 1 \tag{A2}$$

$$f(x) = \mathrm{LOGSIG} = \frac{2}{1+e^{-n_s}}, \quad 0 < f(x) < 1 \tag{A3}$$

The data used for the training of the ANN model were normalized by the following equation:

$$\ln_{\mathrm{Norm}} = 0.8 \times \left(\frac{\ln_{\exp} - \ln_{\min}}{\ln_{\max} - \ln_{\min}} \right) + 0.1 \tag{A4}$$

where $\mathrm{In}_{\exp}$ represents the experimental value obtained from the process, $\mathrm{In}_{\min}$ is the experimental minimum value, $\mathrm{In}_{\max}$ is the maximum experimental value and $\mathrm{In}_{\mathrm{Norm}}$ is the normalized value.