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Nitrite Removal Efficiency from Tofu Wastewater Using Robusta Coffee Grounds as a Biosorbent

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Abstract
Tofu wastewater may contain toxic nitrite ions (NO_2^-) due to inadequate treatment before discharge into aquatic environments. This study investigated the effectiveness of Robusta spent coffee grounds as a low-cost biosorbent for nitrite removal from tofu wastewater using a batch adsorption process. The biosorbent was prepared by carbonising dried coffee grounds at 350°C for 3.5 hours, followed by chemical activation with 0.1 M HCl for two days, washing to neutral pH, and drying. Adsorption experiments were conducted using 100 mL of tofu wastewater with an initial nitrite concentration of 40 mg/L. Biosorbent masses of 0.2, 0.3, and 0.4 g and contact times of 10, 30, 50, 70, and 90 minutes were evaluated. The residual nitrite concentration was determined using UV–Visible spectrophotometry. The results demonstrated that nitrite-removal efficiency increased with increasing biosorbent mass and contact time. The highest removal efficiency of 77.75% was achieved using 0.4 g of biosorbent at a contact time of 90 minutes, reducing the nitrite concentration to 8.9 mg/L. Meanwhile, the highest adsorption capacity of 14.50 mg/g was obtained using 0.2 g of biosorbent at 90 minutes. Kinetic analysis showed that both pseudo-first-order and pseudo-second-order models adequately described the adsorption process, with coefficients of determination ranging from 0.86 to 0.96. However, the pseudo-second-order model produced adsorption-capacity estimates closer to the experimental values, suggesting that chemisorption contributed to the overall removal mechanism. A preliminary equilibrium analysis indicated that the Freundlich isotherm provided a better fit than the Langmuir model, although additional experiments involving different initial nitrite concentrations are required. Two-way ANOVA confirmed that biosorbent mass and contact time significantly affected nitrite-removal efficiency, with contact time identified as the dominant factor. These findings demonstrate that Robusta spent coffee grounds have considerable potential as an environmentally friendly biosorbent for treating nitrite-contaminated tofu wastewater.

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1. Introduction

Population growth contributes positively to economic development by encouraging the establishment and expansion of businesses that meet increasing societal needs. However, the growth of industrial and household-scale production activities also increases the volume of wastewater discharged into the environment. Inadequately treated industrial effluents may introduce toxic and persistent pollutants into surface waters, leading to the deterioration of water quality and disruption of aquatic ecosystems. One pollutant of concern in tofu-processing wastewater is nitrite (NO_2^-), an inorganic nitrogen compound that can be toxic to aquatic organisms.

Tofu production is an important soybean-based food-processing industry that generates considerable quantities of wastewater containing high concentrations of biodegradable organic matter and suspended solids. Previous studies conducted in Banda Aceh reported biochemical oxygen demand (BOD) concentrations of 3,500–4,500 mg/L and chemical oxygen demand (COD) concentrations of 5,000–8,500 mg/L in untreated tofu wastewater, indicating a substantial organic pollutant load [1], [2]. The direct discharge of wastewater with a high organic load can increase the oxygen required for microbial decomposition, reduce dissolved oxygen concentrations, deteriorate water quality, and adversely affect aquatic organisms [3].

The discharge of untreated tofu wastewater is inconsistent with the environmental protection requirements established under Indonesian legislation, including Law No. 32 of 2009 concerning Environmental Protection and Management, as amended by Law No. 6 of 2023 [4], Government Regulation No. 22 of 2021 [5], and Minister of Environment Regulation No. 5 of 2014 concerning Wastewater Quality Standards [6]. According to Appendix XVIII of Minister of Environment Regulation No. 5 of 2014, wastewater generated by soybean-processing industries must meet maximum discharge limits of 150 mg/L for BOD, 300 mg/L for COD, and 200 mg/L for total suspended solids (TSS). The permissible pH range is 6–9, while the maximum wastewater generation rate is 20 m^3 per tonne of raw material [6].

In addition to organic matter and suspended solids, tofu wastewater may contain nitrogen compounds, including ammonia, nitrate, and nitrite, which are generated through the decomposition of soybean proteins and subsequent nitrogen-transformation processes [3]. Nitrite is an intermediate inorganic nitrogen species that may accumulate during nitrification and denitrification. At elevated concentrations, nitrite poses a considerable risk to aquatic organisms. It can enter fish through the gills and oxidise haemoglobin into methaemoglobin, which has a reduced capacity to transport oxygen. This process may impair respiration, ionic balance, growth, and the survival of aquatic organisms [7], [8].

Excessive inputs of inorganic nitrogen, particularly when accompanied by phosphorus, may also increase nutrient availability, stimulate primary productivity and algal growth, and accelerate eutrophication. These processes can ultimately reduce dissolved oxygen levels and further degrade aquatic ecosystems [9]. Saadah et al. reported that untreated tofu wastewater exhibited a 96-h median lethal concentration (LC_{50}) of 8.545% and an acute toxicity unit value of 11.70 in Nile tilapia (Oreochromis niloticus) [10]. The wastewater contained BOD, COD, and TSS concentrations of 871.62, 2,149.66, and 947.82 mg/L, respectively, together with a pH of 4.08 and an ammonia concentration of 3.81 mg/L [10]. These results indicate that the observed toxicity was associated with the combined effects of multiple pollutants rather than nitrite alone. Nevertheless, the findings emphasise the importance of treating tofu wastewater before its discharge into receiving water bodies.

Adsorption is a promising approach for reducing nitrite concentrations in wastewater because of its relatively simple operation and its potential application using low-cost materials. The use of biosorbents derived from organic residues is particularly attractive because it simultaneously addresses wastewater pollution and solid-waste management. Spent coffee grounds are abundantly generated by households, coffee shops, and coffee-processing activities. As a lignocellulosic biomass, spent coffee grounds contain cellulose, hemicellulose, lignin, polyphenols, and carbon-rich compounds. Their surface functional groups, including hydroxyl and carboxyl groups, may provide active sites for interactions with inorganic ions and other pollutants.

Previous studies have demonstrated the potential of spent coffee grounds as adsorbent materials for various contaminants. Activated carbon prepared from coffee grounds has been applied to ammonia removal, with removal efficiencies of up to 84.64% reported under specific operating conditions [11], [12]. Coffee-ground-derived activated carbon has also been used to remove the synthetic dye Rhodamine B [13], while modified coffee-ground hydrochar has shown favourable performance in the adsorption of Cd(II) ions [14]. Furthermore, Irmanto and Suyata demonstrated that activated carbon derived from spent coffee grounds could reduce ammonia, nitrite, and nitrate concentrations in tofu-industry wastewater [15].

Although these studies have established the adsorption potential of spent coffee grounds, research specifically examining nitrite removal from tofu wastewater using Robusta coffee-ground biosorbents remains limited. Previous investigations indicate that the application of coffee-ground biosorbents for nitrite removal is not entirely new; however, further research is required to evaluate their performance under different preparation and operating conditions. In particular, Robusta coffee grounds are relevant because Robusta coffee is widely produced and consumed in Aceh, resulting in a locally available biomass residue that could be converted into a value-added adsorbent.

Therefore, this study investigated the removal of nitrite from tofu wastewater using carbonised and acid-activated Robusta spent coffee grounds as a biosorbent. The effects of biosorbent mass and contact time on nitrite-removal efficiency and adsorption capacity were evaluated through batch adsorption experiments. The adsorption kinetics, preliminary equilibrium behaviour, and statistical significance of the operating variables were also assessed. The utilisation of Robusta spent coffee grounds is expected to provide an environmentally friendly alternative for tofu-wastewater treatment while supporting waste valorisation, organic-waste reduction, and circular-economy principles.

2. Literature Review

2.1 Nitrite in Wastewater and Its Environmental Impacts

Nitrite (NO_2^-) is an inorganic nitrogen compound formed as an intermediate in the nitrogen cycle. During nitrification, ammonia or ammonium is first oxidised to nitrite and subsequently converted to nitrate. Conversely, during denitrification, nitrite is formed as an intermediate product in the reduction of nitrate to nitrogen gas [7], [16]. The accumulation of nitrite in wastewater and receiving water bodies poses a significant environmental risk because of its toxicity to aquatic organisms.

In freshwater fish, nitrite is absorbed primarily through the gills and oxidises haemoglobin to methaemoglobin, which has a limited ability to transport oxygen. This process can impair respiration, ionic regulation, growth, and survival [7], [8]. Nitrite exposure through drinking water may also contribute to methaemoglobinaemia in humans, particularly in infants, who are more susceptible to impaired oxygen transport in the blood [9], [17]. In addition to its direct toxic effects, the continuous input of inorganic nitrogen compounds, particularly in combination with phosphorus, can increase primary productivity, stimulate algal growth, and accelerate eutrophication. These processes may subsequently reduce dissolved oxygen concentrations and deteriorate aquatic ecosystem quality [9].

Tofu-processing wastewater contains organic matter, proteins, and nitrogen compounds originating from soybeans and the whey separated during tofu coagulation [18]. The mineralisation of proteins and organic nitrogen can generate ammonia or ammonium, which may subsequently be oxidised by nitrifying microorganisms to nitrite and nitrate under suitable environmental conditions [3], [19]. Therefore, nitrite concentrations must be controlled before tofu wastewater is discharged into receiving water bodies.

Adsorption using activated carbon, biochar, or agricultural-waste-derived materials is a potential method for nitrite removal. The process is relatively simple, potentially economical, and environmentally sustainable, particularly when the adsorbent is produced from locally available biomass residues. Activated carbon derived from rice straw, for example, has been reported to remove nitrate and nitrite anions from wastewater [20]. Nevertheless, nitrite adsorption performance is influenced by several operational and material-related factors, including solution pH, adsorbent dosage, contact time, surface characteristics, initial nitrite concentration, and the presence of competing ions [21], [22].

2.2 Adsorption Process and Biosorbents

Adsorption is a physicochemical process involving the transfer and accumulation of a substance, known as the adsorbate, from a liquid or gaseous phase onto the surface of a material known as the adsorbent. Adsorption is widely applied in water and wastewater treatment because of its operational simplicity, flexibility, and ability to remove various pollutants under appropriate operating conditions [23], [24].

Depending on the nature of the interaction, adsorption can be classified as physical adsorption (physisorption) or chemical adsorption (chemisorption). Physisorption occurs through relatively weak intermolecular interactions, including van der Waals forces, dipole interactions, and hydrogen bonding. Consequently, physically adsorbed substances are generally more readily desorbed. Chemisorption involves stronger interactions, including chemical-bond formation, electron transfer, complexation, ion exchange, and electrostatic interactions between the adsorbate and active functional groups on the adsorbent surface [25], [26]. The dominant adsorption mechanism depends on the chemical properties of the adsorbate, the structure and surface charge of the adsorbent, surface area, pore size, solution pH, temperature, and the presence of competing substances or ions.

Biosorbents are adsorptive materials derived from biological sources, including living or non-living biomass and biomass-derived materials such as agricultural residues, forestry waste, algae, bacteria, fungi, and other organic residues [25], [27]. Lignocellulosic biosorbents generally contain cellulose, hemicellulose, lignin, proteins, and other organic components that provide active functional groups such as hydroxyl (–OH), carboxyl (–COOH), amine ($–NH_2$), carbonyl (C=O), phenolic, and phosphate groups. These functional groups can bind pollutants through physical adsorption, ion exchange, electrostatic attraction, complexation, and chelation [26].

Agricultural residues and other organic wastes offer potentially economical and environmentally preferable alternatives to commercial activated carbon. Their utilisation can reduce solid-waste generation, provide value-added products from abundant biomass resources, and support circular-economy principles [23], [24]. However, biosorbent performance is influenced by biomass type, surface area, porosity, functional-group composition, activation or modification treatment, regeneration capacity, and the characteristics of the wastewater being treated.

2.3 Potential of Robusta Spent Coffee Grounds as a Biosorbent

Spent coffee grounds are solid residues generated during coffee brewing and contain considerable amounts of lignocellulosic compounds, polyphenols, and carbon-rich materials. The presence of surface functional groups, particularly hydroxyl and carboxyl groups, enables spent coffee grounds to interact with heavy-metal ions, synthetic dyes, and inorganic contaminants. Consequently, spent coffee grounds can be converted into low-cost biosorbents rather than being disposed of as organic waste.

Previous studies have demonstrated the adsorption potential of spent coffee grounds for various pollutants. Aman et al. applied activated carbon derived from spent coffee grounds to ammonia removal from wastewater [11]. Pradhana et al. used coffee-ground-derived activated carbon to adsorb Rhodamine B dye [13], while Fitasari and Ramadani developed H_2O_2-modified coffee-ground hydrochar for Cd(II) adsorption [14]. Irza et al. developed a magnetic biosorbent based on spent coffee grounds for Pb removal [28]. In another application, Sutisna et al. utilised spent coffee grounds as a supporting material for a TiO_2 photocatalytic composite to reduce Cr(VI) in an aqueous waste solution [29].

More directly related to the present investigation, Irmanto and Suyata reported that activated carbon prepared from spent coffee grounds could reduce ammonia, nitrite, and nitrate concentrations in tofu-industry wastewater [15]. These findings demonstrate that the adsorption capacity of spent coffee grounds can be improved through physical, chemical, or thermal activation, thereby increasing their potential application for removing contaminants, including nitrite.

2.4 Previous Studies on Nitrite Adsorption and Research Positioning

The study by Irmanto and Suyata [15] provides an important foundation for the application of spent-coffee-ground activated carbon in removing nitrogen compounds from tofu wastewater. Their findings confirmed that coffee-ground-derived activated carbon could reduce ammonia, nitrite, and nitrate concentrations. However, the available literature on the specific performance of Robusta spent coffee grounds for nitrite adsorption remains limited.

A subsequent investigation by Sartika et al. examined the reduction of COD, BOD, and nitrite in tofu wastewater using activated carbon derived from ground coffee residues [30]. The present study extends these previous investigations by specifically evaluating Robusta coffee-ground biosorbent, which is particularly relevant because Robusta coffee is widely produced in Aceh. The study places greater emphasis on the effects of biosorbent mass and contact time and on the kinetic behaviour of nitrite adsorption. Therefore, the contribution of this research lies not in introducing coffee grounds as an entirely new adsorbent, but in providing a more focused assessment of activated Robusta coffee grounds under the experimental conditions applied in this study.

3. Materials and Methods

3.1 Preparation of the Robusta Coffee-Ground Biosorbent

Robusta spent coffee grounds were initially dried and sieved to obtain a uniform particle size of 100 mesh. The prepared material was then carbonised at 350 °C for 3.5 h. After cooling, the resulting biocarbon was chemically activated by immersion in 0.1 M HCl for two days. The activated material was subsequently washed with water until a neutral pH of approximately 7 was achieved and then dried to obtain the final biosorbent.

3.2 Batch Adsorption Experiments

Batch adsorption experiments were conducted using 100 mL of tofu wastewater for each treatment. The initial nitrite concentration was 40 mg/L. Three biosorbent masses—0.2, 0.3, and 0.4 g—were evaluated at contact times of 10, 30, 50, 70, and 90 min. During each experiment, the Robusta coffee-ground biosorbent was brought into contact with the wastewater using a magnetic stirrer to promote interaction between nitrite ions and the available adsorption sites. At the end of the specified contact time, the solid biosorbent was separated from the liquid phase. The solid residue or adsorption cake was used for morphological analysis, while the filtrate was collected for the determination of residual nitrite concentration.

3.3 Analytical Procedures

The surface morphology of the biosorbent after adsorption was analysed using scanning electron microscopy (SEM). The residual nitrite concentration in the filtrate was measured using a UV–Visible spectrophotometer. The resulting concentration data were subsequently used to determine nitrite-removal efficiency and adsorption capacity for each combination of biosorbent mass and contact time. The experimental preparation and batch conditions follow the procedures reported in the source article.

4. Result & Discussion

This section presents the performance of the Robusta coffee-ground biosorbent in removing nitrite from tofu wastewater under different biosorbent masses and contact times. The discussion focuses on changes in nitrite-removal efficiency, residual concentration, and adsorption capacity, followed by adsorption-kinetic modelling, preliminary isotherm analysis, and two-way ANOVA to determine the significance and relative influence of the investigated operating variables.

Table 1 shows that nitrite removal efficiency increased with increasing contact time at all biosorbent masses. At a biosorbent mass of 0.2 g, the removal efficiency increased from 32.50% after 10 min to 72.50% after 90 min. A similar trend was observed at 0.3 g, where the efficiency increased from 38.75% to 73.75%, and at 0.4 g, where it increased from 51.00% to 77.75%. These results indicate that a longer contact time provides greater opportunity for nitrite ions to diffuse towards the biosorbent surface and interact with the available active sites. Between 50 and 70 min, only a slight increase in removal efficiency was observed at biosorbent masses of 0.2 and 0.3 g, suggesting that a substantial proportion of the active sites had begun to become occupied before a further increase occurred at 90 min.

Table 1. Experimental Data on Nitrite Adsorption by Robusta Coffee-Ground Biosorbent

Biosorbent Mass(g)	Contact Time (min)	Removal Efficiency (%)
0,2	10	32,50
0,2	30	44,25
0,2	50	50,00
0,2	70	51,00
0,2	90	72,50
0,3	10	38,75
0,3	30	44,75
0,3	50	52,25
0,3	70	53,75
0,3	90	73,75
0,4	10	51,00
0,4	30	53,25
0,4	50	56,25
0,4	70	64,25
0,4	90	77,75

Increasing the biosorbent mass also had a positive effect on nitrite removal. At a contact time of 10 min, the removal efficiency increased from 32.50% using 0.2 g of biosorbent to 38.75% using 0.3 g and 51.00% using 0.4 g. After 90 min, the corresponding removal efficiencies were 72.50%, 73.75%, and 77.75%, respectively. A higher biosorbent mass provides a larger adsorption surface area and a greater number of active sites capable of binding nitrite ions. Therefore, within the experimental range investigated, the optimum condition was achieved using 0.4 g of biosorbent and a contact time of 90 min, resulting in the highest nitrite removal efficiency of 77.75%.

Figure 1 demonstrates that nitrite removal efficiency increased with contact time for all biosorbent masses. At 0.2 g, the efficiency rose from 32.50% at 10 min to 72.50% at 90 min, while the use of 0.3 g increased the efficiency from 38.75% to 73.75%. The 0.4 g treatment consistently produced the highest removal efficiency, increasing from 51.00% at 10 min to 77.75% at 90 min. These trends indicate that longer contact times provide more opportunity for nitrite ions to diffuse from the wastewater to the biosorbent surface and interact with the available adsorption sites. The relatively small increase between 50 and 70 min for the 0.2 and 0.3 g treatments may indicate temporary diffusion limitations or the progressive occupation of readily accessible active sites.

The results also show that increasing the biosorbent mass generally improved nitrite removal at each contact time. The 0.4 g biosorbent treatment remained above the 0.2 and 0.3 g treatments throughout the experiment, suggesting that a greater biosorbent mass provides a larger effective surface area and more active sites for nitrite adsorption. The sharp increase observed between 70 and 90 min for all three masses indicates that equilibrium may not have been fully reached before 90 min. Within the experimental range, the best operating condition was therefore obtained using 0.4 g of Robusta coffee-ground biosorbent and a contact time of 90 min, resulting in a maximum nitrite removal efficiency of 77.75%.

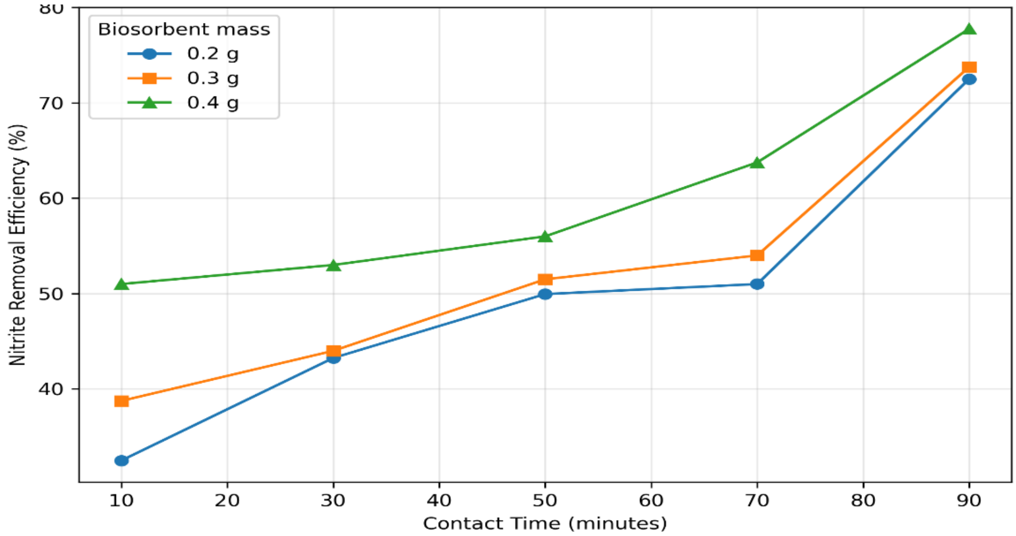

Figure 1: Effect of Contact Time on Nitrite Removal Efficiency Using Robusta Coffee-Ground Biosorbent at Biosorbent Masses of 0.2, 0.3, and 0.4 g

The results showed that increasing the biosorbent mass at each contact time positively affected nitrite removal efficiency. At a contact time of 10 min, the removal efficiency increased from 32.50% with 0.2 g of biosorbent to 38.75% with 0.3 g and reached 51.00% with 0.4 g. A similar pattern was consistently observed at the other contact times. At 30 min, the removal efficiencies were 44.25%, 44.75%, and 53.25%; at 50 min, they were 50.00%, 52.25%, and 56.25%; at 70 min, they were 51.00%, 53.75%, and 64.25%; and at 90 min, they reached 72.50%, 73.75%, and 77.75% for biosorbent masses of 0.2, 0.3, and 0.4 g, respectively.
These findings demonstrate that a greater biosorbent mass consistently resulted in higher nitrite removal efficiency throughout the range of contact times investigated. This trend is consistent with the findings of Rahman et al. (2022) and Aman et al. (2018), who reported that increasing the adsorbent dose or mass was positively correlated with pollutant removal efficiency in batch adsorption systems. The improvement can be attributed to the increased surface area and greater number of functional groups available as active adsorption sites, allowing more nitrite ions to be adsorbed during contact with the biosorbent. Robusta coffee grounds contain cellulose, hemicellulose, and lignin, as well as hydroxyl and carboxyl functional groups that can act as adsorption sites.

Table 2. Complete Experimental Data on Nitrite Adsorption by Robusta Coffee-Ground Biosorbent (C_0 = 40 mg/L; Wastewater Volume = 100 mL per Treatment)

Biosorbent Mass (g)	Contact Time (min)	Removal Efficiency (%)	Ce (mg/L)	qe (mg/g)
0,2	10	32,50	27,00	6,50
0,2	30	44,25	22,30	8,85
0,2	50	50,00	20,00	10,00
0,2	70	51,00	19,60	10,20
0,2	90	72,50	11,00	14,50
0,3	10	38,75	24,50	5,17
0,3	30	44,75	22,10	5,97

Biosorbent Mass (g)	Contact Time (min)	Removal Efficiency (%)	Ce (mg/L)	qe (mg/g)
0,3	50	52,25	19,10	6,97
0,3	70	53,75	18,50	7,17
0,3	90	73,75	10,50	9,83
0,4	10	51,00	19,60	5,10
0,4	30	53,25	18,70	5,33
0,4	50	56,25	17,50	5,63
0,4	70	64,25	14,30	6,43
0,4	90	77,75	8,90	7,78

The initial nitrite concentration of 40 mg/L decreased substantially after adsorption using the Robusta coffee-ground biosorbent. As shown in **Table 2**, the lowest final nitrite concentration (C_e) was obtained using 0.4 g of biosorbent at a contact time of 90 min, reaching 8.90 mg/L. This result indicates that 31.10 mg/L of nitrite was removed, corresponding to the highest removal efficiency of 77.75%. In contrast, the highest final concentration was recorded using 0.2 g of biosorbent for 10 min, with a C_e value of 27.00 mg/L. Under this condition, only 13.00 mg/L of nitrite was removed, resulting in the lowest removal efficiency of 32.50%. Overall, the decrease in C_e with increasing contact time and biosorbent mass demonstrates that longer interaction periods and a greater availability of adsorption sites enhanced nitrite removal from the wastewater.

The adsorption capacity (q_e) increased with contact time at each biosorbent mass. For 0.2 g of biosorbent, q_e increased from 6.50 mg/g at 10 min to 14.50 mg/g at 90 min. Similarly, the adsorption capacity increased from 5.17 to 9.83 mg/g for 0.3 g and from 5.10 to 7.78 mg/g for 0.4 g over the same contact-time range. Although the 0.4 g treatment produced the highest removal efficiency and the lowest residual nitrite concentration, the highest adsorption capacity per unit mass was obtained using 0.2 g at 90 min. This difference occurs because q_e is expressed relative to the mass of biosorbent; therefore, a smaller biosorbent mass may adsorb a greater amount of nitrite per gram even when its overall removal efficiency is lower. These findings confirm that biosorbent mass and contact time influence both the total nitrite removal and the adsorption capacity of Robusta coffee grounds.

Table 3. Pseudo-First-Order and Pseudo-Second-Order Adsorption Kinetic Parameters

Massa (g)	qe eksp. (mg/g)	k1 (/sec)	qe PFO (mg/g)	R^2 PFO	k2 (g/mg·sec)	qe PSO (mg/g)	R^2 PSO
0,2	14,50	0,01045	8,26	0,905	0,00294	15,61	0,878
0,3	9,83	0,00989	5,09	0,956	0,00505	10,43	0,894
0,4	7,78	0,01091	3,23	0,857	0,00899	8,17	0,940

The adsorption kinetics were evaluated using the pseudo-first-order (PFO) and pseudo-second-order (PSO) models. The PFO model was expressed in its linear form as ln $q_e - q_t = In(q_e) - k_1 t$, whereas the PSO model was expressed as $\frac{t}{q_t} = 1/(k_2 q_e^2 + t/q_e)$, where q_t represents the adsorption capacity at time t, and qe represents the adsorption capacity under near-equilibrium conditions, taken at a contact time of 90 min. As summarised in **Table 3**, both kinetic models provided reasonably good fits to the experimental data, with coefficients of determination (R^2) ranging from 0.857 to 0.956. For biosorbent masses of 0.2 and 0.3 g, the PFO model produced slightly higher R^2 values of 0.905 and 0.956, respectively, than the PSO model, which gave values of 0.878 and 0.894. In contrast, at a biosorbent mass of 0.4 g, the PSO model showed a better fit, with an R^2 value of 0.940 compared with 0.857 for the PFO model.

Although the goodness of fit varied among the biosorbent masses, the equilibrium adsorption capacities predicted by the PSO model were consistently closer to the experimental values. The predicted qe values from the PSO model were 15.61, 10.43, and 8.17 mg/g for biosorbent masses of 0.2, 0.3, and 0.4 g, respectively, compared with the corresponding experimental values of 14.50, 9.83, and 7.78 mg/g. By comparison, the PFO model underestimated the adsorption capacities, producing predicted values of only 8.26, 5.09, and 3.23 mg/g. This pattern is consistent with the findings of Ho and McKay (1999), who reported that the PSO model often provides more accurate predictions of equilibrium adsorption capacity in biosorbent-based sorption systems. The relatively close agreement between the experimental and PSO-predicted qe values suggests that chemisorption may contribute to nitrite uptake by the Robusta coffee-ground biosorbent, although physical adsorption and mass-transfer processes may also be involved.

The results demonstrate that Robusta coffee grounds have considerable potential as a natural biosorbent for reducing nitrite concentrations in tofu wastewater. The maximum removal efficiency of 77.75% indicates that spent coffee grounds can be utilised as a value-added adsorbent material while supporting the principles of the circular economy and biomass-based waste management. This adsorption capability is influenced by the lignocellulosic content and the presence of functional groups, such as hydroxyl (–OH) and carboxyl (–COOH) groups, which can interact with nitrite ions through physical adsorption, electrostatic interactions, and hydrogen-bond formation.

Table 4. Two-Way ANOVA of Nitrite Removal Efficiency (%) Based on the Complete Experimental Dataset (α = 0.05)

Source of Variation	**JK**	**db**	**KT**	**Calculated F-Value**	**F table (α=0,05)**	**p-value**	**Remarks**
Biosorbent Mass	295,98	2	147,99	18,16	4,46	0,0011	Significant
Contact Time	1956,06	4	489,01	60,01	3,84	0,0000	Significant
Error (Interaction)	65,19	8	8,15	-	-	-	-
Total	2317,23	14	-	-	-	-	-

The statistical significance of contact time and biosorbent mass on nitrite removal efficiency was evaluated using a two-way analysis of variance without replication. Contact time was treated as Factor A with five levels, whereas biosorbent mass was treated as Factor B with three levels. The analysis was performed using all 15 experimental observations presented in Table 1, following the standard procedure for a two-factor factorial design without replication. Because no replicate measurements were available for each treatment combination, the residual term represented the combined contribution of possible factor interactions and experimental error. Therefore, the analysis assumed that the interaction between contact time and biosorbent mass was negligible and could be used as the error term. The results of the analysis are summarised in Table 4.

The ANOVA results demonstrate that both experimental factors significantly affected nitrite removal efficiency at a significance level of $\alpha=0.05$. For biosorbent mass, the calculated F-value was 18.16, which was substantially higher than the critical F-value of 4.46, with a p-value of 0.0011. Similarly, contact time produced a calculated F-value of 60.01, exceeding the critical F-value of 3.84, with $p<0.0001$. These results confirm that variations in both biosorbent mass and contact time produced statistically significant changes in nitrite removal efficiency. Accordingly, the null hypotheses stating that the mean removal efficiencies were equal across the different biosorbent masses and contact times were rejected.The estimated effect sizes further indicate that contact time was the dominant factor controlling nitrite removal within the experimental range. Contact time accounted for approximately 84% of the observed variation in removal efficiency ($\eta^2 \approx 0.84$), whereas biosorbent mass accounted for approximately 13% ($\eta^2 \approx 0.13$). This finding is consistent with the experimental trend, in which removal efficiency generally increased considerably as the contact period was extended, particularly up to 90 min. Nevertheless, the statistically significant effect of biosorbent mass indicates that increasing the amount of Robusta coffee-ground biosorbent also enhanced nitrite removal by increasing the number of available adsorption sites. Thus, although both factors were influential, contact time exerted a substantially greater effect on nitrite removal efficiency than biosorbent mass under the tested conditions.

The absence of replication should, however, be considered when interpreting these results because the interaction effect between contact time and biosorbent mass could not be estimated separately from experimental error. Additional replicate experiments at each treatment combination would enable a more reliable assessment of experimental variability and determine whether a significant interaction occurs between the two factors.

Tukey HSD post hoc test—contact-time factor: The removal efficiency at 90 min differed significantly ($p<0.05$) from those observed at all other contact times of 10, 30, 50, and 70 min. The efficiency at 70 min also differed significantly from that at 90 min ($p=0.025$); however, it did not differ significantly from the efficiencies recorded at 10, 30, or 50 min. No significant differences were observed among the 10-, 30-, and 50-min contact times ($p>0.05$). This pattern is consistent with the kinetic curve, which showed a relatively gradual increase between 10 and 50 min, followed by a marked rise as the contact time approached 90 min.

Tukey HSD post hoc test—biosorbent-mass factor: No significant pairwise differences were identified among the biosorbent masses at $\alpha=0.05$: 0.2 versus 0.3 g ($p=0.946$), 0.2 versus 0.4 g ($p=0.436$), and 0.3 versus 0.4 g ($p=0.617$). Nevertheless, the mean removal efficiency increased consistently with biosorbent mass, from 50.05% at 0.2 g to 52.65% at 0.3 g and 60.50% at 0.4 g. These findings indicate that, although the ANOVA detected a significant overall effect of biosorbent mass, the differences between individual mass levels, particularly adjacent levels such as 0.2 and 0.3 g, were not sufficiently large to be detected separately by the post hoc test with the available sample size.

5. Conclusion

This study evaluated the effects of contact time and biosorbent mass on nitrite removal from tofu wastewater using Robusta coffee grounds as a natural biosorbent. Based on the experimental, kinetic, isotherm, and statistical analyses, the following conclusions were drawn:

1) Increasing the contact time and biosorbent mass positively affected the reduction in nitrite concentration, and this trend was consistently observed across all 15 treatment combinations investigated.
2) The optimum condition was obtained using 0.4 g of biosorbent at a contact time of 90 min, resulting in a final nitrite concentration of 8.9 mg/L and a removal efficiency of 77.75%.
3) The highest adsorption capacity was achieved using 0.2 g of biosorbent for 90 min, reaching 14.50 mg/g. This finding indicates that a smaller biosorbent mass may allow more efficient utilisation of active adsorption sites per unit mass.
4) The kinetic data showed good agreement with both the pseudo-first-order and pseudo-second-order models, with R2 values ranging from 0.86 to 0.96. However, the pseudo-second-order model produced predicted qe values that were closer to the experimental values, suggesting the contribution of a chemical adsorption mechanism or chemisorption.
5) The preliminary isotherm evaluation indicated that the equilibrium data more closely followed the Freundlich model (R2≈0.81) than the Langmuir model. Nevertheless, further confirmation using a wider range of initial nitrite concentrations is required.

6) The two-way ANOVA based on the complete experimental dataset confirmed that both contact time ($F=60.01; p<0.0001$) and biosorbent mass ($F=18.16; p=0.0011$) significantly affected nitrite removal efficiency. Contact time was identified as the most dominant factor, accounting for approximately 84% of the observed variation ($\eta 2 \approx 0.84$).
7) Overall, the findings demonstrate that Robusta coffee grounds have considerable potential as an environmentally friendly biosorbent for the treatment of tofu wastewater.

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