Optimization of photocatalytic degradation of rhodamine B dye using graphite carbon nitride nanocomposite in visible light and analyzing its experimental validation in kinetics under optimal conditions

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Abstract

In this paper, graphite carbon nitride nanocomposite was synthesized by stabilization of graphitic carbon nitride nanoparticles during the hydrothermal process onto periodic mesoporous organosilica based, and its efficiency was studied in photocatalytic degradation of rhodamine B in LED photoreactor using optimizing the parameters affecting the process in response surface methodology by Box-Benken method in three variables, time (10-50 min), photocatalyst dosage (0.3-0.7 g/l) and light wavelength (472-618 nm). The accuracy and validity of the quadratic model were confirmed with a high F-value, the significance of p-value (less than 0.0001), a small percentage of coefficient of variation, high correlation coefficients (0.98), non-significance of lack of fit and lack of autocorrelation based on D-W test results. The highest efficiency of photocatalytic degradation was observed in the wavelength variable and the wavelength-dose interactive variable. The optimal conditions in this analysis were defined as the time of 50 minutes, dose of nanocomposite 0.7 g/L, and wavelength 472 nm. The predicted mean value of photocatalytic degradation based on this model was 92.2% and its experimental value in validation kinetics under optimal conditions was 90.04%. The difference between the predicted mean value and experimental value in photocatalytic is within the prediction interval of 95% and finally, the model was approved.

Keywords: Nanophotocatalyst, graphitic carbon nitride, periodic mesoporous organosilicas, RSM optimization, Rhodamine B.

1. Introduction

Photocatalytic degradation has been introduced as a very suitable option for wastewater treatment due to its non-toxicity, high selectivity, and long-term stability [1]. To improve the photocatalytic performance, the morphology of many photocatalysts can be designed on a high surface area based. Among these supporting materials for the base, mesoporous silica such as periodic mesoporous organosilica (PMO) is the best selection due to its features such as balanced porosity, a high percentage of organic compounds, high surface area, adjustable pore size, low densities, and versatile organic-inorganic hybrid frameworks [2, 3]. Recently, graphite carbon nitride nanoparticle (CNNP) has a wider band gap compared to bulk materials, so it has a larger negative conduction band and positive valence band, more potential in reduction

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and oxidation and also, and this zero-dimensional semiconductor, usually has a shorter charge-transfer length and have rich surface catalytic active sites [4, 5]. Despite the extremely valuable application and capability of PMO, there has been no study on the photocatalysis process of a catalyst on this based or the construction of heterogeneous photocatalyst or heterogeneous bonding with this support in photocatalytic degradation. This article aims to evaluate the efficiency of photocatalytic decomposition of Rhodamine B organic dye by using graphite carbon nitride nanocomposite (CNMS) in LED visible light photoreactor with RGBW color separation and to model and optimize this process using response surface test (RSM).

2. Materials and methods

In this study, CNMS nanoparticles were synthesized during the hydrothermal process with PMO through the stabilization of desired functional groups on the silicate mesoporous network and CNNP through the synthesis and separation of graphite carbon nitride nanoparticles as processes such as ultrasonication to prevent agglomeration, ultracentrifugation, using filters and freeze dryers. The design and construction of the photoreactor with the ability to control the light spectrum with a certain wavelength (the color of the lamp output) were carried out through the remote control on RGBW LED lamps. The structure of the photoreactor consists of a PVC cube with LED lamps installed on the four side walls. The light source consists of four colors: red, green, blue and white. The range of wavelength spectrum of these lamps for blue, green and red colors is about 472, 545 and 618 nm, respectively.

3. Results and discussion

Effective parameters in the removal of RhB pollutants during the photocatalysis process, including time, photocatalyst dose, and light source wavelength, were investigated using the Box-Benken Design (BBD) method. This method not only predicts reliable results as a function of other variables but also introduces the best mathematical model.

The choice of all three parameters was based on the recommended values in previous studies and also on the results of the preliminary tests (pre-test). First, in mixed light conditions (to investigate the relative effect of wavelength) with a fixed and similar dosage of 0.5 g/liter (the average of previous studies), the removal percentage was measured and compared in 30 and 90 minutes. By comparing the results, the relative importance of wavelength ranges was studied. Also, the percentage of removal in doses of 0.1 and 0.80 (upper limit and lower limit) was done in three time periods of 10, 45, and 90 minutes. By plotting the results, the effective times in these pretests were the first 10 minutes, 30 minutes, and 50 minutes, and at other times, it showed a smoother and non-significance trend.

The similarity of structural and optical properties of CNMS with BCN and PMO in the results of XRD, SAXRD, FTIR, and DRS analysis showed that the structure and framework of CNMS have been preserved and have been affected by both precursors. The crystal structure and plane characterization of BCN, CNMS and PMO can be well analyzed in the XRD and SAXRD (Small angle XRD). The presence of the (002) peak of g-C3N4 in the CNMS indicates the special effect of CNNP on structural properties of the nanocomposite. The (110) and (200) crystal planes remain in the SAXRD pattern of CNMS with a slight change, showing a slight shift. The effect of loading CNNP into PMO during CNMS synthesis is observed by intensifying the peak at 800-810 cm-1, caused by an interaction between symmetric stretching vibrations of Si-O and triazine (-C-N) [6].

4. Conclusions

According to ANOVA results, the accuracy and validity of the quadratic model were accepted with a high F-value (933), high all correlation coefficients (0.98), the significance of the p-value parameter (less than 0.0001), small% C.V. (2.96), And lack of autocorrelation on D-W test results. The wavelength variable (C) and the interaction wavelength and dosage (BC) variable have the most significant impact on photocatalytic degradation. Variable C (wavelength) will have the most and B^2 (quadratic variable of photocatalyst dosage) the least impact on the photocatalytic degradation process. Three-dimensional diagrams play an important role in the interpretation of surface response by depicting interactions between experimental variables and their effects on experimental results. The increase in the response (photocatalytic efficiency) occurs with increasing the dosage due to the increase in the number of active sites on the surface of the catalyst and the increase in effectiveness with the production of more hydroxyl and superoxide radicals. However, by increasing the amount more than the optimal value, the turbidity of the suspension increases and this reduces the penetration of light. The synergistic interaction of radiation wavelength and nano photocatalyst dose, which assigns the highest F value to itself (BC), can be well analyzed in three-dimensional figures. The effect of reducing the wavelength and improving the removal may be affected by the increase in electron-hole pair production at shorter wavelengths. Comparing the photocatalytic efficiency of this study with previous articles shows the higher efficiency and effectiveness of CNMS nanocomposite in removing RhB dye in a shorter time.

References

[1] D. Yang, X. Zhao, X. Zou, Z. Zhou, Z. Jiang, Removing Cr (VI) in water via visible-light photocatalytic reduction over Cr-doped SrTiO3 nanoplates, Chemosphere, 215 (2019) 586-595.
[2] B. Ekka, M.K. Sahu, R.K. Patel, P. Dash, Titania coated silica nanocomposite prepared via encapsulation method for the degradation of safranin-O dye from aqueous solution: optimization using statistical design, Water Resources and Industry, 22 (2019) 100071.
[3] Z. Teng, W. Li, Y. Tang, A. Elzatahry, G. Lu, D. Zhao, Mesoporous organosilica hollow nanoparticles: synthesis and applications, Advanced Materials, 31(38) (2019) 1707612.
[4] L. Wang, G. Zhou, Y. Tian, L. Yan, M. Deng, B. Yang, Z. Kang, H. Sun, Hydroxyl decorated g-C3N4 nanoparticles with narrowed bandgap for high efficient photocatalyst design, Applied Catalysis B: Environmental, 244 (2019) 262-271.
[5] H. Jing, R. Ou, H. Yu, Y. Zhao, Y. Lu, M. Huo, H. Huo, X. Wang, Engineering of g-C3N4 nanoparticles/WO3 hollow microspheres photocatalyst with Z-scheme heterostructure for boosting tetracycline hydrochloride degradation, Separation and Purification Technology, 255 (2021) 117646.
[6] D.P. Sahoo, S. Patnaik, D. Rath, B. Nanda, K. Parida, Cu@ CuO promoted gC 3 N 4/MCM-41: an efficient photocatalyst with tunable valence transition for visible light induced hydrogen generation, RSC advances, 6(113) (2016) 112602-112613.