Modeling of Amicarbazone Herbicide Degradation by Ozonation Process Assisted with UV Light using Neural Artificial Networks

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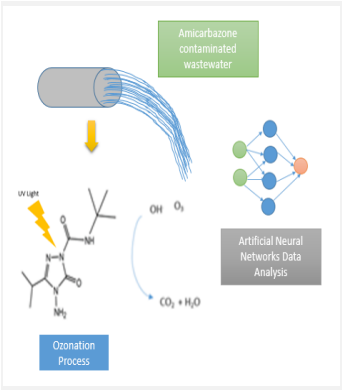

In this work, the degradation of amicarbazone herbicide, in aqueous medium, using the UV-assisted ozonation (UV/O_3) process, was modelled applying the artificial neural networks (ANN). In the first step, the experimental data were treated in order to find outliers. After this, the dataset was randomly splitted into 80% and 20% for training and testing stages, respectively. The independent variables considered were the initial total organic carbon concentration (TOC), initial pH, UV lamp power applied, ozone concentration in the inlet stream of the reactor and the reaction time, in order to predict the TOC (dependent variable). Tests were executed in order to find the best configuration for the ANN. The best results were obtained with 10 neurons in the hidden layer. In this scenario, the coefficient of determination was 0.984, considering the entire database.

Introduction

Herbicides are highly toxic, making the application of conventional processes unfeasible for the treatment of wastewaters containing these types of pollutants. In this context, the advanced oxidative processes (AOP) emerge as an important alternative. AOP are based on the production of free radicals, highlighting the hydroxyl radical (·OH), which has a high standard reduction potential [1]. The generation of hydroxyl radicals can occur using different methods, such as ozone photolysis by UV radiation (O_3/UV) [2].

Meanwhile, it should be noted that the development of mathematical models is vital for different industrial chemical processes. This is due to the importance of simulation for the analysis and development of processes. Mathematical models make it possible, for example, to carry out appropriate simulations, enabling the prediction of various operational conditions and their respective responses [3].

O_3/UV process presents several chemical chain reactions, in addition to involving the mass transfer of ozone from the gaseous phase to the aqueous medium. Thus, it is a complex system to apply phenomenological modeling, including because it requires knowledge of the field of UV radiation.

So, the present work consisted of modeling, using the technique of artificial neural networks (ANN), an empirical modeling method, the O_3/UV process in the degradation of amicarbazone, in an aqueous medium, which is an important herbicide [4].

Material and Methods

In the present work, experimental data, previously obtained by researcher J. P. V. Garcia (2013) [5], were used, involving the degradation of amicarbazone by the O_3/UV process. In these tests, the following variables were studied: initial total organic carbon content: 11.3 – 48.5 mgC/L (TOC_0), concentration of ozone in the inlet stream of the reactor: 0 – 31.6 mg/L ($[O_3]_0$), initial pH: 4.0 – 10.0 (pH_0) and the nominal power consumption of the UV lamps applied: 0 - 75 W (P_{UV}). As a dependent variable (response), the total organic carbon content (TOC) was adopted, measured over the reaction time (90 min). The trials were carried out in a photochemical reactor, operating in a batch regime.

The experimental data were treated to guarantee their quality, aiming to eliminate possible outliers. Then data were randomly divided into two groups: 80% for the training and 20% for the testing sets. To obtain the optimized ANN, the following topology was adopted: an input layer, a hidden layer and an output layer. The number of neurons in the hidden layer was varied, with the aim of obtaining the simplest possible network. To choose the best neural network, different metrics were used, such as the coefficient of determination (R^2). Furthermore, some simulations were carried out, in order to verify whether the optimized models presented overfitting problems. The ANN obtained was generated using the “nnet” function, applying the RStudio® software.

Results and Discussion

The dataset presented the following independent variables: reaction time, initial total organic carbon content, initial pH, nominal power of the UV radiation source and the concentration of ozone in the inlet stream of the photochemical reactor.

In the determination of the optimized ANN, it was observed that the best scenario resulted in 10 neurons in the hidden layer. Figure 1 presents the TOC results, predicted by the ANN model, versus the experimental values for the training (a) and testing (b) sets and for all experimental data (c). It can be

observed that for the training and testing sets, the optimized ANN presented coefficients of determination equals to 0.985 and 0.979, respectively. Furthermore, it can be seen that the ANN was able to satisfactorily predict the behavior of the studied process, explaining around 98.4% of the variation observed for the all experimental values.

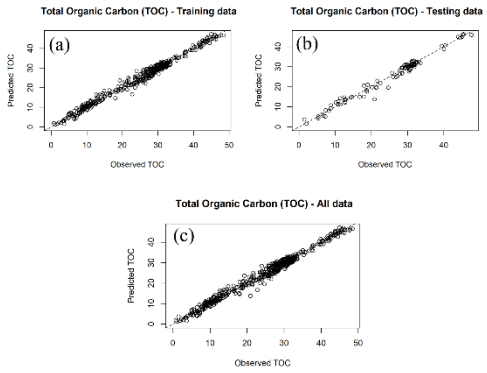

Figure 1. Predicted and observed TOC values for training set (R^2 = 0.985), testing set (R^2 = 0.979) and all data (R^2 = 0.984)

With the aim of analyzing the behavior of the obtained model, some simulations were carried out, under some experimental conditions, of the TOC profile throughout the reaction time, comparing with experimental data. In Figure 2, the simulation behavior of the ANN model can be observed, under the following conditions: (a) TOC_0 = 31.2 mgC/L; $[O_3]_0$ = 18.1 mg/L; pH_0 = 10 and P_{UV} = 75 W and (b): TOC_0 = 30.5 mgC/L; $[O_3]_0$ = 31.6 mg/L; pH_0 = 10 and P_{UV} = 75 W. According to the results presented in Figure 2, it can be seen that the proposed model, via artificial neural networks, was able to represent, in a very satisfactory way, the studied process.

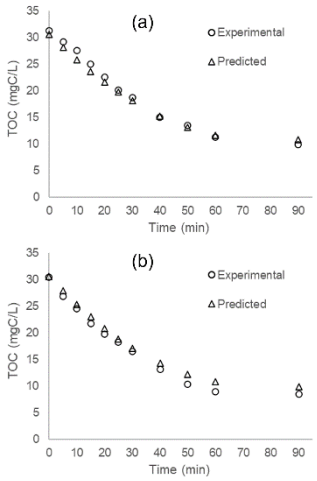

Figure 2. Simulations of the ANN model for TOC, along the reaction time, and experimental values: (a) TOC_0 = 31.2 mgC/L; $[O_3]_0$ = 18.1 mg/L; pH_0 = 10; P_{UV} = 75 W and (b): TOC_0 = 30.5 mgC/L; $[O_3]_0$ = 31.6 mg/L; pH_0 = 10 and P_{UV} = 75 W

Conclusions

The present work consisted of applying the ANN technique to model the amicarbazone degradation by O_3/UV process, using the free software RStudio®. In a first step, the experimental dataset was analyzed in search of outliers, with no anomalous values being found. Then, the ANN technique was applied, using the "nnet" function, adopting the following topology: an input layer, a hidden layer and an output layer. After several analyses, the optimized number of neurons in the hidden layer resulted in 10, with 71 optimized parameters, 60 weights and 11 biases. The optimized ANN presented a coefficient of determination equal to 0.984 between the all experimental TOC values and those predicted by the model.

Acknowledgments

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