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Effect of Immobilized-ZnO Resin and UV Lamp Spacing on Continuous Photocatalyst Reactor for Total-N and Phosphate Removal

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Abstract

The tofu industry generates wastewater with high concentrations of Total Nitrogen (Total-N) and phosphate, posing significant environmental risks if untreated. This study evaluates the performance of a continuous photocatalysis reactor using Resin-Immobilized Photocatalyst Zinc Oxide (RIP-ZnO) to remove Total-N and phosphate from tofu wastewater. The research varied the RIP-ZnO mass (25, 37.5, and 50 g) and the distance between the Ultraviolet (UV) lamp and the liquid surface (0, 10, and 20 cm). Prior to treatment, the wastewater was filtered to remove coarse particles and then processed in a tubular continuous-flow photocatalysis reactor at a constant flow rate of 30 mL/min. Samples were collected at 0, 4, 8, and 12 hours and analyzed for Total-N and phosphate concentrations using spectrophotometric methods based on Standar Nasional Indonesia (SNI) 6989.11:2019 standards. One-way Analysis of Variance (ANOVA) was used to assess the significance of treatment variables. Results showed optimal removal efficiency with 25 g of RIP-ZnO and a UV lamp distance of 0 cm, achieving 65.05% Total-N and 67.22% phosphate removal. The ANOVA test yielded a p-value < 0.05, indicating that both RIP-ZnO mass and UV distance significantly affected removal efficiency. The removal mechanism involved oxidation by Hydroxyl Radicals (•OH) generated by ZnO and ion exchange by the resin. Under optimal conditions, the reactor reduced Total-N from 18.3 mg/L to 6.4 mg/L and phosphate from 6.4 mg/L to 2.1 mg/L.

Keywords: Photocatalysis, Tofu wastewater, Resin-immobilized Photocatalyst Zinc Oxide, Total Nitrogen, Phosphate.

1 | Introduction

The tofu industry generates liquid waste with high organic content due to the continued use of simple production technologies, resulting in low resource utilization efficiency, particularly for raw materials and

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water, and substantial waste generation [1–3]. The liquid waste from tofu production has a Chemical Oxygen Demand (COD) value ranging from 7,000 to 12,000 mg/L, which significantly exceeds the regulatory maximum COD limit of 300 mg/L for soybean processing, as stipulated in Indonesian Regulation PermenLHK No. 5 of 2014. This waste has the potential to pollute aquatic environments if discharged directly without proper treatment [4]. Moreover, the high Total Nitrogen (Total-N) and phosphate content in tofu wastewater can contribute to eutrophication [5]. Tofu wastewater treatment can be performed through biological methods, such as aerobic or anaerobic processes utilizing bacteria, or through physico-chemical methods, such as coagulation-flocculation followed by sedimentation [1]. However, biological methods have limitations, including long processing times and the need for large land areas, whereas chemical methods involve high operational costs and generate hazardous sludge classified as B3 waste, which requires further treatment [6].

The photocatalysis method offers an alternative solution for treating tofu wastewater. Photocatalysis employs semiconductor materials, such as ZnO, to degrade COD and BOD into CO_2 and H_2O through a photocatalytic oxidation mechanism activated by Ultraviolet (UV) light [7]. However, the direct use of photocatalyst powders presents challenges, including difficulties in separating catalyst particles from the treated effluent [8] and low adsorption capacity [9]. To address these limitations, ZnO photocatalysts can be immobilized in resins, forming Resin-Immobilized Photocatalyst-Zinc Oxide (RIP-ZnO), which enhances photocatalytic activity and simplifies the separation process. RIP-ZnO has been demonstrated to effectively reduce COD and BOD levels in tofu wastewater, with its primary components being resin and ZnO [10]. Factors such as catalyst mass in the reactor and the distance between the UV lamp and the water significantly influence the efficiency of the photocatalysis process. While increasing the catalyst amount can enhance pollutant degradation, excessive catalyst mass may reduce removal efficiency due to agglomeration [11]. Additionally, optimal UV lamp spacing is crucial to ensure sufficient energy input for the photocatalytic reaction [12]. Previous studies have demonstrated the effectiveness of RIP-ZnO in batch systems for reducing COD in tofu wastewater. In this study, a continuous photocatalysis reactor is employed to investigate the effects of RIP-ZnO mass variation and UV lamp spacing on the degradation of COD, Total-N, and phosphate. The determination of optimum reactor conditions is expected to support large-scale applications for treating wastewater from the tofu industry.

2 | Research Method

The research was conducted on a continuous scale using tofu industry wastewater, which was collected directly from the source as the research sample. Before experimentation, the wastewater was filtered to remove coarse particles. The resin was modified with a ZnO photocatalyst, forming RIP-ZnO, which was synthesized using the impregnation method and was incorporated into the resin and subsequently dried for use. The wastewater was introduced into a continuous photocatalysis reactor during the process, and the RIP-ZnO mass was added based on the predetermined treatment variations. The UV lamp was positioned at specified distances from the water surface in accordance with the experimental design.

The research utilized several tools, including a continuous photocatalysis reactor, a UV lamp, and supporting equipment. The photocatalysis reactor was specifically designed for processing wastewater, while the UV lamp, with a wavelength of 365 nm, was used to activate the photocatalyst. Supporting equipment included a peristaltic pump to regulate the wastewater flow and a spectrophotometer for analyzing Total-N and phosphate concentrations, following the methods outlined in Standar Nasional Indonesia (SNI) 6989.11:2019. A pH meter was also employed to monitor environmental conditions within the reactor, with measurements based on the same standard. In the phosphate and Total-N tests, additional reagents were used, including Ascorbic Acid ($C_6H_8O_6$) for the phosphate test, and Potassium Nitrate (KNO_3) for the Total-N test.

The effluent was processed in the reactor for varying durations. During the catalytic process, effluent samples were collected at 4-hour intervals (0, 4, 8, and 12 hours) to assess the treatment effectiveness. The flow rate

of tofu wastewater was set at 30 mL/min. The RIP-ZnO composition consisted of a 1:1 ratio of resin to ZnO photocatalyst, which was mixed into 1000 mL of distilled water. The reactor was configured as a system of three tubes, each with a diameter of 3.1 cm and a height of 50 cm. The mass of RIP-ZnO was varied (25, 37.5, and 50 g), as well as the distance between the UV lamp and the water surface (0, 10, and 20 cm). The experimental matrix is shown in *Table 1*.

Table 1. Research variable matrix.

RIP-ZnO Mass (g)	Distance Between Liquid and Lamp (Cm)	Sampling Time (Hour)	Code
25	0	0	A
		4	
		8	
		12	
	10	0	B
		4	
		8	
		12	
	20	0	C
		4	
		8	
		12	
37,5	0	0	D
		4	
		8	
		12	
	10	0	E
		4	
		8	
		12	
	20	0	F
		4	
		8	
		12	
50	0	0	G
		4	
		8	
		12	
	10	0	H
		4	
		8	
		12	
	20	0	I
		4	
		8	
		12	

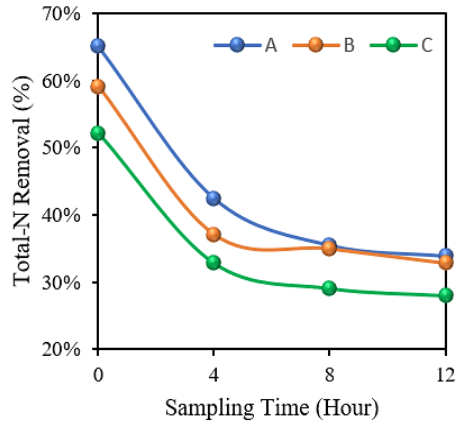
After conducting the experiments based on the planned research matrix, the results will be analyzed statistically using one-way Analysis of Variance (ANOVA) to determine the average effect on the removal efficiency of Total-N and phosphate. Statistical analysis will be performed using Minitab 17, a free version of the software.

3|Results and Discussion

3.1|Total-N Removal

The initial Total-N concentrations in tofu wastewater ranged from 59.5 to 70.35 mg/L. The results indicated that both the resin and RIP-ZnO were effective in reducing Total-N at all sampling time intervals. Variations in RIP-ZnO mass, UV lamp distance from the water in the reactor, and sampling time resulted in differing percentages of Total-N removal. The formation of electron holes, which generate radical species, played a key role in the decomposition of Total-N through photocatalytic oxidation. The percentage of Total-N

removal was observed for RIP-ZnO with mass treatments of 25, 37.5, and 50 g at UV lamp distances of 0, 10, and 20 cm from the water surface.



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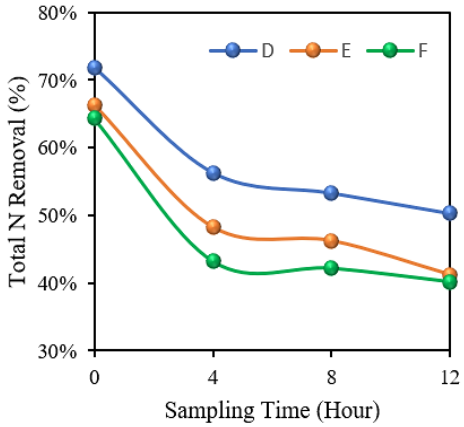

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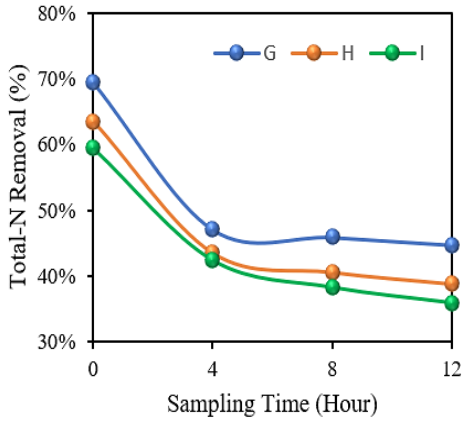


c.

Fig. 1. Total-N removal percentage of Tofu wastewater using RIP-ZnO 25 g; a. RIP-ZnO 37.5 g, b. RIP-ZnO 50 g, c. in a Photocatalytic reactor.

Based on the Total-N removal graph in *Fig. 1*, the highest percentage of Total-N removal occurred at the 0-hour sampling time with a UV lamp distance of 0 cm. The Total-N removal ranged from 65.05% to 71.86% at the 0-hour sampling time with a lamp distance of 0 cm, 59.14% to 66.33% with a lamp distance of 10 cm, and 52.15% to 64.32% with a lamp distance of 20 cm, using RIP-ZnO media at various mass variations. In

contrast, the Total-N reduction using resin as the control medium at the 0-hour sampling time with lamp distances of 0, 10, and 20 cm was 37.31%, 36.82%, and 35.32%, respectively.

At the 0-hour sampling time in the continuous photocatalysis reactor, the highest percentage of Total-N reduction was observed for each mass treatment and lamp distance. However, at the 4-hour, 8-hour, and 12-hour sampling times, the percentage of Total-N removal decreased. For example, at the 0-hour sampling time, the Total-N removal with 37.5 g RIP-ZnO and a lamp distance of 0 cm was 71.86%, which decreased to 50.25% at the 12-hour sampling time with the same mass treatment and lamp distance. These results indicate that as the reactor operation time increases, the efficiency of Total-N removal decreases. This decline in removal efficiency occurs because, at the initial stage, the RIP-ZnO media is in optimal condition, allowing for high degradation capacity. Over time, the media becomes saturated, reducing its ability to degrade pollutants, which leads to a higher concentration of Total-N contaminants in the effluent compared to earlier sampling times, resulting in a decrease in the removal percentage [13]. The reactor operating time is directly related to the duration of photocatalyst irradiation. As the operating time increases, the irradiation period also lengthens, leading to the accumulation of adsorbed products on the photocatalyst. These adsorbed products can block the interaction between UV light, the photocatalyst, and the pollutants that have yet to be degraded, thus reducing the effectiveness of the photodegradation process.

Phytodegradation activity decreases as the photocatalyst becomes saturated with prolonged irradiation time [14]. The reduction in degradation effectiveness is likely due to the active sites of RIP-ZnO becoming saturated with Total-N and other pollutants present in the tofu wastewater, leading to an adsorption-desorption equilibrium on the surface of RIP-ZnO [15]. In this study, Total-N removal reached a stable state starting from the 4-hour sampling time, indicating that RIP-ZnO had reached saturation in removing Total-N from tofu wastewater. The percentage of Total-N removal consistently decreased from the 0-hour to the 12-hour sampling time. The saturation time for RIP-ZnO in Total-N removal is estimated to occur within 40 minutes, where the photocatalytic process for degrading ionic substances reaches its maximum capacity and remains relatively constant with further contact time. These findings suggest that after 40 minutes, no additional ionic substances are removed from the wastewater [16].

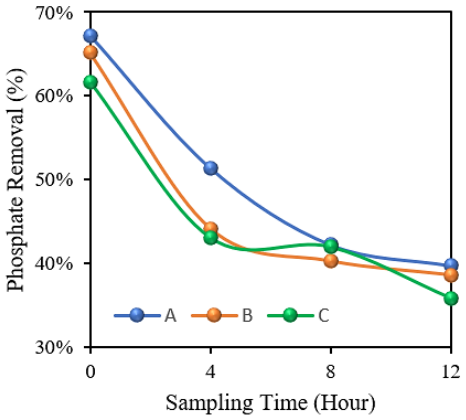
The study also found that the optimal distance between the UV lamp and the water for effective Total-N degradation is 0 cm. At this distance, the percentage of Total-N removal at the 0-hour sampling time with 37.5 g of RIP-ZnO treatment reached 71.86%. However, at a lamp distance of 20 cm with the same RIP-ZnO mass treatment and sampling time, the removal efficiency decreased to 64.32%. These results demonstrate that as the distance between the light source and the water increases, the Total-N removal efficiency in the wastewater decreases. The power radiated over a given area will result in a flux density or power density per unit area. As the distance from the radiation source increases, the flux density decreases. The flux density is directly related to the intensity of the radiation [17]. The closer the distance between the UV lamp and the water, the greater the reduction in Total-N removal efficiency. This phenomenon is because a shorter distance between the lamps, which initiates the photochemical reaction, enhances the photocatalytic activity of RIP-ZnO, as electron pairs are generated more quickly [18]. The experimental results indicate that the optimal RIP-ZnO mass for Total-N removal in the photocatalytic reactor is 37.5 g, where the percentage of Total-N removal reaches 71.86%. This value is followed by 65.05% at 25 g and 69.41% at 50 g, all measured at the 0-hour sampling time with a lamp distance of 0 cm. These results suggest that increasing the mass of RIP-ZnO does not always lead to a proportional increase in Total-N removal efficiency. Increasing the mass of RIP-ZnO can enhance the percentage of Total-N removal by providing more electron holes available to degrade Total-N. However, adding a larger mass of RIP-ZnO may lead to a decrease in removal efficiency due to the potential agglomeration of the photocatalyst, which reduces the active surface area and, consequently, the photocatalytic activity. This decrease in surface area can diminish the overall pollutant removal efficiency [19].

The optimal operating conditions in the continuous photocatalysis reactor for reducing Total-N were achieved with a RIP-ZnO mass of 37.5 g and a UV lamp distance of 0 cm. These conditions allowed for adequate contact between the RIP-ZnO and tofu wastewater without significant clumping, and the 0 cm lamp

distance promoted better formation of electron holes. The best removal efficiency was observed within the 0 to 4-hour sampling time range, where the final Total-N concentration met the quality standard of 19.60 mg/L. However, at the 4-hour sampling time, the Total-N level approached the quality standard with a value of 30.45 mg/L. Statistical analysis using one-way ANOVA was conducted to determine if there were significant differences in the average removal efficiency across various treatment variations. The results of the statistical analysis for the effect of lamp distance on Total-N removal showed a p-value of 0.181, indicating that there is no statistically significant difference in the average Total-N removal between the different lamp distances, as the p-value is greater than 0.05.

4 | Phosphate Removal

The initial measurements revealed that the phosphate concentrations in tofu wastewater ranged from 27.22 to 36.85 mg/L. The results also demonstrated that both resin and RIP-ZnO were effective in reducing phosphate levels across all sampling time variations. Variations in the mass of RIP-ZnO, the distance between the UV lamp and the water, and the sampling time resulted in different phosphate removal efficiencies. The formation of electron holes, which generate radical species, plays a key role in the phosphate degradation process during photocatalytic oxidation. In the continuous photocatalysis reactor, it was observed that extended operating times lead to media saturation, reducing its effectiveness in degrading pollutants in tofu wastewater. *Fig. 2* illustrates the percentage of phosphate removal achieved using RIP-ZnO with mass treatments of 25 g, 37.5 g, and 50 g at UV lamp distances of 0 cm, 10 cm, and 20 cm.



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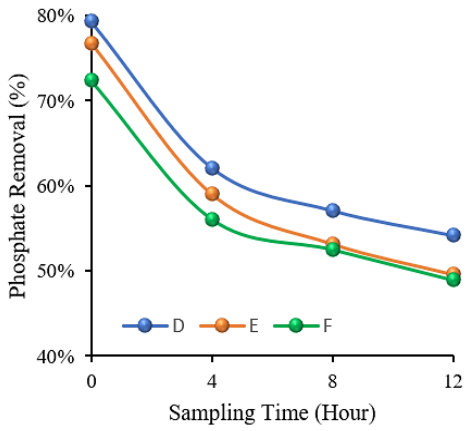

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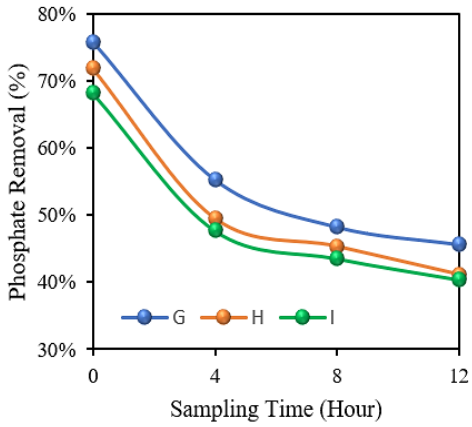

c.

Fig. 2. Percentage Phosphate removal of Tofu wastewater using RIP-ZnO 25 g; a. RIP-ZnO 37.5 g, b. RIP-ZnO 50 g, c. in a photocatalytic reactor.

The initial measurements revealed that the phosphate concentrations in tofu wastewater ranged from 27.22 to 36.85 mg/L. The results also demonstrated that both resin and RIP-ZnO were effective in reducing phosphate levels across all sampling time variations. Variations in the RIP-ZnO mass, UV lamp distance from the water, and sampling time influenced the percentage of phosphate removal. The formation of electron holes, leading to the generation of radical species, plays a key role in the decomposition of phosphate during photocatalytic oxidation. In the continuous photocatalysis reactor, it was observed that prolonged operation leads to media saturation, reducing its effectiveness in degrading pollutants. *Fig. 2* illustrates the percentage of phosphate removal achieved with RIP-ZnO at mass treatments of 25 g, 37.5 g, and 50 g, and UV lamp distances of 0 cm, 10 cm, and 20 cm.

At the 0-hour sampling time in the continuous photocatalysis reactor, the highest percentage of phosphate removal was observed for all mass treatments and lamp distances (*Fig. 2*). However, the percentage of phosphate removal decreased at the 4, 8, and 12-hour sampling times. Specifically, at the 0-hour sampling time, the phosphate removal with 37.5 g of RIP-ZnO and a 0 cm lamp distance was 79.35%, which decreased to 54.15% at the 12-hour sampling time under the same conditions of RIP-ZnO mass treatment and lamp spacing. This trend indicates that as the reactor operates for longer periods, the efficiency of phosphate removal decreases. In the initial stages, both the RIP-ZnO and resin media are in their optimal, unused state, allowing for a high degradation capacity. However, as time progresses, these media become saturated, reducing their degradation capability and leading to an increase in the concentration of phosphate contaminants in the wastewater compared to earlier sampling times, resulting in a decrease in removal efficiency [13]. The operating time of the reactor is directly related to the duration of photocatalyst irradiation. As the operating time increases, the length of irradiation also increases, leading to a greater accumulation of adsorbed products on the photocatalyst. The accumulation of adsorbed products on the photocatalyst surface can obstruct the interaction between UV light, the photocatalyst, and the pollutants that remain undegraded. This obstruction leads to progressively less effective photodegradation. Additionally, it is hypothesized that as the irradiation time increases, the photocatalyst becomes saturated, resulting in a decrease in its photodegradation activity [14]. The decline in degradation efficiency is likely due to the saturation of the active sites on the RIP-ZnO and native resin by phosphate and other pollutants from the tofu wastewater, which leads to the establishment of an adsorption-desorption equilibrium on the surfaces of both the RIP-ZnO and the native resin [15]. Phosphate removal consistently exhibited a decreasing trend from the 0-hour to the 12-hour sampling time. It is estimated that the saturation time of the RIP-ZnO media for phosphate removal occurs at approximately 40 minutes. After this period, the photocatalytic degradation of ionic substances reaches its maximum capacity and remains stable, even with further extension of contact time. This observation suggests that no additional ionic substances are being removed from the wastewater beyond this point.

The optimal distance between the UV lamp and the water for phosphate degradation is found to be 0 cm. At this distance, the percentage of phosphate removal at the 0-hour sampling time with 37.5 g of RIP-ZnO mass treatment was 79.35%. However, this percentage decreased to 72.36% when the UV lamp was positioned 20 cm away from the water, with the same RIP-ZnO mass treatment and sampling time. The finding demonstrates that as the distance between the light source and the water increases, the efficiency of phosphate removal in wastewater decreases. The power radiated over a specific area will have a corresponding flux density, or power density, per unit area. As the distance from the radiation source increases, the flux density decreases. This reduction in flux density is directly related to the intensity of radiation [17]. Consequently, when the UV lamp is closer to the water, the percentage of phosphate reduction is higher. This phenomenon is because a shorter distance between the lamp and the water enhances the initiation of the photochemical reaction, leading to a higher photocatalytic activity of RIP-ZnO, as electron pairs are generated more rapidly [18].

The experimental results indicate that the optimal RIP-ZnO mass for phosphate removal in the photocatalytic reactor is 37.5 g, with a phosphate removal efficiency of 79.35%. At a mass of 25 g, the removal efficiency was 67.22%, while at 50 g, it was 75.82%, all at the 0-hour sampling time with a lamp distance of 0 cm. These results demonstrate that an increase in mass does not always correlate directly with a proportional increase in phosphate removal. While increasing the RIP-ZnO mass can enhance phosphate removal by providing more electron holes for degradation, excessive mass may lead to agglomeration of the photocatalyst, reducing its effective surface area and ultimately decreasing the pollutant removal efficiency [19].

The optimal operating condition for phosphate removal in the continuous photocatalysis reactor was achieved by using a RIP-ZnO mass of 37.5 g and a UV lamp distance of 0 cm. This setup ensures that the RIP-ZnO maintains effective contact with tofu wastewater, minimizing clumping. A UV lamp positioned 0 cm from the water surface facilitates better electron-hole formation. The best removal efficiency was observed in the range of 0 to 4-hour sampling times. However, even under these optimal conditions, the final phosphate concentration did not meet the quality standard of 6.12-11.23 mg/L. The results of the statistical analysis using the one-way ANOVA method tested whether there were significant differences in the averages obtained from various treatment variations and provided an overview of the most effective variants. The statistical analysis of the lamp distance on the percentage of phosphate reduction yielded a p-value of 0.544. Since the p-value is greater than 0.05, it indicates that there is no statistically significant difference between the different lamp distance variables, and the same percentage of phosphate removal was observed across these conditions.

Based on the sampling time, the results of the statistical analysis using the ANOVA method revealed a p-value of 0.000, which is less than 0.05. These results indicate that the sampling time significantly affects the percentage of phosphate removal. When considering all the results for total-N and phosphate pollutant removal, the optimal operating conditions in the continuous photocatalysis reactor for reducing COD, phosphate, and total-N were found to be using a RIP-ZnO mass of 37.5 g and a lamp distance of 0 cm. This configuration ensures optimal contact between the RIP-ZnO and tofu wastewater, minimizing clumping. Additionally, the UV lamp distance of 0 cm enhances the formation of electron holes. The best percentage removal is achieved within the operating time range of 0 to 4 hours.

5 | Conclusion

This study successfully demonstrated the effectiveness of a continuous photocatalysis reactor utilizing RIP-ZnO for the removal of Total-N and phosphate from tofu industry wastewater. The experimental results revealed that the optimal operating conditions for pollutant removal were achieved with a RIP-ZnO mass of 37.5 g and a UV lamp distance of 0 cm. Under these conditions, significant removal efficiencies of Total-N (71.86%) and phosphate (79.35%) were observed, particularly during the 0-hour sampling time. The study further highlighted the critical role of reactor operating time, where the removal efficiencies decreased as the reactor run time increased, likely due to the saturation of the photocatalyst.

Statistical analysis using one-way ANOVA confirmed that variations in sampling time significantly influenced the removal efficiencies, while the effects of UV lamp distance on removal were found to be less impactful. The results also suggested that while increasing the RIP-ZnO mass could enhance pollutant removal, excessive mass may lead to agglomeration, reducing the effective surface area and limiting the overall removal efficiency.

Although the optimal conditions did not fully meet the quality standards for phosphate concentrations (6.12 - 11.23 mg/L), the findings provide valuable insights into the operational parameters that can be optimized for more efficient pollutant removal in continuous photocatalysis systems. Future studies could explore further optimization of reactor design, media configuration, and photocatalytic materials to enhance treatment efficiency and meet environmental discharge standards.

Authors' Contributions

All aspects of the research and manuscript preparation were carried out by the author. The author has read and approved the final version of the manuscript.

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Data Availability

All data supporting the reported findings in this research paper are provided within the manuscript.

Conflict of Interest

The author declares that in this paper, there is no conflict of interest.

Consent for Publication

The author confirms consent for the publication of this work

Ethics Approval and Consent to Participate

This article does not involve studies with human participants or animals conducted by any authors.

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