

Article

Photocatalytic and Photo-Fenton-like Degradation of Cationic Dyes Using $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ Under LED Irradiation: Optimization by RSM-BBD and Artificial Neural Networks (ANNs)

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Abstract: The development of heterostructures incorporating photocatalysts optimized for visible-light activity represents a major breakthrough in the field of environmental remediation research, offering innovative and sustainable solutions for environmental purification. This study explores the photocatalytic capabilities of a $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ heterojunction nanocomposite, successfully synthesized from graphitic carbon nitride ($\text{g-C}_3\text{N}_4$) and tin ferrate (SnFe_2O_4) and applied to the degradation of the cationic dye brilliant cresyl blue (BCB) in an aqueous solution. These two components are particularly attractive due to their low cost and ease of fabrication. Various characterization techniques, including XRD, FTIR, SEM, and TEM, were used to confirm the successful integration of SnFe_2O_4 and $\text{g-C}_3\text{N}_4$ phases in the synthesized catalysts. The photocatalytic and photo-Fenton-like activity of the heterojunction composites was evaluated by the degradation of brilliant cresyl blue under visible LED illumination. Compared to the pure components SnFe_2O_4 and $\text{g-C}_3\text{N}_4$, the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposite demonstrated a superior photocatalytic performance. Furthermore, the photo-Fenton-like performance of the composites is much higher than the photocatalytic performances. The significant improvement in photo-Fenton activity is attributed to the synergistic effect between SnFe_2O_4 and $\text{g-C}_3\text{N}_4$, as well as the efficient separation of photoexcited electron/hole pairs. The recyclability of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ composite toward BCB photo-Fenton like degradation was also shown. This study aimed to assess the modeling and optimization of photo-Fenton-like removal BCB using the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanomaterial. The main parameters (photocatalyst dose, initial dye concentration, H_2O_2 volume, and reaction time) affecting this system were modeled by two approaches: a response surface methodology (RSM) based on a Box–Behnken design and artificial neural network (ANN). A comparison was made between the predictive accuracy of RSM for brilliant cresyl blue (BCB) removal and that of the artificial neural network (ANN) approach. Both methodologies provided satisfactory and comparable predictions, achieving R^2 values of 0.97 for RSM and 0.99 for ANN.



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Keywords: photocatalytic; photo-Fenton-like; heterojunction; nanocomposite; brilliant crystal blue; synergistic effect; RSM; ANN

1. Introduction

Water is a necessary resource for all organisms, humans, animals, plants, and microorganisms, because most of the important processes of the biosphere are related to it [1]. Today, issues over water pollution have increased considerably. This relates to industries like paper manufacturing, cosmetics, textiles, food, pharmaceuticals, and medical diagnostics that frequently produce waste that contains cationic and anionic dyes. These industries release a large amount of toxic substance mixtures, including surfactants, biocidal compounds, solid suspensions, dispersing and wetting agents, dyes, and metal traces, which jeopardize and compromise the existence of a number of living beings. In order to mitigate the effects of these toxic mixtures, several methods have been applied. Some oxidation methods, classified as oxidation processes, play a vital role in the purification of wastewater. Among the methods that present pollutant elimination potential are oxidation, biodegradation, and adsorption. However, these methods have deficiencies in terms of effectiveness and treatment time [2–5].

To alleviate the effects of these toxic mixtures, various methods have been implemented. Among these, advanced oxidation processes (AOPs) prove promising for treating refractory and/or toxic pollutants. AOPs generate highly reactive hydroxyl radicals $\bullet\text{OH}$, which can degrade a wide range of organic compounds, including azo dyes. Photocatalytic and photo-Fenton-like processes, in particular, have garnered notable interest for their efficiency in decomposing dyes, owing to their low cost, moderate reaction conditions, and simplicity of operation [6].

The photocatalysis process relies on the generation of electron–hole pairs (e^-/h^+) under appropriate light irradiation [7], allowing the transfer of photoexcited charges to the surface of the photocatalyst, where they trigger redox reactions to form active species capable of degrading pollutants. The photo-Fenton-like process combines this approach with the Fenton reaction, in which $\bullet\text{OH}$ radicals are produced through the reaction of a Fenton reagent. However, these processes are traditionally effective only under UV irradiation, which constitutes only 5% of solar light, thereby limiting their practical application. It is therefore crucial to develop photocatalysts and photo-Fenton-like catalysts activated by visible light [8].

Currently, there is growing interest in the degradation of organic pollutants under visible light, particularly on graphitic carbon nitride ($g\text{-C}_3\text{N}_4$), due to its moderate band gap (2.4–2.8 eV) [9,10]. Furthermore, $g\text{-C}_3\text{N}_4$ may have several drawbacks, such as a high complexity level of photo-generated supports, low utilization of visible light, difficult recovery, and easy agglomeration [11,12].

To overcome these hindrances, several studies have explored the employment of $g\text{-C}_3\text{N}_4$ composites with different spinel ferrites for enhanced photocatalysis (MFe_2O_4 , $\text{M} = \text{Mn, Zn, Ni, Cu, and Co}$) [13–16]. Moreover, ferrite spinel nanoparticles are magnetic materials with unique characteristics such as a narrow band energy gap, utilization of UV–visible light, elevated electromagnetic performance, cost-effectiveness, feasible preparation, high chemical stability, intriguing adsorption, and innocuous properties [17–19]. Additionally, ferrites can be easily separated from the polluted aqueous solution due to their magnetic properties, and thanks to this separation process, ferrite spinel nanoparticles can be reused, justifying their employment as photocatalysts at affordable prices [20–22]. The major challenge for the utilization of photocatalysts for wastewater treatment lies in

recycling and separation. The association of spinel ferrites and g-C₃N₄ has shown not only an improvement in photocatalytic performance but also a magnetic recycling capacity of the photocatalyst [23,24]. However, spinel ferrites might cause the pollution of treated water due to their minor toxicity [25–29]. In this context, it is worth noting that tin ferrite (SnFe₂O₄) can be considered an advantageous option due to its non-toxic nature and the abundance of Fe and Sn elements on earth [30]. SnFe₂O₄ has been mentioned as being extremely effective in photocatalytic elimination due to its high adsorption of UV–visible light, with a band-gap energy of 1.89 eV [30].

In this work, we present a simple method for synthesizing SnFe₂O₄ by co-precipitation without the need for a calcination step, using NaOH as a precipitating agent. Meanwhile, g-C₃N₄ was synthesized by the calcination of a urea precursor. We successfully fabricated magnetic SnFe₂O₄/g-C₃N₄ nanocomposites with varying mass ratios, demonstrating their potential as visible-light-activated photocatalysts. The synthesized nanocomposites underwent thorough characterization using several techniques to assess their phase, purity, and morphology. This innovative approach introduces a simplified and cost-effective synthesis route that eliminates the high-temperature calcination typically associated with conventional methods, which can be both energy-intensive and costly. The resulting nanocomposites demonstrated enhanced photocatalytic and photo-Fenton activities under visible-light irradiation, effectively addressing the limitations of traditional UV-dependent catalytic processes. This advancement paves the way for more efficient and sustainable photocatalytic applications. First, we compared the performance of photocatalytic and photo-Fenton processes under identical experimental conditions. Then, we studied the impact of various experimental parameters, such as catalyst loading, initial BCB concentration, solution pH, H₂O₂ volume, and contact time, in order to identify the optimal conditions for the most efficient photo-Fenton process. These parameters, frequently explored in the literature, significantly influence the efficiency of the photo-Fenton process, making their optimization essential [31]. As a further step, radical scavenging experiments were carried out to identify the main active species involved in the photo-Fenton mechanism. In addition, this study is one of the first to compare the predictive capabilities of response surface methodology (RSM) and artificial neural networks (ANNs) in the modeling and optimization of brilliant cresyl blue (BCB) dye degradation, enabling the establishment of relationships between input factors and process responses for optimal performance. The practical significance of our results is also underlined by the magnetic properties of the SnFe₂O₄/g-C₃N₄ nanocomposites, which facilitate separation and reuse, reducing operational costs and minimizing environmental impact, while the non-toxic nature of SnFe₂O₄ addresses concerns about secondary pollution. By addressing the limitations of existing photocatalysts and photo-Fenton processes, this study advances the field of wastewater treatment and contributes to the development of sustainable and efficient solutions to mitigate water pollution, with the integration of experimental optimization and advanced modeling techniques (RSM and ANN) paving the way for future research and industrial applications [32].

2. Materials and Methods

2.1. Materials

Urea (CH₄N₂O, Sigma-Aldrich, Munich, Germany), tin(II) chloride dihydrate (SnCl₂·2H₂O, 98%, Sigma-Aldrich, Munich, Germany), iron(III) chloride hexahydrate (FeCl₃·6H₂O, 97%, Sigma-Aldrich, Munich, Germany), sodium hydroxide (NaOH, purity ≥ 98%, Merck & Co., Rahway, NJ, USA), and hydrochloric acid (HCl, 37%, Merck & Co.) were used. The structure of brilliant cresyl blue (BCB) is shown in Figure 1 (its molecular formula is C₁₇H₂₁ClN₄O, its molar mass is 332.84 g·mol^{−1}, and its λ_{max} = 626 nm)

(REACTIFS RAL, Paris, France). Rhodamine B (RhB) and crystal violet (CV) were obtained from Sigma-Aldrich, Munich, Germany, as well as hydrogen peroxide (H₂O₂, 30%, Merck). All analytical-grade reagents were used as received, without any further purification.

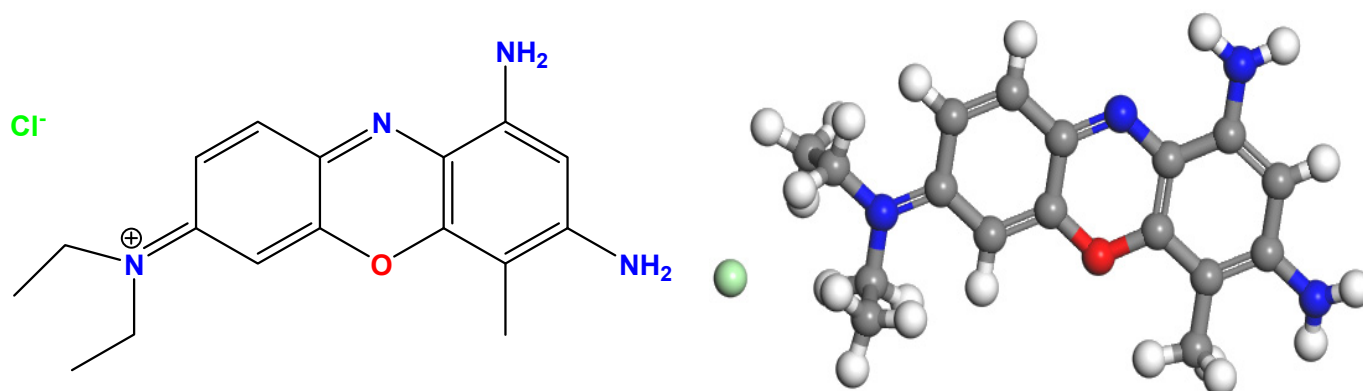
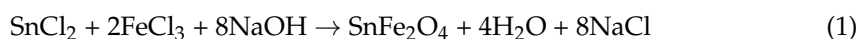


Figure 1. Chemical structure of brilliant cresyl blue (BCB).

2.2. Synthesis of *g*-C₃N₄ and SnFe₂O₄ Nanoparticles

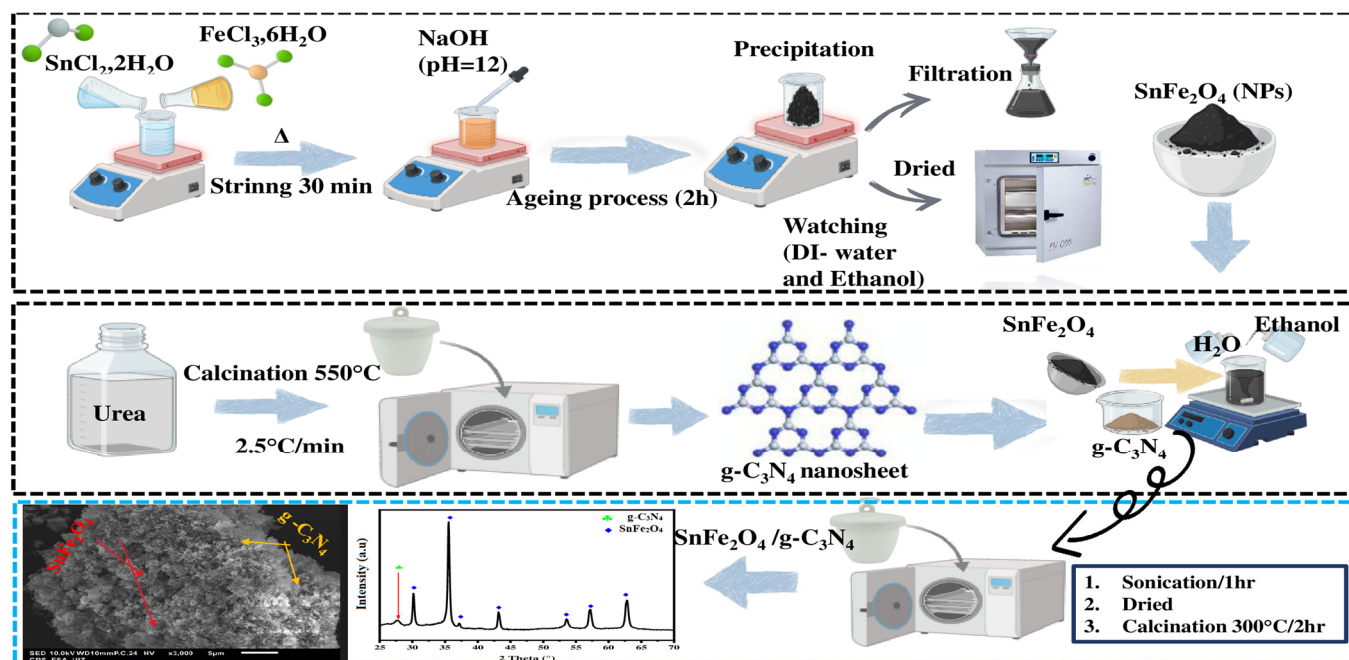
g-C₃N₄ was synthesized by the calcination of a urea precursor. Typically, 20 g of urea was placed in a ceramic crucible and then heated in a muffle furnace at 550 °C with a heating rate of 2.5 °C min^{−1} for 4 h.

SnFe₂O₄ nanoparticles were synthesized using a chemical co-precipitation method. Initially, 1.5 g of SnCl₂·2H₂O and 3.59 g of FeCl₃·6H₂O were dissolved in distilled water at a weight ratio of 1:2. The solution was stirred and heated to 60 °C. Once the desired temperature was reached, a 3 M NaOH solution was added dropwise until complete precipitation was achieved, while maintaining a highly basic pH (≈12). When the pH was maintained at 12, the solution was stirred for 2 h. After cooling to room temperature, the solution was decanted. The resulting SnFe₂O₄ product was washed with ethanol and distilled water until neutralized, then dried in an oven at 80 °C for 12 h. The co-precipitation reaction is described as follows:



2.3. Synthesis of SnFe₂O₄/*g*-C₃N₄ Nanocomposites

SnFe₂O₄/*g*-C₃N₄ nanocomposites with two different proportions of *g*-C₃N₄ (5% and 20% by weight) were fabricated. Variable amounts of *g*-C₃N₄ and SnFe₂O₄ were dispersed in a mixture of ethanol and distilled water. After an ultrasonication period of 60 min, the mixture was heated in a water bath at 80 °C with constant stirring to evaporate the solvents. It was then placed in a muffle furnace for 2 h at 300 °C, forming the desired SnFe₂O₄/*g*-C₃N₄ nanocomposites. These nanocomposites were named SnFe₂O₄/*g*-C₃N₄ (5%) and SnFe₂O₄/*g*-C₃N₄ (20%), depending on the proportion of *g*-C₃N₄ used. The schematic diagram illustrating the preparation of SnFe₂O₄/*g*-C₃N₄ is presented in Scheme 1.



Scheme 1. The synthesis process of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ heterojunction.

2.4. Characterisation

The $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposites were characterized using various analytical techniques to assess their properties and structure. Characterization included X-ray diffraction (XRD) to determine the crystal structure and Fourier transform infrared (FTIR) spectroscopy to study chemical bonding. The morphology and microstructure were analyzed by scanning electron microscopy (Supra 40 VP Gemini Zeiss Column, with a maximum voltage of 20 KV), and the elemental composition of the composite was determined by recording its energy dispersive X-ray (EDX) spectra. Additionally, transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) were used to examine the morphological structure and the presence of the heterojunction structure, respectively. These techniques provided detailed information on the physical and chemical properties of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposites, which is crucial for understanding their potential applications in various fields.

2.5. The Photocatalytic and Photo-Fenton-like Catalytic Activity Test

The photocatalytic activity of the catalysts was evaluated in a glass batch reactor by monitoring the degradation of brilliant cresyl blue (BCB) under irradiation from a commercial 72 W LED lamp ($\lambda \geq 420$ nm, Philips, Amsterdam, The Netherlands). Typically, 30 mg of the synthesized photocatalyst was introduced into a beaker containing 100 mL of an aqueous BCB solution (20 mg L^{-1}). To establish an adsorption–desorption equilibrium, the reaction mixture was kept in the dark for 30 min while being magnetically stirred. During the degradation process, 3 mL of the solution was sampled every 30 min. After centrifugation, the residual solution was analyzed using a UV-Vis spectrophotometer at the maximum absorption wavelength of 626 nm for BCB. For the photo-Fenton-type process, 1 mL of 30 wt% H_2O_2 was added to the solution after stirring in the dark. After completing the degradation of brilliant cresyl blue (BCB), we tested two additional cationic dyes, rhodamine B (20 mg L^{-1}) and crystal violet (20 mg L^{-1}), to evaluate the effectiveness of the catalyst. Furthermore, to explore the active species involved in the photo-Fenton-like reaction, trapping experiments were conducted under experimental conditions similar to those of the previous experiments but with the addition of different trapping agents.

into the reaction system. In this study, ethylenediamine tetra acetic acid (EDTA-2Na), isopropyl alcohol (IPA), and L-ascorbic acid (ASC) were used as trapping agents for holes (h⁺), hydroxyl radicals (•OH), and superoxide anion radicals (O₂•[−]), respectively [33]. The efficiency of degradation (DE%) and the rate constants K (min^{−1}) are determined using Equations (2) and (3) [34], respectively:

$$\text{Degradation Efficiency} = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (2)$$

$$\ln\left(\frac{C_0}{C_t}\right) = K \times t \quad (3)$$

Herein, C₀ and C_t are the concentrations of the BCB dye at the initial and any time during the photo-Fenton process, respectively. K (min^{−1}) is the constant of the first-order reaction.

2.6. Optimization Results Using the Box–Behnken Method

In 1960, Box and Behnken introduced a new experimental design suitable for second-order models, which facilitates the estimation of certain interactions. One of the main advantages of these experimental models is their ability to minimize the number of experiments needed during the testing phase by pre-identifying specific factors or combinations of factors that affect the response of the studied system [35]. The response surface methodology (RSM) is a set of mathematical and statistical techniques by which we can determine the efficiency or optimal conditions of degradation using Box–Behnken (BBD) models [36]. The parameters are defined at three distinct levels: low, medium, and high, represented by the codes −1, 0, and +1, respectively (Table 1). In this study, we evaluate the degradation efficiency of brilliant cresyl blue by examining the effects of four different factors: catalyst loading (X₁), initial concentration of brilliant cresyl blue (X₂), volume of H₂O₂ (X₃), and time (X₄). The degradation rate of BCB was used as the response variable in this study. A total of 30 experiments were conducted in a randomized order, and the results are available in the Supplementary Materials (Table S1). To evaluate the relevance and significance of the mathematical models proposed by RSM, certain analyses are required, including analysis of variance (ANOVA). The selected mathematical model is a second-degree polynomial, which describes the relationships between independent and dependent variables (Equation (4)) [37]. Experimental design, mathematical modeling, and optimization were performed using Desgn-Expert 13.0.5.0 software.

Table 1. Actual and coded values of variables in the Box–Behnken design (BBD).

Coded Factors	Actual Factors	Unit	Low (−1)	Center (0)	High (+1)
X ₁	Catalyst loading	g L ^{−1}	0.2	0.3	0.4
X ₂	BCB concentration	mg L ^{−1}	20	30	40
X ₃	Volume of H ₂ O ₂	mL	0.5	1	1.5
X ₄	Contact time	min	90	120	150

$$\hat{Y} = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i=1}^k \sum_{i < j}^k \beta_{ij} X_i X_j + \varepsilon \quad (4)$$

In this equation, $\hat{Y}$ represents the response function, while β_0 is the polynomial constant that reflects the overall mean effect. The coefficients corresponding to the linear, quadratic, and interaction effects are denoted by β_i , β_{ii} , and β_{ij} , respectively. The coded independent variables are represented as X_i and X_j .

2.7. Artificial Neural Network Methodology

Artificial neural networks (ANNs) are models inspired by the neural architecture of intelligent organisms, which learn and acquire knowledge through experience. ANNs serve as a tool for polynomial regression modeling, facilitating data processing and analysis. An artificial neural network is made up of three main components: an input layer, one or more hidden layers, and an output layer. Each layer is made up of a large number of units called neurons connected by links, to which values called weights are assigned; this enables communication between the neurons [38].

The ANN used in this study was a four-layer neural network trained with the gradient descent backpropagation algorithm. The experimental data obtained was randomly divided into three sets: 20 for training, 5 for testing, and 5 for validation. The Levenberg–Marquardt algorithm updated the weights and biases of the training datasets, while the testing process assessed the extent to which the network had learned to generalize. In addition, during training, errors in the validation data were also observed to check for overlearning.

The optimal structure of the ANN model in this study is presented in Figure 2. A hyperbolic tangent function (tansig) with a sigmoid slope was used as the transfer function (purelin) for the neurons in the hidden layer, while linear functions were used for the neurons in the input and output layers, respectively. The input and output layers comprised 4 and 1 neurons, respectively. To determine the optimal number of neurons in the hidden layer, different topologies were evaluated, with the number of neurons ranging between 2 and 18. Each topology was repeated 10 times to avoid any random association due to the random initialization of weights and biases [39]. All calculations were performed using MATLAB R2024b.

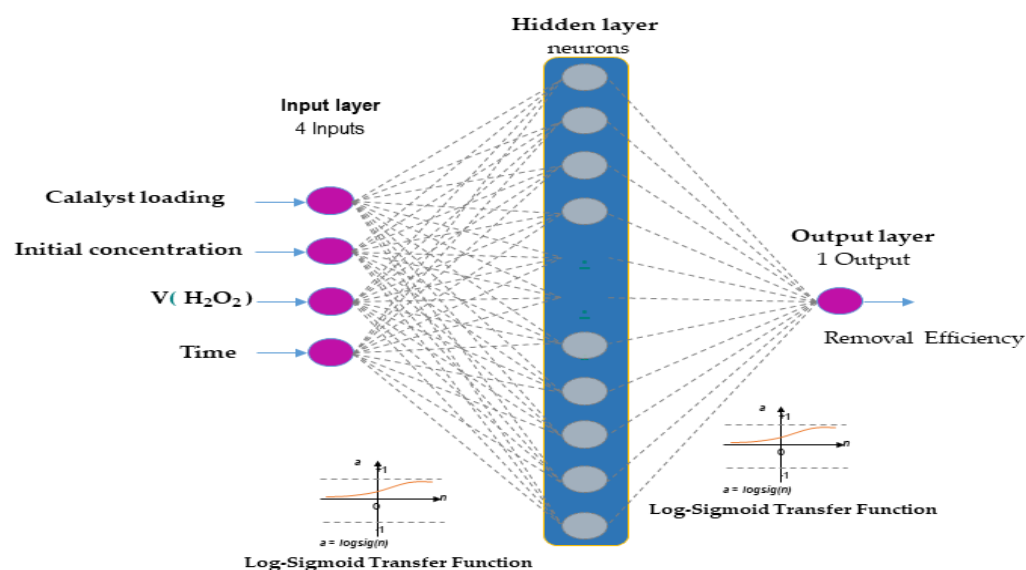


Figure 2. Artificial neural network architecture composed of four input nodes, hidden nodes, and one output node.

2.8. Statistical Analysis

The quality of the fit between the experimental data and the outcomes of the photo-Fenton process was assessed using error metrics including the coefficient of determination (R^2), root mean square error (RMSE), and mean absolute error (MAE). Additionally, these parameters were also employed to compare the RSM-BBD and ANN methods. R^2 represents the proportion of variation in the dependent variable explained by the independent variables and is commonly used to indicate this proportion of variation. RMSE is the square root of the mean squared errors [38,40], and it is an absolute error metric primarily used for model fitting, validation, selection, comparisons, and forecast evaluations [41].

MAE denotes the average deviation between the observed and predicted values, reflecting the average total deviation within the dataset. Table S2 in the Supplementary Materials provides the various equations used to calculate these metrics.

3. Results and Discussion

3.1. Structural Analysis

3.1.1. XRD Analysis

The X-ray diffraction (XRD) patterns of the photocatalysts g-C₃N₄, SnFe₂O₄, and SnFe₂O₄/g-C₃N₄ (20%) are shown in Figure 3. It can be seen that the diffraction peaks of synthesized SnFe₂O₄ via the co-precipitation method correspond well to the standard pattern of a cubic spinel structure SnFe₂O₄ (PDF #11-0614) [42]. In the CN pattern, the intense peak at 27.524°, which correspond to a d-spacing of 0.32 nm, can be indexed to the (002) diffraction plane, associated with the characteristic interplanar stacking of conjugated aromatic networks [43].

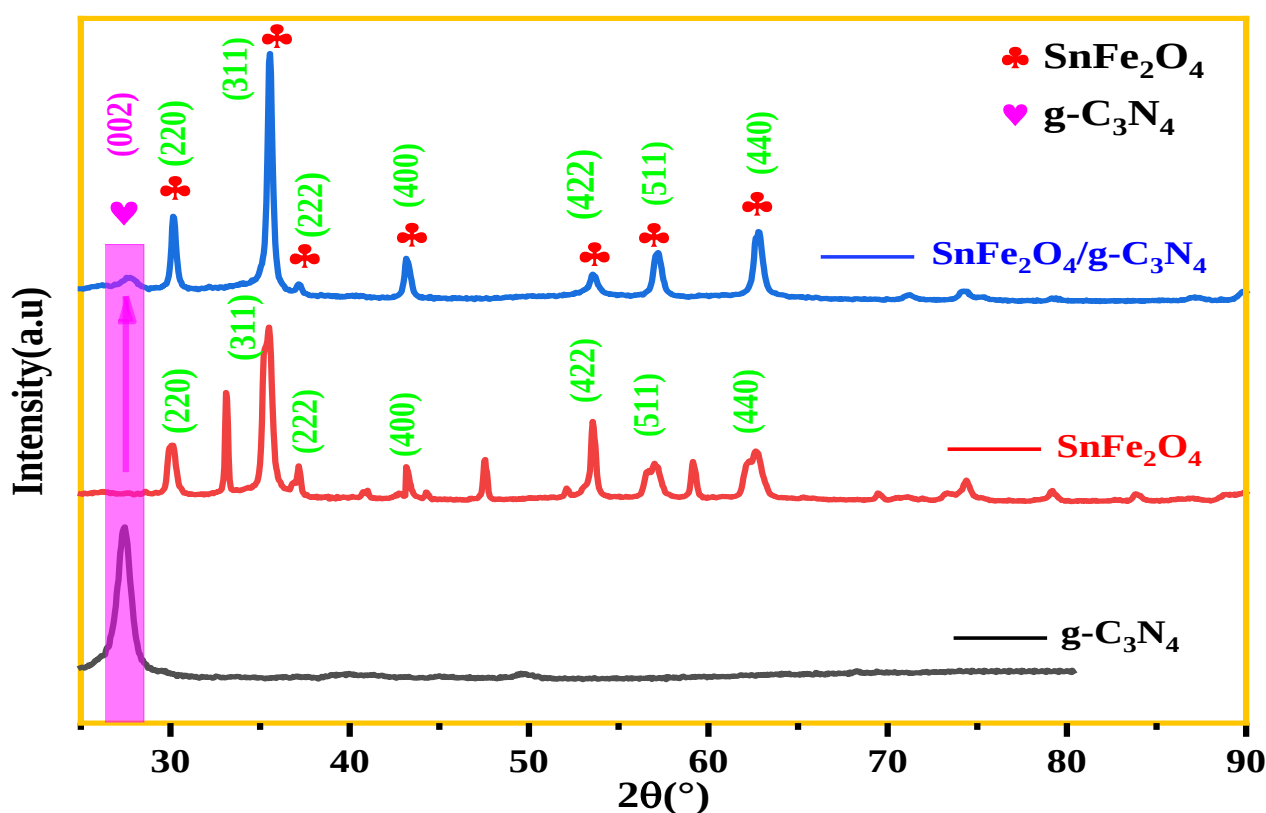


Figure 3. XRD patterns of g-C₃N₄, SnFe₂O₄, and SnFe₂O₄/g-C₃N₄ (20%).

Diffraction peaks of pure SnFe₂O₄ at $2\theta = 30.137^\circ$, 35.65° , 43.165° , 57.155° , and 62.564° can be indexed on the (220), (311), (400), (511), and (440) diffraction planes, respectively, corresponding well to the reflections of the SnFe₂O₄ spinel structure reported in the literature [44,45]. Another detected diffraction peak indicated the presence of an α -Fe₂O₃ phenomena similar to what was previously reported [46,47]. When SnFe₂O₄ is coupled with g-C₃N₄, no impurity phases such as iron oxide were detected or identified, indicating the achieved preparation of SnFe₂O₄/g-C₃N₄ (20%) hybrids. The XRD diffraction patterns of SnFe₂O₄/g-C₃N₄ (20%), thus, show hybrid formation. Similar results can also be found in others systems [42].

3.1.2. FTIR Analysis

The FTIR data of composite samples (gC_3N_4 , SnFe_2O_4 , and $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%)) are illustrated in Figure 4. The spectra analysis of gC_3N_4 has disclosed three predominant adsorption zones: a broad band located between 3000 and 3400 cm^{-1} attributed to stretching vibrations of non-condensed amino groups (N-H), another band with sharp intensity was detected at 804 cm^{-1} corresponding to the characteristic respiration of tri-s-triazine heterocycles, along with bands in the range of $1200\text{--}1700\text{ cm}^{-1}$ arising from the vibrational stretching of the C-N bond inside the tri-s-triazine cycles. In the case of SnFe_2O_4 , a distinctive absorption peak at 564 cm^{-1} was observed, associated to the vibrations of Fe-O bonds, consistent with the formation of the spinel structure of ferrite. Significantly, the main characteristics peaks of $\text{g-C}_3\text{N}_4$ and SnFe_2O_4 are present in the spectrum of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) composite, indicating the accomplished formation of the composite material.

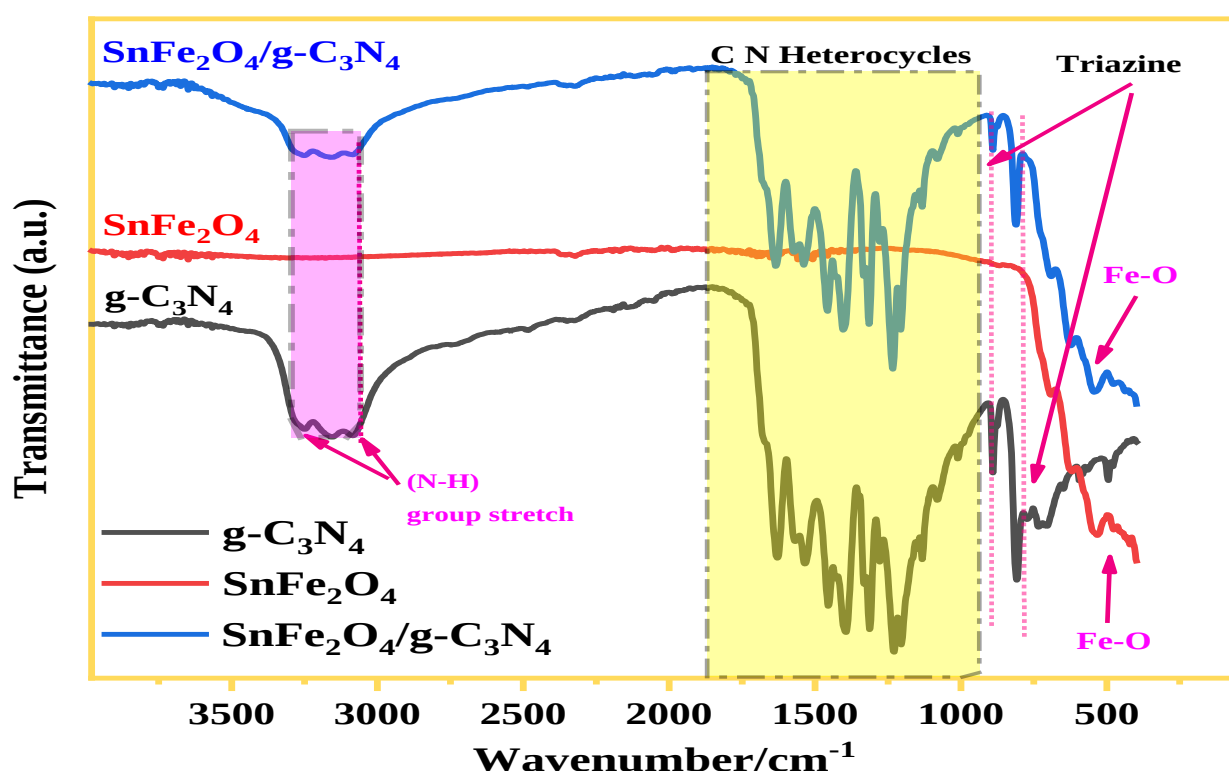


Figure 4. FT-IR spectra of SnFe_2O_4 , $\text{g-C}_3\text{N}_4$, and $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%).

3.2. Morphological Analysis

3.2.1. SEM/EDX Analysis Results

The SEM and EDX characterization of $\text{g-C}_3\text{N}_4$, SnFe_2O_4 , and $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) is presented in Figures 5 and S1. $\text{g-C}_3\text{N}_4$ exhibits a folded and irregular sheet structure (Figure 5a), which enhances electron transmission capacity, improves light absorption performance, and extends the lifetime of photoexcited charge carriers. The spinel structure of SnFe_2O_4 , along with intergranular pores and voids, is visible in Figure 5b. Additionally, the surface of $\text{g-C}_3\text{N}_4$ appears very rough, with a large number of SnFe_2O_4 nanoparticles attached (Figure 5c). Furthermore, the EDX diagram displays the elements of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) composite material, with carbon (C), nitrogen (N), oxygen (O), iron (Fe), and tin (Sn) atoms uniformly distributed on the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) photocatalyst composite (Figure S1), confirming the successful synthesis of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) photocatalyst composite [48].

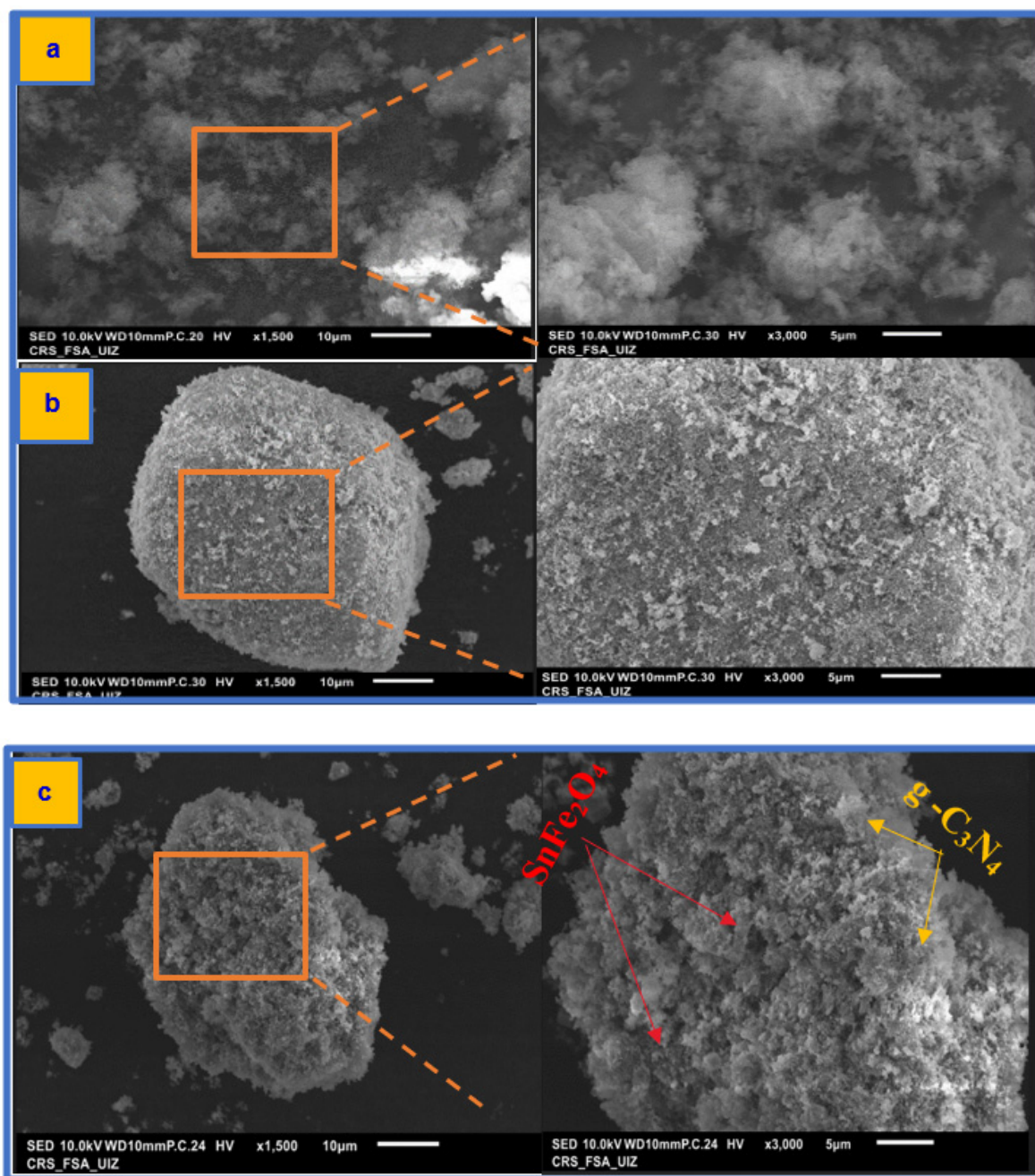


Figure 5. SEM images of (a) $\text{g-C}_3\text{N}_4$, (b) SnFe_2O_4 , and (c) $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%).

3.2.2. TEM Analysis Results

The successful synthesis is confirmed by the TEM image of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) hybrid shown in Figure 6a–c. Two distinct phases are clearly visible: $\text{g-C}_3\text{N}_4$ (light region) and SnFe_2O_4 (dark region), in close contact and forming an intimate interface. This observation demonstrates that the SnFe_2O_4 (SFO) nanoparticles, with an average diameter of approximately 16 nm, have effectively covered the surface of $\text{g-C}_3\text{N}_4$. These results are fully consistent with the analyses by X-ray diffraction (XRD). Furthermore, the HRTEM (high-resolution transmission electron microscopy) image of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) hybrid (Figure 6d) reveals lattice fringes with interplanar spacings of 0.32 nm and 0.253 nm, corresponding to the (002) plane of $\text{g-C}_3\text{N}_4$ and the (311) plane of SnFe_2O_4 , respectively [29,48].

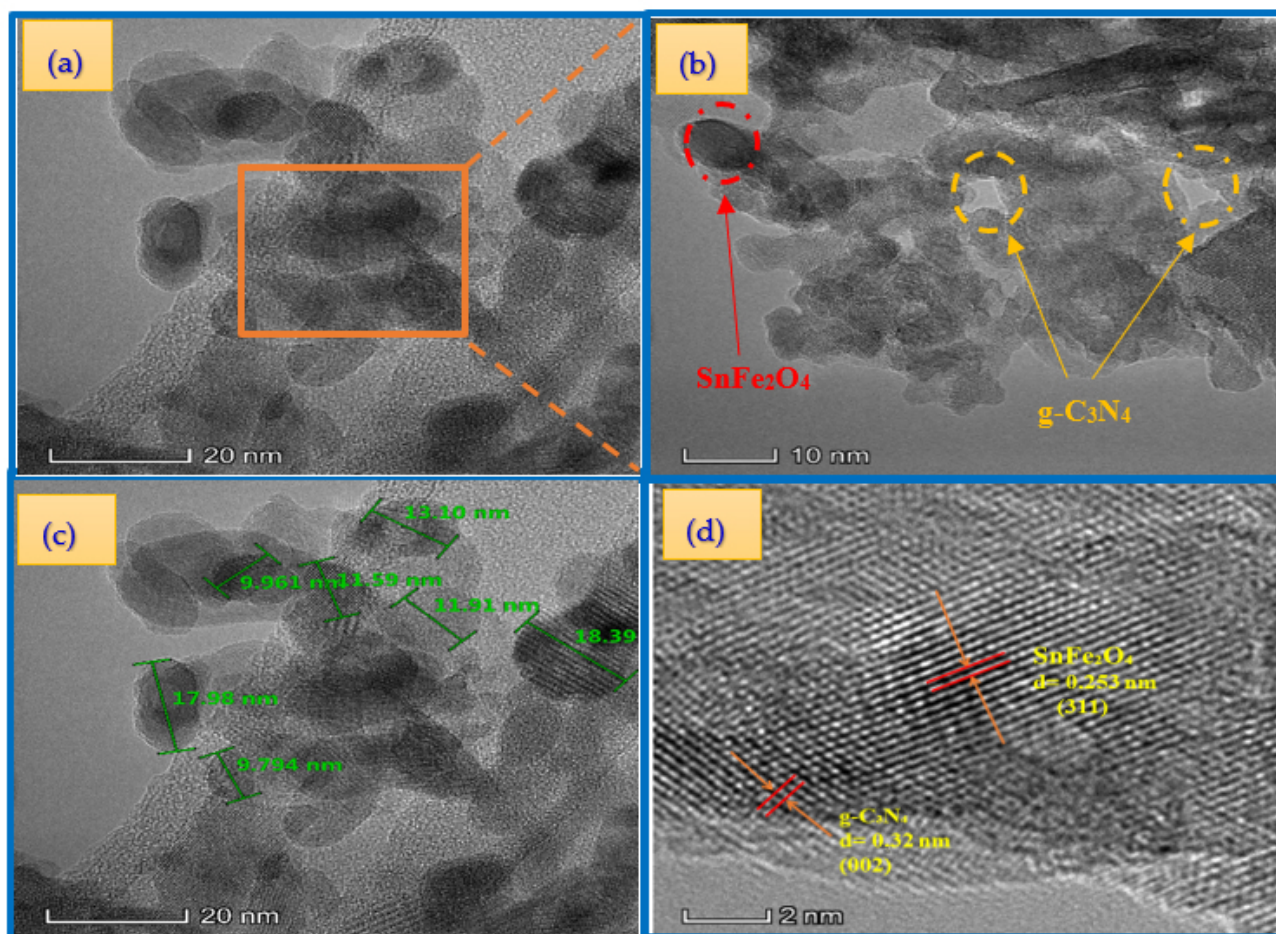


Figure 6. TEM (a–c) and HRTEM (d) images of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%).

3.3. Photo-Fenton-like Catalytic and Photocatalytic Performance

Dye adsorption plays a crucial role in the efficiency of photocatalytic degradation, particularly for semiconductors, where the adsorbed dye molecules act as electron donors, facilitating the transfer of excited electrons to the conduction band of the semiconductor under visible-light irradiation [49]. A study was conducted on the adsorption of the BCB dye on the surface of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ composite by maintaining the dye solution with the necessary amount of catalyst in the dark, without H_2O_2 , for 60 min. At the end of the experiment, negligible adsorption (<10%) was observed (Figure 7a).

The efficiency of the photocatalytic and photo-Fenton-like degradation of BCB over time, using $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposites, was evaluated under visible light. The order of BCB degradation follows this sequence: $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{H}_2\text{O}_2/\text{Vis} > \text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{Vis} > \text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{H}_2\text{O}_2/\text{Dark}$ (Figure 7b). This demonstrates that the combination of the catalyst, visible light, and H_2O_2 offers the best degradation performance. Conversely, the absence of H_2O_2 during photocatalysis significantly reduces efficiency, as the process mainly relies on the separation of electron–hole pairs and the generation of radicals on the catalyst’s surface, which is often hindered by charge recombination.

Figure 8a,b illustrates the progression of the photocatalytic and photo-Fenton-like degradation of BCB over time for the various samples. In all photocatalytic and photo-Fenton-like experiments, the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) composites exhibited significantly enhanced photocatalytic activity for dye degradation compared to pure SnFe_2O_4 nanoparticles, $\text{g-C}_3\text{N}_4$ nanosheets, and $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (5%). Furthermore, the percentage of dye degradation during 150 min of reaction is indicated in Figure 8c,d. Among the tested composites, the one with 20% $\text{g-C}_3\text{N}_4$ showed the highest photo-Fenton-like activity, achieving

a degradation rate of 94.4% during 150 min of reaction (Figure 8d). Additionally, the control experiments confirmed that BCB remains stable during the photocatalytic and photo-Fenton-like processes in the absence of the photocatalyst. This indicates that dye removal is not solely due to surface adsorption but also involves the degradation of adsorbed dye molecules and their release from the catalytic sites.

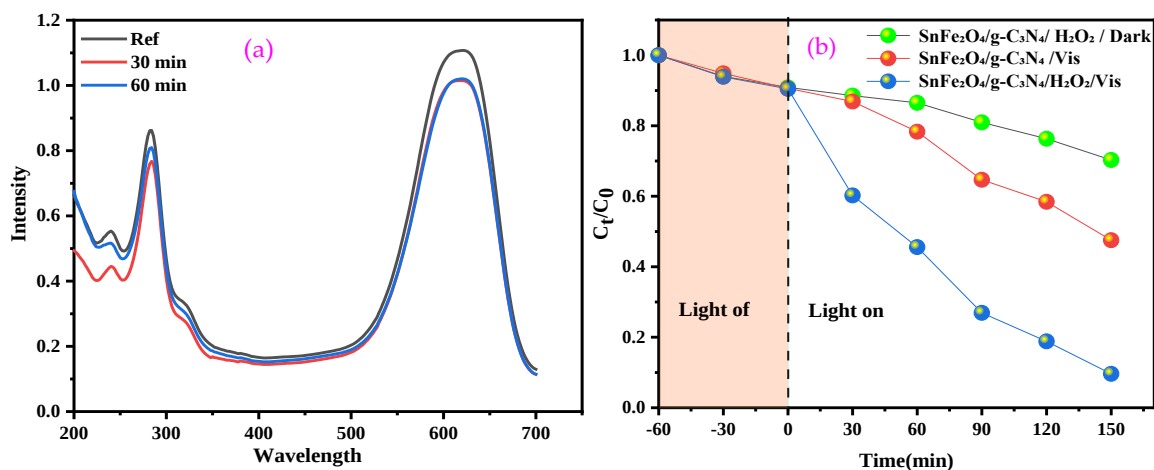


Figure 7. (a) Adsorption of BCB by SnFe₂O₄/g-C₃N₄; (b) Fenton-like, photocatalytic, and photo-Fenton-like of BCB in the presence of SnFe₂O₄/g-C₃N₄.

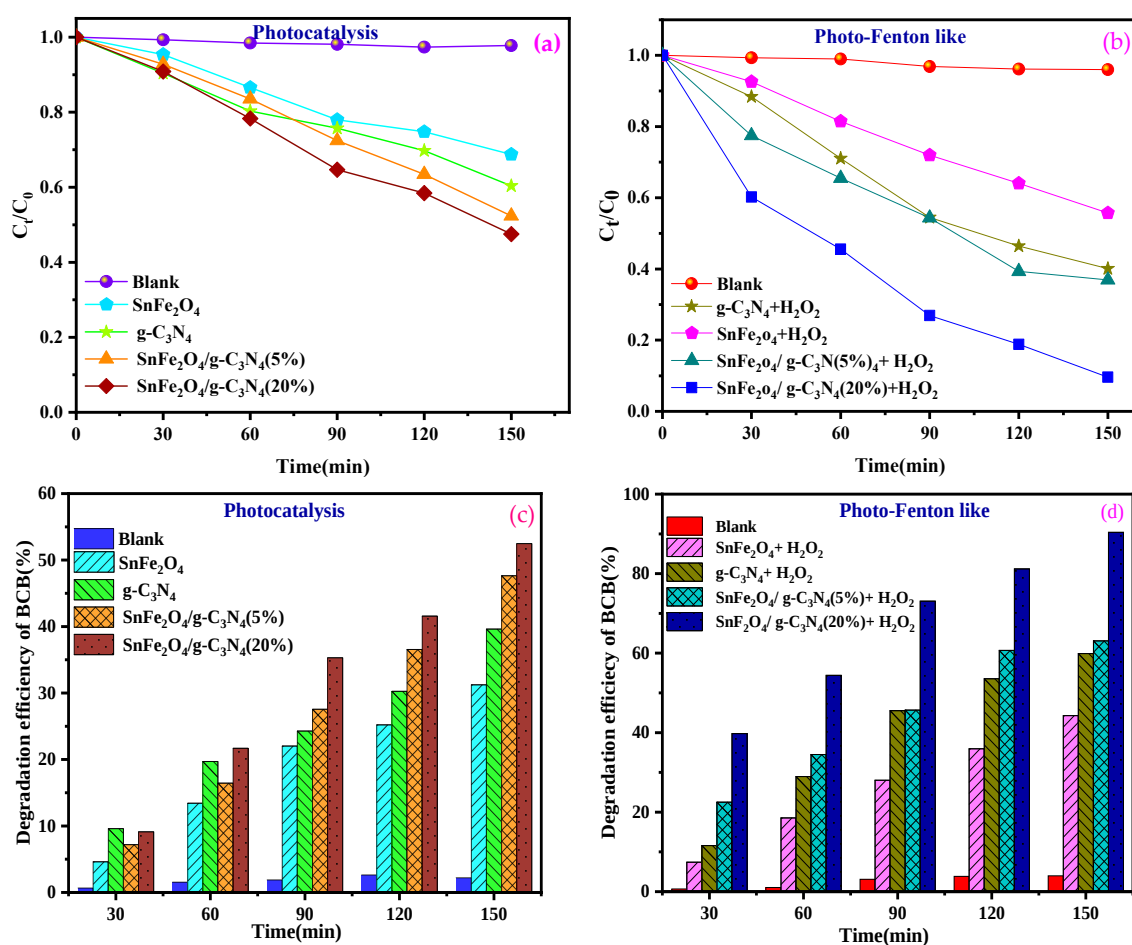


Figure 8. (a,b) Photocatalytic degradation, photo-Fenton-like as a function of time of BCB by the samples, respectively. (c,d) Degradation efficiency of BCB dye for photocatalytic degradation and photo-Fenton-like.

Figure 9a,b shows the plots of $\ln(C_t/C_0)$ versus irradiation time for the photocatalytic and photo-Fenton-like degradation of BCB, respectively, while the reaction rate constants are illustrated in Figure 9c,d. The rate constants for the photo-Fenton-like processes (0.0069, 0.00123, 0.01299, and 0.03975 min^{-1} for SnFe_2O_4 , $\text{g-C}_3\text{N}_4$, $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (5%), $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%), respectively) are higher than those for the photocatalytic processes (0.00129, 0.00195, 0.00412, and 0.00557 min^{-1}) for all catalysts (Figure 9c,d). It has been demonstrated that the Fenton-like photocatalytic reaction with $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) exhibits a reaction rate approximately 7.13 times higher than that of the standard photocatalytic reaction.

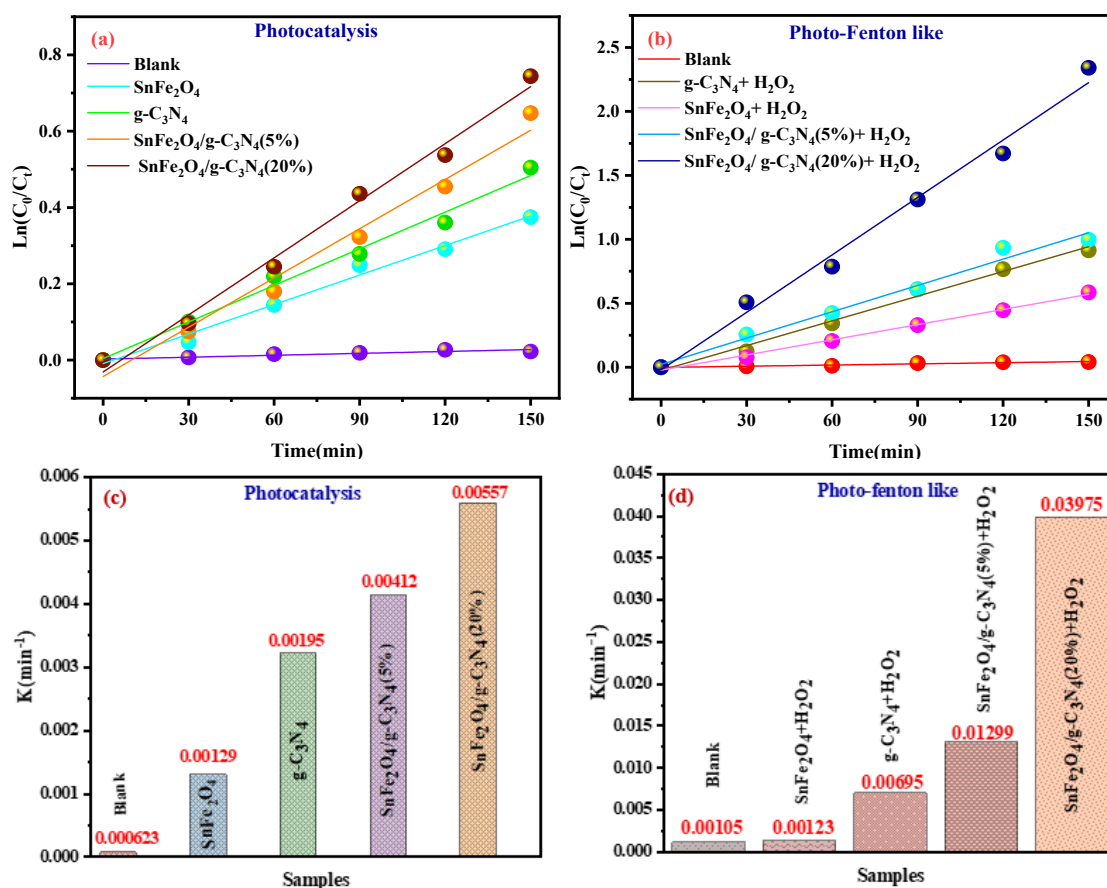


Figure 9. (a,b) $\ln(C_t/C_0)$ graphs as a function of irradiation time; (c,d) kinetic curves of the samples for the photocatalytic degradation and photo-Fenton-like activity of BCB, respectively.

Additionally, further tests were conducted using other cationic dyes, such as rhodamine B and crystal violet, to evaluate the catalytic efficiency of the system. As illustrated in Figure S2b,c, the results revealed that the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) composite also demonstrated remarkable efficiency in degrading these dyes, confirming the versatility of this catalyst for treating wastewater containing various cationic dyes.

Finally, it was observed that the photo-Fenton-like performance of the samples was significantly superior to their photocatalytic performance, suggesting that photo-Fenton-like catalysis is more effective for treating wastewater containing cationic dyes. Furthermore, the degradation capacity of the synthesized photocatalysts was compared to that of other heterojunction photocatalysts, as presented in Table 2. To further explore these performances, additional photo-Fenton-like degradation experiments were conducted using the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) composite as the photocatalyst, studying the effects of the amount of photocatalyst, initial concentration, as well as the H_2O_2 content and pH of the solution on BCB degradation.

Table 2. Comparison of different catalysts.

Catalysts	Pollutant	Initial Conc.	H ₂ O ₂ Dosage	Catalysts Loading	Light Source	% Removal	References
SnFe ₂ O ₄ /g-C ₃ N ₄	BCB	20 mg L ⁻¹	(1 mL in 100 mL)	0.3 g L ⁻¹	LED	94% within 150 min	This study
ZnFe ₂ O ₄ /g-C ₃ N ₄	MB	20 mg L ⁻¹	(1 mL in 50 mL)	0.5 g L ⁻¹	Natural sunlight	99.5% within 60 min	[50]
NiFe ₂ O ₄ /gC ₃ N ₄	RhB	20 mg L ⁻¹	~0.2 M	1 g L ⁻¹	LED	98% within 90 min	[51]
p-CuFe ₂ O ₄ /n-g-C ₃ N ₄	MO	20 mg L ⁻¹	(0.5 mL in 100 mL)	0.1 g L ⁻¹	300 W Xenon lamp	98% within 300 min	[52]
NiFe ₂ O ₄ /g-C ₃ N ₄	MB	10 mg L ⁻¹	(1 mL in 50 mL)	0.5 mg L ⁻¹	300 W Xenon lamp	87% within 240 min	[53]

3.3.1. The Effect of Catalyst Loading

The impact of catalyst loading on the photo-Fenton-like catalytic activity was examined for BCB, as shown in Figure 10a. The degradation efficiency evolved linearly with an increase in catalyst loading from 20 to 30 mg. This phenomenon was attributed to the increased availability of active sites for catalytic reaction. However, above 30 mg of added catalyst, a slight decrease was observed. Presumably, this can be explained by the induced turbidity in the solution due to the elevated catalyst concentration, which results in the diffusion and slight penetration of light. As a result, the optimal catalyst loading for the experiments was fixed at 30 mg.

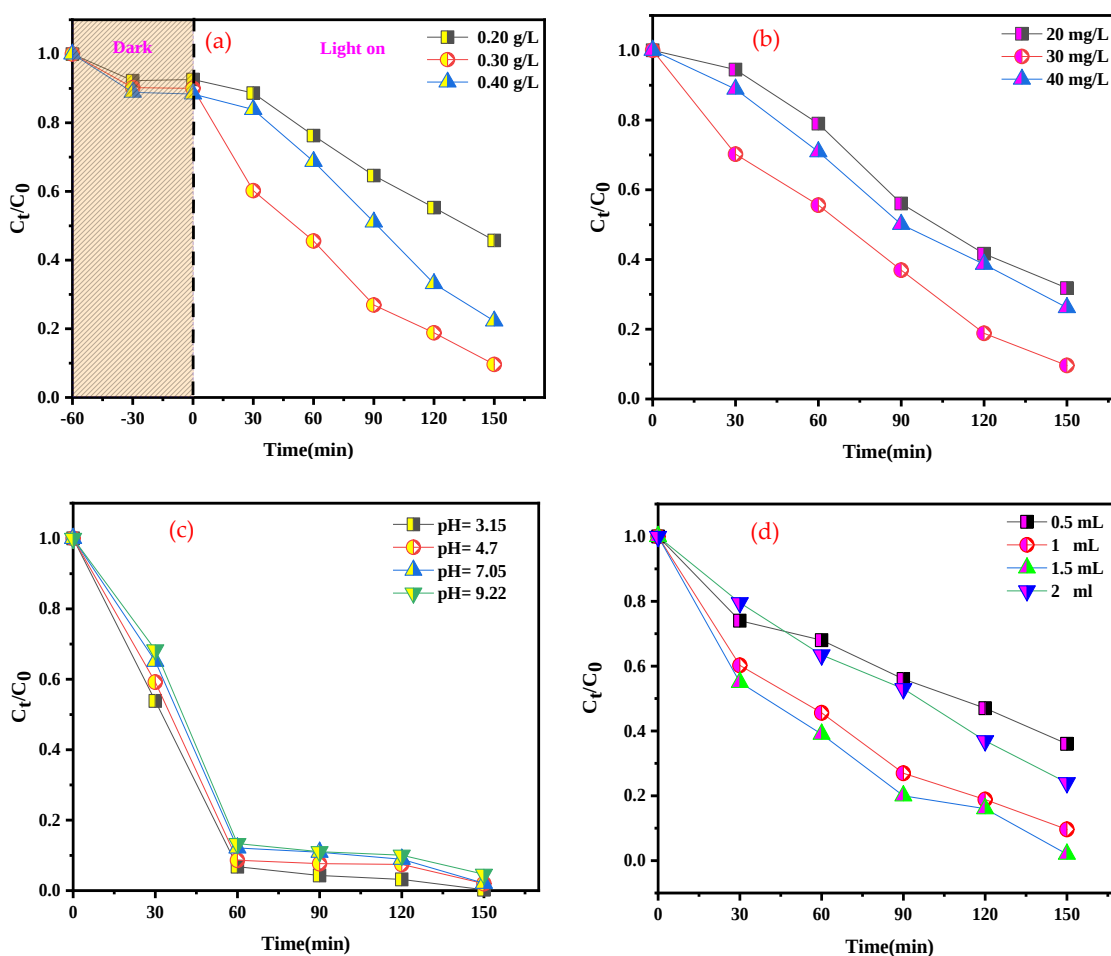


Figure 10. Effects of different catalytic parameters on BCB degradation: (a) catalyst dosage, (b) BCB concentration, (c) pH value, and (d) volume of H₂O₂ (other catalytic parameters: catalyst dosage: 0.30 g L⁻¹; H₂O₂ volume: 1 mL; pH: 5.46; T: 25 °C; BCB concentration: 20 mg L⁻¹).

3.3.2. Impact of the Initial Dye Concentration

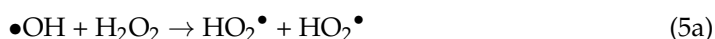
The effect of the initial dye concentration on BCB degradation is shown in Figure 10b. With a range of 20 to 40 mg L⁻¹, the reaction rate progressively decreased as the initial BCB concentration increased. For example, when the initial BCB concentration was increased, nearly 70% of BCB degradation was achieved within 90 min (20 mg L⁻¹). The same degradation percentage was achieved when the initial BCB concentration was 30 mg L⁻¹ within 120 min, while the degradation time increased to 150 min when the initial BCB concentration was 40 mg L⁻¹. It can be inferred that higher BCB concentrations have reverse effects, possibly because dye molecules cover the most available active sites. Consequently, the quantity of H₂O₂ freely available to interact with the active sites of SnFe₂O₄/g-C₃N₄, which is vital for the effective generation of •OH radicals, decreases. This phenomenon has been reported in other studies regarding dye oxidation.

3.3.3. Effect of Initial pH of the Solution

The influence of pH is one of the main parameters affecting the photocatalytic decomposition rate of certain organic compounds, which is crucial for wastewater treatment. To analyze the impact of pH on BCB dye degradation, experiments were conducted by varying the reaction medium pH from 3.15 to 9.22, while maintaining a fixed concentration of BCB dye (20 mg L⁻¹) and catalyst (30 mg L⁻¹). pH adjustments were made using appropriate NaOH (0.1 N) and HCl (0.1 N) solutions. The results presented in Figure 10c highlight the influence of pH variation on the photocatalytic degradation of BCB dye in the presence of H₂O₂. It is well known that the Fenton system requires acidic conditions [54], which is consistent with these results. The highest degradation efficiency was achieved at a pH value of 3.15. It was observed that the BCB degradation rate could reach approximately 75% in 30 min, even at a pH of 9. Beyond this value, a decrease in degradation efficiency was noted. In a basic medium, the increased formation of hydroxyl radicals •OH enhances decomposition efficiency. However, at a pH above 9, the photocatalytic efficiency decreases due to the negative surface charge induced by a high concentration of OH⁻ ions, repelling the BCB from the catalyst surface.

3.3.4. The Effect of H₂O₂ Dosage

Figure 10d presents the effect of H₂O₂ concentration on the degradation efficiency of BCB. It was found that the degradation efficiency increased from 69.5% to 99.8% with an increase in H₂O₂ dosage from 0.5 mL to 1 mL. The accelerated degradation rate is due to the increased number of •OH radicals at a growing H₂O₂ dosage, demonstrating a gradual improvement in degradation efficiency with increasing H₂O₂ concentrations. However, no significant improvement was observed when the volume of H₂O₂ increased from 1.5 to 2 mL, due to the inhibitory effect of H₂O₂. A higher concentration of H₂O₂ promotes the trapping of •OH radicals (Equation (5a,b)), leading to the formation of a new radical (HO₂•) with a markedly lower oxidation potential than that of •OH. Similarly, the recombination of •OH is also explained by Equation (6). This suggests that 1.5 mL constitutes the optimal concentration of H₂O₂ under the current experimental conditions.



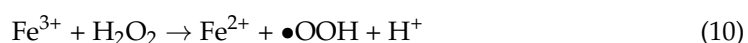
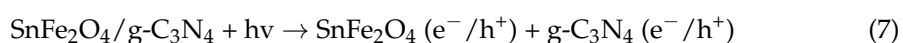
3.4. Potential Photo-Fenton-like Mechanism

The photocatalytic mechanism of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{H}_2\text{O}_2/\text{Vis}$ system was investigated to gain a deeper understanding of the photo-Fenton-like process. This process involves several active species, including hydroxyl radicals ($\bullet\text{OH}$), holes (h^+), and superoxide anion radicals ($\bullet\text{O}_2^-$). The roles of these active species, generated by the nanocomposite photocatalysts, were assessed using various radical scavengers [14].

To evaluate the effectiveness of the $\text{g-C}_3\text{N}_4$ photocatalyst, radical scavenging tests were performed to assess the degradation of BCB dye in the presence of various scavengers: disodium ethylenediaminetetraacetate (EDTA-2Na), L-ascorbic acid (ASC), and isopropyl alcohol (IPA), which capture h^+ , $\bullet\text{O}_2^-$, and $\bullet\text{OH}$ radicals, respectively. The results, shown in Figure S3, indicate that BCB degradation was 91%, 73.5%, and 35.5% with the addition of EDTA, ASC, and IPA, respectively. These data suggest that IPA had the most significant effect of the primary active species in the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{H}_2\text{O}_2/\text{Vis}$ system, highlighting the crucial role of $\bullet\text{OH}$ as the primary active species in the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{H}_2\text{O}/\text{Vis}$ system.

The term “photo-Fenton-like” refers to a process similar to the Fenton reaction but utilizes different catalysts and mechanisms to produce hydroxyl radicals $\bullet\text{OH}$ activated by ultraviolet or visible light. This method falls under advanced oxidation processes (AOPs), which enhance the efficiency of organic pollutant degradation. When exposed to light, photo-Fenton-like catalysts, such as metal ferrites or metal oxides, generate hydroxyl radicals through the interaction of the catalyst with hydrogen peroxide (H_2O_2). These highly reactive radicals facilitate the breakdown of organic contaminants into smaller molecules, as well as carbon dioxide (CO_2) and water (H_2O). By optimizing the generation of active radicals and expanding the options for pollutant treatment, this process serves as an effective alternative to conventional methods.

In a photo-Fenton-like process, the photocatalyst acts as a source of Fe^{3+} , absorbing photon energy equal to or greater than its band-gap energy. This allows electrons (e^-) to migrate from the valence band (VB) to the conduction band (CB), thereby balancing the number of holes in both bands. When exposed to visible light, the semiconductors generate electrons (e^-) and holes (h^+), which then migrate to the semiconductor surface. Here, they interact, respectively, with the electron donor and electron acceptor, as described in the literature [55]. Figure 11 illustrates the schematic of the photo-Fenton degradation of BCB by $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ under visible-light irradiation. In the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4/\text{H}_2\text{O}_2$ system, hydrogen peroxide (H_2O_2) collaborates with Fe^{3+} to create a photo-Fenton reaction, where Fe^{3+} present on the surface of SnFe_2O_4 facilitates the formation of hydroxyl radicals while oxidizing H_2O_2 to produce $\bullet\text{OH}$, which is then reduced to Fe^{2+} . As a highly effective electron acceptor, H_2O_2 directs the energy band electrons to produce $\bullet\text{OH}$ and OH^- , thereby minimizing the likelihood of electron–hole pair recombination. The $\bullet\text{OH}$ radicals convert BCB into byproducts such as CO_2 and H_2O . Additionally, these reactive species (h^+ , $\bullet\text{O}_2^-$, and $\bullet\text{OH}$) oxidize the BCB molecules adsorbed on the active sites of $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ through mechanisms involving π - π stacking and electrostatic attraction, contributing to their degradation [56,57]. The main photo-Fenton reaction process can be briefly described by Equations (7)–(16):



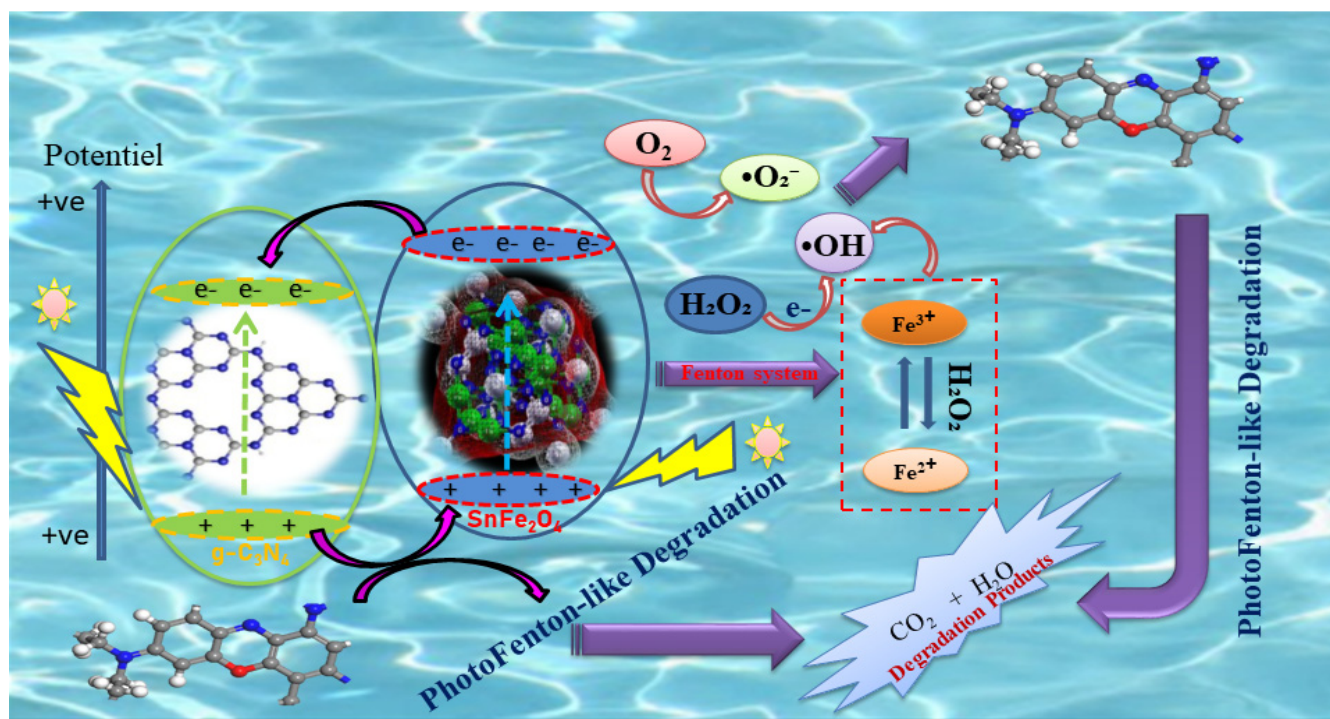
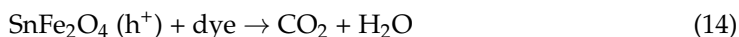


Figure 11. Schematic illustration of photo-Fenton-like activity for BCB dye degradation using a $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposite under eco-friendly LED irradiation.

3.5. The Resistance and Recycling Capacity of the Catalyst

Among the criteria that can determine the robustness of a catalyst is its ability to retain efficiency and be reused over multiple cycles, which helps reduce operating costs and minimize negative environmental impacts. To evaluate the regenerative capacity of our catalysts, repeated cycle experiments were conducted to test their stability. After each regeneration cycle, the degradation efficiency was verified. According to Figure 12a, there is no significant change in the degradation efficiency of BCB by $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ after four recovery cycles. FTIR analysis (Figure 12b) confirms the stability of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ catalyst, as no changes are observed in the bands corresponding to SnFe_2O_4 and $\text{g-C}_3\text{N}_4$ after reuse. These results highlight the excellent stability and reusability of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ catalyst. Furthermore, the $\text{g-C}_3\text{N}_4$ -based catalysts developed here stand out for their efficiency in BCB degradation, ease of preparation, eco-friendly characteristics, and recyclability, which make them highly promising for future industrial applications.

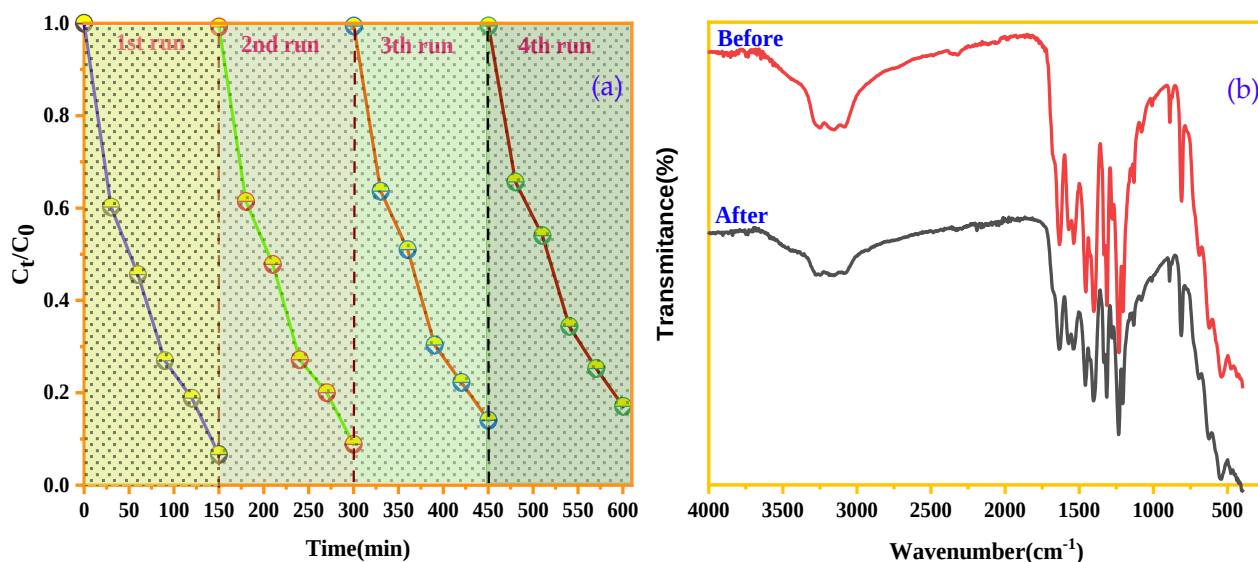


Figure 12. (a) Recycling test of the SnFe₂O₄/g-C₃N₄ (20%) catalyst. (b) FTIR spectrum of the SnFe₂O₄/g-C₃N₄ (20%) before and after BCB degradation.

3.6. Experimental Optimization via the Response Surface Methodology (RSM)

3.6.1. Statistical Model Analysis

The response surface methodology (RSM) is a set of statistical and mathematical techniques by which the efficiency and optimal conditions of degradation can be easily determined via a Box–Behnken model (BBD) [58]. In this study, we assess the performance of BCB degradation by examining the effects of the following factors: catalyst loading (X_1), BCB concentration (X_2), H₂O₂ volume (X_3), and reaction time (X_4). The values of the aforementioned variables were selected and listed in Table S3, and thirty experiments were carried out in order to evaluate the effects of independent variables on the BCB degradation efficiency percentage (%). Table S1 shows the complete experimental design matrix as well as the observed responses (Y) and predicted responses (Y') of the BCB degradation efficiency percentage (%). We have employed a polynomial quadratic model to study the interaction between variables. The regression model predicts the response of BCB degradation efficiency percentage (%) as described below:

$$Y = 62.17 + 6.17X_1 - 7.06X_2 + 13.92X_3 + 21.05X_4 + 1.91X_1X_2 + 3.99X_1X_3 - 0.3050X_1X_4 - 1.18X_2X_3 - 3.63X_2X_4 - 1.75X_3X_4 - 11.57X_1^2 - 0.0424X_2^2 - 1.27X_3^2 + 3.79X_4^2$$

From this equation, we can notice that the catalyst load, H₂O₂ volume, reaction time, interaction between catalyst load and BCB introduced concentration, interaction between catalyst load and H₂O₂ volume, and the quadratic term of reaction time have shown a positive effect on BCB degradation efficiency. However, BCB introduced concentration, interaction between catalyst load and reaction time, along with the quadratic term of the catalyst load have shown a negative impact on BCB degradation efficiency. Analysis of variance (ANOVA) is an experimental design method that enables the analysis of each parameter in order to better define the objective function. In fact, the parameters do not share the same impact degree; some have significant effects while others do not [59]. An ANOVA was employed to assess and confirm the relevance of the quadratic polynomial model. The ANOVA results of BCB degradation efficiency are listed in Table 3: the statistical significance of the model was determined via the p and f values, where a p -value inferior to 5% indicates a statistical significance. The presented results in Table S3 show a strong correlation between the predicted and experimental degradation efficiencies (%),

supported by the R^2 (0.9875) and adjusted R^2 (0.9758) illustrated in Table S4. Figure 13a presents a comparison between the actual response values and their corresponding predicted values calculated by the model. This graph of predicted versus actual values is useful for identifying any values or groups of values that are not easily predicted by the model. The straight line resulting from the plot of experimental and predicted values (Figure 13a) demonstrates a strong correlation between the experimental data and the predictions. Additionally, Figure 13b depicts a random dispersion of residuals across the experiments, indicating that there is no significant difference between the experimental and predicted values. This has proven that the quadratic polynomial model describes, with precision, the relation between process' variables and responses, explaining 99.05% of the total variation of BCB degradation data. Parallely, the ANOVA confirms the significance of the suggested model, with a p -value and f -value less than 0.01% and 66.47%, respectively, showing a satisfying adjustment in the experimental data. Moreover, the low value of the variation coefficient (4.31%) proves the high accuracy and reliability of the experiments. The probability (p -value) determines whether the importance of models should be accepted or not. An inferior p -value to 0.05 indicates the significance of the parameter of the term in the response surface p -value (<0.0001) and confirms that the model was way significant. According to the p -values, the significant factors were catalyst loading (X_1), BCB introduced concentration (X_2), H_2O_2 volume (X_3), reaction time (X_4), interaction between catalyst load, and H_2O_2 volume (X_1X_3), interaction between BCB introduced concentration and reaction time (X_2X_4), the quadratic term of catalyst loading (X_1X_1), and the quadratic term of reaction time (X_4X_4). An adequate precision measures the signal/noise ratio; a higher value than 4 of the latter is desirable [59].

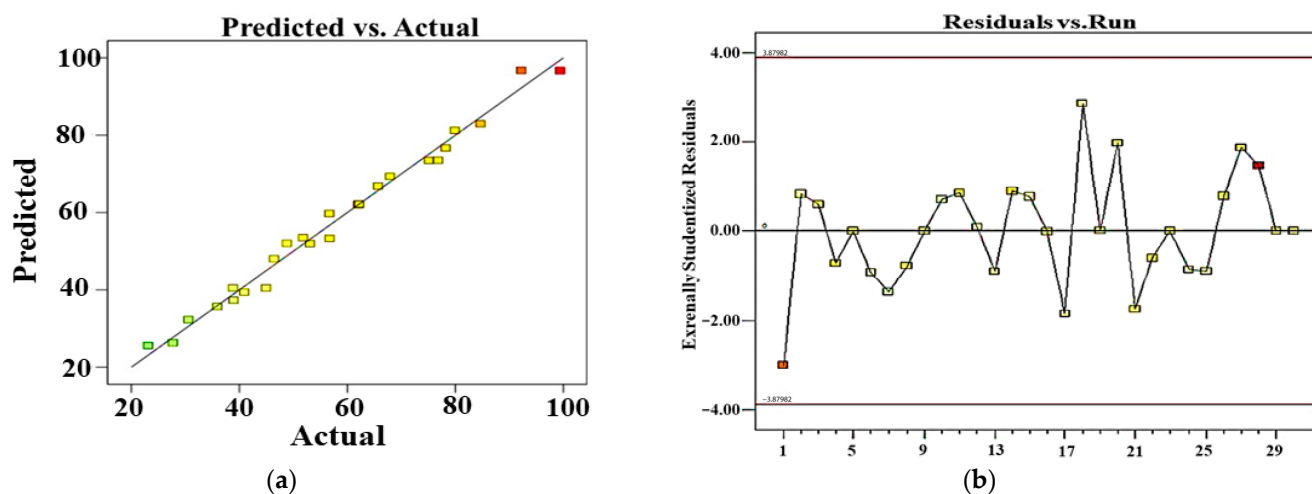


Figure 13. Plots of predicted vs. experimental values (a) and residuals vs. run numbers (b) for the removal of BCB.

Table 3. Analysis of variance (ANOVA) for the Box–Behnken design (BBD).

Source	Sum of Squares	df	Mean Square	F-Value	p -Value	
Model	10,130.59	14	723.61	84.55	<0.0001	significant
X_1	469.48	1	469.48	54.86	<0.0001	
X_2	570.63	1	570.63	66.68	<0.0001	
X_3	2465.48	1	2465.48	288.09	<0.0001	
X_4	5404.27	1	5404.27	631.49	<0.0001	
X_1X_2	14.55	1	14.55	1.70	0.2119	
X_1X_3	55.90	1	55.90	6.53	0.0219	
X_1X_4	0.3721	1	0.3721	0.0435	0.8376	
X_2X_3	5.56	1	5.56	0.6500	0.4327	

Table 3. Cont.

Source	Sum of Squares	df	Mean Square	F-Value	p-Value	
X_2X_4	39.08	1	39.08	4.57	0.0495	
X_2X_4	30.16	1	30.16	3.52	0.0801	
X_1^2	891.82	1	891.82	104.21	<0.0001	
X_2^2	3.943×10^{-6}	1	3.943×10^{-6}	4.608×10^{-7}	0.9995	
X_3^2	17.66	1	17.66	2.06	0.1714	
X_4^2	76.25	1	76.25	8.91	0.0093	
Residual	128.37	15	8.56			
Lack of Fit	128.37	10	12.84	2.271×10^5	<0.0001	significant
Pure Error	0.0003	5	0.0001			
Cor Total	10,258.96	29				

3.6.2. Response Surface and Contour Diagrams

The response surface curves and contour plots illustrating the interaction between the catalytic dose, H_2O_2 volume, BCB concentration, and time of the BCB degradation rate are shown in Figure 14a–c. In the 3D response surface diagrams, steeper response surfaces indicate a stronger influence on the response value [60]. Darker colors in these diagrams correspond to higher BCB degradation efficiency. The 2D contour plots represent the interaction and magnitude of each factor, while the 3D response surface diagrams provide a complementary visualization of the contour lines.

Figure 14a illustrates the effect of the catalyst loading and BCB concentration on degradation efficiency. An initial increase in the catalyst dose enhances degradation due to the greater availability of active sites, as discussed in Section 3.3.1 regarding the effect of catalyst dosage. However, excessive catalyst amounts can reduce efficiency due to increased turbidity or optical masking. Similarly, a lower BCB concentration promotes degradation, while an excessive concentration limits molecule access to the active sites where radical reactions occur. The optimal zones for high efficiency are found at the intermediate levels of both parameters, reflecting a synergy, whereas extreme levels result in antagonistic effects. These findings help identify the ideal conditions to maximize efficiency and guide process optimization.

Figure 14b shows the combined effect of the catalyst loading and H_2O_2 volume on the BCB degradation rate. The rate improves with increasing H_2O_2 volume and catalyst dose, up to an optimal point. This improvement can be attributed to the increased generation of $\bullet OH$ radicals through H_2O_2 decomposition. However, an excess of catalyst may reduce efficiency due to optical masking or charge recombination. Regarding H_2O_2 , a moderate volume is optimal as it generates a maximum number of radicals without producing undesirable by-products or wasting resources. The maximum degradation rate was achieved when the H_2O_2 volume was 1.37 mL. A synergistic interaction is observed in the optimal zones where these two parameters are balanced, whereas excessive or insufficient H_2O_2 volumes, combined with inappropriate catalyst concentrations, decrease efficiency.

Contact time is a critical factor with a direct and significant impact on the efficiency of the photo-Fenton-like process. A higher BCB concentration is degraded with prolonged contact time, which can be attributed to the increased availability of active sites over time. The results in 14c indicate that BCB degradation efficiency reaches an exceptionally high rate of 99% after 150 min of the decolorization process [61].

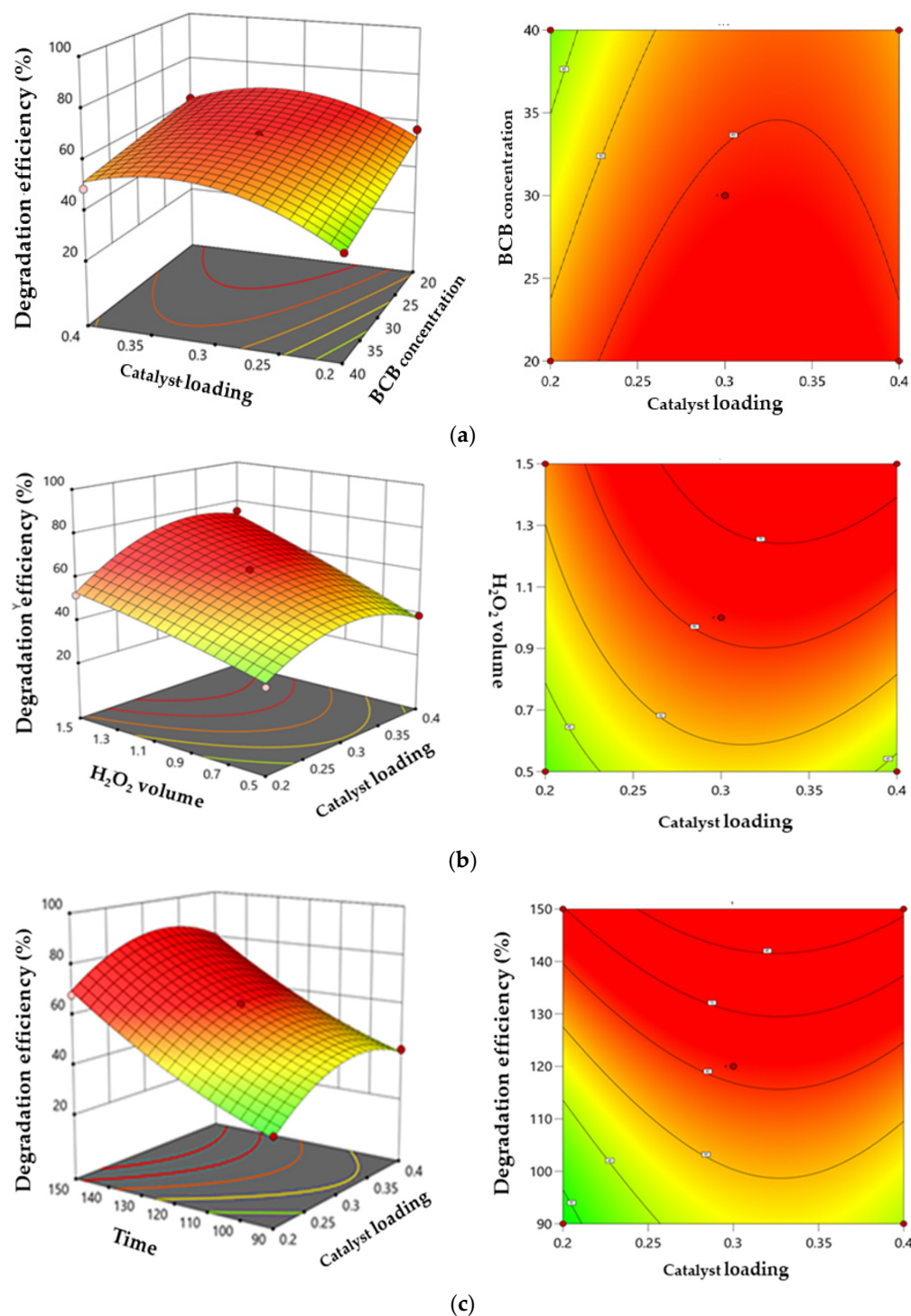


Figure 14. Three-dimensional surface and contour plots illustrating the interactive effects. (a) catalyst loading and BCB concentration. (b) catalyst loading and H₂O₂ volume. (c) catalyst loading and contact time.

3.6.3. Optimal Conditions for the Photo-Fenton-like Process

The developed model allowed to predict the optimal conditions for enhanced degradation with numeric optimization application; the model seeks for the better trade-offs in the factorial space to achieve several objectives [59]. A desired value can be picked for each input and output factor. This document suggests a diverse input optimization option, including range, maxima, minima, target, along with the absence (of response). These options are defined in such a way as to obtain the optimal value for a set of specific

conditions. The input variables and selected response in this study are depicted in Table S5. We have employed a desirability ramp diagram in order to analyze the obtained results. In fact, the desirability is an objective function that varies from zero outside the limits to the fixed target. The key point of numeric optimization consists of maximizing the desirability. The different responses and factors are combined into one single desirability function [62]. Desirability functions were used in order to determine the optimal levels for independent factors. An underlined element illustrates both factors accuracy on response and the magnitude wherein the objective has been achieved. Figure S4 depicts the results of the approach ramp desirability optimization for BCB degradation efficiency. The optimum values of catalyst load, BCB introduced concentration, and H_2O_2 volume were estimated to be approximately 0.29 g L^{-1} , 25.046 mg L^{-1} , 1.37 mL , and 149.657 min . In these conditions, the average obtained degradation was 99.347% within 150 min, with a probability of 1000. Under these predicted conditions, a verification experiment was carried out in order to verify the validity and suitability of the model. The confirmation experiment has led to a degradation (%) of 98.5%, which is too close to the predicted results. To conclude, the developed model was found to be accurate and reliable. This testifies the viability and precision of the model.

3.7. ANN Architecture Optimization

The LM (Levenberg–Marquardt) algorithm was utilized to optimize an artificial neural network (ANN), with 70% of the data designated for training and 15% each for validation and testing. The input parameters, defined according to the Box–Behnken design (BBD), included catalyst loading (X_1), initial concentration of brilliant cresyl blue (X_2), H_2O_2 volume (X_3), and time (X_4), with the output variable being the BCB degradation rate (Y). The network architecture was adjusted by testing different numbers of neurons in the hidden layer (ranging from 2 to 18), revealing a nonlinear relationship between the number of neurons and the coefficient of determination (R^2). The optimal model, with 5 neurons in the hidden layer, achieved an R^2 of 0.9996 (Figure 15a), beyond which a gradual decrease in R^2 indicated overfitting. The final configuration 4-5-1, with four inputs, five hidden neurons, and one output, proved optimal for maximizing generalization capacity while minimizing RMSE. The results also showed a good distribution of residuals (Figure 15b) and a significant increase in the R correlation coefficient, as illustrated in the regression lines.

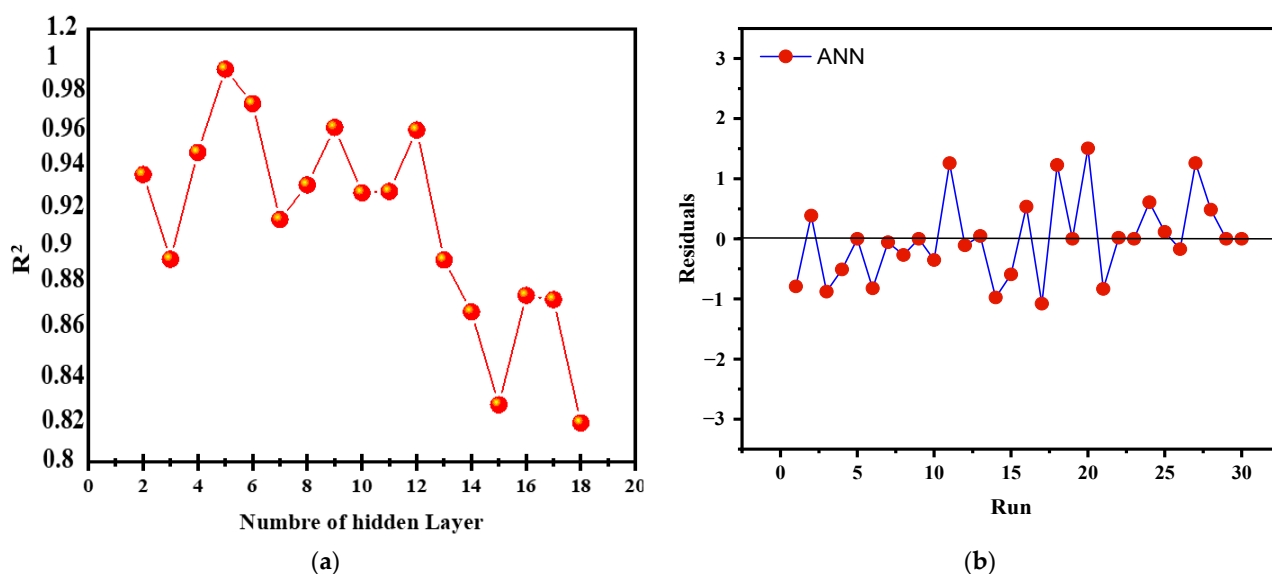


Figure 15. (a) Number of hidden neurons Vs R^2 , and (b) running and residuals of the ANN model.

3.8. Global Sensitivity Analysis

This passage describes how the estimated weights and biases in an optimized neural network (ANN) were used to determine the relative importance (RI) of the input variables in predicting the output, following Garson's formula (Garson, 1991) (Equation (S1)) [63]. According to Figure 16, the highest RI value was obtained by the catalyst dose (34.75%), followed by time (27.89%), the initial concentration of brilliant cresyl blue (23.93%), and finally, the volume of H_2O_2 (13.40%). The percentage of BCB degradation was thus highly dependent on the four studied factors, which were appropriately selected to describe the photo-Fenton degradation process.

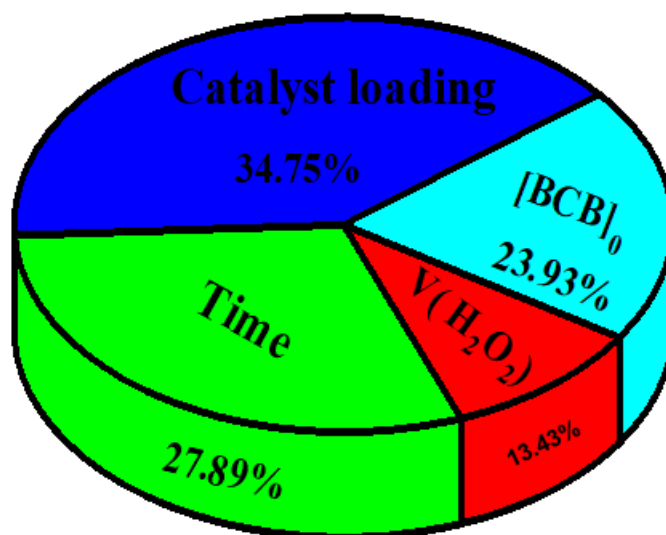


Figure 16. The relative importance of the input variables to the degradation of BCB analyzed by the ANN model.

3.9. Comparison of RSM and ANN

Comparing the ANN and RSM models is essential for determining their effectiveness in complex environments for predicting future events and identifying which model has the greatest impact based on the data and analysis criteria. This comparison helps to understand their ability to account for existing data as well as their potential to extrapolate predictions with new data. The results show that the artificial neural network (ANN) model outperforms the response surface methodology (RSM) in terms of predictive accuracy and overall fit. With determination coefficients (R^2) of 0.9996 for tests, 0.99978 for training, and 0.98 for validation (Figure 17), the ANN offers an almost perfect fit, while the RSM shows R^2 values of 0.9875 and 0.9279 for prediction, indicating a good but slightly lower accuracy. To evaluate these models, a statistical analysis based on RMSE (root mean square error) and MAE (mean absolute error) was performed (Table 4), justifying the relevance of each model while facilitating their comparison. The predicted values from both models were compared to the actual values, and as shown in Figure 18a, they are well aligned with the actual results, demonstrating their accuracy. However, the residuals from the ANN model are closer to the median line, revealing a significantly higher accuracy compared to RSM, as illustrated in Figure 18b. These results are consistent with statistical analyses showing lower RMSE and MAE values for the ANN, confirming its superiority in terms of its predictive and estimation capabilities compared to RSM, a finding also supported by previous research, which highlighted the superior predictive and estimation capabilities of the ANN architecture over the RSM approach [40,64,65].

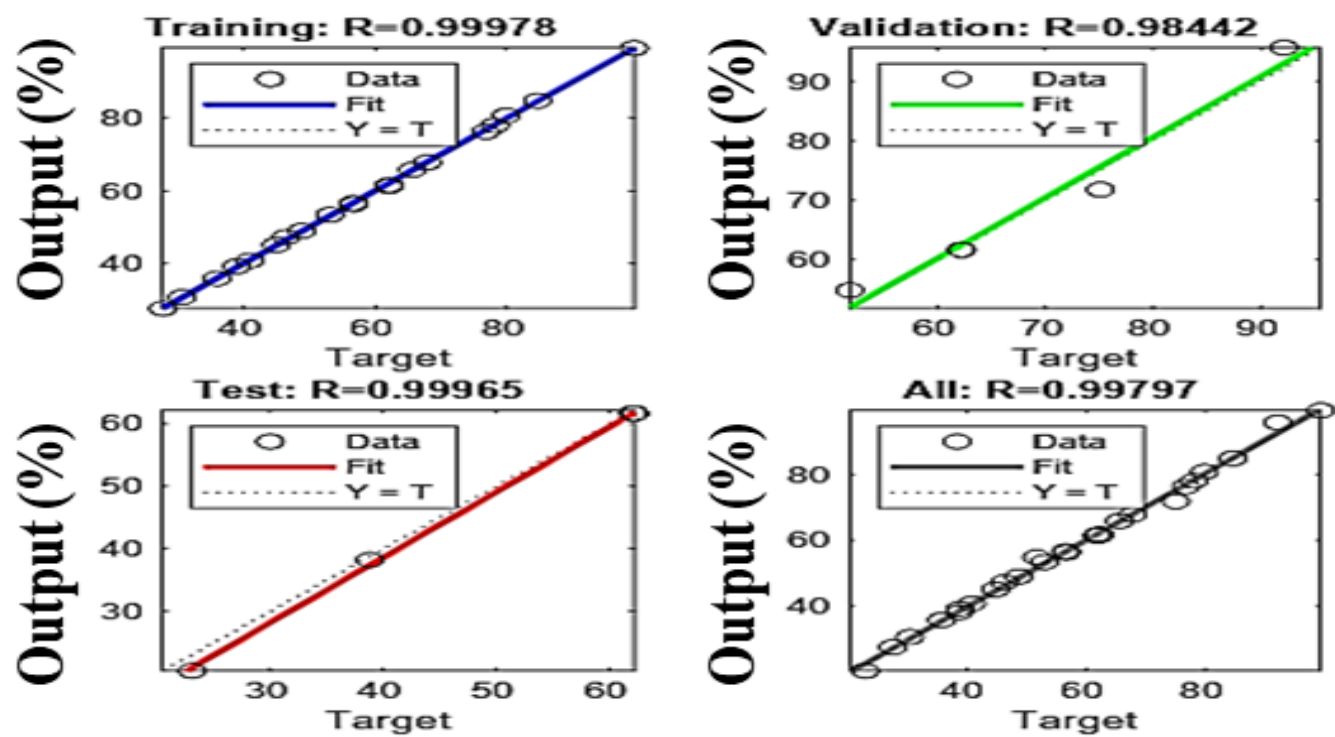


Figure 17. Regression plots of the ANN model.

Table 4. Statistical evaluation of the BBD-RSM and ANN models.

Empty Cell	Statistical Parameters		
	RMSE	MAE	R ²
RSM	1.672	1.013	0.97
ANN	1.22	0.99	0.9996

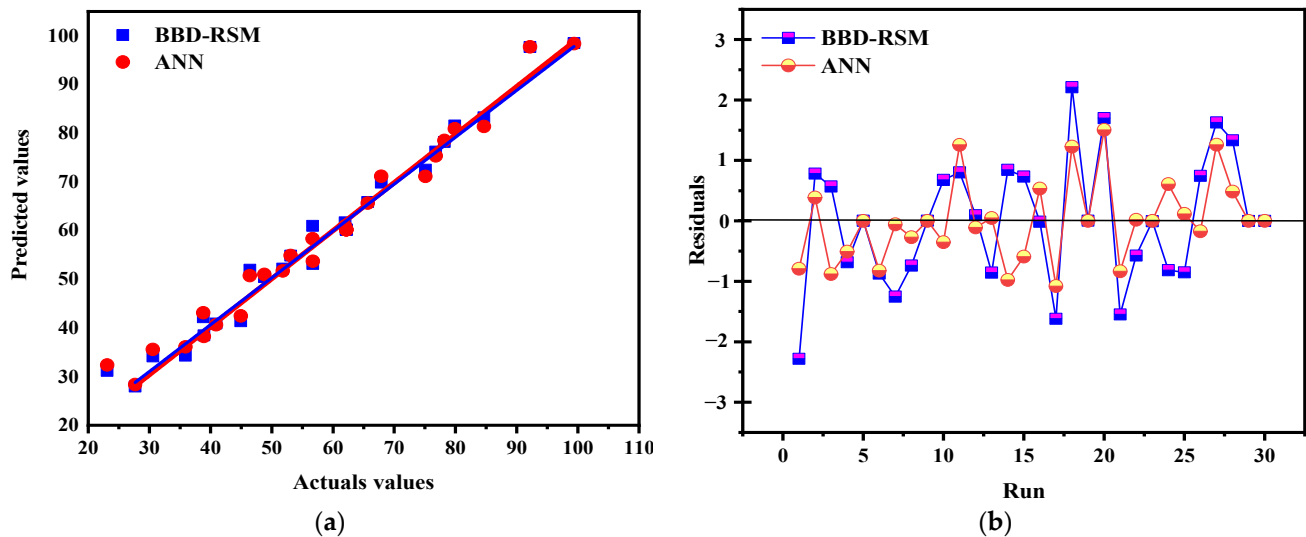


Figure 18. Actuals values vs. predicted values (a); run vs. residuals (b).

4. Conclusions

This present study demonstrates the effectiveness of a visible-light-activated SnFe₂O₄/g-C₃N₄ heterojunction nanocomposite photocatalyst for the degradation of cationic dyes in photocatalytic photo-Fenton-like and photo-Fenton-like processes. Utilizing graphitic carbon nitride (g-C₃N₄) and tin ferrate (SnFe₂O₄) synthesized via co-precipitation, the

composite was meticulously characterized by X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and transmission electron microscopy (TEM), revealing an intimate contact between the two phases. Under visible LED light irradiation, the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ composite exhibited significantly enhanced performance in degrading brilliant cresyl blue (BCB) compared to its individual components, reaching optimal activity. This improvement is due to the efficient separation of electron–hole pairs, facilitated by electron transfer and the synergistic interaction between SnFe_2O_4 and $\text{g-C}_3\text{N}_4$. Using response surface methodology (RSM) with a Box–Behnken design, the optimization of cationic dye removal was carried out by analyzing four variables. The optimal conditions for achieving a BCB removal rate of approximately 99.347% included a catalyst loading of 0.29 mg, a BCB concentration of 25.046 mg L^{-1} , an H_2O_2 volume of 1.37 mL, and a reaction time of 149.657 min. These results are consistent with those obtained from batch optimization. Modeling the experimental data with both BBD-RSM and ANN models demonstrated the effectiveness of each approach in predicting BCB removal. However, the comparative analysis favored ANN, which achieved a higher R^2 value of 0.9996 compared to RSM's R^2 value of 0.97. These findings underscore not only the efficiency of the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposite photocatalyst in breaking down recalcitrant organic pollutants but also its stability and high recyclability. The $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposite photocatalyst proves to be an effective and sustainable solution for treating wastewater contaminated with cationic dyes, with potential applications in ecological environments and in magnetic fields.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/reactions6020023/s1>, Figure S1: EDX spectrum and mapping micrographs, of $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20 %); Figure S2: shows the UV-Vis absorption spectra of different dyes at various time intervals during their degradation via the Photo-Fenton-like process using the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ (20%) composite; Figure S3: Impact of scavengers on the photocatalytic degradation of BCB using the $\text{SnFe}_2\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposite; Figure S4: Desirability Curve for Numerical Optimization of Experimental Variables; Table S1: Experimental design conditions by RSM and ANN (Y(%): actual values (%), RSM; Table S2: Statistical parameters; Table S3: Levels of variables considered for the Box–Behnken design; Table S4: Model Validation Statistics; Table S5: Range of Variables and Selected Responses in the Solutions.

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